1994_16

RADON MEASUREMENT QA - PRACTICE vs PROTOCOLS

Raymond Johnson, Sandy Colon, and Douglas Heim,
Key Technology, Inc.
Jonestown, PA

ABSTRACT

The EPA radon measurement device protocols of July 1992 specify several recommendations for quality assurance, which are now mandated as requirements by several states. These QA requirements include 1) annual calibration of measurement systems, 2) known spike measurements at a rate of 3 per 100 (minimum of 3 per year or a maximum of 6 per month), 3) duplicates at a rate of 10 percent of devices deployed or a maximum of 50 per month, and 4) blanks at a rate of 5 percent of devices deployed or a maximum of 25 per month. While these requirements may have general merit for radon measurements, we propose that they are not all needed, practical, or sensible for a primary charcoal canister laboratory. For example, for over a year, Key Technology has participated in a regular program of 10 spikes a month with Bowser-Morner's radon chambers. Each set of monthly spikes is at a different radon concentration and humidity level. The analyses of these samples can be run in the laboratory either as blinds or as known concentrations. The lab at Key Technology analyzes each of the 10 spikes 3 - 4 times on each of 8 analyzers, for a total of 240 to 320 measurements each month. Over a year of these spike measurements show results that are consistently within 10% (often within 1 - 2%) of expected values. On the basis of these results we contend that an annual system recalibration is unnecessary and would not further improve the quality or confidence in our routine radon measurements.

In addition, we also propose that measurements of field blanks for charcoal canisters have very limited value. The general reason for field blanks is to determine if the device may pick up radon during shipment or storage which would add to the reading for normal test exposures. This reason is not valid for charcoal canisters because charcoal is an equilibrium device rather than an integrating device. Therefore, even if charcoal may pick up radon during storage, it will come to a new equilibrium with the test environment when it is used for radon testing. For example, an open-faced canister containing 110 pCi/L was reused immediately for a new test, as if the canister had never been used. After a three day exposure at 2 pCi/L, the canister was closed and analyzed and the result was 2 pCi/L. Even with a very high "blank" reading the canister achieved equilibrium with the test environment. Key Technology routinely analyzes 25 blanks a month, but they are always less than the lower limit of detection and thus we believe these measurements are not providing useful QA information.

Therefore, we conclude that appropriate QA measurements should be device specific and not applied arbitrarily as general requirements when they will not actually improve the quality of routine measurements. The evaluation of QA programs, by either the EPA or states, should consider what makes practical sense for specific devices, rather than applying general protocols that do not take into account the properties of particular devices, such as charcoal canisters.

INTRODUCTION

The Environmental Protection Agency (EPA), July 1992, protocols for radon measurement devices specify in general for quality assurance, that, *"to limit errors in accuracy, this edition recommends that users*

calibrate their measurement systems at least once every 12 months" (EPA 1992). Specifically for activated charcoal systems, this protocol states that, *"every AC system should be calibrated in a radon calibration chamber at least once every 12 months."* While these protocols are only recommendations for voluntary participation in the EPA Radon Measurement Proficiency Program, they frequently become mandated by inclusion as requirements in state operated programs to qualify radon testing businesses and devices.

Consequently, these protocols are no longer voluntary recommendations, but necessary requirements to satisfy laws, in some states, that specify such requirements to qualify radon testers to perform radon testing in those states. States with these requirements include; Pennsylvania, New Jersey, and New York, for example. Furthermore, EPA, and many states, require detailed Quality Assurance Plans. The primary criteria in reviewing these QA plans is to determine if they properly reflect the QA protocols recommended by EPA. Several states also have radon programs with sufficient staffing to perform on-site inspections of activated charcoal laboratories to assure that protocol requirements and QA plans are being correctly performed.

IS ANNUAL CALIBRATION NEEDED?

While QA requirements for annual calibration may have general merit for radon measurement systems, we propose that such requirements are not necessary, practical, or sensible for primary activated charcoal laboratories. This proposal is based on careful evaluation of daily and monthly quality control (QC) activities which would detect systematic analytical errors and verify performance on a regular basis. The QC activities which will provide these performance checks are 1) daily control charts, and 2) monthly spikes.

Daily Control Charts

Key Technology has 12 gamma spectrometry systems available for analysis of activated charcoal canisters. Eight of these systems are used for routine analyses, with four held in reserve for additional capacity as needed. The morning startup procedure includes a 15 minute analysis of (blank) background counts for each system. In addition, each system is checked with five minute counts of a radon standard. While counting these radon standards, each system is also checked for peak gain and window settings (energy alignment), and adjustments are made as necessary.

The results of these morning QC measurements are immediately plotted on a standard Shewhart means control chart. (Goldin 1984). These control charts are prepared with warning levels at two standard deviations from the mean, and control limits at three standard deviations. The plotted data points are evaluated by normal control chart procedures to determine whether the results indicate acceptable system performance, before conducting any radon testing that day. If a single point falls outside of the control limits, the background or standard measurement is repeated to determine if the initial result was a random statistical fluctuation, or an indication of instrument malfunction. If the second measurement is within the control limits, then measurements will proceed for the day. If the second measurement is also not acceptable, the system is taken off-line for trouble shooting. Data points outside of the two standard deviation warning level would be expected about five percent of the time. Goldin describes methods for evaluating data outside of warning levels, which include evaluation of the number of warning points and trends (Goldin 1984).

These daily control chart data are a means of checking instrument performance and stability every day. The daily data collected over a month also become the basis for the next month's control charts, thus providing for longer term evaluation of performance. Instrumental error should be detected by means of control charts before any measurements are made on charcoal canisters each day. When all control chart data are within the normal range, subsequent measurements that day should be in conformance with those made on previous days.

While daily control charts do not verify a system's accuracy, they do verify stable performance on a daily basis. It is also unlikely that instrument operation failures would occur during a given day and not show up on the following day's control chart data. Also, whenever charcoal sample analyses carry over to the afternoon, background counts are repeated to establish a new background, to account for the normal diurnal variations in radon in the laboratory. These data are also plotted on the control charts and provide an additional source of performance data each day.

Monthly Spike Measurements

For over 15 months, Key Technology has participated in a monthly spike program in the radon chamber of Bowser-Morner, Inc, of Dayton, OH. This program includes 10 charcoal canisters each month exposed for three days in the Bowser-Morner radon chamber. This spike program satisfies the EPA protocol requirement for spikes which specifies that, *"spikes be conducted at a rate of three per 100 measurements, with a minimum of three per year and a maximum required of six per month"* (EPA 1992). The radon concentrations are varied each month at levels of approximately 4, 10, 15, and 25 pCi/L. Humidity levels are also varied from about 25 to 50 percent. These spike tests provide a basis for evaluation of both accuracy (closeness to the chamber radon level) and precision (reproducibility among the 10 canisters, eight systems, and multiple analyses).

These spike samples are returned to Key Technology's laboratory by express mail so that measurements can be performed within one to two days following spiking in the radon chamber. We analyze each of the 10 charcoal canisters three to four times on each of the eight detector systems used for routine analyses. This procedure results in 240 to 320 spike measurements each month. The results of each month's data are averaged for comparison with the true radon values as reported by Bowser-Morner. The results of these spike measurements from June 1993 to June 1994 are shown in Table 1.

The spike data are evaluated by the Percent Difference (%D) method (defined as the difference between the Key Technology value and the Bowser-Morner value divided by the Bowser-Morner value, times 100 to convert from a decimal fraction to a percent.) Table 1 shows that the Percent Differences ranged from 0.0 to 15.5 percent over the past year. By comparison with EPA performance testing criteria (25 Percent Difference), all of these spike tests were acceptable. In fact, for three different months, the measurements by Key Technology were the same as those of Bowser-Morner for a %D of 0.0 percent. For another three months, the %D was less than 3 percent. Larger values of %D were observed for other months, ranging from about 7.5 to 15.5 percent. Differences of 5 to 15 percent are well within the variations that would be expected by the statistics of normal radioactive decay at 1 to 2 standard deviations. Overall, evaluation of over a year's worth of spike results indicates that we may have a small positive bias of about 3 - 4 percent. A correction was made to the computer program to adjust measurements accordingly and the results were verified in a blind test as described below.

Blind Test Results

Spike exposures with Bowser-Morner can be run as known spikes or as blind tests. You can ask Bowser-Morner to report their radon chamber levels at the time spike samples are returned to the laboratory, or you can have the chamber levels withheld until after you have completed the laboratory analyses for blind testing. Such a test was performed in May 1994 as an annual performance test at the request of the New Jersey Department of Environmental Protection.

Table 2 shows the blind test results for five 3-day open face canisters and five 7-day diffusion barrier canisters. The 3-day open face canisters had a Percent Difference ranging from 0.3 to 4.0 percent. The 7-day diffusion barrier canisters had larger differences with %Ds ranging from 0.7 to 12.8 percent. Since the goal for all of these blind tests was the performance criteria established by EPA (%D of less than 25 percent), then all of these blind test results were well within the acceptance criteria.

Annual Calibrations are Not Needed

On the basis of the daily performance tests as recorded in control charts, the monthly spike test data, and random blind testing results, we conclude that an annual recalibration of the charcoal canister analysis systems of Key Technology would not improve the quality or confidence of routine measurements. Therefore, a recalibration would be an unnecessary expense, for which there is no clear benefit, even though an annual recalibration is required according to EPA testing protocols.

This conclusion is also based on the following factors: continuation of use of charcoal canisters manufactured according to original specifications and with the same uniform quality of charcoal as used in the original calibration, maintenance and operation of the same analytical system, and continuous good performance as demonstrated by control charts, and known and blind spikes. If Key Technology was to manufacture new canisters with different charcoal, or different quantities, or some other difference in design features, or if some change was made in the analytical system, then recalibration would be needed immediately.

ARE FIELD BLANKS NECESSARY?

The 1992 EPA protocols for activated charcoal (AC) devices state that, *"Field control detectors (field blanks) should consist of a minimum of five percent of the devices that are deployed every month or 25, whichever is smaller (EPA 1992).* The protocols go on to recommend that, *"large users of ACs should set these aside from each shipment, keep them sealed an in a low radon (less than 0.2 pCi/L) environment, label them in the same manner as the field detectors to ensure identical processing, and send them back to the supplier with one shipment each month for analysis.*

Key Technology has routinely analyzed the minimum 25 field blanks each month for over nine years with the accumulation of over 2500 blank measurements according to the EPA protocol. All of these measurements have consistently shown results that are near or below our lower limit of detection, which is 0.2 pCi/L. With all of this data showing that our charcoal canisters do not take up radon during storage, we believe that it is unlikely that future field blank measurements will show anything different. Therefore, we conclude that field blank measurements have very limited value and should be discontinued.

Some may argue that field blanks also provide a measure of performance accuracy. If a field blank is returned as a blind test to the laboratory and a measured result is reported of 1 pCi/L or greater, then this may indicate an error in sample handling or reporting. Such errors need to be detected, however, we contend that this is not the purpose of field blanks. Evaluation of performance accuracy (for devices, analytical systems, handling, and reporting) should be done by spike samples (either as known spikes, or single or double blind spikes).

The Reason Given for Field Blanks is Erroneous

The main reason we conclude that field blanks are unnecessary for AC devices is that the EPA protocols are based on a misunderstanding of the way charcoal works as a sampling medium for radon. The EPA protocols state that the reason for field blanks is that, *"these control devices measure the background exposure that may accumulate during shipment or storage, and results should be monitored and recorded. If one or a few of the field control detectors have concentrations significantly greater than the LLD established by the supplier, it may indicate defective devices or poor procedures"* (EPA 1992). This concern for accumulation of radon on AC devices while in storage would be valid if charcoal were truly an integrating medium. However, charcoal is not an integrating medium, but an equilibrium medium.

How Charcoal Works

When a charcoal canister is initially opened, there will normally be no radon on the charcoal. Figure 1

shows that radon will then diffuse from the surrounding air onto the surface of the charcoal by concentration gradient diffusion. Radon will accumulate (adsorb)on the charcoal surface until the partial pressure of the radon on the surface reaches equilibrium with the partial pressure of the radon in the air. During the time of initial accumulation of radon, the charcoal is in an integrating mode until equilibrium is achieved, and then the material no longer accumulates or integrates the radon concentration. Equilibrium means that some atoms of radon are diffusing to the surface and some are returning to the air (or decaying) and the net effect is a balance between the radon concentration in the air and the radon on the charcoal.

If the concentration of radon in the air goes up, then the concentration of radon on the charcoal will go up a corresponding amount. The reverse is also true. Therefore, if the radon concentration on the charcoal is greater than the corresponding concentration in the air, the charcoal will give up radon until it reaches a new equilibrium with the air. Consequently, even if charcoal takes up radon while in storage, when it is used for testing it will come to a new equilibrium with radon in the test environment. This was demonstrated by tests in Key Technology's laboratory as shown by the data in Table 3.

Eight sets of two canisters each were selected from canisters coming into the laboratory for analysis. Each set of two was chosen for similar results from the initial tests. After analysis these canisters were then used to test the radon level in Key Technology's laboratory (in duplicate) as if these were new unused canisters. The first set of four was run while the laboratory was operating with an older radon mitigation system and the radon level was about 2 pCi/L. These canisters had initial readings ranging from 12.9 to 110 pCi/L. Even the canister with the highest initial reading of 110 pCi/L achieved equilibrium with the laboratory air at 2 pCi/L in the normal exposure time of three days. The second set of four was run after installing a new radon mitigation system which reduced the laboratory air concentration to about 0.7 pCi/L. Each of these canisters also reached equilibrium with the laboratory air, including four canisters which started with levels below the lower limit of detection at 0.2 pCi/L.

These results demonstrate that even if charcoal picks up radon in storage at levels over 100 pCi/L, it will come to a new equilibrium with radon in the air of the test environment and will give the correct concentration. Therefore, the EPA protocol which states, *"If most of the controls have concentrations significantly greater than the LLD, the average value of the field controls should be subtracted from the reported field detector concentrations and the supplier should notified of a possible problem,"* is based on an erroneous understanding of the behavior of charcoal. Field control blank results should not be subtracted because the charcoal will come to the correct equilibrium no matter what the original blank reading may be.

Field Blanks Are Not Needed

We conclude, therefore, that on the basis of several hundred field blank measurements showing only concentrations less than 0.5 pCi/L, and because even greater concentrations will come to a new equilibrium in the test environment, field blank measurements provide very little QA value and should be discontinued.

DISCUSSION

We have evaluated the EPA protocols for quality assurance generally recommended for all types of radon measurement devices and systems. For specific application to measurements by activated charcoal devices in a primary laboratory, we find that not all of the recommended protocols are equally needed, practical, or sensible. More specifically we find that when a primary charcoal laboratory applies good quality control practices on a daily and monthly schedule, an annual calibration of the detection system would not likely improve the overall quality of routine measurements. The reliability of the each detector system at Key Technology is effectively checked every day by control charts of background and standards counts. Once each month we also conduct an independent check on performance by counting 10 separate spike samples 3 - 4 times

on each detector system. This means a total of 30 to 40 monthly checks on accuracy for each system. Since these spike results are normally within only a few percent of the true radon chamber levels, and well within the normal variation expected by the statistics of radioactive decay, we contend that a recalibration annually (or any other particular frequency) is not needed.

In addition we have reviewed the EPA protocol for analysis of field blanks for activated charcoal. The EPA protocol is based on the erroneous premise that charcoal is an integrating medium. In fact, charcoal integrates or collects radon by surface adsorption only up to the point at which the quantity of radon on the surface reaches equilibrium with the quantity in the air of the test environment. An open faced charcoal canister will normally reach this equilibrium condition in about 12 hours. Tests by Key Technology show that even when charcoal starts out with a very high blank reading (over 100 pCi/L), it will come to equilibrium with the test environment in 2 to 3 days. Consequently, the EPA protocol which says blank readings should be subtracted from field readings is simply wrong and based on a misunderstanding of the way charcoal works. Furthermore, after over 2500 field blank measurements, Key Technology has not found any high blank readings. On the basis of both of these observations, we propose that field blanks offer very little QA value and should be discontinued.

CONCLUSIONS

Our overall conclusion is that general QA requirements should NOT be applied arbitrarily to all radon measurement devices. Rather, requirements for QA should be specific for each type of radon measurement system and should be designed for the particular characteristics and needs of that system. QA measurements should only be conducted when they verify or improve the quality of radon measurements. If such measurements do not lead to quality improvements, then they should not be conducted, even if they are part of general QA recommendations in EPA protocols. The evaluation of QA programs, by either the EPA or states, should consider what makes practical sense for specific devices, such as activated charcoal systems.

We propose that annual calibration of activated charcoal systems does not make practical sense, if the normal daily control charts, monthly spikes, and random blind test continue to show satisfactory performance in terms of accuracy and precision. Furthermore, the analysis of field blanks for activated charcoal is virtually useless as a QA measurement because charcoal is an equilibrium material, not an integrating material.

REFERENCES

Goldin, A. Evaluation of Internal Quality Control Measurements and Radioassay, Health Physics, 47:361-374;1994.

U.S. Environmental Protection Agency, Indoor Radon and Radon Decay Product Measurement Device Protocols, Washington, DC; U.S. EPA Office of Radiation Programs; EPA 520-402-R-92-004; 1992.

Table 1. Key Technology Analysis of Monthly Spike Samples from Bowser-Morner's Radon Chamber

Month	Radon - pCi/L*		Percent** Difference
	Bowser-Morner	Key Technology	
Jun 93	26.2 +/- 0.6	26.9 +/- 1.2	+ 2.6
Jul 93	25.7 +/- 0.4	25.2 +/- 1.2	- 1.8
Aug 93	7.4 +/- 0.2	8.0 +/- 0.4	+ 8.1
Sep 93	25.4 +/- 0.6	25.8 +/- 1.0	+ 1.5
Oct 93	16.3 +/- 0.6	16.3 +/- 0.8	0.0
Nov 93	16.5 +/- 0.4	18.4 +/- 0.9	+ 11.2
Dec 93	5.1 +/- 0.1	5.1 +/- 0.3	0.0
Jan 94	25.4 +/- 0.6	27.4 +/- 1.1	+ 7.9
Feb 94	9.3 +/- 0.2	10.0 +/- 0.6	+ 7.5
Mar 94	24.4 +/- 0.8	28.2 +/- 1.2	+ 15.5
Apr 94	15.5 +/- 0.4	15.5 +/- 0.8	0.0
May 94	27.4 +/- 0.6	28.0 +/- 1.3	+ 2.2

* Radon levels are reported at +/- 1 standard deviation, based on replicate analyses, not counting statistics
** %D is calculated as the difference between the Key Technology and the Bowser-Morner value divided by the Bowser-Morner value and multiplied by 100 to convert to a percentage.

Table 2. Key Technology Blind Test Performance Results on Activated Charcoal

Canister No.	Radon - pCi/L* Bowser-Morner	Key Technology	Percent** Difference
3 - Day Open Face AC			
1	27.4 +/- 0.6	26.9 +/- 0.5	- 1.8
2	"	28.0 +/- 1.3	+ 2.2
3	"	26.3 +/- 0.6	- 4.0
4	"	26.9 +/- 0.4	- 1.8
5	"	27.3 +/- 0.4	- 0.3
7 - Day Diffusion Barrier AC			
1	27.3 +/- 0.6	29.9 +/- 1.0	+ 9.5
2	"	26.9 +/- 0.9	- 1.8
3	"	28.3 +/- 0.8	+ 3.7
4	"	30.8 +/- 1.7	+ 12.8
5	"	27.1 +/- 1.0	- 0.7

* Radon levels are reported at +/- 1 standard deviation, based on replicate analyses, not counting statistics
**%D is calculated as the difference between the Key Technology and the Bowser-Morner value divided by the Bowser-Morner value and multiplied by 100 to convert to a percentage.

Table 3. Activated Charcoal Equilibrium Tests by Key Technology

Original Result pCi/L	Re-Exposure Result pCi/L
102	1.7*
110	2.0
102	2.3
102	2.0
19.8	1.7
20.2	2.1
12.9	2.2
28.0	1.8
6.6	0.7**
6.7	0.6
15.0	0.7
16.0	0.6
15.5	0.9
18.8	0.9
<0.2	0.6
<0.2	0.7
<0.2	0.7
<0.2	0.8

* Canisters exposed in the laboratory at Key Technology at an average radon level of about 2 pCi/L
** Canisters exposed in the laboratory at Key Technology at an average radon level of about 0.7 pCi/L

How Charcoal Works

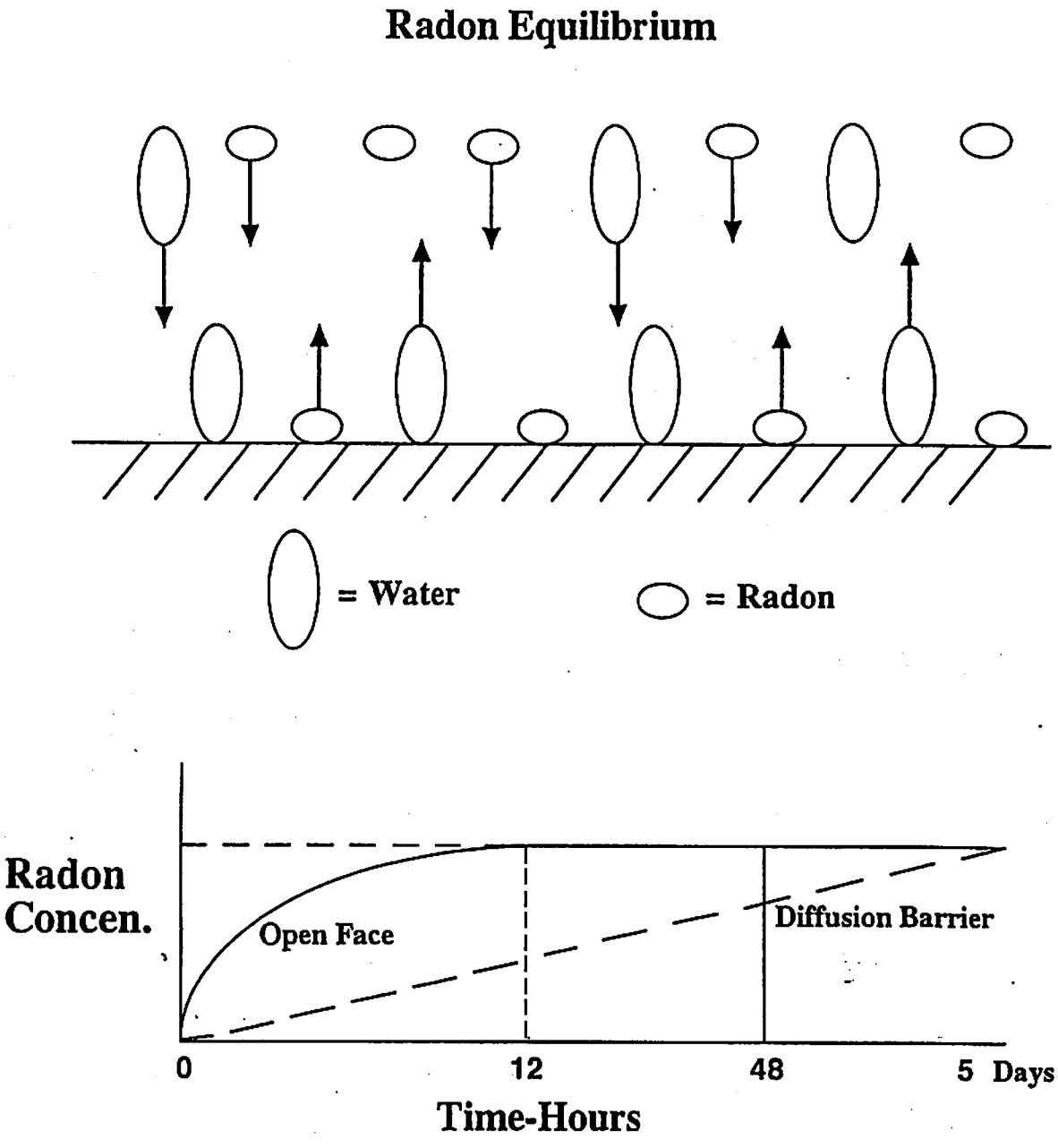

Figure 1. How Charcoal Works as an Equilibrium Material