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ANALYSIS OF THE GAMMA RAYS FROM GRANITE AND THE IONIZATION BALANCE OF THE ATMOSPHERE

BY

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With 1 figure.

Scope of Present Study and Experimental Procedure.

More than four years ago the author pointed out (1) that the observed difference in the ionization of hermetically sealed vessels over land and over water cannot be accounted for if one accepts the recently reported low values for the radium uranium and thorium content found by Robley D. EVANS and Clark GOODMAN (2) as representative for the various classes of rocks.

It seemed worth while to investigate this discrepancy. The large variations existing between specimens of the same type of rock taken from different localities makes it hazardous to draw conclusions on the average content of any type of rock, and to derive figures for the gamma ray ionization to be expected in individual cases.

Such comparisons are meaningless if they are not made with very homogeneous types of rocks.

Among the numerous rock specimens investigated, EVANS and GOODMAN recommended to the author the Quincy granite, the diabase from the Palisades, N. J, and the granites from Fitchburg, Rockport and Chelmsford, Mass. as specially suitable for a quantitative comparison.

I decided to study the ionization produced by the granite from Quincy, Mass. first.

The experiments were performed in the quarry of Swingle Co, in Quincy in August 1945 and were continued with a large quantity of crushed granite (400 lb) from the same source at Fordham University.

The ionization apparatus was constructed upon the author's and Mr. O. H. GISH's suggestions by the Department of Terrestrial Magnetism (Carnegie Institution), Washington, D.C. in 1942 and improved for field experiments in 1943. It will be described in detail by O. H. GISH and K. L. SHERMAN elsewhere.

Fig. 1 shows the main features of this ioniziation meter. Three ionization chambers (of 1.6, 4.9 and 13.1 litre volume), hermetically closed and filled with dry, aged nitrogen at atmospheric pressure can be used alternatively, in connection with a Lindemann electrometer which serves here only as a null instrument. The electrometer housing and the guard ring systems of the three chambers are kept at 135 volts. Only one of the three chambers is shown in the figure.

If the electrometer is set on top of one of the chambers and a contactor connects the inner electrode plus the electrometer needle with the 135 volt source, the needle is at its zero position. As soon as the contactor is withdrawn at the beginning of each measurement, the needle begins to drift and the rate of the migration of the needle is proportional to the ionization current flowing between the discharging inner electrode and the grounded wall of the chamber.

Since the guard ring is at all times at practically the same potential as the inner electrode of the chamber (differing at the most by about $\pm$ 0.1 volt) all leakage losses are here completely avoided.

Instead of observering, as usual, this displacement of the needle per unit time ("drift method") it is much more accurate to add gradually a compensating potential of from 0 to 3 volts to the guard ring potential by connecting it (and the housing of the electrometer) with a potentiometer circuit and adjusting the voltage continously so as to keep the electrometer needle always nearly at the zero point.

This compensating voltage (acting by its induction effect on the floating system) is read in successive intervals of a few minutes on a precision voltmeter. If this voltage is divided by the number of seconds and multiplied by a carefully determined calibration coefficient one obtains directly the ionization (q) expressed in number of ions (I) produced in the chamber per cm^3 and per second.

The advantage of using three geometrically similar cylindrical brass chambers as ionization vessels is described in my previous paper (1). It is only of importance if one desires to determine separately the ionization produced by cosmic rays, by local gamma rays and from the walls of the chamber (background).

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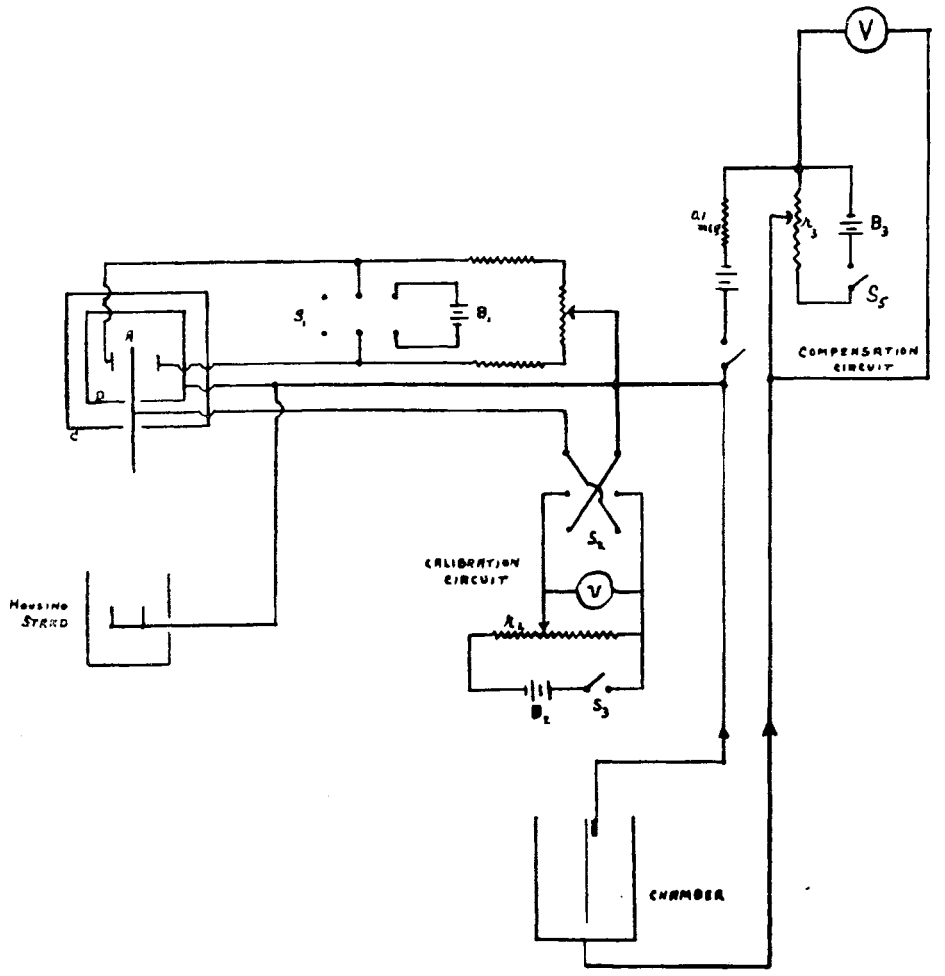
Ionization -Meter (Gish -Sherman -Hess)



B_1 = 45 V. R_3 = 960 Ω
B_2 = 1.5 V.
B_3 = 3.0 V.
B_v = 135 V.

Fig. 1.

For the present purpose only one chamber is necessary and the choice was made according to the activity of the samples to be studied.

Computation of the Gamma Ray Ionization from Radium, Thorium and Potassium.

A point-shaped source of gamma rays (quantity m), placed at a distance r from the center of an ionization chamber produces an ionization of

$$q = K \cdot \frac{m}{r^2} \cdot e^{-\mu \cdot r}$$

where K is a constant ("Eves constant") which has to be determined by using known amounts of radium, thorium or potassium; μ denotes the coefficient of absorption of the gamma rays in the medium between the source and the ionization chamber. Eves constant for radium, slightly diffiering for vessels of different size and materials, was determined by the author and Eva M. BALLING (3) for the chambers actually used for the present study. The results vere reported at the Meeting of the Amer. Geophys. Union in Washington, 1945. For brass vessels, filled with nitrogen at atmospheric pressure we obtained

$$K = 4.8 \times 10^9 \text{ I per gram radium}$$

at unit distance.

If the radium is distributed in a large volume, as for instance in rocks, the gamma ray ionization produced can be determined by integration for a few simple cases. For example, if the ionization vessel is placed on even ground and its content of radium, thorium and potassium is known (according to chemical analysis or other methods) we have for the integrated effect of the gamma rays from radium, thorium and potassium the three formulas:

$$q_1 = \frac{2\pi K_1}{\mu_1} \cdot m_1, \quad q_2 = \frac{2\pi K_2}{\mu_2} \cdot m_2, \quad q_3 = \frac{2\pi K_3}{\mu_3} \cdot m_3$$

where the subscripts 1, 2, 3 refer to the respective values of radium, thorium and potassium.

In a monograph (4) on the ionization balance of the atmosphere I have shown that the gamma ray effects of radium itself, of radium B, uranium X etc. are negligible and therefore a simplified treatment with only one significant absorption coefficient μ for each term is permissible.

It is obvious that μ_1, μ_2, μ_3 must be known for the hardest component of gamma rays in each case (RaC'', ThC''). The absorption coefficient of the gamma rays from potassium is, according to L. H. GRAY and C. T. P. TARRANT (5), practically equal to the one of the gamma rays from RaC''.

Consideration of the softer components of gamma radiation (larger coefficients μ) would give only small and insignificant increases in the values of q_1 and q_2.

If an apparatus is surrounded on all sides by a very thick layer of a material containing radium, thorium and potassium the formulas have to be changed into $q_1 = \frac{4\pi K_1}{\mu_1} \cdot m_1$ etc. For rocks a layer of 50 cm could be considered for the use of these formulas (infinite thickness). With crushed rocks (density 1.3 to 1.5 g/cm³) layers about twice as thick would be advisable.

The absorption coefficient μ_1 of the gamma rays from RaC'' was directly determined by putting a sealed radium tube in the center of a cylindrical box containing crushed granite and measuring the ionization at a large distance with different thicknesses of the absorbing layer of crushed granite. Miss Eva BALLING obtained thus a value for the absorption coefficient of crushed granite (density 1.34 g/cm³) $\mu = 0.057\,\mathrm{cm}^{-1}$. The mass absorption coefficient resulting ($0.0426\,\mathrm{cm}^2/\mathrm{g}$) agrees well with the usual mean value for the gamma rays from RaC'' (0.045).

In the formulas given above m_1, m_2, m_3 denote the contents of radium, thorium and potassium per cubic centimetre of rock. On account of the fact that these contents are usually given in grams of Ra, Th, K per gram of rock it is convenient to transform these formulas by introducing

$$M_1 = m_1/\rho, \quad M_2 = m_2/\rho, \quad M_3 = m_3/\rho$$

where M_1, M_2, M_3 denote the radium, thorium and potassium content per gram of the rock sample.

Thus we have, immediately above an infinite plane surface of rock

$$q_1 = \frac{2\pi K_1}{(\mu_1/\rho)} \cdot M_1, \quad q_2 = \frac{2\pi K_2}{(\mu_2/\rho)} \cdot M_2, \quad q_3 = \frac{2\pi K_3}{(\mu_3/\rho)} \cdot M_3$$

This has the further advantage that we need only the mass-absorption coefficients μ_1/ρ etc. which are much better known and practically constant for all materials.

Eves Constant for Thorium and Potassium.

The constant K_2 for thorium has never been determined. For this reason, in my previous article on the radioactivity of rocks (1) I had to make the bold assumption that the contribution of the gamma rays from thorium and its products to the total ionization of the atmosphere near the ground is equal to the ionization from the uranium-radium products. A.S. EVE (6) used the same assumption in 1911.

The lack of data on thorium was due to the fact that no radium-free thorium standard was available which was old enough to be in equilibrium with mesothorium and radiothorium. After a futile waiting of about two years I finally decided to use a powdered sample of a thorium mineral, free of uranium as a standard, upon a suggestion from the Swiss geologist H. HIRSCHI.

The National Bureau of Standards in Washington recently provided for this purpose a sample of Thorite (104,6 g) in a sealed glass tube, 18 cm long and 2 cm wide. This sample contains 9.61% thorium and only 0.5% uranium.

The gamma radiation from this tube was easily measurable with the ionization meter at distances from 30 to 100 cm. The absorption curve taken with layers of from 0 to 22 mm lead showed after the first 6 mm a homogeneous hard gamma-radiation with an absorption coefficient of

$$\mu = 0.472 \text{ cm}^{-1} \text{ in lead}, \quad \mu/\varrho = 0.0418 \text{ cm}^2/\text{g}$$

which is characteristic for ThC' '. The soft component (at zero thickness giving 23% of the total ionization) gave $\mu/\varrho = 0.71$ cm²/g.

Preliminary experiments (May 1946) give (after correcting for the small gamma ray effect of the 0,5% uranium content) for Eves constant for thorium (K_2) the value

$$K_2 = 742 \text{ I per gram thorium, at unit distance.}$$

The corresponding value K_3 for potassium can only be estimated: according to MUELHOFF (see EVANS and GOODMAN (2)) one gram of potassium gives off 23 beta rays per second. GRAY and TARRANT (5) found that three photons are emitted for every 100 disintegration electrons. From this it follows that 1 g potassium emits 0.69 gamma ray photons of about 2×10^6 e-volts energy which corresponds to an emission of 2.2×10^{-6} ergs per sec.

The total number of ions produced by these photons (N) would be

$$\frac{2.2 \times 10^{-6}}{5.1 \times 10^{-11}} = 4.32 \times 10^4 \text{ pairs of ions/sec.}$$

Thus $$N = \int_0^\infty K_s \frac{e^{-\lambda r}}{r^2} \cdot 4\pi r^2 \, dr. = \frac{4\pi K_s}{\lambda} = 4.32 \times 10^4 \text{ I}$$

where λ denotes the absorption coefficient of gamma rays in air. Taking this as 4.5×10^{-5} cm^{-1} (HESS) one obtains Eves constant for potassium

$$K_s = 0.154 \text{ I per gram at unit distance.}$$

Another way of deriving K_s was used in my previous publication (1): if we adopt GRAY and TARRANT'S data as the most reliable, one gram of potassium would be equivalent (in its gamma ray effect) to 1.6×10^{-11}g radium. Thus, for our brass cylinders the value

$$K_s = 1.6 \times 10^{-11} \times 4.8 \times 10^9 = 0.077 \text{ I/g}$$

results. The two values differ considerably. Since however the potassium content of most granites is only from 0.02 to 0.04 grams per gram of rock it is of no great importance if we choose the higher or the lower value. A rough mean value of $K_s = 0.12$ I/g at unit distance will be accurate enough for our present purpose.

Computation of the Ionization Produced by the Gamma radiation from Quincy Granite.

With the foregoing data one can compute separately the ionization produced by the uranium-radium, the thorium products and by potassium for any type of rock.

EVANS and GOODMAN (2) give for Quincy Granite, taken from Swlngles quarry (l.c. table 6, Nr. $12A_2$) the uranium content as $(2.7 \pm 0.5) \times 10^{-6}$ grams per gram of granite. This in equilibrium with radium corresponds to $0{,}95 \times 10^{-12}$ g radium per g of rock. Therefore

$$q_1 = \frac{2\pi K_1}{(\mu/\rho)} \cdot M_1 = \frac{6.28 \times 4.8 \times 10^9}{0.045} \cdot 0.95 \times 10^{-12} = 0.64 \text{ I}$$

Similarly the thorium content of Quincy granite as given by EVANS and GOODMAN $M_2 = (8.1 \pm 2.0) \times 10^{-6}$ g Th/g leads to a value

$$q_2 = \frac{6.28 \times 742}{0.041} \cdot 8.1 \times 10^{-6} = 0.92 \text{ I}$$

For the potassium content, choosing a mean value of 0.03 gK/g we get

$$q_s = \frac{6.28 \times 0.12}{0.045} \cdot 0.03 = 0.50 \text{ I}$$

The sum of the three components of ionization therefore is

$$q = q_1 + q_2 + q_s = 2.06 \text{ I}$$

This would represent the total ionization to be expected over a large surface of Quincy granite.

For comparison with the actual observitions see table 1. Surrounding an ionization chamber on all sides with an infinite layer of the same material we could, of course, expect a value of q twice as high as the one given, i.e. 4.12 I.

Observations in the Quarry.

These observations were carried out by the Rev. F. A. BENEDETTO, S. J. and the author in the quarry of Swingle Co, Quincy (Mass.) in August 1945. The granite quarried there is very homogeneous; the shade of coloration of this granite differs slightly but we could not detect any difference in the gamma radiation from the "dark grain", medium and "light grain" specimens.

First the ionization chambers with the panel box were set up directly on the horizontal surface of a large slab of granite 8×4 ft, 3 ft high. Then the apparatus was placed on a card table and the table set up on top of the same block of granite. The results are listed in the following table 1. The figures given represent the net ionization produced by the gamma rays from the rock, after subtracting the average ionization by cosmic rays, air radiation (radioactive products in the atmosphere) and the residual ionization from the inner walls of the chambers.; these three effects together amounted to 3.21 I for the large chamber, 3.36 I for the medium chamber, according to numerous experiments over water, made on a wooden pier in New York, performed in the summers of 1944 and 1945.

In another experiment, the apparatus standing on the card table was set up directly on the ground which consisted of crushed granite with a slight admixture of humus. The ionization observed here was only slightly smaller (5.18 I) than above the solid block of granite (5.57 I).

In order to find out whether these unexpected high ionization values were due to softer rays penetrating through the 2.5 mm brass walls of the chambers, a special experiment was performed in which the apparatus, standing on the card table, was placed in a large iron boat, resting on the ground, over crushed granite.

Such boats are used in the quarry for lowering a crew of workers into a deep pit in order to drill the holes for subsequent blasting. The walls of the iron boat were about 1.2 cm thick. The shape of

Table 1.

Observations in Swingle's Quarry, Quincy, Mass.

Ionization Produced by Gamma Rays from Quincy Granite.

Apparatus standing on large granite block (8 × 5 × 3 ft)	5.95 I
Apparatus on card table, set up on the same block of granite ..	5.57 I
Apparatus on card table, set up on the ground (crushed granite)	5.18 I
Apparatus on card table, standing in iron boat above crushed granite	4.29 I

Computed Gamma Ray Ionization (Taking Evans' and Goodman's Figures for the Uranium and Thorium Content and 3% Potassium Content).

Uranium-radium series	0.64 I
Thorium series	0 92 I
Potassium	0.50 I
total	2.06 I

the boat is a half cylinder (horizontal axis), open on one side, of about 6 ft diameter. The mean ionization in the boat (4.29 I) was only slightly less than on the ground (5.18 I). The absorption coefficient in iron, computed from this experiment (0.16 cm^{-1}) indicates that only gamma rays were involved. Naturally, this coefficient is not very accurate since the boat was open on one side while the other end consisted of perforated iron plating. However the coefficient is of the right order of magnitude, indicating that our results are not falsified by softer radiations emitted by the granite.

A comparison with the figures computed from the uranium, thorium and potassium content shows a very large discrepancy: the observed ionization is more than twice as great as the computed value.

This finding, now substantiated in a very well defined special case (Quincy granite) corroborates the conclusion drawn in the author's 1941 paper (1).

It seemed advisable to investigate this discrepancy further by experiments with crushed granite.

Laboratory Experiments.

In the autumn of 1945 the Swingle Co. in Quincy shipped, upon my request, 400 lb of crushed dark grain granite in bags to Fordham. This quantity was used for another set of measurements in which the ionization chamber — the medium or the small one — was surrounded on all sides with a thick layer (21 cm) of crushed granite while the local gamma rays from the walls of the room were completely absorbed in a layer of 10 cm of iron. First the chamber, with the electrometer on top was set up in the empty "iron house" built up from iron bars on the bottom and on all sides and leaving only the top open for the electrometer and microscope.

The ionization observed in this case (4.01 I) was due to cosmic rays (filtered through four floors and ceilings), local gamma rays from the ceiling of the room above the iron house and the residual radiation.

Then the space between the iron bars and the ionizaton chamber was filled completely with crushed granite. The average thickness of this layer was 21 cm. The total ionization observed in this case was 9.71 I (average of experiments of many days).

Thus the effect of the 21 cm layer of crushed granite was $9.71-4.01=5.70$ I. A graphical intergration of the open cone on top of the chamber shows that in the experiment the solid angle subtended by the granite layer amounted to $4\pi\times 0.85$ sterradians. A complete concentric layer (spherical shell, 21 cm thick) therefore would give $5.70/0.85=6.68$ I.

Since the mean absorption coefficient of gamma rays in the crushed granite was determined directly (0.057 cm^{-1}) one can now extrapolate to infinite thickness by using the formula (V. F. Hess see (7)).

$$q_\infty = \frac{4\pi K M}{\mu/\rho}(1 - e^{-\mu d})$$

where $d=21$ cm. This gives $q_\infty=9.5$ I, as the extrapolated value of the ionization produced by an infinite layer (see table 2).

The comparison with the computed value (4.12 I) shows again very decisively that the observed ionization cannot be explained from the reported values of the uranium and thorium content of the granite.

Table 2.

Laboratory Experiments with 400 lb of Quincy Granite.

(Ionization chamber placed in the center of an iron housing, wall 10 cm thick; concentric layer of crushed granite, 21 cm thick around the chamber, except for open cone on top of the chamber).

Ionization produced by a layer of 21 cm of crushed granite (directly observed)	5.70 I
Corrected for open cone on top	6.68 I
Extrapolated for granite layer of infinite thickness	9.50 I
Values computed, taking the uranium content as $(2.7 \pm 0.5) \times 10^{-6}$ g per gram, thorium content as $(8.1 \pm 2.0) \times 10^{-6}$ g per gram, potassium content 0.03 g per gram:	
Uranium-radium series	1.28 I
Thorium series	1.84 I
Potassium	1.00 I
total:	(4.12 ± 0.9) I.

Discussion.

Since both the field experiments and the laboratory measurements in the iron house show that the ionization actually produced by the gamma rays from Quincy granite is at least twice as great as the computed values one is confronted with a serious difficulty.

At first one may be tempted to assume that "back-scattering" of cosmic rays on solid ground may be responsible for the observed surplus ionization. It would be conceivable that this effect, well known from experiments with Geiger-Mueller counters is responsible for part of the difference in ionization of closed vessels placed over land and over water. However, this effect would be expected to be of much smaller magnitude since the total ionization produced by cosmic rays amounts to only 2 I at sea level.

Furthermore the back-scattering of cosmic rays is certainly not less over other soil materials (gravel, sand) while the author, from his experiments at Spray Beach, N. J. (August 1944) found that dune sand produces an ionization of only 0.5 I. This is in good agreement with expectation since dune sand contains 10 times less radioactive materials than volcanic rocks, such as granite.

Thus only two possibilities remain: Either the uranium and thorium content, reported by Evans and Goodman is much too low —

one hesitates to make this assumption in view of the great care and improved technique employed by these authors — or one has to assume that granite (and perhaps most of the other rocks) emits a penetrating radiation of unknown origin.

One might think of penetrating radiation accompanying the fission produced by cosmic rays in the uranium and thorium contained in the rocks, but such an effect would be much too small, on account of the rarity of such events.

Another remote possibility would be artificial radioactivity induced by cosmic radiation: it would be rather easy to test this possibility by performing the "iron house experiment" in a deep mine. Independent analyses of the uranium, thorium and potassium content of the Quincy granite are also planned.

Summary.

With a very sensitive ionization apparatus devised by O. H. Gish and the author, experiments were carried out to determine the average ionization produced by the penetrating rays from the uranium, thorium and potassium in a very homogeneous sort of granite quarried at Quincy, Mass. (U.S.A.). The uranium-radium, and thorium content of this granite was most carefully determined by R. D. Evans and Cl. Goodman in 1941. The potassium content was estimated as 3 per cent.

The ionization measurements performed in the quarry at Quincy and later experiments carried out with 400 lb of crushed granite from the same source at Fordham University show that the ionization actually observed above a plane surface of this granite amounts to almost 6 ion pairs per ccm. sec., while the value computed from the known amounts of uranium, thorium, and potassium content is only slightly above 2 ion pairs. Similar discrepancies were found in the laboratory experiments. They cannot be explained by assuming that part of the excess radiation is due to secondaries produced by cosmic rays impinging on the surface.

It must be concluded either that the reported values for the uranium and thorium content are much too low (which is unlikely) or that a hitherto unknown penetrating radiation is given off by granite.

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