

## THE RADIOACTIVITY OF PHILIPPINE WATERS

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In recent years there have been so many investigations of the radioactivity of waters that comparative data are now available from many different parts of the world. It is the purpose of this paper to present the results of similar work on the radioactivity of typical Philippine waters.

Up to the present time our work on radioactivity has been limited to the determination of radium emanation and actual radium content of typical springs and deep, drilled wells. As yet, no attempt has been made to study the radioactivity of the deposits or residues from springs, or of typical rocks, or of the gases evolved from springs or wells. All of the work was done on Luzon, part of it at high altitudes in the mineral-bearing mountainous region and the remainder practically at sea level.

Owing to the difficulties of travel, equipment for provincial expeditions had to be reduced to a minimum; in consequence comparatively few data on the chemical composition of the waters were secured.

### Experimental Part

The testing apparatus was a Spindler and Hoyer aluminum leaf electroscope used in previous radioactive measurements<sup>1</sup> in the Philippines. The instrument was provided with the usual equipment of tripod, shaking vessel, and circulation system necessary for field work.

Owing to the high humidity generally encountered in the Philippines, a small tube of calcium chloride was fastened inside the leaf chamber of the electroscope. In all determinations, in the field or in the laboratory, a calcium chloride tube (generally of the Bender and Hobein type) was introduced into the circulation system, so that all air and gases were

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<sup>1</sup> J. R. Wright and O. F. Smith: *Phys. Zeit.*, **15**, 31-39 (1914); *Philippine Jour. Sci.*, **9A**, 51-77 (1914); *Phys. Rev.*, **5**, 459-482 (1915); **7**, 49-61 (1916).

dried before they entered the ionization chamber. It was found impossible to obtain reliable readings without this precaution. Our results indicate that drying is also necessary at lower temperatures in order to ensure correct results with the type of instrument used. With the precautions noted, the natural leak was usually lower in laboratory work, and frequently lower in field work, than the figures for the natural leak furnished by the instrument makers. The electroscope was charged by means of a bank of storage cells in the laboratory; in the field, by means of an ebonite rod.

The electroscope was standardized by means of a radium bromide solution of known strength secured<sup>1</sup> from the Bureau of Standards, Washington, D. C. A definite portion of this solution was kept sealed in a proper vessel (essentially a "Curie<sup>2</sup> tube") for over one month. The emanation was then removed from the solution by boiling and by circulation of air and drawn into the evacuated ionization chamber.

The standardization was checked with a known quantity of standard radium bromide solution put directly into the shaking vessel used in field determinations. After the closed vessel had been allowed to stand one month to ensure radioactive equilibrium, a determination was made by the usual field method. Two different determinations showed satisfactory agreement with each other and with the results previously obtained. This procedure is much simpler than the usual method of standardization, and since determinations both with the standard solution and with water samples of unknown activity are thus performed under identical conditions, the chance for error is minimized. Our data indicate that this method can be relied upon for accurate results.

The shaking apparatus of Schmidt<sup>3</sup> was employed in field

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<sup>1</sup> Our thanks are due the Bureau of Standards, Washington, D. C., for the standard radium solution used in this work.

<sup>2</sup> P. Curie: "Dosage du radium par la mesure de l'emanation dégagee," *LeRadium*, 7, 65-70 (1910).

<sup>3</sup> H. W. Schmidt: "Über eine einfache Methode zur Messung des Emanationsgehaltes von Flüssigkeiten," *Phys. Zeit.*, 6, 561-566 (1905).

work. Determinations were made as soon as practicable after a sample had been taken; usually within thirty minutes and in no case after more than three hours.

Samples of typical waters were brought to the laboratory, sealed up in proper vessels, allowed to stand for one month, and tested for radium content. Instead of Curie tubes, 400 cc Jena flasks, equipped with aspirator tubes, were used to hold the water. About 250 cc of water were used in a determination. In some cases, when larger quantities of water were available, as much as 15 liters were evaporated to 250 cc, acidified with hydrochloric acid, sealed up in Jena flasks, and tested.

Though the object of this work was primarily to get reliable comparative data rather than exceedingly accurate absolute values, it is probable that the error was not great. The radium solution used for standardization may be considered accurate within 5 percent. Duplicate determinations made with the standard solution checked within 1.5 percent. Duplicate determinations made in the field on the same water checked within the limits of observation error. We have reason to believe that, except for isolated cases in which proper samples could not be secured because of the nature of the source, the maximum error in field determinations was not greater than 7 percent. The probable error was, therefore, much smaller and hence was well within the limits of accuracy to be expected for this class of work.

No attempt was made in the field to determine anything but radium emanation content. That we were actually dealing with radium emanation was shown by the fact that when the gases from a number of the waters studied were allowed to remain in the ionization chamber for long periods of time the typical decay curve of radium emanation was obtained.

The radioactivity of over ninety Philippine waters was determined, the following data being typical:

TABLE 1.—RADIOACTIVITY OF PHILIPPINE WATERS. (SOURCES DESIGNATED BY \* HAVE REPUTATIONS FOR MEDICINAL OR HEALING PROPERTIES)

No.	Date 1916	Location (Province, town, and barrio)	Source	Geological formation <sup>1</sup>	Radium emanation <sup>2</sup> Curies per liter $\times 10^{12}$	Remarks
1	June 7	Batangas, Batangas	Artesian well	—	2106	Non-thermal
2	June 6	Batangas, Batangas	Crater lake, Taal volcano	Volcanic sinter	3	—
3	April 9	Bulacan, San Miguel de Mayumo, Sibul Springs	Sibul Springs*	Limestone	1284	Non-thermal mildly sulphuretted
4	June 9	Bulacan, San Miguel de Mayumo, Sibul Springs	Sibul Springs*	Limestone	1293	Non-thermal mildly sulphuretted
5	June 20	Laguna, Calamba Pansol	"Pansol" Springs*	Volcanic basalt	Neg.	Series of large hot and cold springs
6	June 11	Laguna, Calamba Pansol	"Pansol" Springs*	Volcanic basalt	Neg.	Sample from cool portion (31° C)
7	April 19	Laguna, Los Baños	Hot Spring, * near sanitarium	Igneous conglomerate cemented by tuff	539	Sample from hot portion (45° C)
8	June 29	Laguna, Majayjay, Olla	"Olla" spring	Loose volcanic agglomerate	528	Temperature 70° C; probably from same source as water for sanitarium
9	June 29	Laguna, Majayjay, Malinoa	"Sinabac" spring	Igneous conglomerate	1297	Temperature 22° C; probably (see page 531) from a river in vicinity
10	June 24	Laguna, Nagscarlan	"San Diego" spring*	Igneous conglomerate	526	Temperature 25° C
11	June 21	Laguna, Pagsanjan, Maulaun	Small artesian well	—	880	Temperature 25° C
12	June 21	Laguna, Pagsanjan, Pinagsanjan	"Bumbungan" spring*	—	146	Temperature 32° C
13	June 30	Laguna, Pakil	Spring "Baño" spring	Igneous conglomerate	365	Temperature 31° C
14	June 22	Laguna, San Pablo	Spring "Bañadero"	—	713	Temperature 25.5° C
15	June 22	Laguna, San Pablo, Maganpun	Spring "Baño"	—	606	Non-thermal
16	June 22	Laguna, San Pablo, Santa Maria	Spring "Años"	—	324	Non-thermal

Temperature about 40° C. Located in bed of river Añosan; sample probably mixed with river water

17	June 22	Laguna, Santa Cruz	Large artesian well, No. 459	—	Nil	Temperature 36° C
18	May 30	La Union, San Fernando	Municipal spring	Andesite	242	Non-thermal
19	June 1	La Union, Santo Tomas	Artesian well*	—	Trace	Non-thermal
20	April 29	Mountain, Baguio	Camp John Hay No. 1 spring	Igneous, fined grained rock	194	Non-thermal; supplies Camp Hay, elevation about 3,500 ft.
21	April 29	Mountain, Banaue	Small spring near school house	—	381	Non-thermal; elevation 3,400 ft.
22	May 14	Mountain, Bontoc	Spring adjacent to municipal spring	Volcanic tuff	Neg.	Non-thermal; elevation about 2,800 ft.
23	May 10	Mountain, Cervantes	Hot spring on river bank, opposite town	—	Nil	Temperature 50° C; elevation about 2,400 ft.
24	Dec. 18	Mountain, Itogon	Hot springs*	Andesite	Nil	Temperature 45°
25	Dec. 18	Mountain, Kiangnan	Spring E, of school, on Nueva Viscaya road	—	1058	Non-thermal; elevation about 2,700 ft.
26	May 29	Mountain, Klondike	Klondike springs*	Igneous conglomerate cemented by tuff	Trace	Temperature 50° C; elevation about 600 ft.
27	May 15	Mountain, Mainit	"Mainit" spring*	Andesite siliceous and calcareous sinter	Nil	Sample from boiling saline spring, elevation 4,000 ft.
28	May 8	Mountain, Mancayan	Spring on Balili trail	Trachyte andesite	114	Non-thermal, elevation about 4,700 ft.
29	May 12	Mountain, Sagada, Teteupan	Small spring at slide	Decomposed andesite	263	Elevation 4,000 feet
30	May 31	Mountain, Sagada, Teteupan	Artesian well, calle Espanol	—	137	Temperature about 35° C
31	June 19	Rizal, Parañaque	Artesian well, plaza	—	632	Temperature about 35° C

<sup>1</sup> Most of the data in this column were furnished by V. E. Lednicky, Chief, Division of Mines, Bureau of Science.

<sup>2</sup> If there was no change in the leak when radioscope was filled with gas, emanation content is designated as "nil;" when increase was so slight that its magnitude was uncertain, emanation content is designated as "neg." (negligible); when increase was positive, but too small to warrant calculation, emanation content is designated as "trace."

<sup>3</sup> Tested for radium content only. Negative results with 250 cc.

In order to examine waters for actual radium content, 250 cc samples of twenty typical waters, from which the emanation had been removed, were acidified, sealed up in proper containers, and allowed to stand at least one month. In no case was there any indication of radioactivity at the end of that time.

In addition, 15 liters of Batangas water (No. 1), 15 liters of Los Baños water (No. 7), and 5 liters of Sibul Springs water (No. 3) were evaporated to small bulk and similarly tested. The first two showed no emanation, and the third showed a trace.

The radioactivity of the waters studied, therefore, was due primarily to emanation derived from the materials in the ground with which the water had been in contact.

### Discussion of Results

The work has not proceeded sufficiently to justify many conclusions. The typical Philippine water supplies studied possess no abnormal features, so far as their radioactivity is concerned. Though some of them are moderately high in radium emanation content, none show an excessive amount, compared with waters from other countries reported in the literature.<sup>1</sup>

Since hot water is a poorer solvent of gases than cold water, it is to be expected that the radioactivity of hot springs should, in general, be low. With the exception of the Los Baños water (No. 7), most of the thermal waters studied in the course of this work showed little or no activity.<sup>2</sup>

In general, the average activity of igneous rocks is greater than that of the sedimentary,<sup>3</sup> and it is to be expected that water from the former material should show the higher emana-

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<sup>1</sup> Cf. Schlundt and Moore: *Bull. U. S. Geol. Surv.*, **395**, 29 (1909); *Jour. Phys. Chem.*, **18**, 662 (1914); D. Isitani: *Proc. Tokyo Math. Physic. Soc.* (1912), ff., etc.

<sup>2</sup> The average temperature of Philippine ground waters in the lowlands is about 28° C.

<sup>3</sup> Cf. F. W. Clarke: "Data of Geochemistry," *Bull. U. S. Geol. Surv.*, **166**, 122 (1916).

tion content. Thus Sahlbom<sup>1</sup> found that the water from sedimentary deposits was much lower in activity than that from primary rocks; further, that wells bored in the acid rocks showed the highest activity. In the Philippines the relatively small number of determinations made and the frequent difficulty of determining the actual water-bearing stratum, since this is frequently not the same as the geological formation exposed at the place where the water emerges, make generalizations at this time inadvisable.

In some cases at least, the radioactive material from which the water derived its activity must have been confined to a rather limited area. Thus it was pointed out<sup>2</sup> that "Olla" Springs (No. 8) were in reality only seepage water derived from a river about a hundred meters distant. The "spring" water must obviously have acquired its activity in the course of a short journey underground.

So far as the available analytical data on the waters tested are concerned, there is no apparent general relation between the chemical quality of a water and its radioactivity. This is not surprising, since the emanation content appears to be due, not to dissolved radium, but to contact with radioactive materials, sometimes within a very restricted area.

There was no sharply defined rainy season<sup>3</sup> during 1916 in the places visited. With the exception of the determinations made in April and December, the tests for radioactivity were conducted during months of considerable rain.

Though no systematic study of the relation between the radioactivity and the variation of flowing wells and springs has been made,<sup>4</sup> it may be of interest to point out that Sibul

<sup>1</sup> N. Sahlbom: *Arkiv. Kemi, Min. Geol.*, 6, No. 3, 1-52 (1915); through *Chem. Abstracts*, 10, 1134 (1916).

<sup>2</sup> By Mr. B. E. Lednicky, Chief, Division of Mines, Bureau of Science.

<sup>3</sup> For the distribution of rainfall in the Philippines according to locality and season, see A. J. Cox: *Philippine Jour. Sci.*, 6A, 287-296 (1911).

<sup>4</sup> The flow of many springs and deep wells in the Philippines changes greatly in quantity with the tide, though there is no appreciable variation in chemical quality. See note on the tidal variation of springs and deep wells in the Philippine Islands. W. G. Heise: *Philippine Jour. Sci.*, 11A, 125-127 (1916).

Springs (Nos. 3 and 4) were tested on two different occasions (as indicated in Table I), once in the middle of the dry season, that is, after two or three practically rainless months, and once during a period of frequent rains. The results of the two determinations, which are practically identical, indicate that deep-seated springs, such as Sibul Springs, may show surprisingly small seasonal variations in radioactivity. It should be pointed out that Sibul Springs show a comparatively slight variation in flow throughout the year, so that the inference from the two isolated determinations just discussed is not considered to be at variance with the findings of others. Thus Ramsey<sup>1</sup> found a greater emanation content in certain springs during period of wet weather and great flow, and Steichen<sup>2</sup> observed an increase in activity in certain Bombay hot springs during the dry season, while the flow of water was considerably reduced. As pointed out by the latter investigator, local conditions may well account for the differences noted.

For the sake of completeness, we have noted in our data all sources of water supply which are popularly considered to have special medicinal virtues. Obviously, either popular opinion is a poor guide to the medicinal value of a water, or else the medicinal properties of water are not to any great extent due to radium emanation content. The waters with perhaps the greatest reputation, namely, Los Baños and Sibul Springs (Nos. 7 and 3), have relatively high radium emanation contents, yet many others regarded as highly, such as Marilao, Pansol (Nos. 5 and 6), Santo Tomas (No. 19), and Klondike (No. 26), contain little or no emanation. Moreover, many waters high on the list, such as Batangas (No. 1), Kiangan (No. 25), Pagsanjan (No. 11), and in general the spring waters near Mt. Banajao (Nos. 8-16), are regarded with entire indifference.

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<sup>1</sup> R. R. Ramsey: "The Variation of the Emanation Content of Certain Springs," *Phil. Mag.*, **30**, 815-818 (1915).

<sup>2</sup> A. Steichen: "The Variation of the Radioactivity of the Hot Springs at Tuwa," *Phil. Mag.*, **31**, 401-3 (1916).



### Summary

The radioactivity of about ninety different Philippine waters, chiefly from springs and flowing wells, has been studied. The highest radium emanation content encountered in a deep-well water was equivalent to  $2100 \times 10^{-12}$  grams of radium (per liter); the highest in a spring water was equivalent to  $1300 \times 10^{-12}$  grams of radium.

A test for the actual radium content of about twenty typical sources showed that the radioactivity encountered was due to emanation absorbed from materials with which the ground water had been in contact and was not due to dissolved radium salts. One sample of water showed a scarcely detectible trace of activity due to radium salts in solution; all the others tested gave negative results.

This paper describes a part of an extensive study of Philippine waters, full details of which will be found in the *Philippine Journal of Science*.

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