ATOMIC ENERGY CONTROL BOARD

ELLIOIT LAKE TECHNICAL NOTE NO. 1

SAMPLE BAG RADON AVERAGING UNITS

Dilworth, Secord, Meagher and Associates Limited
Acres Consulting Services Limited

January 1978

DSMA Report No. 1012/968
"The Atomic Energy Control Board is not responsible for the accuracy of the statements made or opinions expressed in this publication and neither the Board nor the author(s) assume(s) liability with respect to any damage or loss incurred as a result of the use made of the information contained in this publication."
1. INTRODUCTION

This Technical Note is issued to document information which is of sufficient reference value as to merit something more than transmission by letter, but which is not properly a part of a fully formal report. The subject of the Note is the pump-and-bag samplers presently being introduced into the radon survey program at Elliot Lake.

Since these averaging samplers have not previously been applied to the radon survey program, a brief description of the units, and their merits and limitations, appears to be worthwhile.

Concerning the format of a Technical Note, this is seen as being relatively informal, but sufficiently ordered as to present a logical picture of the subject. Further such Notes may be issued in the event that suitable subjects arise.

2. REASONS FOR ADOPTION

The results of a large number of radon, and WL, readings at Elliot Lake to date shows that readings are very variable. One reasonably typical house showed values ranging from 0.001 WL to 0.162 WL in 15 readings, with a logarithmic mean of 0.021 WL. The house next door varied from 0.008 WL to 0.06 WL, also in 15 readings, with a log mean of 0.011 WL. The spread of readings throughout the survey as a whole is comparable to the spread observed at a 'typical' house. As a result, it is difficult to conclude, with confidence, that a given house is or is not acceptable on the basis of a reasonable number of individual readings, or grab samples.

Confidence can, of course, be increased by increasing the number of grab samples. Unfortunately, a large number of such samples is expensive, both in terms of man-hours and in terms of schedule.

The best solution to this averaging problem would be an integrating instrument which continuously read, and recorded, WL values with little or no attention over a time period of a week or a month. The problem with this approach is that, though the labour costs are low, the instruments are expensive. That is, one instrument may not be too costly as a single item, but if a hundred are required in order to
survey a number of houses in a reasonable schedule, then the total capital cost is high.

The bag sampler is attractive in that it avoids the two extremes of cost. It is cheap per unit. It does require attention about twice a week, but this is not a serious manpower requirement. Its major attraction is that it does give a genuine average value. Its accuracy is not perfect, but it is adequate for the task and it is almost certainly better than that of an average of a number of grab samples.

3. DESCRIPTION AND COST

The pump-and-bag sampler concept is far from new. It consists of a low, and constant, volume pump which fills a sample bag over some period of time. The contents of the sample bag then contain an average concentration of whatever is of interest during that time, at least in the case of normal pollutants. The system has been used successfully for a variety of normal atmospheric contaminants and was the subject of a report on its use for radon by C.W. Sill in 1969. A copy of this report is attached for reference.

In the case of an active gas, such as radon, allowance for its decay must be made. Some error will be inherent in this allowance because the radon concentrations will vary with time. Both the allowance and errors are discussed in a following section. A further error will be introduced if the air pump flow rate varies with time and so a reliable pump and flow control orifice are required. The actual flow rate is not important as long as it remains constant and is low enough as to not overfill the sample bag in the time it is unattended.

The units presently in use at Elliot Lake consist of a small pump, a flow control valve or orifice (integral with the pump or separate) and a collection bag, all mounted in a garbage pail to form a handy unit. Commercially available, low-volume, battery operated pumps have been used, but the battery packs have not been satisfactorily reliable. Simple 110V AC aquarium pumps, with a pipe restriction to limit airflow, appear to be a better choice.
Commercially available 5-layer aluminized polyvinyl fluoride collection bags, which have excellent radon retention, have been adopted and two 40-l bags will fit inside the garbage can.

The cost of components for the unit is then about:

- Pump $15.00
- Bags, 2 x $20 - 40.00
- Valve 20.00
- Fittings 10.00
- Garbage Can 15.00
- TOTAL - $100.00

4. PROCEDURE FOR USE

The unit is placed in a house for 3 or 4 days, as convenient. At the end of this time the radon concentration in the bag is measured using a lucas cell. The bag is emptied, flushed with nitrogen, and replaced. The average radon concentration is obtained by applying the appropriate correction factor from the following section.

This allows the average radon concentration in a house, over a period of a week, to be measured in only two units, with a sampling error of less than ± 40%. Compared to grab sampling, the saving in manpower is considerable and the sample includes evenings, nights and weekends.

5. CORRECTION FOR DECAY OF RADON DURING SAMPLING

If the radon concentration is constant at B Bq (Becquerels)/m³, and the sampling rate is R m³/s, then in a small time dt, we collect B.Rdt Bq in the bag.

If a time t seconds elapses to the end of sampling, this amount of radon will decay by a factor of \( e^{-\lambda t} \), where \( \lambda \) is the decay constant for radon.

The total activity remaining in the bag after sampling for a time T seconds

\[
\int_{0}^{T} B.Rdt e^{-\lambda t} Bq = \frac{B.R}{\lambda} \left[ 1 - e^{-\lambda T} \right] Bq.
\]
The volume of gas collected is \( RT \) m\(^3\).
and so the concentration of radon in the bag at the end of \( T \) seconds
\[
= \frac{B}{\lambda T} \left[ 1 - e^{-\lambda T} \right] \text{ Bq/m}^3.
\]
For small \( T \), this reduces to
\[
\frac{B}{\lambda T} \left[ 1 - (1 - \lambda T) \right] \text{ Bq/m}^3 = B \text{ Bq/m}^3 - \text{as for a stable gas.}
\]
The concentration in the bag is less than \( B \) by a factor \( D \) where
\[
D = \frac{1 - e^{-\lambda T}}{\lambda T}.
\]
The value of \( 1/D \) (the decay correction factor) is shown against various sampling periods in Table 1.
If the concentration is not constant, the average concentration calculated from the concentration in the bag will be in error by a factor depending on when the radon entered the bag. The maximum error can be estimated by considering two extreme cases, which will give the maximum possible errors.

CASE 1 All the radon in the bag enters at the start of the sampling period, and only radon-free air enters the bag for the rest of the sampling period.

Let \( S \) Bq enter the bag at the start of sampling and so, the concentration in the bag at the end of \( T \) seconds will be
\[
\frac{Se^{-\lambda T}}{RT} \text{ Bq/m}^3.
\]
This concentration will be corrected by \( 1/D \) to give the estimated average concentration \( \bar{A} \text{ Bq/m}^3 \) over the sampling period, so
\[
\bar{A} = \frac{Se^{-\lambda T}}{RT} \cdot \frac{1}{D}.
\]
The true value of the average concentration \( A \), is
\[
A = \frac{S}{RT}.
\]
Therefore, the average concentration is underestimated by a factor of
\[
\frac{\bar{A}/A}{\frac{S}{RT} \cdot \frac{e^{-\lambda T}}{D} \cdot \frac{RT}{S} = e^{-\lambda T}}.
\]
The value of this factor is shown in Table 1 against various sampling periods.
CASE 2  All the radon in the bag enters at the end of the sampling period, and only radon-free air entered the bag for the rest of the sampling period. Let \( S \) Bq enter the bag at the end of sampling, and so the concentration in the bag at the end of \( T \) seconds will be
\[
\frac{S}{RT} \text{ Bq/m}^3.
\]

This concentration will be multiplied by \( 1/D \) to give the estimate average concentration \( \bar{A} \) Bq/m\(^3\) over the sample period, so
\[
\bar{A} = \frac{S}{RT} \cdot \frac{\lambda T}{1 - e^{-\lambda T}}.
\]

The true value of the average concentration \( A \) is
\[
A = \frac{S}{RT}
\]

so the average concentration is overestimated by a factor of
\[
\frac{\bar{A}}{A} = \frac{\lambda T}{1 - e^{-\lambda T}} = \frac{1}{D}.
\]

The value of this factor is shown in Table 1 against various sampling periods.
<table>
<thead>
<tr>
<th>Sample Period (Days)</th>
<th>Decay Correction Factor $1/D$</th>
<th>Extreme Error Factor Overestimate</th>
<th>Extreme Error Factor Underestimate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.09</td>
<td>1.09</td>
<td>0.91</td>
</tr>
<tr>
<td>2</td>
<td>1.19</td>
<td>1.19</td>
<td>0.83</td>
</tr>
<tr>
<td>3</td>
<td>1.30</td>
<td>1.30</td>
<td>0.75</td>
</tr>
<tr>
<td>4</td>
<td>1.41</td>
<td>1.41</td>
<td>0.68</td>
</tr>
<tr>
<td>5</td>
<td>1.52</td>
<td>1.52</td>
<td>0.61</td>
</tr>
<tr>
<td>6</td>
<td>1.64</td>
<td>1.64</td>
<td>0.55</td>
</tr>
</tbody>
</table>

AN INTEGRATING AIR SAMPLER FOR DETERMINATION OF $^{222}$Rn

CLAUDE W. SILL
Health Services Laboratory, U.S. Atomic Energy Commission, Idaho Falls, Idaho

(Received 3 July 1968; in revised form 12 August 1968)

Abstract—The actual radiological hazard from radon emitted from tailings piles remaining from processing uranium ores is extremely difficult to assess. Exhalation of radon from the ground varies markedly with time and is particularly dependent on the condition of the surface and the prevailing meteorological conditions. Using present techniques, the sampling required to permit a realistic estimate of the dose sustained by a significant population in a year's time is economically prohibitive. A sampler has been developed to collect air continuously at 10 ml/min for 48 hr to provide an integrated sample which can be analyzed to determine the average radon concentration to which people at that location were exposed during the same period of time. A small aquarium aerator pumps filtered air through a precision needle valve with a digital indicating handle into a collapsible 40 l. bag made of laminated 3 mil Mylar. The pump can be used either directly from an a.c. line or from a battery with a small power inverter. Each station is resampled at 3-week intervals for a calendar year, amounting to approximately 10% of the total time available. A 1 yr program involving 37 stations in four different cities has been started.

During the processing of uranium ores for the recovery of uranium, the radioactive daughters of the uranium remain almost completely with the insoluble portion of the ore. The radioactivity of the tailings material averages about 700 pCi/g for each nuclide in the uranium chain, amounting to about 58,000 Ci of each nuclide, or over 125 lb. of $^{224}$Ra. By the end of 1970, the projected total of such tailings in the U.S. will amount to approximately 90 million tons contained in 35 different piles. Recent calculations\(^1\) suggest that the dose to the segmental bronchi from $^{222}$Rn in equilibrium with its short-lived daughters amounts to 13.8 rad/yr from a concentration of 10.1 pCi/l. Because of the large mass of tailings materials and its associated $^{224}$Ra content, and the ability of radon to diffuse out of the solid pile, the radiological dose to the lungs from the gaseous radon and its particulate daughters might actually exceed that from the longer-lived nuclides in the tailings material itself.

In an exploratory survey, 36 samples from four different cities were analyzed for $^{222}$Rn by a modification of a conventional method\(^2\) involving absorption of the radon on activated carbon, elution at 500°C, and counting in a scintillation chamber.\(^3\) These preliminary samples were obtained by allowing an evacuated 16 l. stainless steel tank to fill spontaneously to atmospheric pressure. The highest value of 38.3 pCi/l. was obtained directly over tailings under nocturnal inversion conditions and dropped to 5.6 pCi/l. for an afternoon sample at the same location. Five other samples taken within about 0.5 mile from the respective piles were between 1 and 3 times the maximum permissible concentration (MPC) of 3 pCi/l. permitted by Federal regulations for continuous exposure to the general public in uncontrolled areas.\(^4\) The remaining samples from residential areas were not significantly different than background samples taken at approximately the same time several miles away from tailings areas. The preliminary results clearly demonstrated the need for further more sophisticated measurements to determine the average yearly concentrations leaving the piles and in unrestricted areas. However, the actual radiological hazard from radon emitted from tailings piles is difficult to assess. Exhalation of radon from the ground varies markedly with time and is particularly...
dependent on the condition of the surface and the prevailing meteorological conditions.\(^{(4-11)}\)

Normally, the radon content in the air near the ground increases during the nocturnal inversion period and decreases during the day-time convective period when atmospheric dilution is at a maximum. Consequently, the radon concentration changes significantly from one hour to the next.

Federal regulations permit the radiological dose received by those in uncontrolled areas from licensed facilities to be averaged over a period of 1 yr.\(^{(4)}\) Consequently, a reasonable sampling program should logically cover at least a 1 yr period to sample radon under all meteorological conditions prevailing during the same time period over which the radiological dose is averaged. If separate samples were taken by conventional techniques employing evacuated flasks, which sample literally for only a few seconds at a time, such a program would become almost prohibitively expensive in terms of numbers of samples and analyses required if many sampling stations were involved.

In contrast, if the sample itself could be averaged by being collected continuously at a constant flow rate for several hours at a time, a single analysis would give an average value for the entire period sampled. Such integrated samples would obviously permit air to be sampled for a greater fraction of the total time available for a given cost, and permit a much more realistic evaluation of the radiological dose to be made.

A sampling period of 48 hr was selected as being about the longest time that could be used without introducing an undesirably large uncertainty due to radioactive decay of the 3.823 day \(^{222}\text{Rn}\) during the sample collection period. If the activity were calculated to the midpoint of the collection period, the error from this cause could not exceed about 15\% if all the radon were collected during the first or last 2 hr, a most unlikely occurrence. With the variations in radon concentrations normally encountered, the error is not expected to exceed about 5\%. Sampling for 2 full diurnal cycles should smooth out the marked diurnal fluctuations and give a good average value for the entire period. Sampling will be repeated at each location every 3 weeks to cover 10\% of the total time available for exposure and to sample every calendar month, every season, and every usual meteorological condition occurring within 1 calendar year with only 17 samples per station.

Because only 61. of air is required to detect activities higher than about 0.01 pCi/l in a 1 hr count, approximately 301. of air should be adequate for duplication and radiological decay during shipment. If the sampling period is to cover 48 hr, the flow rate must not exceed 10 ml/min. Consequently, the present investigation was initiated to develop equipment for collecting air at such a low flow rate, which must remain reasonably constant, in outdoor locations under all kinds of weather conditions and frequently at locations remote from a source of electric power. Available equipment capable of doing the job was too expensive to permit large numbers of stations to be sampled simultaneously to meet the desired schedules.

**SAMPLING EQUIPMENT**

**Pump**

The pump finally used was a small 4 W 117 V aquarium aerator called the Silent Giant (model 120, Aquarium Pump Supply Inc., Prescott, Ariz.) that employs a cup and plunger compression chamber sealed by a flexible web and operated by an electro-magnet. The pump also employs an efficient method of back-pressure compensation so that the flow rate is not affected significantly by conditions of air use or depth of water in the aquarium that is helpful in the present application. Under conditions of free flow, the pump delivers about 3 l./min of air. With a needle valve restricting the flow to 0-125 ml/min the pump builds up a pressure of about 6 psi.

As supplied commercially, the air inlet to the pump consists of several small holes in the interior can in which the pump mechanism is contained. To provide a single specific air inlet so that the incoming air can be filtered free of \(^{226}\text{Ra}\) and also to permit a tube to be run directly into the air stream to be sampled while the pump itself is protected inside a weather shelter, the interior can was reencased in a plastic housing made conveniently from 3.5 in. OD cylindrical tubing 4 in. long and \(\frac{1}{4}\) in. thick. Both ends were sealed with flat plates \(\frac{1}{4}\) in. thick, one end of which had been previously
drilled and tapped to provide the necessary air inlet and outlet and electrical connections. When sampling at locations remote from a source of electric power, the pump can be operated from a 12 V storage battery through a small power inverter (Model 50-103, Newark Electronics Corp., Chicago, Ill.) used to operate an electric shaver from an automobile storage battery.

Flow regulator

The first samplers used a small variable-area type flow meter and a fairly coarse needle valve to control and indicate the air flow obtained. Even with fairly expensive models, the flow meter itself appeared to contribute considerably to the instability of the air flow due to sticking floats, etc. Consequently, the flow meter and needle valve assembly were replaced by a precision extra-low-flow needle valve with a direct reading digital handle (Model 8502, Brooks Instrument Division, Hatfield, Penn.). Under the pressure of 6 psi supplied by the aquarium pump, a flow rate of about 125 ml/min is obtained with the valve wide open. The valve requires 15 turns from fully open to fully closed and can be read to about 1/10 of a turn. The flow rate can be quickly and easily calibrated by water displacement after which the same volume can be reproduced to within ±1% by resetting the digital indicator.

Sample container

A collapsible container is necessary for use with the present low-volume pump to prevent significant changes in flow rate due to changing back pressure as the sample is collected. It is well known that rubber, polyethylene and many other kinds of plastics are extremely permeable to radon gas. In addition, natural rubber generally contains sufficient 224Ra to interfere seriously in the determination of radon. Bags made of Mylar have been used for years as meteorological balloons in weather science and are known to be capable of flying long distances without losing significant amounts of various gases such as oxygen, nitrogen, helium, etc. The present investigation shows that the same material is perhaps the best presently available for retention of radon.

The bags used were fabricated from a double layer of ½ mil Mylar laminated together to eliminate pin holes (GT-21, G. T. Schjeldahl Company, Northfield, Minn.). To make the bags, a strip of the laminated material of the size desired is folded over and the side and both ends are sealed by inserting a strip of sealing tape (GT-400) between the two faces and ironing the tape area with a hand sealing iron at approximately 350°F until the milky appearance disappears from the tape indicating that a good bond has been achieved. A dill valve available from bicycle or hardware stores is placed in the face of the bag prior to scaling. The bags used in the present study have inside dimensions of 17½ in. x 31½ in. with the bag lying flat, and hold approximately 40 l. when filled completely. The bags are made somewhat larger than the volume to be contained to provide room for expansion at temperatures and pressures other than those at which the sample is collected, e.g., mountain passes, aircraft, etc.

The complete sampling equipment is shown in Fig. 1 connected to the small electric shaver inverter and storage battery for use in locations remote from a source of electric power. For sampling in remote or inaccessible locations where weight and bulk must be kept to a minimum, such as in a uranium mine, the small 10 amp-hr motorcycle battery shown provides sufficient capacity for an 8 hr integration over a shift. For sampling in remote areas above ground, a regular automobile storage battery of about 70 amp-hr capacity is necessary for a 48 hr sample integration. When 115 V power is available, the electric plug is simply removed from the inverter shown and plugged into the source of A.C. power.

In operation, the entire equipment shown in Fig. 1 is placed in a weather shelter and the piece of gum rubber tubing shown on the right is allowed to extend out of one end into the air stream to be sampled. A small funnel is inserted in the end of the tubing to prevent rain from being drawn in and saturating the filter, resulting in a marked decrease or complete stoppage of the air flow. The air is then drawn through a double 1 in. filter (in the conical holder) consisting of a glass-fiber roughing filter followed by a membrane filter (GA-6 Gelman Instrument Company, Ann Arbor, Mich.)
AN INTEGRATING AIR SAMPLER FOR DETERMINATION OF $^{222}$Ra

into the plastic case. The pump then draws the air through holes in the bottom of the can, and expels it through the coiled tube, the needle valve and into the Mylar bag. In actual use, the Mylar bag is contained in a cardboard box, 10.5 in. x 12 in. x 30 in., inside the weather shelter to protect the bag from abrasion, air motion, etc., and to serve as a container in which to ship the gas sample back to the laboratory. Current cost of the equipment shown is about $55 for the a.c.-operated system, about $110 for the battery-operated system and about $6 each for the Mylar bags.

PERFORMANCE

The operating characteristics of the pump and needle valve were determined by measuring the volume of water displaced from an inverted 10 ml. graduated cylinder in 60 sec under various conditions. A flow rate of 9.6 ml/min could be reproduced to within 0.1 ml/min or about 1% by setting the indicating handle back to the same number from either fully open or fully closed provided that the final adjustment was always made from the same direction.

Effect of voltage

At a flow rate of 9.6 ml/min, changing the voltage from 135 to 105 V changed the flow rate by less than about 1%. The flow rate drops to about 50% of its initial value at 90 V and decreases rapidly at lower values. In fact, this same pump makes an excellent low-cost transfer pump for laboratory use, using a variable transformer to change the flow rate continuously and smoothly by changing the voltage in the lower voltage ranges. Accordingly, care should be taken that the voltage at the end of long runs of wiring required to reach remote locations is not permitted to drop so low that the flow rate becomes susceptible to normal voltage fluctuations. However, for the year-long program for which this equipment was developed, several thousand feet of no. 14 electric wire were used to reach most of the sampling stations on and around the mill tailings piles with a resultant voltage drop of less than 1 V. Because of the very small power consumption of the pumps, longer runs of even smaller wire can be used to reach even more remote locations before the voltage drop becomes significant.

When battery operated, the pump and inverter together draw about 0.7 amp which will use about half the total capacity of a 10 amp-hr motor cycle battery in 8 hr or of a 70 amp-hr automobile battery in 48 hr. During an 8 hr period, the voltage of a new motorcycle battery dropped from 12.5 to 11.6 V with less than about 2% change in flow rate. The automobile battery will undoubtedly act similarly, but either battery should be recharged before taking another sample.

Effect of temperature

The effect of temperature is more difficult to assess and interpret. Initially, the outlet from the pump was connected to the Mylar bag through a flow meter. The entire assembly of pump, needle valve and flow meter were placed in a steel ammunition box only slightly larger than the assembly itself, the lid was closed tightly and the flow meter was read through a single small opening in the box periodically over a period of 2 days. The box was placed outside near the west wall of the laboratory building so that it would become cold during the night, warm up slowly during the forenoon hours when the box was in the shadow of the building, and finally become extremely hot during the afternoon when the hot summer sun shined directly on the box for several hours. Since the change in flow rate observed was less than 10-15% at most, no attempt was made to define temperatures quantitatively except to say that the steel box was so hot all afternoon that it could not be touched comfortably for longer than a few seconds at a time. At night, the air temperature was approximately 35°F. These temperature extremes are thought to be certainly greater than those that need be tolerated if the pump assembly is placed in a ventilated weather shelter to protect it from the direct rays of the sun.

In another attempt to evaluate the effect of temperature, the flow rate was determined by water displacement while the needle valve and pump were alternately heated and cooled. After an initial adjustment of the flow rate to 9.6 ml/min at 24°C, the flow rate decreased to 7.1 ml/min or about 26% when the needle valve was heated with an infrared lamp hotter
Fig. 1. Equipment used for taking integrated air samples for determination of $^{222}\text{Rn}$.
CLAUDE W. SILL

than could be touched comfortably for any significant period of time. When the needle valve was cooled as much as possible with an aerosol spray used to cool electric circuits, the flow rate increased to 10.6 ml/min or about 10%. Since the flow rate was measured by water displacement from a large tray of water at room temperature, the changes observed obviously reflect variations in mass transfer rather than volume. Both changes would undoubtedly have been smaller had the exit air remained more nearly at the temperature of the air being sampled. Since respiratory processes are more dependent on volume transfer than on mass transfer, it appears that the effect of temperature on the pump is not very different than its effect on the volumes breathed by humans. Again, no attempt was made to make a precise determination of a characteristic which is fundamentally imprecise in usage. It is unlikely that any reasonable temperature variation will introduce more than a 10-15% uncertainty between the volume sampled by the pump and that breathed by people.

DIFFUSION OF $^{222}$Rn THROUGH MYLAR

The use of Mylar bags as containers for air samples for the determination of $^{222}$Rn depends on their ability to retain radon for the several days required for sampling, transportation to the laboratory and analysis. To determine possible losses from diffusion, pin holes, faulty seams or whatever cause under actual conditions of use, a bag was filled with approximately 30 l. of air containing sufficient $^{222}$Rn to give an initial counting rate of about $4 \times 10^5$ gamma counts/min of daughter activity at equilibrium under the counting conditions employed. The bag of air was placed horizontally on a stretcher in a 9 ft cubical steel room used for whole-body counting and an 8 in. $\times$ 4 in. NaI(Tl) detector was placed over the center of the bag at a distance of about 12 in. The bag was allowed to stand overnight to ensure ingrowth of the radon daughters to complete equilibrium. After rechecking the experimental setup the following morning, the door to the vault was closed and sealed to prevent any traffic or air motion inside the vault that could possibly change the orientation of the bag and thereby change the counting efficiency. A 1 min. count was made immediately and was repeated at approximately the same time on each successive day for the next 6 days during which time the door to the vault was never opened. An additional measurement was obtained on the 15th day which had not been planned but which was available since the bag had not been moved from its original position in the meantime. However, the vault had been opened after the last planned experimental point.

The decrease in activity obtained is shown in Fig. 2. The dashed line shows the expected decrease of 3.823 day $^{222}$Rn from the initial activity due to radiological decay alone. The solid curve was drawn through the intermediate points which it fits better than anything that can be drawn to include the first and/or last points. A least-squares best fit of all the data gives an apparent half life of 3.60 days, corresponding to a maximum loss of about 1% per day from all causes other than radiological decay. However, it is interesting that the first and last points obtained while the vault was open show no detectable loss in 15 days and are distinctly different statistically than the 6 intermediate points that were obtained while the vault was sealed. It is entirely possible that electrostatic attraction of the charged ions being measured to the non-conducting walls of the Mylar bag is biased in some small unknown way by a stagnant environment. In any event, the slopes of the 2 groups of points are closer to each other than to the best fit of all the points and loss of $^{222}$Rn must be less than 0.5% per day which is more than adequate for the intended use. Even if the losses increase somewhat when the bags are flexed as expected, the overall loss should still be entirely negligible compared to the other uncertainties present.

Although the diffusion of $^{222}$Rn through rubber and most plastics is known to be several orders of magnitude higher than with Mylar, short pieces of gum rubber tubing are used to make connections because of its ready availability and because of its elasticity to ensure that the connections are air tight. The quantity used is so small and the residence time of the air in the tubing is so short that losses from this source are completely negligible. For example, measurement showed a loss of
AN INTEGRATING AIR SAMPLER FOR DETERMINATION OF $^{222}\text{Rn}$

![Graph showing the decay of $^{222}\text{Rn}$](image)

**Fig. 2.** Decay of $^{222}\text{Rn}$ contained in a Mylar bag.

about 5% per hr from a 2 ft piece of the $\frac{1}{4}$ in. tubing used. Since the bore volume is about 4 ml, loss from this source is only 0.03% for the entire 48 hr integration.

Lucite was used to re-encase the pump mechanism because of its low cost and ease of fabrication. To determine if this plastic would cause significant loss of radon from a given sample and contaminate a subsequent one, the following experiment was performed. Thirty liters of air containing 40 pCi/l. of $^{222}\text{Rn}$ was pumped from a Mylar bag containing 401. of the gas into an empty bag over a period of 48 hr. Analysis of the air in each bag showed that the concentrations of $^{222}\text{Rn}$ present before and after pumping did not differ by more than could be expected from the experimental uncertainties involved. The pump was then flushed quickly with 1.5 l. of aged air in about 10 min with the valve wide open to determine how much of the residual gas in the void space could be removed in a short purge. Aged air was then pumped from a Mylar bag into an empty one at 10 ml/min for 48 hr to determine the quantity of radon that would diffuse back into a subsequent low-level sample during the lengthy collection period used in the field. The radon found in the purge and subsequent eluate was about 2.5 and 0.5% respectively, of that taken in the original 30 l. of gas of which 1% is attributed to the free volume space of about 300 ml. This contamination would increase the concentration of a subsequent 30 l. sample by a maximum of 1.2 pCi/l. which can be reduced to 0.2 pCi/l. by a short purge with the valve wide open. In practice, the contamination will be even less due to decay between uses. Certainly, pumps used over tailings or in other areas of high radon concentrations should be purged before being used for sampling in low-level areas unless sufficient time is allowed for adequate radiological decay.

**PRACTICAL APPLICATION**

In cooperation with the U.S. Public Health Service and the health departments of the states of Colorado and Utah, a 1 yr program involving 57 stations in four cities was based on the philosophy and equipment described above and is nearing completion. Results of this
CLAUD3 W. SILL

program will be described in another publication. However, a few comments concerning the success of the program itself seem appropriate. The philosophy of employing 48 hr integrated samples to increase the reliability and decrease the cost of long-term measurements worked extremely well. Excellent coverage of the four study areas was obtained at minimum cost and results are clear, consistent, and singularly free of any significant anomalies. The sampling equipment performed almost as well in the field as it had in the laboratory with a few exceptions. Of 18 complete samplers in use, five of the $15 pumps burned out and had to be replaced. This was probably due to the low flow rate which caused a much higher back pressure than that recommended by the manufacturer. A more frequent problem encountered was the occasional sticking of the needle valves which would not retract when the handle was turned to the open position. On prolonged use, a very small amount of corrosion occurs that tends to fix the needle in its existing position so that the spring-loaded return is insufficient to break it loose and open the valve. This problem is easily rectified by removing the needle and wiping it gently with a dry cloth before each 48 hr run.

The Mylar bags have held up very well on repeated usage with each bag having been used an average of 8 times without showing signs of significant deterioration. The infrequent breaks that develop where the Mylar has been creased sharply, and even an occasional 6 in. tear, are easily and rapidly repaired with the sealing tape and hand iron used. The one really frustrating problem encountered in the entire program was the occasional obstinate refusal of some of the bags to fill up in the proper time even though it could be shown that air was entering the bag at the proper rate. The only reasonable explanation is the presence of pin holes in the bags or rubber tubing, minute leaks in the seams or valves or a faulty connection to the valve. Obviously, it would not take much of an opening to permit the gas to bleed off as fast as it is collected, considering the very low rate involved. Some of the leaks are extremely difficult to find. If a bag is partially filled with air and then tested in the conventional manner by immersion in a large container of water, some of the leaks, particularly in the seams, will close off when pressure is applied and then open up again when the pressure is relaxed to the conditions under which the samples are collected. Leaks from very small pin holes that can be observed if covered with a very thin layer of water or soap solution will stop if immersed under a greater depth of water. However, most leaks are readily detected by simple immersion in a vessel of water and this method is used routinely to leak test all bags in the laboratory after the analysis has been completed before they are returned to the field.

The same philosophy and sampling equipment will undoubtedly find many other applications in air pollution control.

Acknowledgements—The author acknowledges the many timely suggestions and assistance that he has received from his associates in the laboratory. Particular thanks are due M. Wilhelmsem, D. Parker and P. R. Boren for assistance in procurement of the instrumental components, to D. G. Olson and D. R. Percival for much of the data, and to P. R. McKnight and W. E. Bush for solving many of the initial problems of field implementation.

REFERENCES
12. S. D. Shiferar, Jr. and C. W. Sill, Evaluation of atmospheric radon in the vicinity of uranium mill tailings, to be published.