

THE RADIUM CONTENT OF SEA-SALT SPECIMENS COLLECTED ON CRUISE IV OF THE *CARNEGIE*.

By C. W. HEWLETT.

The following is an account of an investigation which was carried out in the laboratory of the Department of Terrestrial Magnetism at Washington to determine the amount of radium in some samples of sea salt collected by the *Carnegie* in the Atlantic and Pacific oceans, 1915-1917.¹ The method of investigation was that first used by Joly and described by him in volume 32 of the *Philosophical Magazine*, 1911. The sea salt was mixed with two to three times its weight of a fusion mixture of sodium and potassium carbonates, and this mixture was heated to about 1,000° C. in an electric furnace. At this temperature carbon dioxide bubbled off from the mixture and presumably carried with it any radium emanation previously present in the mixture. The gases from the furnace were then introduced into the ionization chamber associated with an electroscope, and investigated for radioactivity.

Description of Apparatus.—The electrical furnace was tubular, about 1.5 inches internal diameter and the heated length about 12 inches. The fusion mixture was made of equal molecular weights of the anhydrous carbonates of sodium and potassium. A steel tube, 17 inches long and 1.5 inches external diameter, was inserted in the electric furnace. The ends of this tube were closed with perforated rubber stoppers. One rubber carried a tube which conducted the furnace-gases away for investigation, while the other held a short glass tube about 1 cm. in diameter. This tube extended about 1.5 inches outside the stopper and its end was closed with a flat piece of glass, so that a view could be had inside the furnace. Into the side of this tube another tube of smaller diameter was joined, and a piece of rubber tubing controlled by a pinch cock was attached to this smaller tube. By means of this arrangement air could be admitted to the furnace for washing it out after all the carbon dioxide had been driven off. The end

¹ The investigation here described was carried out under the immediate direction of Dr. W. F. G. Swann.

portions of the steel tube were kept cool by dripping water on them. A collar of cotton was wound around each end and water, dripping from two siphons into a large vessel supported above the apparatus, fell on these collars. This then dripped into two funnels and was carried off to the waste. The rubber stoppers were protected from radiation from the furnace by perforated brass plugs placed between the stoppers and the furnace.

The gases leaving the furnace first passed into a flask containing soda lime, where chlorine, carbon dioxide, and water vapor were partially absorbed. They next passed into a rubber bladder, where they collected until the heating was completed. These gases were then sucked into a glass flask containing silver and copper shavings, the connections being made in such a way that the gases in passing from the rubber bag to the flask, passed back through the flask of soda lime. The gases were then allowed to remain in the flask containing the metal shavings for 10 minutes, so that any remaining gases harmful to the insulation inside the ionization chamber might be used up. The gases in the flask were then admitted to the previously exhausted ionization chamber. It was usually necessary to fill the ionization chamber twice in order to secure a thorough wash-out of the furnace. The volume of the steel tube was about 300 cc., of the unoccupied portion of the soda-lime flask, about 100 cc., of the flask containing the metal shavings, about 500 cc., and of the ionization chamber about 1,200 cc. The volume of the connecting tubes was negligible.

A nickle boat whose volume was about 30 cc. was used to contain the melt of sea salt and fusion mixture. Further, this nickle boat was contained in a nickle shield to protect the iron tube in case the melt boiled over. The samples of sea salt to be investigated were sealed up in thin walled glass tubes for periods of time varying from 3 to 13 days, so even if the salt had no emanation when sealed up, it should have had, when tested, from 0.4 to 0.9 of its equilibrium value.

The electroscope used was a Wulf single quartz fiber instrument adjusted to a sensibility of 20 divisions per volt. The ionization chamber was charged to 100 volts above the case of the electroscope, and the method used for measuring the ionization current was that used on the *Carnegie* for measuring the ionization current in determinations of the radioactivity of the atmosphere.

Experimental Procedure.—From 5 to 12 grams of sea salt were

mixed with from 2 to 3 times its weight of the fusion mixture and heated in the furnace. The furnace was brought up to its highest temperature slowly, and the total time of heating was about 2 hours. As soon as the rubber bag had collected a sufficient amount of gas, the ionization chamber was filled, allowing the gases, however, to reside in the flask containing the metal shavings for 10 minutes on their way to the chamber.

These gases were then tested for radioactivity while more gas was collecting in the rubber bag. After these gases had been satisfactorily investigated, they were sucked out of the ionization chamber, the rest of the gases were admitted, and the furnace was washed out. The pressure in the ionization chamber was always adjusted to atmospheric pressure before observations were made. The ionization chamber contained a dish of phosphorus pentoxide for drying the gases. This was found to have quite a good effect on the insulation.

A word might be said in regard to the procedure on board the *Carnegie* in collecting the sea salt. When the date of collection is given over several days, as in the first item in the table at the end of this paper, from 1 to 1.5 liters of sea water were collected in equal amounts on all the days from the first to the last given, inclusive. When a single date is given, the sea water was taken only on that date. The water was then put in a copper vessel and evaporated to dryness. The residue in the copper vessel was then scraped out, closed up in a test tube, and forwarded to the laboratory in Washington.

Upon undertaking the present investigation it was found that the samples of sea salt were damp. Each sample was evaporated to dryness in a porcelain evaporation dish, and was then ground up in a mortar. A portion of each sample was sealed up in a thin-walled glass receptacle for the present investigation, the rest of each sample being carefully sealed up in a test tube for future work. These thin-walled glass tubes were sealed with bits of paraffin wax, so that they would open up in the furnace without explosion.

Results.—Upon admitting the gases to the ionization chamber, quite a large saturation current was observed, but this current decreased rapidly with the time and usually by the end of one hour it had disappeared. This initial effect was very troublesome till the flask containing the metal shavings was introduced into

the path of the gases. It is suspected that this effect is due to very large ions brought over from the furnace. This initial effect being variable in its magnitude, it was impossible to allow for it in the calculations, so that it was necessary to observe with each set of gases in the chamber for nearly an hour, or even longer, to be sure that all the heavy ions had been removed by the field.

Fifteen samples of sea salt were investigated for radium emanation, but in none of these samples was any emanation found. Various other observers have found values ranging around 10^{-12} curie of radium emanation per gram of sea salt, but the samples in these cases have usually been collected much nearer to land than the bulk of those on the *Carnegie*. The average amount of salt used in a determination was about 8 grams, so that if the radium content were as high as that found by former observers 8×10^{-12} curie of emanation was to be expected. Experiments with known amounts of radium emanation showed that 8×10^{-12} curie of emanation should cause a drift of the electroscope fiber of from 0.5 to 1.5 divisions per minute. A drift of 0.05 division per minute could be detected, so that it would seem that the sea salt tested in these experiments contained not more than 10% of the radium emanation which has been found by other observers.

Experiments with known amounts of radioactive material were made in various ways: By putting a fairly large amount of the unsealed material together with some fusion mixture into the furnace and heating it; by sealing a certain amount of radioactive material for a definite time and then putting this in the furnace with some fusion mixture; and finally by placing with a usual charge of sea salt and fusion mixture a small amount of radioactive material, containing a small amount of emanation of the order of magnitude of that expected from the sea salt. Some of this same sample of sea salt had been previously tested for emanation, and it was found to have none. This test with the radioactive material was made to eliminate the possibility that such a small amount of emanation might be prohibited from coming off by the large amount of material with which it was mixed.

As examples of the method of observation the two determinations, with the sea salt and carbonates alone, and then with a sample of the same sea salt, carbonates, and radioactive material, will suffice.

EXAMPLE 1.

Date: Aug. 28, 1917. 9.61 grams of sea salt sealed up at 11^h 27^m a. m., Aug. 25, 1917, mixed with 17 grams of fusion mixture. This mixture was placed in the furnace and direct current turned on as follows:

| | | h m | | First Gases Admitted to: | |
|-------------|------------|-----|----------|--------------------------|----------|
| 42 volts at | 2 18 p. m. | | | Flask | |
| 72 " | " 2 32 | | | (metal shavings) | Chamber |
| 96 " | " 2 46 | | | h m | h m |
| 120 " | " 3 14 | 3 | 36 p. m. | 3 | 53 p. m. |
| 96 " | " 3 47 | 3 | 54 | 4 | 04 |
| 0 " | " 4 18 | 4 | 05 | 4 | 15 |

Preliminary Drift of Electroscope-Reading.

| Read- ing | Time | | Time for 2-di- vision drift | |
|--------------|------|-----|--------------------------------|-------|
| | h | m s | m s | |
| 51 | 3 | 21 | 20 | p. m. |
| 50 | | 22 | 47 | 2 25 |
| 49 | | 23 | 45 | |
| 51 | 3 | 24 | 10 | |
| 50 | | 25 | 20 | 2 30 |
| 49 | | 26 | 40 | |
| 51 | 3 | 27 | 00 | |
| 50 | | 28 | 10 | 2 15 |
| 49 | | 29 | 15 | |
| Mean | | | 2 | 23 |

Drift of Electroscope-Reading with First Gases in the Ionization-Chamber.

| Read- ing | Time | | Time for 2-di- vision drift | | Read- ing | Time | | Time for 2-di- vision drift | |
|--------------|------|-----|--------------------------------|-------|--------------|------|-----|--------------------------------|-------|
| | h | m s | m s | | | h | m s | m s | |
| 51 | 4 | 19 | 20 | p. m. | 51 | 4 | 41 | 00 | p. m. |
| 50 | | 20 | 20 | 1 50 | 50 | | 42 | 30 | 2 45 |
| 49 | | 21 | 10 | | 49 | | 43 | 45 | |
| 51 | 4 | 21 | 30 | | 51 | 4 | 44 | 10 | |
| 50 | | 22 | 40 | 2 05 | 50 | | 45 | 25 | 2 30 |
| 49 | | 23 | 35 | | 49 | | 46 | 40 | |
| 51 | 4 | 23 | 50 | | 51 | 4 | 47 | 00 | |
| 50 | | 24 | 55 | 2 00 | 50 | | 48 | 20 | 2 50 |
| 49 | | 25 | 50 | | 49 | | 49 | 50 | |
| Mean | | | 1 58 | | Mean | | | 2 42 | |

Second Gases Admitted to:

| Flask (metal shavings) | | Chamber | |
|---------------------------|----------|---------|----------|
| h | m | h | m |
| 4 | 15 p. m. | 4 | 54 p. m. |
| 4 | 55 | 5 | 05 |

Drift of Electroscope-Reading with Second Gases in the Ionization-Chamber.

| Read- ing | Time | | | Time for 2-di- vision drift | |
|--------------|------|----|------|--------------------------------|----|
| | h | m | s | m | s |
| 51 | 5 | 14 | 30 | p. m., Aug. 28 | |
| 50 | | 14 | 45 | 0 | 35 |
| 49 | | 15 | 05 | | |
| 51 | 5 | 15 | 20 | | |
| 50 | | 15 | 35 | 0 | 35 |
| 49 | | 15 | 55 | | |
| 51 | 5 | 16 | 15 | | |
| 50 | | 16 | 35 | 0 | 40 |
| 49 | | 16 | 55 | | |
| | | | Mean | 0 | 37 |
| 51 | 9 | 51 | 10 | a. m., Aug. 29 | |
| 50 | | 52 | 10 | 2 | 15 |
| 49 | | 53 | 25 | | |
| 51 | 9 | 53 | 50 | | |
| 50 | | 55 | 00 | 2 | 34 |
| 49 | | 56 | 24 | | |
| 51 | 9 | 56 | 45 | | |
| 50 | | 58 | 18 | 2 | 35 |
| 49 | | 59 | 20 | | |
| 51 | 9 | 59 | 40 | | |
| 50 | 10 | 01 | 00 | 2 | 35 |
| 49 | | 02 | 15 | | |
| | | | Mean | 2 | 30 |

| Read- ing | Time | | | Time for 2-di- vision drift | |
|--------------|------|----|------|--------------------------------|----|
| | h | m | s | m | s |
| 51 | 10 | 12 | 35 | a. m., Aug. 29 | |
| 50 | | 13 | 55 | 2 | 20 |
| 49 | | 14 | 55 | | |
| 51 | 10 | 15 | 18 | | |
| 50 | | 15 | 20 | 2 | 30 |
| 49 | | 17 | 48 | | |
| 51 | 10 | 18 | 05 | | |
| 50 | | 19 | 08 | 2 | 10 |
| 49 | | 20 | 15 | | |
| 51 | 10 | 20 | 35 | | |
| 50 | | 22 | 00 | 2 | 37 |
| 49 | | 23 | 12 | | |
| | | | Mean | 2 | 24 |

Second Gases Cleared Out of Ionization-Chamber, and Drift Again Observed.

| Read- ing | Time | | | Time for 2-di- vision drift | |
|--------------|------|----|------|--------------------------------|----|
| | h | m | s | m | s |
| 51 | 10 | 36 | 45 | a. m. | |
| 50 | | 37 | 40 | 2 | 00 |
| 49 | | 38 | 45 | | |
| 51 | 10 | 39 | 10 | | |
| 50 | | 40 | 40 | 2 | 55 |
| 49 | | 42 | 05 | | |
| 51 | 10 | 42 | 20 | a. m. | |
| 50 | | 43 | 30 | 2 | 25 |
| 49 | | 44 | 45 | | |
| 51 | 10 | 45 | 10 | | |
| 50 | | 46 | 05 | 2 | 25 |
| 49 | | 47 | 35 | | |
| | | | Mean | 2 | 26 |

Since the time for a drift of 2 divisions is as long when the gases which were driven off from the sea salt were in the ionization-chamber as when the ordinary room gases were in the ionization-chamber, it follows that the emanation driven off from the amount of the sea salt is smaller than the electroscope would detect.

The following is a record of the test made with a portion of the same sample of sea salt as the above test, but this time a small amount of carnotite containing an amount of radium emanation of the order of magnitude of that looked for in the sea salt, was added.

Second Gases Admitted to:

| Flask (metal shavings) | | Chamber | |
|---------------------------|----------|---------|----------|
| h | m | h | m |
| 1 | 31 p. m. | 2 | 53 p. m. |
| 2 | 54 | 3 | 04 |
| 3 | 05 | 3 | 15 |

Drift of Electroscope-Reading with Second Gases in the Ionization-Chamber.

| Read- ing | Time | | | Time for 2-di- vision drift | | Read- ing | Time | | | Time for 2-di- vision drift | |
|--------------|------|----|----------|--------------------------------|----|--------------|------|----|----------|--------------------------------|----|
| | h | m | s | | | | h | m | s | | |
| 51 | 3 | 31 | 00 p. m. | | | 51 | 4 | 16 | 00 p. m. | | |
| 50 | | 31 | 45 | 1 | 40 | 50 | | 16 | 55 | 2 | 00 |
| 49 | | 32 | 40 | | | 49 | | 18 | 00 | | |
| 51 | 3 | 33 | 00 | | | 51 | 4 | 18 | 20 | | |
| 50 | | 33 | 45 | 1 | 45 | 50 | | 19 | 05 | 1 | 30 |
| 49 | | 34 | 45 | | | 49 | | 19 | 50 | | |
| 51 | 3 | 35 | 00 | | | 51 | 4 | 20 | 10 | | |
| 50 | | 36 | 00 | 1 | 55 | 50 | | 21 | 15 | 2 | 03 |
| 49 | | 36 | 55 | | | 49 | | 22 | 13 | | |
| Mean | | | | 1 | 47 | 51 | 4 | 22 | 50 | | |
| | | | | | | 50 | | 23 | 55 | 2 | 15 |
| | | | | | | 49 | | 25 | 05 | | |
| | | | | | | 51 | 4 | 25 | 25 | | |
| | | | | | | 50 | | 26 | 18 | 1 | 40 |
| | | | | | | 49 | | 27 | 05 | | |
| | | | | | | 51 | 4 | 27 | 20 | | |
| | | | | | | 50 | | 28 | 35 | 1 | 55 |
| | | | | | | 49 | | 29 | 15 | | |
| | | | | | | Mean | | | 1 | 54 | |

Second Gases Cleared Out of the Ionization-Chamber and Drift Again Observed.

| Read- ing | Time | | | Time for 2-di- vision drift | | Read- ing | Time | | | Time for 2-di- vision drift | |
|--------------|------|----|----------|--------------------------------|----|--------------|------|----|----------|--------------------------------|----|
| | h | m | s | | | | h | m | s | | |
| 51 | 4 | 35 | 30 p. m. | | | 51 | 4 | 40 | 35 p. m. | | |
| 50 | | 36 | 25 | 2 | 13 | 50 | | 41 | 33 | 2 | 00 |
| 49 | | 37 | 43 | | | 49 | | 42 | 35 | | |
| 51 | 4 | 38 | 10 | | | 51 | 4 | 43 | 00 | | |
| 50 | | 39 | 10 | 2 | 00 | 50 | | 44 | 05 | 2 | 25 |
| 49 | | 40 | 10 | | | 49 | | 45 | 25 | | |
| | | | | | | Mean | | | 2 | 10 | |

By the time the final readings with the second gases were taken, most of the activity due to the active deposit from the emanation first admitted had, of course, disappeared. The object of sealing the specimen of carnotite for some days before use, was to make the radium content a more definite quantity. Thus, if a specimen contained no emanation when sealed, it would acquire 0.7 of its equilibrium amount in 7 days.

Computation of these results show that a drift of 0.1 division per minute at the end of one hour corresponded to 1.7×10^{-12} curie of radium emanation in the ionization-chamber. This drift could easily be detected, and 1.7×10^{-12} curie of emanation is of the order of magnitude of the amount to be looked for in one gram of sea salt according to previous investigations.

The following table contains the data in regard to the samples of sea salt investigated:

| Date Collected | Lat. | Long. (E. of Gr.) | Grams Sea Salt | Grams Fusion Mixture |
|-----------------------|------|-------------------|-------------------|-------------------------|
| May 24-30, 1916 | 35 S | 186 | 10.00 | 30 |
| Sept. 10-16, 1916 | 44 N | 221 | 6.87 | 17 |
| June 27-July 3, 1916 | 10 N | 180 | 5.31 | 19 |
| June 20-26, 1916 | 5 S | 188 | 6.09 | 18 |
| July 4-11, 1916 | 18 N | 165 | 7.36 | 23 |
| Aug. 27-Sept. 9, 1916 | 49 N | 191 | 6.02 | 21 |
| Dec. 7-13, 1916 | 19 S | 235 | 9.04 | 18 |
| July 12-16, 1916 | 16 N | 150 | 7.77 | 20 |
| Aug. 20-26, 1916 | 44 N | 160 | 7.76 | 15 |
| Jan. 2-8, 1917 | 20 S | 248 | 8.28 | 17 |
| Jan. 10-16, 1917 | 16 S | 234 | 9.61 | 17 |
| Jan. 22-29, 1917 | 35 S | 219 | 12.31 | 17 |
| Dec. 27, 1915 | 59 S | 270 | 7.10 | 17 |
| Jan. 26, 1916 | 55 S | 20 | 6.56 | 17 |
| Dec. 26, 1915 | 59 S | 270 | 8.18 | 17 |

In none of these cases was any radium emanation detected.

In conclusion, mention should be made of Observers H. F. Johnston, I. A. Luke, B. Jones, and A. D. Power, who collected the samples of salt. The author also wishes to express his thanks to Doctors Day and Sosman of the Geophysical Laboratory, for the loan of an alundun-tube heating-element, to Doctor Dorsey, of the Bureau of Standards, for the loan of an analyzed specimen of carnotite, and to the Bureau of Mines for supplying an analyzed specimen of pitchblend.