

Improved Calibration and Moisture Correction Factors
for Carbon Canisters Used for Radon Monitoring

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ABSTRACT

The U.S. Environmental Protection Agency's standard operating procedure for the charcoal canister method of determining airborne radon-222 concentration has some serious moisture correction problems. The problems with that methodology show up especially if the canisters are used under conditions of very high or very low humidities. Furthermore, in using their humidity adjustment curves, slight weight gain difference requiring the use of different calibration curves can cause the calculated radon activity to differ by as much as a factor of 2.6. The authors have developed an improved methodology whereby the same canisters, counting equipment and similar system calibration procedures can be used to obtain more consistently reliable results.

We, as others, have found that the rate at which radon is absorbed on the charcoal in the canisters is affected by the amount of water and radon already

absorbed in the charcoal. We have found that under low humidity conditions and starting with oven dried canisters, the radon concentration in a canister will reach an equilibrium value which is directly proportional to the average ambient radon concentration. Furthermore, we find that a single moisture correction factor curve, based on canister weight gain, can be used to correct the measured canister activity to the low moisture condition whereupon the average radon concentration can be directly calculated.

Thus we recommend the use of oven dried canisters and an exposure time sufficient for the canisters to approach equilibrium with the environmental radon, which is three to seven days in our test. Our paper also shows how to obtain the moisture correction factor curve from procedures similar to the EPA recommended calibration procedures.

KEY WORDS

Radon Monitoring, Charcoal Canister, Humidity Effects, Exposure Time, Effective Half-Life

INTRODUCTION

The U.S. Environmental Protection Agency's standard operating procedure for the activated charcoal canister method of determining airborne radon-222 concentrations has some serious problems. This is especially true if the canisters are employed under conditions of very low or very high humidity. The authors have developed an improved method in which the same canisters can be employed, but more reliable results can be obtained under all humidity conditions.

EPA PROCEDURES

The EPA procedures are given in a 1987 publication of their Eastern Environmental Radiation Facility (EERF) (EPA 1987). This publication, entitled "EERF Standard Operating Procedures for Rn-222 Measurement Using Charcoal Canisters," is based on the work of A.C. George of The U.S. Department of Energy's Environmental Measurement Lab (George 1984). These procedures describe the specifications for the standard EPA canisters, which are now available from several commercial sources. They also describe recommended detector calibration, radiation counting, and radon concentration calculation procedures. Canisters are weighed and employed as received from the manufacturer. No effort is made to remove the several grams of water absorbed on the charcoal during the manufacture of the canisters. After exposure, the canisters are again weighed to determine the water absorbed during the exposure period. The canisters are employed only once and are then discarded.

Airborne radon concentrations are determined using two types of calibration curves. The first is a water gain versus calibration factor for a two day canister exposure time and covers a weight gain from zero to 19 grams of water. The second calibration curve plots exposure time versus humidity

adjustment factors for conditions of low (20%), medium (50%) and high (80%) relative humidity. One uses the low humidity curve if the canister gains less than 1.0 grams, the medium humidity curve if the canisters gains between 1.0 and 4 grams, and the high humidity curve if the canister gains greater than 4 grams. The problems with this technique are as follows: Canister weight gained is in part a function of the amount of moisture already on the canister at the beginning of the exposure period, but this is not determined in the EPA procedures and may vary with manufacturers and batch to batch from the same manufacturer. Weight gain is also a function of exposure time, and the procedures, as written, allow for an exposure time of up to six days. Furthermore, if the humidity is sufficiently low, the canisters may actually lose weight during exposure, and the procedures do not take this into account. Also, depending on the exposure time and the initial moisture content, it is possible under low humidity conditions to end up with a weight gain large enough to require use of the medium humidity curve and under medium humidity conditions end up using the high humidity curve. These curves are sufficiently different such that 0.1 gram difference in weight requiring a shift in calibration curves could change the final results by as much as a factor of 2.6. Such potential errors are too large to make this an acceptable procedure.

PENN STATE CONCEPT

In our procedures, the same standard EPA canisters are employed and they are exposed and counted using the EPA procedures. The changes in procedure include predrying the canisters to nearly constant weight by opening them and heating them for six to eight hours at a temperature of 65°C. The canisters are also reused after sufficient time is allowed for the decay of any absorbed radon (>30 days). The big change involves the radon concentration calculation

procedures. These procedures are based in part upon work of Cohen and Nason, (Cohen and Nason 1986) who demonstrated that under low humidity conditions the movement of radon gas through a diffusion barrier and onto charcoal follows the well known activation equation, and that when a canister containing radon absorbed under dry air conditions is then exposed to low humidity air having a low radon concentration, the canister activity will exponentially decrease like the well known radiation decay equation. Under these two conditions, the radon gas has the same effective decay constant, which is the sum of the true decay constant of radon-222 and a constant determined by the rate at which radon passes through the canister diffusion barrier. Thus, while the true half-life of the radon-222 is about 92 hours, the effective half-life of the radon on the canisters used in this work and exposed under these conditions is only about 30 hours.

When one exposes canisters to air above a relative humidity of about 20% for longer than one day, the amount of radon absorbed on the cartridge is found to be less than that obtained under dry air conditions. The greater the humidity, the greater the reduction in radon activity. This reduction may be due to a reduction of the number of charcoal absorption sites available to the radon and/or to the partial blocking by the water molecules of the pores leading into the interior of the charcoal granules. We have determined that one can use procedures similar to those used to obtain the EPA calibration curves to generate a single moisture correction factor curve, based on canister weight gained above its dry weight, to correct for the loss in canister efficiency due to humidity effects.

EXPERIMENTAL EQUIPMENT AND TEST PROCEDURE

All of the results reported in this study involve the exposure of opened charcoal canisters in a small radon calibration chamber assembled by the

authors. The canisters are 10 cm (4 inch) diameter, open face EPA design (F&J Specialty Products, Inc., P. O. Box 660065, Miami Springs, Florida 33266-0065). The chamber was constructed from a 19 liter (five gallon) steel pail, sealed with a steel lid and containing a shelf for the positioning of a charcoal canister in the center of the chamber. Due to the small chamber size only one canister is placed in the chamber per test to reduce the possibility of a lowering of the chamber radon concentration due to radon adsorption by the use of multiple canisters.

Dry air is produced by drawing air through a desiccant and then pumping it through a Pylon model 1025 radon source (Pylon Electronic Development Company, Ltd., 147 Colonnade Rd., Ottawa, Ontario, Canada K2E 7L9). The radon output generated by this source is 12.6 Bq min^{-1} (340 pCi min^{-1}). The radon concentration in the air leaving the source is a direct function of the airflow rate through the radon source. The radon containing air then passes through a rotometer to measure the airflow rate. During a test, periodic checks are made of the rotometer reading to insure that the flow rate stays constant and at the desired level. The radon containing air is then pumped through an inlet located at the bottom of the exposure chamber and then out the top of the chamber lid and into a ventline. In order to confirm the calibration of the Pylon radon source and the rotometer, air samples from the exposure chamber are periodically collected and assayed using a scintillation cell and counting system which has been calibrated against radon-222 gas emitted from a National Institute of Standards and Technology traceable radium-226 solution.

Constant relative humidity conditions are obtained by placing on the bottom of the exposure chamber one or more open containers of water having sufficient liquid to last through the exposure period. The humidity inside

the chamber is controlled by the amount of liquid surface area exposed to the chamber interior. The humidity and temperature of the air leaving the chamber are determined regularly during a test using an OMEGA Rh-201°C Humidity and Temperature Gauge (OMEGA Engineering, Inc., P. O. Box 4047, Stanford, CT 06907-0047). As needed, a water cooled condenser is used to maintain uniform air temperature in the chamber.

The following procedures are used to produce successive measurement from a canister for each experimental run. Every one to three days the exposure chamber is opened, the open canister is quickly removed and its lid replaced and sealed. The exposure chamber is also quickly resealed to help maintain test conditions. The canister is then counted using a standard NaI(Tl) detector connected to a Nucleus PCA computer based pulse height analyzer (The Nucleus Inc., 791 Emory Valley Road, P. O. Box 2561, Oak Ridge, TN 37831-2361).

In a half hour or less after the canister is removed from the exposure chamber, the canister is again opened and reinserted into the exposure chamber. The time during which the canister is out of the exposure chamber is not included in the determination of its exposure time. Since the time it is out of the chamber is small compared to the total exposure time and to the half-life of radon-222, no other correction needs to be made to account for the fact that the canister was periodically removed from the chamber during a test run.

PROCEDURES AND DEFINITION OF TERMS

1. Decay Factor

The standard decay factor (DF) is determined from the end of the exposure period to the beginning of the counting period and is calculated as follows:

$$DF = e^{-(\lambda_r t_d)} \quad (\text{dimensionless}) \quad (1)$$

where:

λ_r = the decay constant of radon-222 (time⁻¹)

t_d = the time (in the same units as λ_r) from the end of the exposure period to the start of the counting period (time)

2. Counting time correction factor.

The counting time correction factor (CTF) is calculated as follows:

$$CTF = \frac{1 - e^{-(\lambda_r t_c)}}{\lambda_r} \quad (\text{minutes}) \quad (2)$$

where t_c = counting time in the same units as λ_r .

When counting time is much shorter than the half-life, such as a 15-minute count time compared to the 3.8-day half-life of radon-222, then:

$$CTF = T_c \quad (\text{minutes})$$

3. System calibration factor in dry air.

The system calibration factor in dry air CF(d) is determined by exposing, in the calibration chamber, the canisters to a known radon concentration (RN) in dry air. The equation used to determine each point on the calibration curve is as follows:

$$CF(d) = \frac{C(d)}{(CTF)(RN)(DF)} \quad (\text{cpm} \times \text{Bq}^{-1} \times \text{L}) \text{ or } (\text{cpm} \times \text{pCi}^{-1} \times \text{L}) \quad (3)$$

where:

$C(d)$ = the net counts in the region of interests (ROI) for the canister exposed to radon gas in dry air for time t_e (days) (the gross counts minus the background counts for that day)

RN = exposed radon concentration ($Bq \times L^{-1}$) or ($pCi \times L^{-1}$)

The experimentally determined values of $CF(d)$ are then fitted to the following equation to determine the correct values for $CF(\infty)$ and λ_s .

$$CF(d) = CF(\infty)(1 - e^{-\lambda_s t_e}) \quad (\text{cpm} \times Bq^{-1} \times L) \text{ or } (\text{cpm} \times pCi^{-1} \times L) \quad (4)$$

where:

$CF(\infty)$ = the dry air calibration factor for an infinite exposure time

$\lambda_s = \lambda_r + \lambda_d$ ($days^{-1}$)

λ_r = the decay constant of radon-222 ($days^{-1}$)

λ_d = the diffusion constant of radon into or out of the canister ($days^{-1}$)

Figure 1 is a plot of three data sets. Canisters A-157 and A-71 were tested under dry conditions and canister A-122 was tested with the relative humidity varying in the range of seven to eight percent relative humidity. The best fit expression for this combined data set is:

In SI units:

$$CF(d) = 2780(1 - e^{-0.605 t_e}) \quad (5)$$

and in conventional units:

$$CF(d) = 103(1 - e^{-0.605 t_e}) \quad (6)$$

Thus $CF(\infty) = 2780 \text{ cpm} \times Bq^{-1} \times L$ ($103 \text{ cpm} \times pCi^{-1} \times L$)

since $\lambda_s = 0.605 \text{ days}^{-1}$ the effective half life for this combined data set is 1.15 days. (27.5 hours)

Assuming no humidity effects, one can now determine the proper CF(d) for the actual exposure time of a canister. But since humidity will always be present under real world conditions, one must add a humidity correction factor.

4. System calibration factor in humid air.

The system calibration factor in humid air CF(h) is determined by exposing, in the calibration chamber, the canisters to a known radon concentration (RN) in air of a known humidity. The percent humidity in the chamber is directly related to the increase in canister mass during the exposure period. The equation is as follows:

$$CF(h) = \frac{C(h)}{(CTF)(RN)(DF)} \quad (\text{cpm} \times \text{Bq}^{-1} \times \text{L}) \text{ or } (\text{cpm} \times \text{pCi}^{-1} \times \text{L}) \quad (7)$$

where C(h) = the net counts for the canister exposed to humid air of radon concentration RN

Figure 2 shows a plot of CF(h) as determined for seven different percent relative humidity conditions (%RH).

5. Humidity correction factor.

The humidity correction factor (HF) corrects the system calibration factor obtained from the dry air exposure for loss in radon absorption ability due to moisture absorption as follows:

$$HF = \frac{CF(h)}{CF(d)} \quad (\text{Dimensionless}) \quad (8)$$

Note: The value of this factor should vary between one and zero.

This factor is determined for at least three humidity conditions over the expected humidity range and a curve is generated of this factor versus canister mass gain during exposure. A polynomial least squares fit of the data is then obtained. Thus for any mass gain, the correct loss in absorption efficiency can be obtained from this curve.

Figure 3 shows the plot of 62 humidity factors calculated from 12 different sets of canister measurements. Different charcoal canisters and different humidity conditions were employed for each of the data set. Over the period of time these measurements were taken, ambient temperature varied from a low of 16°C (61°F) to a high of 31°C (89°F). Only data collected from the third day on was used in generating this curve. Despite the potential for errors from various sources such as canister variations, temperature effects and fluctuation in system radon-222 flow rate, Fig.3 shows that there is a remarkably small amount of scatter of the data about the fitted line.

The best fit polynomial for the data in Fig.3 was found to be

$$HF = 1.072 - 8.63 \times 10^{-2}(w) + 3.32 \times 10^{-3}(w^2) - 5.76 \times 10^{-5}(w^3) \quad (9)$$

(dimensionless)

where W = increase in canister weight (grams)

Since the HF cannot exceed unity, it can be shown by the above equation that HF will remain unity until a weight gain increase of about 0.8 grams.

One could obtain a very similar HF curve if only the data from the 28-30% RH, the 47-51% RH, and the 79-87% RH curves in Fig.2 were employed in conjunction with the data from a single 0% RH curve. Thus the amount of effort needed to obtain the HF correction factor is not much more than is required to generate the EPA recommended Adjustment Factor Curves (EPA 1987).

6. Calculation of radon concentration from a canister exposed to an unknown radon concentration.

With the various calibration factors calculated previously, the environmental radon concentration $Rn(u)$ to which a canister has been exposed can now be determined from the count rate recorded in the two regions of interest as follows:

$$RN(u) = \frac{C(u)}{CTF \times CF(d) \times HF(u) \times DF} \quad (\text{Bq} \times \text{L}^{-1}) \text{ or } (\text{pCi} \times \text{L}^{-1}) \quad (10)$$

where:

$C(u)$ = net counts in the ROI for the sample radon canister.

$HF(u)$ = humidity correction factor based on increased canister mass.

OTHER OBSERVATIONS AND TESTS

As seen in Fig.2, the radon concentration changes most rapidly with exposure time over the first two to three days. In developing the humidity factor curve, it was found that there was considerable fluctuations in the factors calculated from the data collected over the first two to three days. It appears that during this period of time, the amount of radon adsorbed on the charcoal is most sensitive to variations such as canister or temperature

differences. Yet most procedures for charcoal canisters recommend only a one or two day exposure time. With exposure times of three days or more the humidity factor appears to be independent of canister differences and temperature differences (over the temperature range tested). When one normalizes the data to the dry exposure conditions, the humidity factor is also independent of exposure time.

These charcoal canister procedures were also tested under fluctuating radon concentration conditions more typical of home radon measurements. Because of the decay of radon-222 adsorbed on the charcoal, charcoal canisters are not true integrators of the radon dose. Also used in these tests were E-Perms using short term electrets. (Rad Elec, Inc., 5330J Spectrum Drive, Frederick, MD 21701) This type of detector operates as a gas filled ion chamber and is calibrated to act as a true radon dose integrator. Once each day from the third day to the seventh day of exposure, the detectors were closed, removed from the test room and measured. Then they were quickly returned to the test room and reopened. The results of one of these tests are shown in Table 1. For the E-Perms the average radon concentrations were calculated for both the total exposure time and the exposure time between successive readings. As can be seen from this table, there is good agreement between the results obtained from the charcoal canister and those obtained from the two E-PERM detectors.

CONCLUSIONS

The charcoal canister procedures developed at Penn State have the following advantages over the EPA recommended procedures. They have a better theoretical basis for the correction factor. They are better able to handle low humidity conditions. The use of oven dried charcoal canisters allows the

use of a single correction factor determined from the weight gain of the canister, to account for humidity corrections. Thus they avoid the large discontinuity in determined values related to canister weight gains of 1.0 and 4.0 grams. They avoid uncertainties resulting from one or two day exposure times and they allow for the reuse of the charcoal canisters.

REFERENCES

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TABLE AND FIGURE CAPTIONS

TABLE 1. Comparison Between Two E-Perm Detectors and A Charcoal Canister
Employing The Penn State Method

FIGURE 1. Radon Adsorption in Charcoal Canisters Under Low Humidity
Conditions as a Function of Time

FIGURE 2. Radon Adsorption in Charcoal Canisters as a Function of Time and
Humidity

FIGURE 3. Humidity Factor (HF) as a Function of Charcoal Canister Weight Gain

RADON CONCENTRATIONS $8q \times L^{-1}$

Exposure Time (days)	E-PERM Detectors				Charcoal Canister Value
	Ave. over total time		Value between measurements		
	#1	#2	#1	#2	
3.10	0.80	0.75	0.80	0.75	0.92
3.91	0.80	0.76	0.80	0.81	0.87
4.82	0.80	0.77	0.83	0.83	0.76
5.67	0.82	0.79	0.91	0.88	0.87
6.45	0.81	0.78	0.75	0.76	0.79
Ave. $\pm 1\sigma$	0.81 ± 0.01	0.77 ± 0.02	0.82 ± 0.06	0.81 ± 0.05	0.84 ± 0.07





