THE VARIATION WITH METEOROLOGICAL CONDITIONS OF THE AMOUNT OF RADIUM EMANATION IN THE ATMOSPHERE, IN THE SOIL GAS, AND IN THE AIR EXHALED FROM THE SURFACE OF THE GROUND, AT MANILA.

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In the Philippine Journal of Science of February, 1914, the authors published the results of an extensive series of observations on the amount of radium emanation in the atmosphere of Manila. The well-known charcoal-absorption method was used and the main part of the investigation was preceded by numerous tests on certain points concerned with the accuracy of the method. The work at Manila was supplemented by an investigation of the variation of the emanation content of the atmosphere with altitude, observations being taken on Mt. Pauai, elevation 2,460 meters, and the results compared with those for sea level.[1]

Although the especial points of the investigation were those above mentioned we felt justified at that time, since the period involved extended over almost a year and a half, in pointing out certain evident relations between the emanation content of the atmosphere and the different meteorological factors. The data at our command was not considered sufficient, however, to justify the drawing of more than very general conclusions. During the past year observations have been taken with the object of determining more definitely to what extent the amount of radium emanation in the air is dependent on weather conditions. At the same time tests have been made on several other points which have a more or less direct bearing on the question.

The experimental method used was described in detail in our previous paper, but for the sake of completeness a brief summary of the description will be given, together with such modifications as experience had shown to be advisable.

The method had its origin in the discovery by Rutherford that charcoal made from the shells of coconuts possesses the property of absorbing radioactive emanations. Eve[2] and Satterly[3] independently applied the discovery to the determination of the radium-emanation content of the atmosphere.

[1] Phys. Zeit., *15*, 31, 1914.
[2] Phil. Mag., *14*, 724, 1907.
[3] Phil. Mag., *16*, 584, 1908.

To attain this end air was passed at a known rate and for a definite length of time through a tube containing coconut charcoal, which absorbed the emanation from the air. At the same time air was bubbled through a solution of radium bromide, containing a known amount of radium, and the emanation from the solution and the air collected in another charcoal tube. The emanation absorbed in the charcoal was then driven off by heating, collected over water in aspirators, and finally measured by passing into an ionization chamber connected either with an electroscope or an electrometer. By this method quantitative determinations of the amount of radium emanation in the atmosphere can be obtained, and since the method is a comparative one the results should be independent of variations in the pressure, humidity, and nucleation of the air, which is a serious objection to the extensively used active-deposit method.

Since in the present investigation the object was to determine the variation of the emanation content of the air with weather conditions it was essential that all observations should be taken under identical experimental conditions and in the same manner. Having had two years experience in taking similar observations we were able to attain this end with certainty. Throughout the thirteen months covered by this series of experiments the arrangement of apparatus and the method of procedure in taking observations was never varied. Consequently, the results should at least show quantitatively the variation of the amount of emanation in the atmosphere.

Experimental Procedure.

Collecting.—The air to be tested was drawn in through a tube, projecting from a second story window of the Bureau of Science, by means of a motor driven oil pump. By placing several large bottles, having a total capacity of about 50 liters, in series with the pump a constant rate of flow could be maintained for any desired length of time. The rate of flow was determined by means of accurately calibrated oil manometers across glass capillary tubes of fairly small bore. The air stream was regulated by means of easily adjusted pinch cocks on rubber tubing.

To extract the dust from the air a tube containing cotton wool was placed between the intake and a large distributing bottle. From the distributing bottle the air was divided into two exactly equal parts, one part passing through the branch containing the radium bromide solution and the other part through an exactly similar system, with the exception of that part connected with the standard solution. In both branches the air was thoroughly dried by bubbling it first through sulphuric acid and then passing it over calcium chloride.

The coconut charcoal was contained in electro-silica tubes having a uniform bore of 1.5 cm. In each tube there was placed 70 grams of finely granulated coconut charcoal, which was firmly packed and held in place by long plugs of asbestos wool. Experience had shown the importance of keeping the charcoal firmly and uniformly packed if the same fraction of the total amount of emanation passing through the tubes was to be absorbed in every case. Two of these tubes were placed in series in each branch of the collecting system. Tests, which have been described in detail in an earlier paper, had proven that two such tubes in series absorb practically all the emanation passing through them during a 20-hour run. Each set of tubes was permanently labeled and their relative absorptive powers carefully determined for the conditions under which they were to be used.

The bottle containing the radium bromide solution was so arranged that it could be heated by immersing in a solution of sodium chloride. A large spherical condenser was attached to the bottle to prevent loss of the radium bromide during the process of heating. The standard used throughout the entire series of determinations contained 6.28×10^{-10} grams of radium. Preceding each test the solution was put into the so-called "steady state" by bubbling air through the boiling solution for one hour and then through the cool solution for two hours. Our preliminary work had shown conclusively that bubbling air through the cool solution did not remove all the acculumated emanation, nor remove it as rapidly as formed even after the solution had been put in the "steady state." Under the conditions maintained throughout the tests approximately 80 per cent. of the emanation was removed from the solution. This is absolutely independent of the question of what per cent. of the total amount passing over the charcoal is absorbed. In comparative tests the only point of importance in regard to the absorption is that each branch shall absorb the same fraction of the total amount passing through it.

Testing.—The testing apparatus used was a Spindler and Hoyer aluminum-leaf electroscope with an ionization chamber attached. The aluminum leaf in these electroscopes has a fine quartz fiber attached to one edge which makes it possible to obtain very accurate readings with the aid of the reading microscope. The ionization chamber was 38 cm. high and 7.8 cm. in diameter, giving a volume of 1,820 c.c. It was provided with two outlet tubes so that the chamber could easily be exhausted and refilled with the air containing the emanation. The electroscope with the attached ionization chamber had an electrical capacity of 8.7 e.s. units, the range of the scale of 100 divisions being approximately from 368 to 302 volts. The potential gradient, therefore, was sufficient to produce saturation currents for the degree of ionization dealt with in the experiments. The natural leak was exceedingly constant and had a value of 0.016 division per minute.

The ionization chamber was permanently attached through one opening to a mercury manometer and through the other opening to a Geryk oil pump and to two aspirator bottles, all connected in parallel, so that any one could be put in direct connection with the chamber. Between the aspirators and ionization chamber were placed two small U tubes, one containing calcium chloride and the other phosphorus pentoxide, permitting all the air passing into the chamber to be thoroughly dried.

After a few preliminary experiments we adopted the following method of taking measurements on the emanation collected. The charcoal tubes were placed in a tubular electric furnace and connected in parallel to one aspirator. The tubes were heated to a bright red heat. The temperature for the different determinations was practically the same, equal currents being always passed through the electric furnace for the same length of time. The tubes were then rapidly but thoroughly flushed until the aspirator was filled down to a certain mark. In order to prevent absorption of the emanation the water in the aspirator bottle was heated by an immersed electric coil. The air containing the emanation was then passed into the ionization chamber through the calcium chloride and phosphorus pentoxide tubes, care being taken to thoroughly flush the tubes with air so that all the emanation would be carried into the ionization chamber. The chamber had been made with the necessary volume to accommodate all the gas driven off from 140 grams of charcoal with a liberal margin for flushing. The electroscope readings were always taken over practically the same region of the scale, the reading being started as nearly as possible thirty minutes after introducing the emanation into the chamber. The deflection of the aluminum leaf for the following thirty minutes was then recorded. By this method the reading was always obtained over approximately the same portion of the decay curve for radium emanation, thereby making the electroscope readings directly comparable.

Amount of Radium Emanation in the Atmosphere at Manila.

The theory underlying the calculations of the radium-emanation content from observations by the charcoal-absorption method has been given by several writers on the subject. If M represents the radium equivalent in grams of the emanation in one cubic meter of free air, then

$$M = \frac{M^1 \lambda T}{V} \frac{d}{d_1},$$

where M^1 is the number of grams of radium in the standard solution, λ the radioactive constant of radium, d the electroscope reading due to the emanation from V cubic meters of air, and d_1 the electroscope reading corresponding to the emanation formed by M^1 grams of radium in the time interval T.

In the deduction of the above equation it is assumed that all the accumulated emanation has been removed from the solution previous to a run, and that all the emanation formed during the time of collecting is removed from the solution and passes through the collecting system. Our preliminary experiments had shown that this assumption is not justified for the conditions of temperature, rate of flow of air stream, etc., under which our tests were made. In which case it is necessary to multiply the above equation by some factor having a value less than unity. If we represent this factor by α the equation then becomes

$$M = \frac{\alpha M^1 \lambda T}{V} \frac{d}{d_1}.$$

A long series of determinations, with conditions maintained as rigidly constant as possible, gave for this factor a value of 0.792. Extreme care was taken throughout all our tests on the emanation content to maintain conditions the same as those existing during the determination of the value of α.

Table I. gives the results of a series of observations for the period from July, 1913, to July, 1914, together with such meteorological data as is necessary to show any existing correlation. The meteorological data is taken from the reports of the Manila Observatory which is situated about 400 meters from the Bureau of Science, consequently the two sets of data practically coincide as to location. All the radioactive determinations given in Table I. were taken under identical experimental conditions. The standard solution was put into the "steady state" by bubbling air at the rate of 0.5 liter per minute through the boiling solution for one hour and then through the cool solution for two hours. At the end of the two hours the air stream was started through the collecting system, the rate of flow being maintained constant at 0.5 liter per minute. All observations extended from 1 P.M. to 9 A.M., a period of twenty hours, making the volume of air tested in every case equal to 0.6 cubic meter. As will be seen later it is extremely important that the observations should be taken over the same part of the day if the results are to have a high comparative value.

TABLE I.

Variation of the Radium Emanation Content with Meteorological Conditions.

Date.	Pressure (Mean), (Mm.)	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), (%).	Wind. Prevailing Direction.	Wind. Total Movement, (Km.).	Wind. Total Movement During the Three Days, (Km.).	Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams $\times 10^{12}$).	Weather Remarks.
July 8, 1913	757.94		80.0	WSW	267.0		....			Fair during July 8 and during
July 9, 1913	757.60	Decreasing	81.6	WSW	321.0	922.0	11.8	13.3	35.8	day of July 9. Very heavy
July 10, 1913	757.47		92.9	WSW	334.0		1.5			shower during night of 9th.
July 22, 1913	755.30		91.4	WSW	468.0		28.5			Very heavy rains during the
July 23, 1913	753.64	Variable	89.3	WSW	564.5	1381.0	14.1	45.0	20.5	past 10 days.
July 24, 1913	753.91		89.4	SSW	348.5		2.4			
July 27, 1913	756.90		88.2	WNW	218.0		13.8			Very heavy rains during the
July 28, 1913	753.74	Decreasing	91.0	WSW	476.0	1742.0	5.0	72.7	14.5	past 18 days.
July 29, 1913	750.80		90.1	SW	1048.0		53.9			
Aug. 3, 1913	757.80		86.1	SW	290.0		1.3			Heavy typhoon weather for
Aug. 4, 1913	757.12	Decreasing	93.0	SW	332.0	830.5	23.0	24.7	23.6	preceding three weeks, but only
Aug. 5, 1913	756.36		89.7	SW	208.5		0.4			partly cloudy with occasional showers beginning with Aug. 2.
Aug. 17, 1913	758.60		85.9	SE	109.5		9.7			
Aug. 18, 1913	759.26	Variable	87.7	Variable	94.0	367.5	1.8	54.7	39.5	
Aug. 19, 1913	757.26		86.0	SW	164.0		43.2			

TABLE I.—*Continued.*

Date.	Pressure (Mean), (Mm.).	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), %	Wind.			Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.)	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10[12]).	Weather Remarks.
				Prevailing Direction.	Total Movement (Km.).	Total Movement During the Three Days, (Km.).				
Aug. 24, 1913	757.51		88.6	Variable	246.5		16.5			Partly fair during Aug. 22 and
Aug. 25, 1913	758.77	Variable	88.8	SW	335.5	979.0	5.3	29.1	19.6	23. Heavy showers during night
Aug. 26, 1913	757.34		89.0	SW quad.	397.0		7.3			of 24th.
Aug. 31, 1913	757.98		84.3	SW	224.0		19.2			Fair during the day time of
Sept. 1, 1913	755.98	Decreasing	83.0	W quad.	149.0	652.0	0.8	25.1	49.7	Aug. 31, but heavy showers dur-
Sept. 2, 1913	753.53		85.9	SW	279.0		5.1			ing the night.
Sept. 7, 1913	758.48		84.0	WSW	271.0		....			Occasional heavy showers dur-
Sept. 8, 1913	757.74	Decreasing	81.2	WSW	530.0	1472.0	....	105.8	17.3	ing the period from Sept. 4 to
Sept. 9, 1913	756.39		88.8	SW, WSW	671.0		105.8			Sept. 8.
Sept. 21, 1913	757.76		82.9	W quad.	117.5		0.8			Light afternoon showers for the
Sept. 22, 1913	758.62	Increasing	82.0	SW quad.	159.0	392.0	....	0.8	53.4	period from Sept. 18 to Sept. 21
Sept. 23, 1913	759.83		87.8	SW quad.	115.5		....			inclusive.
Sept. 28, 1913	759.57		82.4	W quad.	136.0		....			Light showers during the pre-
Sept. 29, 1913	759.16	Decreasing	84.8	W	113.0	390.5	5.1	12.0	52.0	ceding week with no heavy rains.
Sept. 30, 1913	758.46		82.4	W quad.	141.5		6.9			
Oct. 12, 1913	753.95		88.6	N quad.	124.0		4.7			Occasional heavy showers with
Oct. 13, 1913	751.87	Variable	91.6	SSW	459.0	1067.0	13.1	66.3	17.1	considerable fine drizzling rain
Oct. 14, 1913	752.36		93.3	SSW	484.0		48.5			intervening during Oct. 10 and 11.

TABLE I.—*Continued.*

Date.	Pressure (Mean), (Mm.).	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), (%).	Wind.			Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10¹²).	Weather Remarks.
				Prevailing Direction.	Total Movement, (Km.).	Total Movement During the Three Days, (Km.).				
Oct. 19, 1913	760.24	Variable	81.5	E quad	70.5		0.3			Only very light showers since the 14th. Fair most of the time with light winds.
Oct. 20, 1913	760.60		86.3	SW, NW	55.0	201.5	0.5	0.8	77.1	
Oct. 21, 1913	760.24		83.7	W, WSW	76.0		....			
Oct. 26, 1913	760.28		76.5	NNW, SSW	94.0		....			Fair for the past week with a few light showers and light winds.
Oct. 27, 1913	760.03	Variable	78.4	W	112.0	264.5	....	....	92.2	
Oct. 28, 1913	760.88		81.8	W quad.	58.5		....			
Nov. 2, 1913	760.35		82.4	NW	95.0		....			Fairly heavy local showers during the past week.
Nov. 3, 1913	760.58	Increasing	88.1	W quad.	81.0	248.5	4.6	7.2	49.9	
Nov. 4, 1913	760.61		93.1	NNE	77.5		2.6			
Nov. 9, 1913	760.64		77.5	NE	167.5		....			Considerable rain during the preceding week, but fair on the 9th.
Nov. 10, 1913	760.60	Decreasing	87.2	N quad.	99.0	414.0	3.4	3.4	75.4	
Nov. 11, 1913	760.59		77.6	NE, WSW	147.5		....			
Dec. 7, 1913	762.08		78.5	NE	106.5		....			Light showers on the 5th and 6th. Fair on the 7th.
Dec. 8, 1913	761.22	Variable	79.2	ENE	119.0	373.0	1.8	2.4	59.1	
Dec. 9, 1913	761.39		82.6	NNE	147.5		0.6			

TABLE I.—*Continued.*

Date.	Pressure (Mean), (Mm.).	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), (%)	Wind. Prevailing Direction.	Wind. Total Movement, (Km.)	Wind. Total Movement During the Three Days, (Km.).	Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.)	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10^{12}).	Weather Remarks.
Dec. 14, 1913	760.13		77.9	NNE	171.0		....			Afternoon showers during the preceding week. Showers during night of 13th.
Dec. 15, 1913	759.85	Variable	83.3	WNW, WSW	80.0	309.0	....	9.3	137.9	
Dec. 16, 1913	760.50		89.5	Variable	58.0		9.3			
Dec. 22, 1913	760.02		79.8	WNW, SE	114.5		....			Fair during the 21st and 22nd. Cloudy during morning of 23rd.
Dec. 23, 1913	760.27	Increasing	86.3	SE	71.5	270.0	3.0	3.0	70.8	
Dec. 24, 1913	760.33		80.2	W quad.	84.0		....			
Jan. 4, 1914	764.59		74.6	E quad.	155.5		....			Fair during the four days immediately preceding.
Jan. 5, 1914	764.34	Decreasing	78.2	E, SE	188.5	538.0	....	....	89.1	
Jan. 6, 1914	764.19		76.2	SE	194.0		....			
Jan. 18, 1914	762.93		72.0	E quad.	169.0		....			Fair.
Jan. 19, 1914	761.90	Decreasing	79.1	E quad.	161.0	531.0	....	....	145.1	
Jan. 20, 1914	761.57		76.1	ESE	201.0		....			
Feb. 1, 1914	761.02		75.0	SE quad.	195.0		....			Fair.
Feb. 2, 1914	761.12	Increasing	76.3	ESE	128.0	427.0	....	....	122.4	
Feb. 3, 1914	761.66		79.2	SE	104.0		....			

TABLE I.—*Continued.*

Date.	Pressure (Mean), (Mm.).	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), (%).	Wind.			Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10[12]).	Weather Remarks.
				Prevailing Direction.	Total Movement, (Km.).	Total Movement During the Three Days, (Km.).				
Feb. 15, 1913	763.23		74.2	ESE	195.5		....			Fairly heavy showers during the period from Feb. 13 to Feb. 15.
Feb. 16, 1914	763.09	Decreasing	66.4	E quad	229.5	624.5	....	....	94.1	
Feb. 17, 1914	762.96		70.2	ESE	199.5		....			
Mar. 8, 1914	759.23		69.6	ESE	264.5		....			Exceptionally clear and warm during the last few days.
Mar. 9, 1914	759.31	Increasing	70.1	ESE	228.5	755.0	....	....	108.6	
Mar. 10, 1914	759.71		70.6	SE	262.0		....			
Apr. 21, 1914	761.88		71.6	SE	218.5		....			
Apr. 22, 1914	760.51	Decreasing	69.6	NNE, WSW	189.0	610.5	1.5	1.5	68.9	
Apr. 23, 1914	759.87		73.4	SE	203.0		0.3			
May 6, 1914	758.36		644.	SE	206.5		....			
May 7, 1914	759.37	Variable	66.9	SE, W	226.5	630.0	....	2.0	70.1	
May 8, 914	758.96		71.5	E quad.	197.0		2.0			
May 24, 1914	758.52		74.9	WSW	137.5		....			
May 25, 1914	758.66	Variable	78.3	Variable	155.5	499.0	2.3	2.3	97.4	
May 26, 1914	757.02		78.9	WSW	206.0		....			
June 7, 1914	759.41		79.5	Variable	98.0		....			Very heavy shower during the early evening of the 8th.
June 8, 1914	758.97	Decreasing	78.6	WNW	141.0	380.5	6.4	6.4	63.5	
June 9, 1914	758.92		77.7	W	138.5		....			

Table I.—*Continued.*

Date.	Pressure (Mean), (Mm.).	Nature of Variation of Pressure During the Three Days.	Humidity (Mean), (%).	Wind.			Rain, 24 Hours Beginning Midnight, (Mm.).	Rain for the Three Days, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams $\times 10^{12}$).	Weather Remarks.
				Prevailing Direction.	Total Movement, (Km.).	Total Movement During the Three Days, (Km.).				
June 21, 1914	758.66		85.0	Variable	83.0		....			Very heavy rain during the period from June 18 to June 21.
June 22, 1914	758.84	Variable	83.8	Variable	103.5	374.0	....	....	54.5	
June 23, 1914	758.82		79.5	SE	187.5		....			
July 8, 1914	755.48		90.5	SW quad.	282.0		21.3			Just clearing off after almost a week of typhoon weather with very heavy rain and winds. No rain during test.
July 9, 1914	756.80	Increasing	84.7	WSW	327.0	1288.0	0.9	22.2	19.2	
July 10, 1914	757.05		81.6	WSW	679.0		....			
								Mean	63.4	

The mean value of the radium equivalent per cubic meter of the atmosphere at Manila, as given by the 29 observations recorded in Table I., is 63.4×10^{-12} grams. If we take the mean of all our observations for Manila we obtain a value of 76.7×10^{-12} grams, which is probably nearer the true value. Any mean determined as above, however, is subject to wide fluctuations, since the variation with weather conditions is so great that a wide range of values, even of a large number of determinations, might be obtained by bunching the observations during a definite season of the year. A better idea of the average value is obtained by taking the mean of the monthly means for an entire year. In Table II. the results by months are given, the numbers in brackets in the last column indicating the number of individual observations entering into the value for each month. The mean of the monthly means has a value of 71.0×10^{-12} grams.

TABLE II.

Annual Variation of the Radium Emanation Content.

Month.	Pressure (Mean), (Mm.).	Humidity (Mean), %	Wind, Total Movement for Month, (Km.).	Rain, (Total), (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, Grams $\times 10^{12}$.
July, 1913	756.26	86.2	10,374.6	570.6	23.6 [3][1]
Aug., 1913	756.93	87.0	8,843.5	349.1	27.6 [3]
Sept., 1913	757.67	85.4	8,664.5	365.5	43.1 [4]
Oct., 1913	758.51	83.4	4,152.0	119.7	62.1 [3]
Nov., 1913	761.04	81.7	3,667.5	31.1	62.7 [2]
Dec., 1913	761.32	80.9	4,021.0	37.8	89.3 [3]
Jan., 1914	763.24	76.2	4,925.5	3.5	117.1 [2]
Feb., 1914	762.26	73.8	5,255.5	7.3	108.3 [2]
Mar., 1914	760.77	68.6	6,344.0	6.1	108.6 [1]
Apr., 1914	760.17	70.8	5,921.0	53.4	68.9 [1]
May, 1914	758.42	72.6	6,137.5	84.0	83.7 [2]
June, 1914	757.62	81.7	6,714.0	367.9	59.0 [2]
July, 1914					19.2 [1]
Mean of monthly means					71.0

The following average results have been obtained for the radium-emanation content by Eve[2] at Montreal, Satterly[3] at Cambridge, and Ashman[4] at Chicago:

Eve .... 60×10^{-12} gram Ra. per cubic meter.
Satterly .... 105×10^{-12} gram Ra. per cubic meter,
Ashman .... 96×10^{-12} gram Ra. per cubic meter.

[1] Number in bracket shows the number of observations entering into the monthly mean.
[2] Phil. Mag., *16*, 622, 1908.
[3] Am. Journ. Sci., 119, 1908.
[4] Phil. Mag., *20*, 1, 1910.

Eve and Satterly used the charcoal-absorption method; Ashman the condensation method. Eve's value is the mean of the monthly means for a period from July to April, Satterly's value is the mean of individual observations taken during the months of March to August, inclusive, while Ashman's is the mean of six determinations, the time of year not specified.

In comparing our results with those of Eve and Satterly it should be remembered that they both assumed that all the emanation was obtained from their standard by bubbling air through the cool solution, and consequently did not use in their calculations any factor corresponding to α. If their results are subject to a correction of approximately the value which we found for α, then our mean is considerably greater than that of Eve for Montreal and slightly less than that of Satterly for Cambridge. No great importance is here attached to the fact that our mean value for Manila seems to be nearer to that for Cambridge than Montreal, except in so far as it may throw light on the question as to what extent the value of the emanation content is dependent on the distance that the air tested has traveled over land or water.

Variation of the Emanation Content with Meteorological Conditions.

From a study of the individual observations of Table I. a distinct correlation is observed between the values of the emanation content and certain of the meteorological factors. For each observation of the emanation content we have given the meteorological data for three days, including the two days during which the observation was taken and the preceding day. That a distinct relation exists between the emanation content, the rainfall and the wind movement is readily seen. In every case a period of heavy rains accompanied by high winds corresponds to a low value of the radium-emanation content, while fair weather with light winds gives correspondingly high values. A good idea of the magnitude of this variation is obtained from the ratio of the maximum to the minimum, which has approximately the value of ten to one. Since in Manila the temperature is fairly constant throughout the year the variations in rainfall and wind movement must necessarily be the chief causes of the wide difference in the values of the atmospheric radioactivity. Which one of the two factors is the more important is somewhat doubtful, since they are generally so correlated that it is difficult to determine exactly what part of the total effect is due to either one.

Not only is there a close relation between the emanation content, the rainfall and the wind movement for the individual observations, but also for the means for the months, as shown in Table II. Taken in conjunction with Table I. it is seen that the observations scattered throughout the month are fairly typical for the entire period. This is especially true for the months in which three or more observations were taken. It is to be regretted that during certain months the rush of other work prevented our taking as large a number of observations as we should have liked. The month of January gives the highest value for the activity, the minimum value for the rainfall, and a low value for the wind movement. July shows the lowest activity, the maximum rainfall and the highest wind movement. A similar relation exists for almost every month of the year. The month of June from Table II. would appear to be an exception, but by referring to Table I. it is seen that the two observations for the month were taken at periods of very little rainfall and comparatively low wind movement, and consequently are not typical of the entire month as far as rain and wind is concerned.

One other factor, namely the direction of the wind, may possibly have an appreciable effect on the value of the emanation content. The location of Manila is such that for the greater part of the year the prevailing wind is from off the sea. Even those which are apparently land breezes, with the exception of those from the northeast quadrant, have at the most passed but a comparatively short distance over land. During the rainy season the prevailing winds are westerly and the radium-emanation content is low, but just as soon as the rainfall and the wind movement decrease the radioactivity increases, even though the direction of the wind remains the same. Certain observations, however, seem to indicate that the highest values are obtained on days when the air currents are from the land, but the results are so uncertain as to practically prohibit the drawing of any definite conclusions. A more detailed discussion of this point will be given in connection with certain observations in which the factor of rainfall is absent.

Diurnal Variation of the Radium-Emanation Content of the Atmosphere.

In our earlier work on the subject we had noticed that there seemed to be a large variation between consecutive observations taken over different parts of the day. No definite attempt had been made, however, to determine accurately the diurnal variation.

Several observers have investigated the question of the daily variation of the radioactivity of the atmosphere by the active-deposit method. Simpson[1] in Lapland made four determinations a day (3–5 A.M., 10–12

[1] Phil. Trans. Roy. Soc., A-V, CCV., 61, 1905.

A.M., 3–5 P.M., and 8.30–10.30 P.M.) for a period of almost a year. His results showed a maximum for the early morning hours and a minimum about noon, the ratio of the maximum to the minimum being approximately 3 to 1. Gockel[1] at Freiburg likewise found an increase in the early morning and at times a depression about noon. Blackwood[2] at Manila found a maximum about 4 A.M. and a minimum around 6 P.M., the ratio of the means being approximately seven to one. These three observers obtained the active deposit on a negatively charged wire stretched horizontally in free air.

In order to eliminate the variable factor of wind force, Dike[3] used a modification of the above method. The air to be tested was drawn at a definite rate through a wooden pipe over one end of which was fitted a piece of wire gauze. The gauze was well insulated and charged to a high negative potential. The radioactive substances were collected on the gauze and tested in the usual manner. As a mean of six sets of observations, each extending over approximately twenty-four hours, a decided maximum was found in the early morning which decreased to a strong minimum during the early evening. The maximum had a value approximately eighteen times the minimum.

Hess[4] using a method somewhat similar to Dike's found an early morning maximum which decreased to a minimum about 11.30 A.M. rising again to a secondary maximum at 4 P.M. and then rapidly falling to a decided minimum around 6 P.M. The greatest variation found was only about 20 per cent. from the mean value.

From the work of these observers it seems quite certain that the emanation content is considerably greater in the early morning hours than during the late afternoon, even for widely scattered points on the earth's surface. The active-deposit method fails, however, to throw much light on the cause of this variation. Simpson drew the following conclusions from his results:

1. For the whole year temperature has a marked effect, but very little effect during any one month.

2. The radioactivity increases with increase in the humidity and decreases with decreasing humidity.

3. The radioactivity is greater with a falling barometer than with a rising barometer, although the activity is not necessarily higher with a low barometer than with a high one.

4. The radioactivity is greater for winds from the land than for winds from the sea.

[1] Phys. Zeit., 5, 591, 1904.
[2] Phil. Journ. Sci., IX–A, No. 4, 1914.
[3] Terr. Mag., 125, 1906–07.
[4] Sitz. d. k. Ak. d. Wiss., *119*, 145, 1910.

Hess likewise concluded that a decrease in pressure was accompanied by an increase in the active deposit, and vice versa.

The question of whether all these factors directly affect the emanation content or whether certain of them cause merely a variation in the amount of active deposit collected led us to take some observations by the charcoal-absorption method. Since the object of these tests was to determine the causes rather than the nature of the diurnal variation no attempt was made to take observations of extremely short duration.

The first series of tests were taken with the object of determining the relation existing between the emanation content for day and night periods. The day tests extended in every case from 6.30 A.M. to 6.30 P.M., and the night from 6.30 P.M. to 6.30 A.M. The corresponding day and night observations were always made within the 24 hour period in order that the individual readings might have a direct comparative value. The results are given in Table III. With one exception all the night obser-

TABLE III.

Diurnal Variation of the Radium Emanation Content.

Day determinations. Time.—6.30 *A.M. to* 6.30 *P.M.*

Night Determinations. Time.—6.30 *P.M. to* 6.30 *A.M.*

	Date.	Pressure Variation.	Humidity (Mean), (%).	Wind.		Rain for 12 Hours, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10^{12}).
				Prevailing Direction.	Total Movement for the 12 Hours, (Km.).		
Day Determinations	Dec. 29, 1913.....	Variable	67.5	Variable	97.0	....	68.3
	Jan. 2, 1914......	"	65.5	N	125.5	....	51.8
	Jan. 12, 1914.....	"	70.6	Variable	118.5	....	66.1
	Jan. 14, 1914.....	"	76.4	N	85.0	....	59.5
	Jan. 16, 1914.....	"	75.4	Variable	65.0	....	36.2
	Jan. 27, 1914.....	"	63.4	Variable	127.5	....	68.0
	Jan. 29, 1914.....	"	68.2	Variable	99.0	....	68.0
	Feb. 24, 1914.....	"	68.4	W. quad.	118.0	....	85.5
	Mean....................................				104.4	....	62.9
Night Determinations	Dec. 29–30, 1913..	Variable	80.8	NE	75.0	....	66.9
	Jan. 2–3, 1914....	"	80.9	N	27.5	....	52.4
	Jan. 12–13, 1914..	"	89.1	Calm	29.5	....	165.0
	Jan. 13–14, 1914..	"	85.1	Calm	40.5	....	111.3
	Jan. 15–16, 1914..	"	86.0	Calm	21.0	....	133.8
	Jan. 26–27, 1914..	"	79.0	ESE	65.0	....	108.3
	Jan. 28–29, 1914..	"	78.2	ESE, Calm	48.0	....	174.9
	Feb. 24–25, 1914..	"	84.0	SE	32.0	....	173.5
	Mean....................................				41.1	....	123.3

TABLE IV.

Diurnal Variation of the Radium Emanation Content.

Day Determinations. Time.—11 A.M. to 5 P.M.

Night Determinations. Time.—11 P.M. to 5 A.M.

	Date.	Pressure Variation.	Humidity (Mean), (%).	Wind. Prevailing Direction.	Wind. Total Movement for the 6 Hours, (Km.).	Rain for 6 Hours, (Mm.).	Radium Emanation per Cu. M. Expressed in its Radium Equivalent, (Grams × 10^{12}).
Day	Feb. 27, 1914	Decreasing	57.9	E. quad.	98.0	.....	45.8
Day	Mar. 25, 1914	Decreasing	42.9	Variable	117.5	.....	26.1
Day	Mar. 30, 1914	Decreasing	53.3	WSW, SE	73.5	.....	34.1
	Mean				96.3	.....	35.3
Night	Feb. 26–27, 1914	Decreasing	90.0	Calm	12.5	.....	132.0
Night	Mar. 24–25, 1914	Decreasing	78.0	Calm	19.0	.....	116.7
Night	Mar. 30–31, 1914	Variable	81.0	ESE, Calm	8.0	.....	104.5
	Mean				13.2	.....	117.7

vations gave higher values than those for the day. The ratio of the mean value for the night to the mean for the day is 1.95.

A few observations were also taken in which the time of collecting was shortened to 6 hours, the results of which are given in Table IV. Since all the observers using the active-deposit method seem to agree that the radioactivity reaches a maximum in the early morning hours and falls to a minimum some time during the afternoon we chose for our 6-hour observations the intervals from 11 A.M. to 5 P.M., and 11 P.M. to 5 A.M., respectively. As in the 12-hour observations corresponding day and night tests were always taken within the 24-hour period. Again it is seen that the night values are all consistently higher than the day values, the ratio of the mean for the night to the mean for the day being 3.31.

From the above results it appears quite evident that there is a decided daily variation of the emanation content of the atmosphere at Manila. Moreover, the variation found by the charcoal-absorption method seems to be in good agreement with the results obtained by the active-deposit method. This would seem to indicate that the active-deposit method is fairly reliable for determinations of the variation at any given locality. That observations taken at widely different altitudes would possess the same comparative value does not necessarily follow, since changes in pressure, nucleation, etc., would in that case introduce undetermined factors. The charcoal-absorption method, moreover, lends itself much more readily to a study of the causes of the existing variation.

The data in Tables III. and IV. show quite conclusively the effect of the total wind movement. The observations were all taken during the dry season and the factor of rainfall is entirely eliminated. Without exception the total wind movement for the day observations was considerably greater than for the corresponding night determinations. For the 12-hour tests the total wind movement for the day is 2.54 times that for the night. This is approximately of the same value as the ratio for the radium-emanation content. The fact that the two relations happen to have nearly the same numerical value has probably no direct significance, but it certainly shows that the question of atmospheric circulation, independent of rainfall, has a very decided influence on the variation of the emanation content. For the 6-hour tests the total wind movement for the day is approximately 7.32 times that for the night, or roughly twice the ratio for the emanation content. From these two series of observations we are able to obtain a fair idea concerning the extent to which the high wind movement is responsible for the decrease of the radioactivity of the air. A study of Table I. in the light of the above results leads to the conclusion that at Manila the fluctuation in the total wind movement is probably responsible for almost one half of the observed variation of the radium-emanation content.

It was also thought probable that the direction of the wind during the different parts of the day might have considerable influence on the above variation. A careful study of the hourly data for the periods of observation fails to show, however, any definite relation. Consequently, we are led to the conclusion that for Manila the important factor, in regard to the wind, is not whether it has passed for a long distance over land or sea, but almost entirely one of rapidity of circulation. That the same relation would be found to exist for a point differently situated with regard to water and land does not necessarily follow.

If the circulation of the air rather than its direction is the important question then evidently the decrease of the emanation content due to the wind may be explained as due almost entirely to the mixing of air of low and high emanation content. Moreover, it seems as if all air in rapid circulation is such a mixture, regardless of whether it has passed for a long distance over water or not. This is easily explained if we assume that the air in rapid circulation over land is a mixture of air of high and low altitude, since without doubt the emanation content of the atmosphere decreases with altitude. Only when the air is very still does the amount of emanation it contains begin to approach that exhaled from the ground for the given locality. We are perfectly cognizant that the above explanation may be applicable only to our particular location.

Manila is so situated that practically all land breezes have passed within a short distance over low mountain ranges, which will tend to give a mixture such as assumed above.

The correlation between the radioactive constituents of the atmosphere and the temperature, humidity, and pressure has been the subject of much investigation, but the results are decidedly discordant. For this reason we thought it advisable to include in our tables the data showing the variation in humidity and barometric pressure. The temperature for Manila is so nearly constant for the year that it has been omitted. A careful study of all our results fails to give any convincing evidence that the emanation content is directly dependent to any appreciable extent on either the humidity or the pressure. In the course of this study we plotted the pressure-time curves for the results given in Tables III. and IV., but were absolutely unable to discover any definite relation for either an increasing or decreasing pressure. Since most of the previous investigations on this phase of the question have been made by the active-deposit method it seems probable that whatever relation was found to exist was due to a variation of the collecting distance rather than to any actual change in the emanation content. That the results obtained by the active-deposit method are influenced by the humidity of the air has been shown by the work of Simpson, Blackwood and others.

Supply of Radium to the Air from Soil Gas.

Exhalation.—It has been fairly conclusively shown that the earth's crust is the source of the radioactive substances in the atmosphere. A large amount of work by numerous observers has also demonstrated that the radioactive substances are fairly uniformly distributed throughout the surface of the earth. Consequently it is to be expected that more or less emanation is being exhaled from the earth's surface at every point. That the rate of exhalation will vary with the physical and chemical conditions of the surface is to be expected. In order to determine the effect of weather conditions on the rate of exhalation we made a few tests on the emanation exhaled from the surface of the ground at Manila. As far as we have been able to learn the only direct tests made on the exhalation are those of Joly and Smyth at Dublin. Joly and Smyth,[1] working together, made determinations at two different locations, obtaining as mean values 839×10^{-12} and $4,087 \times 10^{-12}$ curie per square meter respectively. Smyth,[2] independently, obtained as a mean of 98 observations, taken at a point about 12 meters from the Geological Laboratory of Trinity College, a value of $2,637 \times 10^{-12}$ curie.

[1] Proc. Roy. Dublin Soc., XIII., 148, 1911.

[2] Phil. Mag., 24, 632, 1912.

In order to obtain tests on the emanation exhaled a collecting system somewhat similar to that used by Joly and Smyth was devised. The collector consisted of a cylindrical vessel, 20 cm. in diameter and about 30 cm. deep, open at one end and closed, with the exception of a 4 cm. neck, at the other. The open neck was fitted with a rubber stopper from which portions had been cut to allow the inflow of air. Through the stopper and along the axis of the cylinder was passed a brass tube which supported at its lower end a brass disk, 19.4 cm. in diameter. This disk hung about 1.5 cm. above the open end of the cylindrical vessel. When a test was to be made the collector was placed on a level portion of the campus about 7 meters from the north wall of the Physics Laboratory, and pressed very lightly into the ground. The brass tube was then connected, through the charcoal tubes, to a motor driven pump. When the pump was started the air was drawn down into the cylinder through the openings in the rubber stopper, passing over the edges of the brass disk and along the surface of the ground and then up through the brass tube to the charcoal tubes. It was assumed that by this means practically all the emanation exhaled from the confined portion of the surface would be swept into the collector and carried to the charcoal (see Fig. 1).

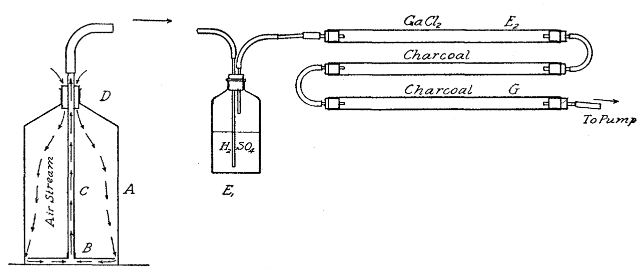

Fig. 1.

A, brass cylinder; *B*, brass disk; *C*, brass tube; *D*, rubber stopper with portions cut out; E_1 and E_2, drying system; *G*, coconut charcoal tubes.

The rate of flow of the air stream was measured by a gauge and was kept fairly constant at about 0.5 liter per minute, the period of collecting being generally one hour. Two charcoal tubes, each containing 70 grams of charcoal, were connected in series. Judging from our preliminary experiments two such tubes should absorb practically all the radium emanation passing through them during a test. The emanation collected in the charcoal was tested in the usual manner.

The results are given in Table V. and are expressed in terms of the radium equivalent of the emanation exhaled in one hour from a surface of one square meter. Although the number of our observations is limited,

TABLE V.

Radium Emanation Exhaled from the Ground.

Date.	Radium Emanation in Air Exhaled per Hour from 1 Square Meter of Ground Surface Expressed in its Radium Equivalent, Grams $\times 10^{12}$.	Weather Remarks.
June 18, 1914..	1642.0	Dry, except for very light showers during the past two weeks.
June 20, 1914..	785.2	Fairly heavy rain during day and night of June 19th. Light rain at intervals during morning of 20th. Light rain during time of collecting.
July 9, 1914...	694.0	Heavy rain and high winds for the last 6 days. A very heavy shower fell for about 15 minutes during the time of collecting.
Aug. 18, 1914..	1122.3	Fair for last three days. A few light showers during the nights. No real heavy rains for almost three weeks.

nevertheless, they show quite plainly the relation of the exhalation for the dry and wet seasons. The observation of June 18 was taken just before the breaking up of the dry season, while that of July 9 followed a week of exceptionally heavy rain. The result of June 18 is 2.37 times that of July 9. This shows approximately to what extent the radioactivity of the air is apt to vary due to rain alone. This is absolutely independent of the question whether rain falling through the air carries down with it any of the emanation. Whether the variations in the wind force effects the rate of exhalation from the ground is still an open question. Since in the above tests the air current remains constant, they throw no light on this phase of the question. The number of our tests is not sufficient to give more than a general idea of the mean value of the radium emanation exhaled at Manila, although our results are in good agreement with those of Joly and Smyth. Without doubt the months of the dry season would show a value as great or greater than that of June 18.

Emanation Content of Soil Gas.—Since the effect of rain is to decrease the rate at which the radioactive emanations are exhaled from the surface of the ground there should be a corresponding increase in the emanation content of the soil gas below the surface. Joly and Smyth found this to be the case for their observations at Dublin, but Satterly[1] failed to discover any correlation between the fluctuations of the emanation content and weather conditions, except for one pipe which always gave low results after rain and high results in fine weather. It was this apparent discrepancy in the results of experimenters working under practically

[1] Proc. Cambridge Phil. Soc., *16*, 514, 1912.

identical conditions which led us to extend our work so as to include a few observations on soil gas.

To obtain the ground air we made three pipes of brass tubing, 9 mm. inside diameter, and 130 cm., 75 cm., and 37 cm. in length. One end of each pipe was fitted with a pointed brass plug. For a length of about 6 cm. from the closed end, holes, sloping slightly downward, were drilled. The pipes were then driven into the ground at a distance of about two meters from a window of the Physics Laboratory to such depths that the mean positions of the portion in which the holes were drilled were approximately 120, 70, and 30 cm., respectively, below the surface of the ground.

The air to be tested was pumped directly into an accurately calibrated gas bottle of one-liter capacity, care being taken in every case to pump out all the air in the pipes previous to taking an observation. Connected in parallel with the gas bottle was a manometer by means of which we could be absolutely certain that the bottle was always filled with gas at atmospheric pressure. After a sufficient time had elapsed to permit the decay of the thorium emanation the air collected was passed into the ionization chamber and tested in the usual manner.

TABLE VI.

Radium Emanation in Ground Air.

Date.	Radium Emanation per Cubic Meter of Ground Air Expressed in its Radium Equivalent, (Grams $\times 10^{12}$).			Weather Remarks.
	Pipe No. 1, Depth 30 Cms.	Pipe No. 2, Depth 70 Cms.	Pipe No. 3, Depth 120 Cms.	
June 15, 1914..			311.5	Dry, except for very light showers, during the period from June 5 to June 15.
June 16, 1914..	32.9	209.1		Generally fair with no rain.
June 17, 1914..	29.4	208.1	269.7	Continued fair weather.
June 19, 1914..	69.5	244.2	284.1	Fairly heavy rain accompanied by high wind during the night of June 18 and continuing during June 19.
June 22, 1914..	44.0	261.6	319.0	Heavy rains up till afternoon of June 20, followed by light showers during June 21 and June 22.
June 24, 1914..	40.7	249.2	314.0	Fair during the last two days. A fairly heavy shower for about 15 minutes when collecting air from Pipe No. 3.
July 14, 1914..	64.0	297.8	321.5	Very heavy rains and high winds during the period from July 4 to July 10 Intervening four days partially fair.
Aug. 19, 1914..	35.1	274.2	314.0	Fair the last four days. A few light showers during the nights. No real heavy rains for almost three weeks.

The results of the observations are given in Table VI. Generally about two hours elapsed between the different observations taken on any given day. It is seen that the values for the 30 cm. pipe fluctuate decidedly with the amount of rainfall, being greater immediately after a period of heavy rains. The 70 cm. pipe shows a slight increase for the rainy weather while the 120 cm. pipe is but little affected. All the results tend to show, however, that the effect of rain is to stop up the soil capillaries, thus retarding the rate of escape of the ground gas, and thereby permitting the emanation in the soil air to approach nearer its equilibrium value.

Summary of Results.

1. The variation of the amount of radium emanation in the atmosphere at Manila has been determined for a period of about thirteen months. The annual and diurnal variation has been studied in connection with the principal meteorological factors. The effect of weather conditions on the rate at which radium emanation is exhaled from the surface of the ground has been investigated with the object of determining its connection with the emanation content of the atmosphere. The relation between the rate of exhalation and the radioactivity of soil gas at different depths has also been investigated.

2. The variation of the radium-emanation content of the atmosphere has been found to follow quite closely the variations in rainfall and wind movement. The ratio of the maximum to the minimum for the year was found to be approximately as 10 to 1. The mean of the monthly means gives for the radium equivalent of the emanation per cubic meter of air a value of 71.0×10^{-12} grams. The month of January shows the highest monthly mean for the radium-emanation content, the minimum value for the rainfall, and a low value for the total wind movement. The month of July gives the lowest monthly mean for the emanation content, the maximum value for the rainfall, and the highest total wind movement. Every other month of the year shows a very similar relation. No direct connection has been discovered between the emanation content and atmospheric pressure or humidity. The effect of the direction of the wind seems at best very indefinite.

3. A decided diurnal variation has been found to exist, the emanation content being considerably greater during the night than during the day. Observations for the interval from 11 P.M. to 5 A.M. gave a mean value 3.31 times greater than the mean value for the interval from 11 A.M. to 5 P.M. This variation has been found to be closely related to the variation in the total wind movement during the period, a high value of the wind movement corresponding to a low value of the emanation content.

4. The rate at which radium emanation is exhaled from the surface of the ground shows a decided decrease after periods of heavy rain. This decrease has been found in some cases to be almost 60 per cent. of the rate of exhalation for fair weather.

5. The radium-emanation content of soil gas has been determined for depths of 30, 70, and 120 cm., respectively, and the variation with weather conditions studied. The variation in the radioactivity of the gas from the 30 cm. pipe was found to follow closely the variation in the emanation exhaled, a decrease in the exhalation resulting in a corresponding increase in the emanation content of the ground gas. The 70 cm. and 120 cm. pipes showed only slight variations with the weather conditions. The average value of the emanation content for the gas collected from the 120 cm. pipe was found to be 304.5×10^{-12} grams per liter, or over 4,000 times the mean value for atmospheric air. The mean value for the 30 cm. pipe was only about one seventh that for the 120 cm. pipe.