

Average radon concentrations for an extended period can be determined using a programmable gas bag sampler and Lucas cells.

A Programmable Gas Sampler to Measure the Average Radon Concentration in a Test Chamber or in the Field

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Abstract: A Tedlar gas sampling bag was used to retain samples of the test environment of interest. Samples, typically hourly, were transferred into a gas sampling bag. The volume of each sample was determined by a programmable timer or any computer capable of executing a compiled basic program. The program automatically adjusted the sample volume of each increment to exactly compensate for radioactive decay of radon referred to the end of the exposure period. At that time the radon activity collected in the sampling bag was the exact average for the whole period. A single Lucas cell was used to report the average concentration for typically 2 and 3 day exposures. The results compared with calculated predictions and with the average of a series of grab samples taken during the measurement period. The benefit of this method was the large work load reduction needed to determine the average concentration from a series of grab samples and the ability to employ Lucas cells, which are inherently more stable and less expensive than active measurement equipment normally used for cham-

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The radon measurement industry is primarily interested in radon risk to inhabitants in homes, schools, and work places. Since health risk accumulates with time, longer term measurements are more important than instantaneous radon assays. Accurate measurements of the average radon concentration over days and years is complicated by the variation of radon concentration with time. Three mechanisms that most perturb the radon concentration are the well known diurnal radon concentration cycle, dilution by drafts, and normal radioactive decay.

Radon measurements are usually performed by three types of detectors. They are electronic instru-

ments that continuously sample the radon concentration, record each value in memory, and calculate the average at the end of the exposure. Integrators such as Eperms, or alpha track devices, or semi-integrators such as activated charcoal detectors as described by George (1984) are used. The reader is referred to EPA (1993) for a discussion on detectors used for radon measurements.

For calibration purposes electronic continuous radon detectors are frequently used to define the radon fields that the other detectors are exposed to. The quality of these test exposures depends on the calibration and stability of the continuous radon measuring device used to define the radon field. Good calibration facilities deploy more than one continuous monitors to confirm the results, and the devices must be calibrated at least annually. This imposes an unacceptable cost of much more than \$10K to management of modest calibration chambers (George 1990).

A better type of continuous monitor should contain a radon source that could be used to more frequently standardize calibration instruments to eliminate both short and long term errors. Some Lucas cells (Lucas 1964), are available with internal radium sources such

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as the Pylon 3150A calibrator.† Evacuated Lucas cells are frequently used for short term "grab" samples to determine near instantaneous radon concentrations. If it were possible to accumulate an air sample composed of many smaller samples taken over time and compensated for decay, one or more Lucas cells could be used to measure the average radon concentration over several days with accuracy and precision rivaling electronic instruments and exceeding the continuous monitors by the ability to normalize to a radium source sealed within a Lucas cell. The purpose of this paper is to describe such a system.

THEORY

For any sample i of air or gas containing radon the amount of radioactivity is

$$A_i = C_i \times V_i \quad (1)$$

where C is the radon concentration, and V is the sample volume. After time T :

$$A_T = A_i e^{-\lambda T} \quad (2)$$

For n samples pumped to some container and summing from one to n :

$$\Sigma A_T = \Sigma A_i e^{-\lambda(T-t_i)} \quad (3)$$

The total of A sub T increments is composed of samples of uncertain radioactive content since radon varies with time. The total is further perturbed since each increment undergoes a different amount of radioactive decay leading to a total sample that does not reflect the original exposure. The purpose of this paper is to describe a method to exactly compensate for these uncertainties. What was required is a system of samples that retains the original individual radioactivity at a given time such as the end of a test exposure. This is easily accomplished by increasing each sampling volume to contain additional radon activity to exactly compensate for radioactive

decay to a reference time such as the end of the test. At time T :

$$\Sigma A_i = \Sigma C_i e^{-\lambda(T-t_i)} \times \Sigma V_i e^{\lambda(T-t_i)} \quad (4)$$

$$\Sigma A_i = \Sigma A_i e^{-\lambda(T-t_i)} = \Sigma A_i e^0 \quad (5)$$

Let:

$$V_T = \Sigma V_i e^{\lambda(T-t_i)} \quad (6)$$

The concentration of radon in the sampling container at the end of the exposure is:

$$C_T = \Sigma A_i / V_T \quad (7)$$

However, the sampling volume is known and can be used to adjust the physical concentration to the total concentration due to the average concentration for the test exposure:

$$C_i(\text{avg}) = \frac{\Sigma A_i}{V_T} \times \frac{V_T}{\Sigma V_i} = C_T \times$$

$$\frac{V_T}{\Sigma V_i} = C_T \times \text{CONSTANT} \quad (8)$$

The constant is the ratio of the total adjusted volume to the unadjusted volume. For a pump set for constant flow rate, the same constant is the total adjusted flow time to the total, baseline, unadjusted flow time:

$$\text{Constant} = \frac{\Sigma_{0 \rightarrow n} e^{\lambda(T-t)}}{n} = \frac{V_T}{nV_i} \quad (9)$$

HARDWARE

From the foregoing, what was needed was a means to control

sample volume. A computer used to drive a sampling pump was a logical choice. Software could automatically adjust each sampleflow duration for any number of total increments comprising the total sample.

Fig. 1 is a schematic representation of the sampling system. A small pump capable of a flow rate between 0.3 and 0.5 L min⁻¹, was convenient. This could be a battery-powered diaphragm pump or a small air pump capable of reproducible flow rates used to aerate fish tanks. If a fish tank air pump was used, the pump must be placed within the sampled environment. Diaphragm pumps with separate inlet and outlet connections allow greater flexibility. A check valve is not shown but may be needed if the sampled environment is at a higher pressure than the sampling bag. For significant pressure differentials, an electrically operated valve may be required.

A flow meter was used to adjust flow and to verify consistent flow rate so as to not over fill the gas sampling bag. Plastics are known to be permeable to gases including radon. A 20-L 4-ml-thick Tedlar gas sampling bag was obtained from the Cole-Parmer Instrument Co.‡ The bag was tested before use. The bag was filled with air containing

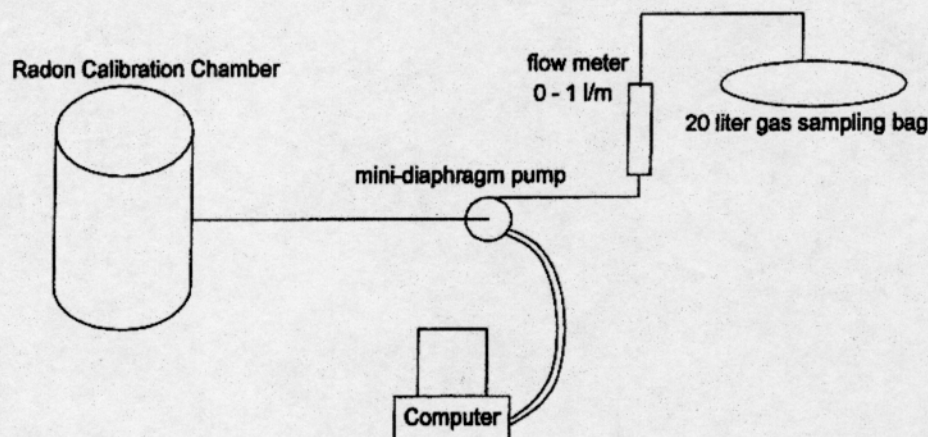


Figure 1. Gas bag sampling device.

‡ Cole-Parmer Instrument Co., 625 East Bunker Court, Vernon Hills, IL 60061, catalog 1999-2000.

† Pylon Electronic Development Co., 147 Colonnade Road, Ottawa, Ontario, Canada, K2E 7L9.

about 463 Bq m^{-3} (12.5 pCi L^{-1}) of radon. Four minutes after filling, an evacuated 0.3-L Lucas cell was used to draw a sample from the bag. This was repeated once per day for the next 5 d. Within a measurement uncertainty of 2.7%, no loss was observed not accounted for by radioactive decay. The Cole-Parmer catalog contained gas permeability data for plastic tubing. The data were for carbon dioxide, hydrogen, oxygen, and nitrogen. Different plastic tubing had permeability values that varied by more than a factor of 1,000, which required care in selection of tubing material. The authors used copper refrigeration tubing where ever possible. The approximately 1-mm bore tubing had an internal volume of less than 4 cc per meter of length. For a nominal flow rate of 0.3 L m^{-1} and minimum time of 20 s, the tubing trapped less than 4% of a minimum sample increment. The trapped volume was not lost, but transferred by the next sample; so the dead volume uncertainty was less than 4% divided by n samples.

Any computer with an internal speaker that is able to run a compiled basic program could be used. The author employed a 286 computer assembled in 1986; an audio output was needed but a sound card was not. Simplicity and economy were motivating factors in the design of the sampling system. Employing an electrical signal taken from the computer speaker terminals and using the standard DOS operating system minimized changes to the computer. A small external interface was designed to drive the air pump, Fig. 2, so tone control was pump control. The band pass of the input transformer may be limited. The authors suggest testing with tones of different frequency for solid operation of the driver; 600 Hz was used for the driver. The driver exhibited a 2-s dwell time after the termination of the audio signal, and this was compensated for by software. All of the

components were readily available from any Radio Shack store.

SOFTWARE

Fig. 3 is a printout of the Quick Basic 4.5 source code that was used to operate the sampler. The code was compiled, with all libraries, and the exe. File was placed on the C drive of a computer. Typing the name of the execute file loaded, started the program, and displayed the options shown as input statements on Fig. 3. The user could choose to print out the starting time and date, cycle time for each incremental sample, an identifier for the measurement, and to select the duration of the test in days. The operating kernel was the Basic statement: sound frequency, duration, where the frequency could be varied from 32 to 32,767 Hz, and the duration from 0 to 65535 ticks. To convert ticks to seconds divide by 18.2 ticks per second. The statement

$$t = 18.2 \times 20 \times \text{EXP}(.693 \times \{\text{dys} - [\text{numr}/(60 \times 24)]\}/3.84), \quad (10)$$

calculated the number of BASIC ticks required for any increment. During the last cycle the tone was turned on for the minimum time of 20 s; since $(\text{dys} - \text{numr}) = 0$. For any other time, the cycle time was increased to exactly compensate for radioactive decay of radon. The half life of radon was taken to be 3.84 d, and λ was $0.693/3.84$ per day.

DISCUSSION

The quality of this sampling system was related to the quality of the Lucas cells and associated readout electronics. Two Pylon 300A cells and a Pylon AB-5 instrument were tested by filling the cells from a fixed concentration of radon contained in the gas bag. The time difference for filling the first to last cell was less than 2 min; hence the radon concentration in the cells was taken to be the same. Two minutes of radioactive decay is too small a difference to measure. The 2 cells were counted sequentially by normal mounting and dismounting the individual cells from the Pylon AB-5 instrument. After the first counting cycle the cells were remounted and counted again a day later. This was repeated for 3 counting cycles for each of the 2 Lucas cells over a period of two days. Correcting for decay, cell No. 841 had a measured repeatability of $\pm 0.8\%$ and cell No. 842 was 2.3%. Much of the error may have been due to counting uncertainty. Apart from errors associated with the original cell calibrations, it was concluded from this test and from the earlier mentioned gas bag leakage test that it was realistic to expect measurement uncertainties to be less than about 3% for radon fields of slowly varying concentration. Longer term errors resulting from changes in the Lucas cells or the AB-5 instrument were reduced by using

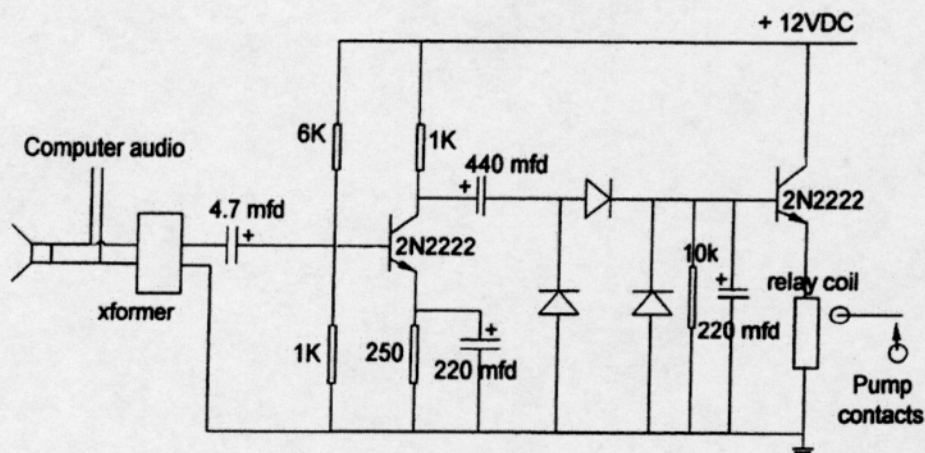


Figure 2. Pump driver.

```

CLS
DIM id AS STRING
numr = 0: min = 0
INPUT "To printout cycles type 'P' or type enter for no printout"; p$
IF p$ = "p" OR p$ = "P" THEN p = 1 ELSE p = 0
CLS
INPUT "Enter gas bag accumulation time in days", dys: CLS
INPUT "Identify this test"; id$: CLS
LOCATE 1, 1: PRINT DATES$,
PRINT USING "# day sample"; dys;
PRINT id$
LOCATE 15, 15: PRINT "Type lower case 'x' to end test early"
IF p = 1 THEN LPRINT DATES$
IF p = 1 THEN LPRINT USING "Sample is # days"; dys;
IF p = 1 THEN LPRINT id$: LPRINT "Start time"; TIMES$: LPRINT

total = dys * 60 * 24
TIMER ON
ON TIMER (60) GOSUB Display
DO WHILE INKEY$ <> "x": LOOP
END

Display:
Oldrow = CSRLIN           'Save current row.
Oldcol = POS(0)           'Save current column.
LOCATE 3, 3: PRINT "The time is"; TIMES$
alarm$ = MID$(TIMES$, 4, 5)
GOSUB tone
LOCATE Oldrow, Oldcol 'Restore row & column.

RETURN

tone:
numr = numr + 1: min = min + 1
LOCATE 4, 3: PRINT "minutes are"; min
IF min >= 60 THEN min = 0 ELSE RETURN
t = 18.2 * 20 * EXP(.693 * (dys - (numr / (60 * 24)))) / 3.84
LOCATE 6, 1: PRINT "Cycle hr."; numr / 60,
LOCATE 6, 30: PRINT USING "t= ###.##"; t
IF p = 1 THEN LPRINT "Cycle hour ="; numr / 60,
IF p = 1 THEN LPRINT USING "Cycle length in sec. = ##.##"; t / 18.2
tc = (t - 36.4) 'to correct for 2 seconds lag at end of each cycle
SOUND 600, tc
IF numr >= total THEN END

RETURN

```

Figure 3. Program software.

a calibration cell consisting of a sealed radium doped Lucas cell, Pylon model 3150A. The calibration cell was routinely counted before and after every measurement set, and the average used to adjust the results of radon determination. Long term changes in the calibration cell were corrected for buildup of ^{210}Po . Several years worth of Lucas cells intercomparison measurements were made with the USDOE Environmental Measurements Laboratory Chamber (George et al. 1992–1995) and more recently other radon chambers that compare with the USEPA radon chamber in Las Vegas, NV (Bowser-Morner 1997).

The TCS radon test chamber is a one-pass flow-through system with an adjustable air change time of >15 min to <4 min. Radon is derived from a flow through 102

kBq radium cell, Pylon model 1025. Flow meters monitor the air flow through the Pylon radon cell nominally operated at 0.8 L m^{-1} and in the much larger dilution stream. Pressure measurements are used to insure the chamber is operated slightly above ambient pressure to eliminate unmonitored dilution as any leakage is to the outside. Routine air flow, humidity, and temperature measurements are made. A radon calibration system has been in service since 1985; several months before TCS started a commercial radon measurement business. Over this extended period, reproducible flow rates and corresponding radon concentrations were routinely observed. This consistency was helpful in evaluating the gas bag sampling system. Tests were conducted over 2 to 4 day

accumulations by drawing 1 or more Lucas cell samples directly from the test chamber each day. At the end of the exposure, Lucas cell samples were also drawn from the gas bag. The measured radon concentration ratio of the test chamber to the gas bag closely compared with the value predicted by calculations. A Honeywell continuous radon monitor was also deployed in the test chamber. The standard deviation of the hourly data derived from the Honeywell greatly exceeded the uncertainty associated with the Lucas cell data taken at the same time; so the Honeywell data could not be used for a proof of principal. The predicted ratio was calculated with a spread sheet program such as Lotus. The sample duration for each hourly step at constant flow rate was adjusted by:

$$\text{EXP}[\lambda(T-t)]. \quad (11)$$

Where λ is $0.693/(\text{radon half life})$, T is the total exposure time, and t is the time remaining for the test. The ratio is the sum of the above adjusted value divided by the number of steps. Calculated ratios are 1.24 and 1.34 for 48- and 72-h exposures. The true average exposure in the radon test chamber is therefore the concentration determined in the gas bag by one or more Lucas cell samples times the ratio appropriate for the duration of the exposure to compensate for the total extra air volume.

The gas bag sampling system is routinely used to determine average radon concentrations during 2-, 3-, and 4-d radon "spike" exposures for activated charcoal detectors. Spike exposures are defined in EPA (1993). The following is a partial copy of a printout made during a 3-d exposure:

Use of the gas bag sampling system in unknown radon fields of varying concentration is also possible. Three model radon fields were tested mathematically. The first was a sinusoidally varying field with one maximum of 185 Bq m^{-3} and a minimum of 37 Bq m^{-3} per 24-h period:

08-02-1999

Sample is 3 days

Spike G cans start 8/2/99

Start time 13:51:48

Cycle hour = 1

Cycle hour = 2

Cycle hour = 3

Cycle hour = 70

Cycle hour = 71

Cycle hour = 72

Cycle length in sec. = 34.1

Cycle length in sec. = 33.9

Cycle length in sec. = 33.6

Cycle length in sec. = 20.3

Cycle length in sec. = 20.2

Cycle length in sec. = 20.0

$$\text{Bq m}^{-3} = 111 + 74\sin(t \text{ pi}/12). \quad (12)$$

The second was a monotonically decreasing radon concentration starting at 370 and declining to 37 Bq m⁻³ over 48 h:

$$\text{Bq m}^{-3} = -(333/48)t + 370. \quad (13)$$

The last case was six sequential fields of constant radon concentration as follows:

Radon Concentration Bq/m3	Duration hrs
370	14
259	5
111	5
555	4
740	8
222	12

For the sinusoidal and triangular cases the mathematical samples were taken on the hour. For the six box case the sampling was retarded by 15 min except for the final sample to not exactly correspond to the changing field. The results were as follows:

Field Description	Ratio
TCS chamber	1.235
Sinusoidal	1.227
Triangular	1.226
Six boxes	1.205

The TCS chamber ratio was measured by taking the average of a series of Lucas cell grab samples directly from the radon chamber and dividing by a single Lucas cell sample taken from the gas bag at the end of the exposure. For the three constructed fields, the mathematical average of each field was compared with the associated predicted concentration in the gas bag due to hourly increments. Sinusoidal changes probably reflect the real environmental changes best. The results indicate the programmable gas sampling system can be used accurately for any normal varying radon field.

CONCLUSION

Lucas cell detectors, normally classified as grab samplers, could be used for radon measurements extended over several days. When Lucas cells were used with a programmable gas bag sampler, measurement precision better than 5% were not difficult to achieve. Absolute accuracy depended on the calibration of the Lucas cells, the radon concentration variation with time, and the number of sampling

steps during a determination. For slowly varying radon fields, it was concluded the programmable gas bag system could reliably be used to determine the average radon concentration over several days with a precision of <5%. Further, the system could be used to measure the average radon concentration anywhere with an additional uncertainty of about 3%.

The cost of all of the components was estimated to be <300\$ exclusive of the Lucas cell system.

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