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A SIMPLE AMD INEXPENSIVE SYSTEM FOR MEASURING CONCENTRATIONS OP ATMOSPHERIC RADON-222

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#### A SIMPLE AND INEXPENSIVE SYSTEM FOR MEASURING CONCENTRATIONS OF ATMOSPHERIC RADON-222

Gary E. Jones and Lowell M. Kleppe

June 27, 1966

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Contents

Abstract	v
I. Introduction	1
II. Radon Collection and Analysis	
A. Sampling and Analysis Techniques	2
1. Sampling System Components	2
2. Sampling Procedure	4
3. Calibration $\ldots$ $\ldots$ $\ldots$ $\ldots$ $\ldots$ $\ldots$	5
4. Performance and Sources of Error	6
B. Data and Discussion	2
1. Data from the San Francisco Bay Area	2
2. Other Data	3
III. Summary	7
Acknowledgments	7
References	8

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- v -

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#### ABSTRACT

A radon-222 sampling system has been designed and built which uses a charcoal adsorption technique for collecting and analyzing that gas in a simple, inexpensive, reliable way. Results of measurements made with the system, although only preliminary, are in agreement with data obtained by other investigators.

#### I. INTRODUCTION

-1-

Radon-222 (half-life 3.83 days) is the first daughter product of radium-226 in the uranium-238 decay chain and, as such, is distributed throughout the natural environment. Various investigators since the early part of this century have demonstrated the ubiquity of radon, and their measurements have indicated the variability of concentrations of the noble gas both from point to point near the surface of the earth and with time at given locations on the earth. Israël and Krebs have presented a summary of such information accumulated over the past several years.<sup>1</sup>

Our present interest in radon-222 has arisen from attempts to find geographical regions with low inherent terrestrial radiation fields. Low background locales provide opportunities for measuring internal background levels in radiation-detection instruments and for the direct measurement of cosmic ray components within the natural environment. In areas where the underlying rock and soil contain little radioactivity, radon and its daughters in the atmosphere may contribute significantly to the natural radiation field; knowledge of radon concentrations and their variations within such regions is requisite, then, to permit evaluation of information acquired from radiological measurements made in those areas. This report describes a system used for collecting  $Rn^{222}$  and for determining concentrations of the gas at several points of interest in California. Although measurements have not been extensive to date, they have permitted a qualitative description of the contribution of  $Rn^{222}$  and its daughters to radiation levels within those regions.

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#### II. RADON COLLECTION AND ANALYSIS

#### A. Sampling and Analysis Techniques

The traditional method of collecting radon is to draw air through activated coconut charcoal chilled to the temperature of dry ice (-78°C). At this low temperature, the charcoal has been shown to adsorb radon very efficiently.<sup>2,4</sup> Probably the most sensitive method of determining the amount of Rn<sup>222</sup> collected is to desorb the gas at 300°C, transfer it to a scintillation chamber, and count the individual a particles produced in the decay of the radon and two of its daughters. The sensitivity of such systems has been reported as being four to five counts per minute per pico-curie (pCi) of Rn<sup>222</sup> being counted.<sup>3</sup> The purposes of the program to be described in this report, however, did not require such a sensitive system, nor did time permit the construction of equipment necessary for transferring and counting directly the radon collected.<sup>4</sup> Therefore, while the collection method was the traditional one, the manner of determining the amount of Rn<sup>222</sup> collected was less sensitive than that described above.

#### 1. Sampling System Components

The collection system built for measurements to satisfy our interests consisted of a linear array of elements through which air was drawn at a determined rate (see Fig. 1). Those elements were, from the inlet end of the array: a gross filter for removing most of the air particulate material; a flow rate indicator; a trap containing Indicating Drierite for gross water removal; a glass U-tube water freeze trap; a glass condenser water freeze trap; a glass U-tube containing 30 grams of activated coconut charcoal; and, finally, a vacuum cleaner modified for attaching tubing. The two water freeze traps and the charcoal radon trap were immersed in a slush consisting of a one-to-one mixture of  $CHCl_3$  and  $CCl_4$  with crushed dry ice. Tubing used was 0.5-in. i.d. by 0.125-in. walled Tygon. Rubber stoppers were bored to receive 15-mm o.d. Pyrex glass tubing for connecting U-tubes to the Tygon hoses. To minimize leakage in the system, all joints were wrapped thoroughly with plastic "electrician's" tape. Since the system was operated at or near atmospheric pressure, no further precautions were necessary for the prevention of leakage. (See also the discussion of components below.)

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Fig. 1. Schematic diagram of components and assembly of  $Rn^{222}$  collection system.

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Power for running the vacuum cleaner air pump was taken from 117-V ac lines whenever available. However, many field measurements were made at locations without a source of line power. For sampling in such areas, a portable supply consisting of a transistorized 12-V dc to 117-V ac converter and one 12-V automobile battery was used.

#### 2. Sampling Procedure

The procedure for collecting a sample was as follows. Equipment was transported to the site of interest and assembled. (This can be done by one man.) The dry-ice slush was mixed in a 4-liter Dewar flask, and the three traps were placed in the mixture and connected to the system. All joints were wrapped thoroughly with plastic tape and visually inspected. At a noted time, the vacuum supply was turned on and the flow rate adjusted to the desired level (usually 10 liters per minute). This adjustment ordinarily required only about 15 seconds, and variation in the average flow rate due to this adjustment was not of importance. For most samples a collection time of 100 minutes was used, although in areas of higher activity, 60 minutes was sufficient for collecting quantities of Rn<sup>222</sup> large enough to provide acceptable counting statistics. In most cases, then, between 0.5 m<sup>3</sup> and 1.0 m<sup>3</sup> of air was sampled. The sampling was terminated by shutting off the vacuum supply and recording the time.

Following collection of a sample, the charcoal trap was removed from the system, and the charcoal with adsorbed radon was immediately poured into a round tin box approximately 3.5 in. in diameter by 0.75 in. deep. The lid was placed on the box and the assembly allowed to warm to near room temperature before being sealed with plastic tape. Very little radon is lost by permitting the charcoal to warm in such a manner. <sup>5</sup> After being sealed with tape, the can and sample were left undisturbed for at least 3 hours while the radon daughters equilibrated with their parent. Also, during this interval, any radon daughters (down to lead-210) which were trapped during sampling decayed to an insignificant level, the longest-lived daughter down to Pb<sup>210</sup> being Pb<sup>214</sup>, with a half-life of about 27 min. Activity from the decay of Pb<sup>210</sup> (half-life 22 years) accumulates very slowly and was not considered in the analysis.

At the completion of the equilibration period, the can with enclosed sample was placed on a  $4 \times 2$ -in. NaI(T1) crystal optically coupled to a 3-in.

-4 -

Dumont 6363 phototube and counted with a 100-channel pulse-height analyzer. Counts accumulated in channels 23 through 30 (0.54 through 0.68 MeV) were summed and assumed to be due to the 0.61-MeV  $\gamma$  ray of bismuth-214, the third daughter of Rn<sup>222</sup>, and to the  $\gamma$  continuum. Since some Bi<sup>214</sup> is found in the tin boxes, in the charcoal, and in the counting equipment, a 30-g sample of charcoal in a typical tin box with no radon adsorbed other than that arising from radium in the charcoal was counted similarly to provide a background correction. Assuming the continuum to be the same, then, subtracting the background correction from the counts accumulated under the Bi<sup>214</sup> peak per unit time gave the count-rate due to Bi<sup>214</sup> produced by the decay of Rn<sup>222</sup> adsorbed on the charcoal during sampling. This counting rate was then related to the amount of Rn<sup>222</sup> being counted, and after correction for decay, division by the volume of air sampled gave the Rn<sup>222</sup> concentration at the time of collection.

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#### 3. Calibration

A 100-g (nominal) sample of uranium ore with a uranium and radium content precisely determined by the New Brunswick Laboratory of the U.S. AEC, New Brunswick, New Jersey, was sealed in one of the tin boxes described above and allowed to stand for several radon half-lives (more than 30 days). This assayed and carefully weighed sample was then counted for various lengths of time in the manner discussed above and the count rate under the Bi<sup>214</sup> peak determined. Calculations showed that the sample contained  $3.270 \times 10^4$  pCi Rn<sup>222</sup> in equilibrium with the 0.098 g of uranium-238 present. The count rate observed under the Bi<sup>214</sup> peak was  $3674\pm66$  (standard deviation) counts per minute. The simplest calibration value, then, was 0.112±0.002 (standard deviation) counts/min per pCi Rn<sup>222</sup>. Further corrections have not been applied, this value being considered sufficiently accurate for the purposes of the present investigation. Such corrections would have included those for continuum differences between calibration and field sample spectra and for geometry differences between the two. Density variation between the uranium ore standard and the charcoal adsorbed samples was not accounted for because of the relative thinness of the samples (about 0.5 in.) and the relatively high energy of the  $\gamma$  ray of interest (0.61 MeV). The configurations of the two types were nearly identical, 30 g of charcoal filling the boxes about as much as the 100-g standard.

#### 4. Performance and Sources of Error

The simplicity of design and operation of the radon-analysis system described above contributed significantly to the reliability of the system for use in the field. Nonetheless, several questions and some difficulties were encountered during the design and use of the equipment. A brief discussion of the performance of each element in the collection system is given below, followed by a discussion of the sources of error that must be considered in interpreting the results of measurements made with the system.

The gross filter used at the input end of the array consisted of type H-70 filter paper (Hollingsworth and Vose, Cambridge, Massachusetts mounted on a suitable filter head. The total area of the filter was approximately 28 in.<sup>2</sup>, a large area being used to reduce drag on the system. This type of paper is not an absolute filter and served only to remove the larger dust particles from air being pulled through the system. Absolute filtration was not necessary, since any radon daughters carried into the charcoal trap on dust passing through the filter would decay to a very low level before the sample was counted, as discussed earlier. The filter paper was changed periodically to prevent increased drag due to dust accumulation. No difficulties were encountered with the filter assembly.

The flow-rate indicator used in the first system constructed was manufactured and calibrated by the Fischer and Porter Co., Warminster, Pa. (Model OA3565A). It was of the floating-ball type, correction curves for atmospheric pressure and temperature variations being supplied by the manufacturer. The range of the indicator was zero to 11.75 liters/min at 14.7 psia and 70°F (air). Most samples were collected at 10 liters/min or less, ensuring operation away from the ends of the meter range at all times.

A second collection system was built in which the flow indicator was obtained from salvage stock. This meter was also of the floating-ball type but was uncalibrated. For our use, we calibrated it by connecting it in series with the precalibrated meter and flowing air through both. The order in which the meters were connected relative to the direction of flow was reversed and the calibration checked to ensure that pressure differentials across the two were not significant. No difficulties were encountered with either flow-rate indicator, both of which were readable to within about 0.5%.

Indicating Drierite (W. A. Hammond Drierite Co., Xenia, Ohio) was used as a first water trap for gross removal of water from air before

-6-

permitting it to pass over the charcoal. Water removal is necessary because the efficiency with which charcoal adsorbs radon is seriously decreased if the charcoal absorbs moisture. The Drierite (about 1.5 pounds) was poured into a 1-quart polyethylene bottle and hoses were inserted into the bottle through two holes drilled in the lid for that purpose. One of the hoses (the input) was long enough to reach to within about 1 in. of the bottom of the bottle; the other (the exhaust) was pushed to within about 0.5 in. of the top surface of the Drierite. To seal the trap, the screw lid was tightened, and all joints were wrapped with plastic tape.

Difficulties were encountered in using such a trap in that the flow rate in the system was altered occasionally when the assembly was mechanically disturbed. Increased packing of the Drierite clogged the orifice of the input hose in some cases, and also reduced flow further by decreasing available pore spaces in the trap. Such susceptibility necessitated taking care to ensure that no noticeable disturbances occurred during the collection of a sample. Similarly, the physical arrangement of the tubing connected to the trap necessitated great care to ensure that no leakage occurred at this point in the system. Both of these necessities were rigidly adhered to throughout the sampling program.

Next in the array were two glass water traps immersed in the  $CHCl_3:CCl_4: CO_2(s)$  slush to freeze out any water not removed by the Drierite. The first of these was a Pyrex glass U-tube 1 in. in diameter and the second a condenser trap consisting of two concentric glass tubes, as illustrated in Fig. 1. No difficulties were encountered in using these components, but, again, care was necessary in connecting tubing to the U-tube through the rubber stoppers. A very tight fit was requisite for preventing leakage. In most cases, the three water traps were successful in removing all water from the flow before the air entered the charcoal trap. In a few samples, dampness in the charcoal was noticed, but this was discovered, any one charge of Drierite was used for the collection of only two samples (about 200 min sampling time). Frequent changing of the Drierite eliminated the problem of water entering the charcoal trap.

The charcoal trap, too, consisted of a Pyrex glass U-tube 1 in. in diameter. Thirty grams of activated coconut charcoal (Fisher Scientific Co., Fair Lawn, N.J.) of size 6-14 mesh was poured into a tube and a small piece

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of cotton placed in the exhaust side of the trap to prevent the charcoal's being carried out of the U-tube by the air flow. This amount of charcoal produced a column about 7 in. long. Hoses were connected to this trap in the same way as to the water trap described above.

It was very important to assure that all the air passing through the charcoal came into intimate contact with the charcoal granules. Any air passing through the trap without contacting a charcoal surface would carry radon atoms completely through the trap, resulting in a measured concentration lower than that actually present. We found that at flow rates much above 10 liters/min, the charcoal was displaced by the air flow and "bumping" occurred at the bend in the U-tube. For this reason, a flow rate of 10 liters/min or less was used consistently throughout the sampling program. No "bumping" occurred at these low flow rates, and maximal contact between the air column and the charcoal granules was obtained.

A vacuum cleaner modified to permit attaching the Tygon tubing was the last element in the array. The one we used was of the canister type with an adjustable motor speed which allowed us to vary the flow rate through the system as needed. The power sources for the vacuum cleaner have been described above. No problems arose in using the vacuum cleaner with 117-V ac line power. However, when it was powered by the portable supply described earlier, one feature was noted. Flow rate through the system was very sensitive to slight changes in the speed of the vacuum cleaner motor which, in turn, was very sensitive to the charge level of the 12-V automobile battery in the portable supply. A single battery permitted operation for about 100 min before a noticeable change in flow rate was observed. However, to preclude the possibility of an unnoticed decrease in flow during the collection of a sample, two 12-V batteries connected in parallel were usually employed.

A second vacuum cleaner was not available for the assembly of the second system mentioned above. In its place, two small 117-V ac motorblowers were used, one at each end of the system. These functioned satisfactorily and behaved essentially the same as the vacuum cleaner.

The mixture used for chilling the charcoal and water traps was chosen in part because of its widespread use in such systems.  $CHCl_3$  and  $CCl_4$  are toxic compounds, however, and a mixture of less toxic and less expensive materials was investigated, namely a slush of dry ice and isopropyl alcohol. Upon being cooled by the dry ice, however, the alcohol acquired a syrupy

-8 -

consistency, and the bulk of the dry ice collected in the bottom of the Dewar flask. For these two reasons, the original mixture was used throughout the program. Five to 10 pounds of dry ice was sufficient to provide a thick slush with the mixing characteristics necessary for consistently achieving good thermal contact with the coolant.

Many of the possible sources of error in measurements made with the system we used for collecting and analyzing Rn<sup>222</sup> have been mentioned above. Among these were errors due to calibration, errors in flow-rate measurements, leakage in the system, the presence of water in charcoal traps, and the flow characteristics in the system. All these possible sources of error were carefully monitored throughout the sampling program, and their effects were minimal. However, other possibilities for introducing error existed and should be discussed.

Since the concentrations of Rn<sup>222</sup> present in most natural environments are rather low, it was necessary that we ensure that all radon atoms entering the system were retained only in the charcoal trap. Any loss of radon in elements of the system preceding the charcoal trap could be significant. Similarly, the charcoal trap had to be capable of retaining all radon passed into it. Should the charcoal become saturated, aberrant results would again be obtained. Shleien has shown the retention of radon on Indicating Drierite to be very slight at room temperature, and the same author demonstrated that the adsorption of radon on the glass of the water and radon traps is likewise of no significance.<sup>5</sup> In testing a similar system for retention of radon in a chilled charcoal trap, Pearson, Rimbey, and Jones have shown the retention to be better than 99.2% with 25 g of charcoal.  $^{6}$  To reaffirm this value for the system being described in the present report, one atmospheric sample was taken in which two radon traps were connected in series in the collection apparatus. The first trap through which air flowed showed the atmospheric  $Rn^{222}$  concentration to be 115±6 pCi/m<sup>3</sup>. The activity (above background) in the second trap was  $2\pm 3$  pCi/m<sup>3</sup>. Thus, the retention of radon in the first trap was better than 95%. In view of the findings of Pearson et al., we consider the retention to be essentially 100% and do not correct for losses of radon passing through the chilled charcoal trap.

Another source of extraneous radon was radium-226 in the collection system itself. While we have not measured the amount of radon in our samples arising from this spurious source, the measurements by Shleien

- 9 -

have shown that materials such as those used in our system contain very little radium. <sup>5</sup> Also, from the results of a sample we took in oceanic air, which exhibits very low  $Rn^{222}$  concentrations, the upper limit to the amount of radon generated within our system in a typical sampling interval could be placed at 2±6 pCi (see Table I, sample 11). Therefore, we do not correct for error introduced by radon produced in the collection system itself.

After the majority of the data to be discussed below had been collected, several improvements were made in our air-sampling equipment. First, both U-tube water and charcoal traps were replaced by concentric-tube condenser traps similar to the condenser water trap illustrated in Fig. 1. Their replacement obviated the use of rubber stoppers, thereby greatly reducing the likelihood of leakage. Also, since the charcoal in the condenser-type trap was in a single vertical column, better contact of air with the charcoal granules was ensured.

A second improvement was the construction of Drierite traps consisting of closed Plexiglas cylinders with removable lids and with brass hose fittings for connections into the sampling system. The use of such a trap greatly reduced the possibility of leakage around tubing leading to and from the Drierite container and very much simplified changing and recharging that element.

A third improvement was the reduction in the lengths of tubing between components of the system by using copper elbows at points where long sections of tubing were required in the earlier systems. The copper fittings were of proper size for connecting tightly to the tubing used, and, as before, all joints were wrapped securely with plastic tape. These three improvements greatly increased the reliability and ruggedness of the system and, at the same time, decreased its physical size by about 50%. Samples taken with the improved system are labeled "N1" in the tables that follow.

- 10 -

Sample number	Appr. time (PST)	Date 1965	Location	Rn <sup>222</sup> conc. (pCi/m <sup>3</sup> ± std. dev.)	Notes
2	1300	7/15	Bldg. 72,		
	· · · · · · · · · · · · · · · · · · ·		room 105	$108 \pm 5$	Sys. 1
4	1100	7/16	72-105	$115 \pm 6$	Sys. 1
8 .	1100	8/9	72-105	85 ± 5	Sys. 1
9	1100	8/9	72-105	$106 \pm 5$	Sys. 2
33	1330	8/29	72-105	$158 \pm 7$	Sys. 1
	•				
1.2	1500	8/10	Outside Bldg. 72	39 ± 6	Sys. 1
18	1500	8/16	Outside Bldg. 72	60 ± 5	Sys. 1
19	1500	8/16	Outside Bldg. 72	63 ± 5	Sys. 2
20	2030	8/16	Outside Bldg. 72	49 ± 4	Sys. 1
21	2030	8/16	Outside Bldg. 72	54 ± 6	Sys. 2
5	0930	8/2	Low-Bkgd. Room	46 ± 3	Sys. 2
13	1500	8/10	72-LBR	82 ± 5	Sys. 2
22	1400	8/18	72-LBR	45 ± 5	Sys. 1
23	1400	8/18	72 Roof	17 ± 5	Sys. 2
10	1430	8/9	2412 Carl'n	30 ± 6	Sys. 2
11	1430	8/9	Berk'y Mari	na 2±6	Sys. 1
1.4	1200	8/12	2412 Carl'n	31 ± 5	Sys. 2
15	1200	8/12	Berk'y Mari	na 7 ± 2	Sys. 1
38	1200	1/21 (1966)	In tunnel, Bldg. 46	377 ± 7	N1
39	1400	1/21 (1966)	Outside tunne Bldg. 46	el, 122 ± 5	N1

Table I. Data from the San Francisco Bay region.

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#### B. Data and Discussion

Atmospheric radon samples were collected at several locations in the State of California. A summary of those measurements and a discussion of the implications of the information are presented below. We stress the fact that the total number of samples acquired at any location was small, and any statements concerning the implications of these data must be viewed with this in mind. Pearson and Moses have shown that atmospheric  $Rn^{222}$  concentrations may vary over two orders of magnitude within a single day;<sup>7</sup> thus, any single measurement, or even a series of measurements made at the same time of day, as ours often were, may not provide complete information about the environmental radon concentration at any one location.

#### 1. Data from the San Francisco Bay Area

A number of samples were collected in and around Building 72 (Health Physics Building) of the Lawrence Radiation Laboratory, Berkeley, California, as well as at two locations in the city of Berkeley itself. Of these latter, one site was about 2.5 miles east of San Francisco Bay and was at an elevation of about 200 feet above sea level. This site is referred to as "2412 Carleton" in Table I. The other was located on the shoreline of the Berkeley Marina at an elevation of about 6 feet above sea level. The Health Physics Building of the Lawrence Radiation Laboratory is about 3.5 miles from the Bay and is at an elevation of about 800 feet. The results of these measurements are presented in Table I.

As can be seen from the data, concentrations inside the building are, in general, higher than those in the outside atmosphere, in agreement with the results of many other workers. This increase is due to radon entering the room from the decay of radium in the materials of the walls and being retained in the room; radon entering the outside atmosphere from the soil, however, is swept away and diluted. Samples 12 and 13, as well as samples 22 and 23, indicate that this is true even in the low-background counting facility, the materials for which were chosen for their low inherent radioactivity contents. <sup>8</sup> Samples 18 and 19, along with 20 and 21, demonstrate the reproducibility obtained between the two systems described earlier in this report. Samples 18 through 21 also seem to indicate that the radon concentrations in the atmosphere do not vary greatly from early afternoon to early evening at this location. However, no such generalization can be made from the meager

#### - 12-

data available. Meteorological variations, both short-term (hourly) and long-term (seasonal), play a great role in determining the amount of radon in the atmosphere, as shown by Pearson and Moses, <sup>7</sup> Pearson and Jones, <sup>9</sup> and many others. In this work, we have made no attempt to record extensive meteorological data; we have noted only qualitative weather descriptions, and these were not sufficiently detailed to provide any useful correlations among our measured  $Rn^{222}$  concentrations and atmospheric conditions at the time of sampling.

One aspect of the local meteorology should be mentioned, however. The prevailing winds in this region are from the northwest, and so the air sampled had traveled, first, a great distance over the Pacific Ocean and then over a short extent of land (from 3 to possibly 50 miles) before entering our sampler. The data from samples 10, 11, 14, and 15, considered with the values determined at Building 72 of the Lawrence Radiation Laboratory, demonstrate the existence of a gradient of Rn<sup>222</sup> concentration, the concentration increasing the farther the air has traveled over land. In view of the finding that air over oceans is largely radon-free, our results represent very well the type of data to be expected from such measurements. <sup>10</sup> It should be noted that at the times of taking samples 10, 11, 14, and 15, winds were directly from the west and thus had not passed over any great extent of land before entering the collection apparatus.

Samples 38 and 39 were the results of measurements made near Building 46 of the Lawrence Radiation Laboratory. Sample 38 was taken inside a concrete-lined tunnel located in the side of a hill adjacent to that building, the tunnel penetrating the hill for a distance of about 120 ft. Sample 39 was acquired immediately after number 38, but the collector was placed in the open air just outside the tunnel entrance. While the outside atmospheric concentration was typical of those made in similar areas, the concentration inside the tunnel was found to be about three times as high. Again, this represents  $Rn^{222}$  accumulation, since air movements at the end of the tunnel where the sample was taken are practically nonexistent.

#### 2. Other Data

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Samples of atmospheric  $Rn^{222}$  were collected at several other sites in California; the results of these measurements are presented in Table II.

Sample number	Appr. time (PST)	Date 1965	Location	Rn <sup>222</sup> conc. (pCi/m <sup>3</sup> <del>+</del> std. dev.)	Notes
			· · ·	· · · · · · · · · · · · · · · · · · ·	
16	1200	8/13	Inside Red Mt. Mine	362 ± 6	Sys. 1
17	1230	8/13.	Outside Red. Mt. Mine	$101 \pm 6$	Sys. 2
24	1130	8/20	Inside Red. Mt. Mine	339 ± 9	Sys. 1
25	1130	8/20	Inside Red, Mt. Mine	$321 \pm 14$	Sys. 2
26	1230	8/20	Inside Red. Mt. Mine	576 ± 18	Sys. 2
27	1300	8/20	Inside Red, Mt Mine	238 ± 15	Sys. 1
29	1200	8/25	Inside Red. Mt. Mine	193 ± 8	Sys. 1
28	1615	.8/24	Fern Patch	76 ± 7	-
30	1645	8/25	DC-1	227 ± 9	-
31	1100	8/27	DC-11	63 ± 7	-
32	1200	8/28	Asbestos Mine	132 ± 9	-

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Table II. Other data.

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The location called "Red Mountain Mine" refers to an abandoned magnesite mine located in the Diablo Mountains about 30 miles south of Livermore, California. This particular mine has been of interest to the members of the Lawrence Radiation Laboratory (Berkeley) (LRLB) Health Physics Group who have been investigating natural environments with low inherent radiation fields. Red Mountain is composed largely of the mineral serpentine, a substance containing very little natural radioactivity. <sup>8</sup> Also, the distance to the surface from within one horizontal adit of the mine varies from less than 5 ft to more than 350 ft, providing natural cosmic-ray shielding continuously varying with distance into the mountain. With a portable scintillation counter, for example, the observed count rate outside the mine entrance is typically 40 counts per second (cps) to 50 cps, while inside the mine, rates as low as 2 to 5 cps are obtained.

No uranium has been detectable in the materials of the mine's walls by using the LRLB gamma-scintillation system (capable of detecting less than  $10^{-8}$  g of uranium per g, using a 1-kg sample<sup>8</sup>); yet the total background radiation levels in the mine as measured with the portable scintillation counter vary with time at the same site in the adit. It was suspected that fluctuations in Rn<sup>222</sup> daughter product concentration in the air circulating through the mine could be the cause of the background variation, and several Rn<sup>222</sup> measurements have been made in the mine to investigate this possibility. The data are incomplete, but several interesting observations have been made.

Sample number 17, when compared with sample 16, which was taken nearly simultaneously, as well as with the other samples from within the adit, indicates that  $\operatorname{Rn}^{222}$  concentrations inside may be higher than in the atmosphere outside the mine. It is possible that enough  $\operatorname{Ra}^{226}$  is contained in the serpentine to supply this excess and yet not be detected by  $\gamma$ -scintillation techniques, this possibility being enhanced when consideration is given to the physical state of the walls of the mine. The large number of cracks and the surface roughness increase the effective emanating area of the mine considerably. Possibly enough radon is generated in the walls and diffuses into the cavity to increase concentrations to the observed levels.

A second possibility is that outside atmospheric concentrations increase greatly during certain times of day (e.g., at night), and radon carried into the mine at such times diffuses into the walls and fissures to be released during the low-activity periods of the external atmosphere. Data are not

- 15-

available to prove either of these or other alternatives. Regardless of the mechanism, however, variations in radon concentration and concomitant changes in radon decay-product activity probably do account for a large portion of the observed gamma background fluctuations observed within the mine. Further studies are in progress to determine the source of the apparent radon "excess" measured in the mine.

Samples 28 through 33 were taken during an intercalibration experiment in cooperation with investigators from several other laboratories. The data are presented here only as examples of radon concentrations at other locations than those discussed above. Their paucity obviates any detailed analysis.

The "Fern Patch" site is in the Santa Cruz Mountains of the Coast Range of California. The soil in the region exhibits anomalously high potassium-40 concentrations, but the atmospheric Rn<sup>222</sup> content measured is only typical of such a coastal area. DC-1 refers to a sample collected over a granodiorite outcrop at an elevation of about 5400 ft near Shaver Lake in the Sierra Nevada. DC-11 is a site over a granitic outcrop (elevation 8200 ft) near Courtwright Reservoir, also in the Sierra. The "asbestos mine" site was in an open-pit asbestos mine located near Copperopolis, California, in the foothills of the Sierra Nevada. More data would be necessary for making any statement concerning Rn<sup>222</sup> concentrations at these locations.

-16 -

#### III. SUMMARY

A portable  $\operatorname{Rn}^{222}$  gas sampler developed by the Health Physics Group of the Lawrence Radiation Laboratory, Berkeley, has provided a simple, effective, economical way of determining atmospheric  $\operatorname{Rn}^{222}$  concentrations. Measurements made with the system are reliable and reproducible. They have indicated that atmospheric radon concentration dynamics in the San Francisco Bay area follow a pattern that has been found typical of such coastal regions. The sensitivity of the system, although balanced against cost and ease of operation, is sufficient to measure  $\operatorname{Rn}^{222}$  concentrations as low as about 25 pCi/m<sup>3</sup> reproducibly.

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-18-

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