ORAU

April 25, 2014

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SUBJECT: PILOT STUDY REPORT FOR RADON EXHALATION MEASUREMENTS, OAK RIDGE, TENNESSEE DCN: 2052-TR-01-0(RFTA 11-016)

Dear Dr. McLaughlin:

Oak Ridge Associated Universities (ORAU), operating under the Oak Ridge Institute for Science and Education (ORISE) contract, is pleased to provide the enclosed final report that details the results of the Radon Exhalation Measurements Pilot Study. Comments on the draft version have been incorporated.

Please feel free to contact me, via my information below, or Tim Vitkus, at 865.576.5073, if you have any questions or comments.

Sincerely,

Nickolas Altic
Health Physicist
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NAA:fs

Enclosure

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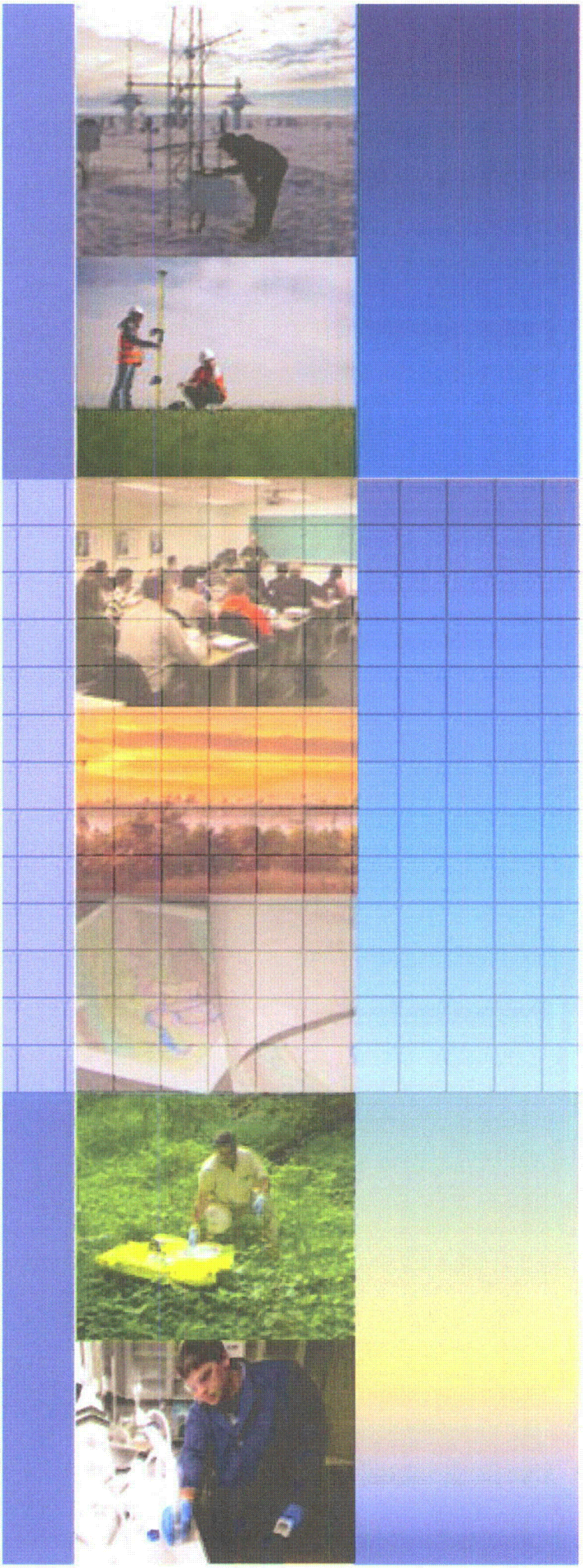

PILOT STUDY REPORT FOR RADON EXHALATION MEASUREMENTS

Nickolas Altic

Prepared for the U.S. Nuclear Regulatory Commission

ORAU
OAK RIDGE ASSOCIATED UNIVERSITIES

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PILOT STUDY REPORT FOR RADON EXHALATION MEASUREMENTS OAK RIDGE, TN

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U.S. Nuclear Regulatory Commission

FINAL REPORT

APRIL 2014

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ORAU

PILOT STUDY REPORT FOR RADON EXHALATION MEASUREMENTS OAK RIDGE, TN

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FINAL REPORT

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ACRONYMS

AA	alternative action
CFR	Code of Federal Regulations
CV	coefficient of variation
DER	duplicate error ratio
DQO	data quality objectives
GPS	global positioning system
LAACC	large area activated charcoal canister
MDF	minimum detectable flux
PSQ	principal study question
RSS	ranked set sampling
VSP	Visual Sample Plan

PILOT STUDY REPORT FOR RADON EXHALATION MEASUREMENTS OAK RIDGE, TN

1. INTRODUCTION

Radon is a naturally occurring inert radioactive gas generated by the decay of radium. The three naturally occurring isotopes of radon are radon-219, radon-220, and radon-222—with radon-222 having the longest half-life of 3.8 days. Radium-containing material can introduce radon to the surrounding atmosphere by two interrelated processes: emanation and exhalation. The process of emanation occurs when a recoil radon atom reaches the interstitial space of the material, where it is considered to be in the unbound state. The emanation coefficient is defined as the ratio of the unbound to bound radon atoms. The emanation coefficient is material-dependent and also depends on the physical properties of the material. Once radon is in the interstitial material space, it is free to migrate towards the surface and is governed by two transport mechanisms: diffusion and active transport (Straden 1984). Exhalation refers to the unbound radon crossing the surface of the material into the atmosphere. Radon flux density (commonly referred to simply as radon flux) is the activity of radon exhaled over a surface area per unit of time, and commonly has units of $pCi/m^2 s$ or $Bq/m^2 s$.

Conventional uranium mill ore extraction processes yield large amounts of leftover material, or uranium mill tailings. After uranium extraction is complete, the mill tailings are placed in large piles for storage. These piles contain considerable concentrations of radium-226, thereby serving as sources for radon-222. The radon concentration present in mill tailings can be up to 1,000 times higher than the concentration in natural soils (Ferry et al. 2002). Because radon has been classified as a Group 1 human carcinogen by the International Agency on Cancer Research, regulatory agencies have enacted limits on the radon releases from mill tailing sites (IACR 1988).

In order to reduce radon emission, the uranium mill tailing impoundments are covered with earthen material. Current radon exhalation measurement methodologies are designed to simply satisfy regulatory requirements and are not useful for evaluating radon exhalation over time to detect trends. As discussed in Section 1.3, there are a number of environmental factors that impact radon exhalation that introduce varying degrees of fluctuation in the exhalation rate at a fixed location. This variability resulting from environmental factors may mask increasing trends in the exhalation

rate. If environmental factors could be accounted for, then unbiased measurements could be performed periodically in order to assess radon exhalation over time and detect any trends of regulatory concern.

1.1 REGULATORY REQUIREMENTS

The United States Code of Federal Regulations (CFR), Title 40, Part 61 and Appendix A, Criterion 6(1) sets the standards for emission of hazardous air pollutants. Subparts T and W set the standards for radon flux emission from mill tailing disposal sites and operating mill tailings sites, respectively. Radon flux emissions are limited to an average of 20 $pCi/m^2 s$ for both subparts. Method 115 in Appendix B Part 61 describes monitoring methods to demonstrate compliance with Subparts T and W. Radon flux measurements to quantify radon emission must be made annually per the regulations. In order to determine an accurate measurement of radon flux, Method 115 recommends that a measurement set, consisting of 100 sample locations, be made for each region of the tailing pile. The method states that water-covered areas do not require monitoring because the radon flux is assumed to be zero due to the water barrier.

1.2 MEASUREMENT THEORY

Radon flux from soils is commonly measured by the accumulator technique. This technique is performed by placing an inverted cup on the surface of interest of known surface area and collecting radon for a specified period of time. Radon flux can then be calculated based on the concentration rate of change with respect to time. Accumulators have two basic design types: closed and flow-through (Zarhorowski and Whittlestone 1996). The closed accumulator design does not exchange air between the sample chamber and the external environment. For the flow-through design, there is constant air exchange between the sample chamber and the ambient environment. The constant air exchange creates an equilibrium of radon concentration in the detector, from which radon flux can be calculated. There are several types of commercially available detectors to measure the change of radon concentration inside the accumulator.

The presence of an accumulator on the ground does not measurement surface can perturb the radon exhalation rate. Radon atoms may diffuse from the cup to the ambient atmosphere where the accumulator cup meets the soil. This phenomenon, referred to as back-diffusion, can reduce the concentration in the cup, thereby causing an underestimation of the radon flux. Two-dimensional

diffusion models have been developed to account for this back-diffusion (Mayya 2004, Aldenkamp et al. 1992). Moreover, Aldenkamp et al. proposes that the only way to estimate unperturbed radon flux is to measure radon concentration *in situ* (1992). For a fixed accumulator size, back-diffusion is directly proportional to radon flux. There are sampling methods that have been used for eliminating back-diffusion. These methods include the use of flow-through accumulator designs and/or short accumulation times (Mayya 2004 and Ferry et al. 2000). However, while these methods reduce back-diffusion, they may also limit radon concentration build-up in the accumulator, which may result in an increased minimum detectable concentration. Therefore, an *a priori* knowledge of radon flux must be known in order to determine if a correction for back-diffusion is required. When measuring areas with an expected high level of radon flux, back-diffusion effects should be mitigated. Because the radon exhaltion at uranium mill tailing sites are higher than those expected from natural soil, the methods for reducing back-diffusion previously discussed may be applicable.

1.3 FACTORS AFFECTING RADON EXHALATION

There are numerous interrelated factors that affect radon exhalation from the soil surface including soil type, atmospheric pressure, soil moisture (i.e., rainfall), soil temperature, and wind. It is difficult to quantify the change in radon exhalation based on these factors because of their interrelation (i.e., a precipitation event is generally associated with a drop in pressure). Therefore, the study also evaluated these factors, both independently and in combination, in order to gauge the significance of the impact of each factor to the radon flux.

Soil type plays an especially important role in radon exhalation. Radon exhalation is dependent on a number of individual soil parameters including porosity, radon diffusion coefficient, radium-226 concentration, and soil moisture. Sandy soils will trend towards higher radon exhalation rates than soils consisting of primarily clay. Uranium mill tailing sites are located primarily in the western portion of the United States where the soils tend to be more porous and have a lower moisture content, which will generally result in an increase in radon exhalation.

Schery et al. reported a negative correlation between atmospheric pressure and surface radon flux, but could not prove causality for the correlation (1984). Schery et al. found it difficult to show causality of pressure effects on radon exhalation for a diurnal time scale because several other

meteorological variables were changing on the same time scale. Their paper reported that pressure changes over a long time scale did occur when no other meteorological variables changed on the same time scale, during which radon exhalation was found to decrease with increasing pressure. In a similar study, Ferry et al. performed radon exhalation measurements on a simulated tailings pile. The pile consisted of a 0.8 m thick layer of mill tailings covered by a 1-meter thick layer of compacted soil. Exhalation measurements were initially collected on the uncovered pile. A much larger variation in atmospheric pressure was observed in that study than what was observed during this work—pressure ranged from approximately 960 hPa to 990 hPa for the Ferry et al. study. Radon exhalation rates ranged from approximately 0 to 12 $Bq/m^2 s$ (0 to 324 $pCi/m^2 s$). The tailings pile was exposed to the environment and the large variation in exhalation rate occurred after a rain event. At the beginning of the measurement period, there was little rainfall recorded and the radon exhalation was fairly constant, indicating that the impact atmospheric pressure has on radon exhalation is small.

Soil moisture, largely a function of precipitation, impacts the diurnal radon flux from the air-soil interface. Periods of precipitation result in decreased radon exhalation rates (Ferry et al. 2000, Schery et al. 1984). Schery et al. postulated that precipitation functioned to seal pores in the top 10 cm of soil, thereby forming a cap that lowers surface radon emission (1984). Method 115 (discussed previously) restricts radon flux measurements on tailings piles after a rain event. Interestingly, Straden et al. found that an increase in moisture content in concrete, shale, and soil led to an increase in radon emanation of up to a factor of 20, depending on the material (1984). The increase in radon exhalation was less pronounced for concrete than for the other materials tested. Moisture increases the amount of radon available for transport to the atmosphere but decreases diffusion through the source material. For a mill tailing site, the source is covered by soil; therefore, a rain event will affect primarily the cover layer by decreasing diffusion and not influence emanation in the ore.

There are conflicting reports about the degree to which temperature affects radon exhalation (Schery et al. 1984, Stranden et al. 1984). Schery et al. concluded that at best there was a weak positive correlation while Straden et al. found a significant increase in radon exhalation as temperature rose (1984). The variations in results may be explained by their measurement methodologies. Schery et al. measured radon flux on the soil while Straden et al. measured exhalation from individual samples in a laboratory setting. Intuitively, one might posit that radon

flux would increase with increasing temperature because the rate of diffusion is directly proportional to temperature.

Wind is another meteorological factor that has been weakly correlated to radon exhalation. Schery et al. was able to detect a slight enhancement of surface radon flux with wind speeds up to 7 m s^{-1} (1984). However, these effects were just within the limits of detection and are much less significant than other atmospheric effects.

2. OBJECTIVES

At the U.S. Nuclear Regulatory Commission's (NRC's) request, Oak Ridge Associated Universities (ORAU), working under the Oak Ridge Institute for Science and Education (ORISE) contract, developed and implemented the methodology to assess and quantify the impacts of atmospheric pressure and soil moisture on radon flux within a pilot-scale system. The study involved the development of a radon flux monitoring process capable of determining flux changes on a diurnal scale. The ability to measure radon flux on a diurnal scale is needed because atmospheric pressure can vary throughout the measurement period required of other exhalation monitors. This study also evaluated how the in-house developed continuous radon flux monitor compared to two other well-known radon flux monitors (i.e., the E-PERM and activated charcoal canister).

The intended use of this system is to assess the change in radon flux over time at a uranium mill tailing site. The primary purpose for the pilot study at ORAU's Oak Ridge campus was to develop a continuous radon flux monitor and work out any issues with the measurement system prior to field deployment. The pilot study was divided into two phases. Phase I involved the development of the continuous radon flux monitoring system. Phase II entailed testing the system under varying environmental conditions (e.g., atmospheric pressure and soil moisture).

3. DATA QUALITY OBJECTIVES

The data quality objectives (DQO) process provides a formalized method for establishing performance and acceptance criteria for plans designed to collect environmental data. DQO definition, implementation, and assessment are iterative processes, because review of comprehensive

data sets (i.e., historical data plus newly collected data) may result in the formation of new decisions, requiring the seven DQO steps to be repeated. The seven steps of the DQO process are as follows.

1. State the problem.
2. Identify the decision.
3. Identify inputs to the decision.
4. Define the study boundaries.
5. Develop a decision rule.
6. Specify limits on decision errors.
7. Optimize the design for obtaining data.

3.1 STATE THE PROBLEM

The first step of the DQO process was to state the problem in a broad sense so that the focus of the project was unambiguous. Environmental variables such as atmospheric pressure, soil moisture, etc., can affect the variability in radon flux from soil, especially in combinations on the diurnal scale. There is a need to measure the impact these variables have on radon exhalation, in order to assess the unbiased radon exhalation rate at a uranium mill tailing impoundment over time.

3.2 IDENTIFY THE DECISION

The second step in the DQO process identified the principal study question (PSQ) and alternate actions (AAs) or outcomes that may result based on the answers to the PSQ. A decision statement is then made by combining the PSQ and AAs into a decision statement. Table 3.1 presents the PSQ and AAs and the resulting decision statement.

Table 3.1. Pilot Study Design Process	
Principal Study Question	**Alternative Actions**
Can ORAU develop a measurement system/process to assess radon exhalation that takes into account environmental factors within a pilot-scale system? Additionally, will this device be suitable for field use at a uranium mill tailing impoundment to evaluate radon exhalation over time?	1. This type of system can be developed and is suitable for field use. 2. This type of system can be developed, but is not suitable for a long term measurement period. 3. This type of system cannot be developed. Current measurement methodologies must be relied on to assess radon exhalation over time.
Decision Statement	
Determine that a radon flux measurement system/process is or is not capable of assessing the impact on the flux from the various environmental factors.	

3.3 IDENTIFY INPUTS TO THE DECISION

The third step in the DQO process determined what information was needed to resolve the decision statement produced in Step 2. For this project, the source of information needed to resolve the decision statement came from two primary sources: peer-reviewed literature and testing of a radon flux measurement system.

The first information source was from published literature, which included peer-reviewed journals as well as a previous ORAU/ORISE document (ORAU/ORISE 2011). These literature and report sources were used to aid in the design of the system. The data collected by the system were also compared with the data reported in the literature.

The second source of information was the data generated during testing of the system. The data generated described how three different flux monitors (discussed in Section 5.3) respond under the same conditions, relative to each other. The system also tested under varying environmental conditions, which would provide proof-of-concept that the system would be suitable for field use,

thus demonstrating that the impacts of environmental conditions on radon flux can be assessed and subsequently account for measurement location-specific variability.

3.4 DEFINE THE STUDY BOUNDARIES

In the fourth step of the DQO process, the target population of interest, spatial, and temporal boundaries were established. The target population for the testing phase of this project was the study of the radon flux variability that results from changing environmental conditions. In terms of physical space, this study was limited to the ORAU-managed South Campus in Oak Ridge, Tennessee.

3.5 DEVELOP A DECISION RULE

The purpose of step 5 in the DQO process was to integrate the previous DQO steps into a single statement that describes a logical basis for choosing among alternative actions. If a system as described in the PSQ cannot be established, then ORAU will determine the next best solution to the problem statement.

3.6 SPECIFY LIMITS ON DECISION ERRORS

The largest possible source of error in this study was from inconsistent response from the continuous radon monitor. In order to limit this error, side-by-side measurements were made with an E-PERM and large area activated charcoal canisters (LAACCs). Average radon flux values as measured by the continuous flux monitor were compared with values obtained with the E-PERM and the LAACC (the E-PERM provides an average flux during the measurement interval).

3.7 OPTIMIZE THE DESIGN FOR OBTAINING DATA

Details of the pilot study are outlined in Section 4.

4. MATERIALS AND METHODS

4.1 MATERIALS

Materials used for this study are outlined below

4.1.1 Radon Flux Monitors

Details of each radon flux monitor are described below.

4.1.1.1 Continuous (Model 1029)

The radon flux monitor developed consisted of two main parts: an accumulator and a continuous radon monitor. The accumulator was made of a fairly thick plastic container intended to prevent diffusive losses of radon through the accumulator wall. Mounting straps were fastened to the top of the accumulator to hold the radon monitor in place. An aluminum collar was fastened to the edge of the accumulator to allow the monitor to be pressed into the soil. All seams were filled with a compound to prevent radon loss.

The second portion of the flux monitor consisted of the continuous radon monitor, which was a Sun Nuclear Model 1029. The Model 1029 has the capability to record and digitally store radon concentration measurements at intervals specified by the user. This particular model also has the ability to record atmospheric pressure, relative humidity, and temperature. Per the manufacture, the measurement range for radon is 0.1 to 9,999 pCi l^{-1}, with an accuracy of 25% (Sun Nuclear 2010). The method of determining radon flux using this monitor is presented in Appendix A. A picture of the Model 1029 radon exhalation monitor placed on the small exhalation bed is shown in Figure 4.2.

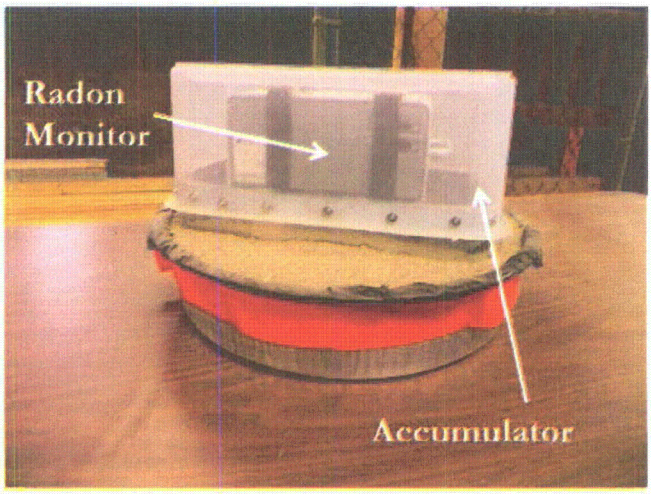

Figure 4.2. Model 1029 Radon Exhalation Monitor

The minimum detectable flux (MDF) was determined to be 0.06 pCi/m^2 s, based on the manufacturer's specifications and a half-hour measurement interval. The MDF was calculated by:

$$MDF = \left(\frac{dC}{dt}\right)_{min}\left(\frac{V}{S}\right) = \left(\frac{27.4Bq/m^3}{0.5h}\right)\left(\frac{0.01m^3}{0.07m^2}\right)\left(\frac{27.027\,pCi\,h}{3600\,s\,Bq}\right) = 0.06\frac{pCi}{m^2 s}$$

Where:

$(dC/dt)_{min}$ = minimum detectable rate of concentration increase

V = net volume in the accumulator

S = surface area of the accumulator

4.1.1.2 Activated Charcoal

A LAACC was fabricated in order to compare the Model 1029 radon flux monitor to a widely accepted standard. The LAACC was fabricated based on the design presented in *Radon Flux Measurements on Gardinier and Royster Phosphogypsum Piles near Tampa and Mulberry, Florida* (EPA 1986). Radon is adsorbed onto the charcoal, and then analyzed by gamma spectroscopy. The gamma spectroscopy results, charcoal weight, measurement time, and canister surface area were used to calculate the radon exhalation rate. The MDF of the activated charcoal canisters was 0.01 pCi/m^2 s based on a measurement time of 24 hours. The MDF was calculated by:

$$MDF = \frac{(MDC)(w)}{(t)(S)} = \frac{\left(0.23\frac{pCi}{g}\right)(180g)}{(24h)(0.05m^2)}\left(\frac{1\,h}{3600\,s}\right) = 0.01\frac{pCi}{m^2\,s}$$

where,

MDC = minimum detectable concentration of gamma spectroscopy system

w = mass of charcoal used per measurement

S = surface area of LAACC

t = measurement time

4.1.1.3 E-PERM

This type of flux monitor uses a modified E-PERM H electret ion chamber that features a 180 cm^2 diffusion window. The rate of discharge of the negatively charged electret is related to the radon flux from soil. The manufacture reports a MDF of 0.24 and 0.08 $Bq/m^2 s^1$ (6.5 and 2 $pCi/m^2 s^1$) for a measurement time of 8 and 24 hours, respectively, using long-term electrets (Stieff et al. 1996).

4.1.2 Radon Exhalation Bed

In order to provide a consistent radon flux, an exhalation bed was constructed. A layer of finely ground uranium ore was spread over the bottom of a Hardigg case with dimensions of 84 cm wide by 53 cm in length. Approximately 12 kg of uranium ore was used. A 10-centimeter thick simulated cover layer of fill dirt was then spread on top of the uranium ore. The layer of fill dirt allowed for the radon generated from the ore to diffuse to the surface and create a uniform flux at the soil/radon flux monitor interface. A sheet of porous landscaping fabric was placed between the ore and fill dirt allowing for easy recovery of the ore. The landscaping fabric was chosen so that radon could freely diffuse from the layer of uranium ore to the layer of fill dirt. Figure 4.1 presents a schematic of the radon exhalation bed.

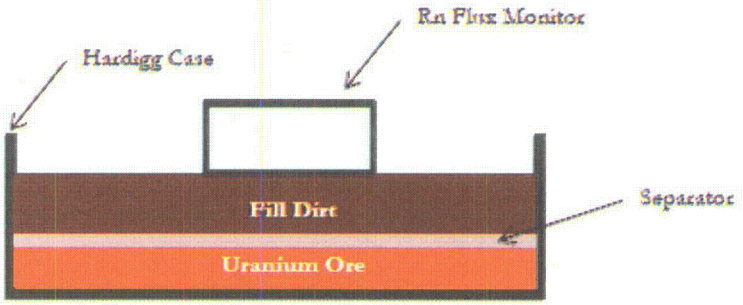

Figure 4.1. Schematic of Radon Exhalation Bed

A smaller radon exhalation bed was also constructed. The smaller bed allowed for a higher concentration of uranium per unit area while keeping sufficient fill dirt for uniform exhalation. The higher quantity of ore per area allowed for a much higher radon exhalation rate to be achieved than with the larger bed. The schematic for the smaller bed was the same as shown in Figure 4.1 only that the Hardigg case was replaced with a circular metal pan with a diameter of 39 cm and a depth of 9.5 cm. The smaller bed also allowed for the quantity of uranium ore to be easily varied.

4.1.3 Atmospheric Chamber

In order to vary atmospheric pressure by factors more than can be observed naturally, an atmospheric chamber, shown in Figure 4.3, was constructed to artificially vary the ambient pressure around the exhalation bed. The atmospheric chamber consisted of a 55-gallon steel drum fitted to a vacuum pump and a pressure gauge. A seal was placed around the drum lid in order maintain

pressure inside the drum. Fans were mounted inside the chamber in order to ensure proper air mixing in the chamber.

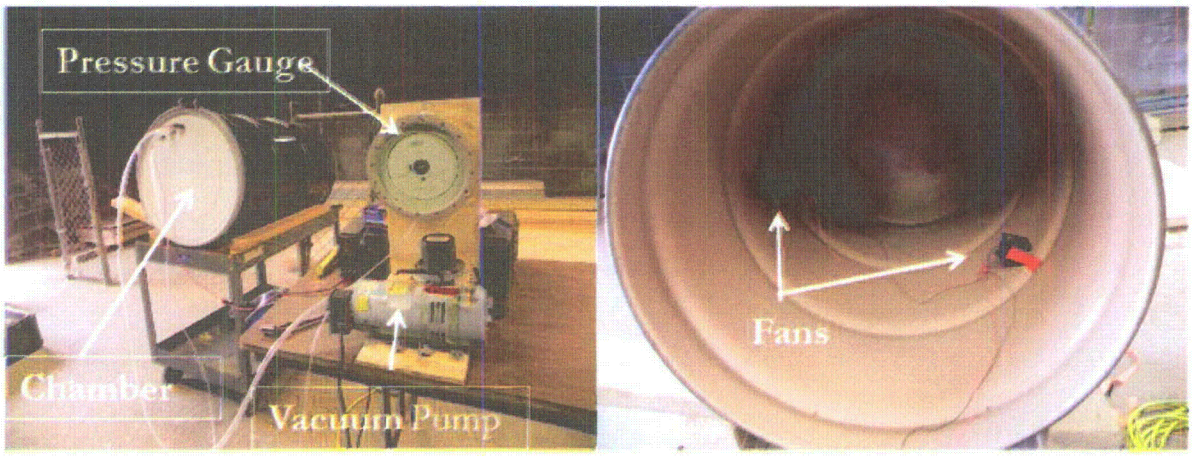

Figure 4.3. Picture of the Atmospheric Chamber

4.2 METHODS

4.2.1 Investigation of Atmospheric Pressure Effects

Influence of atmospheric pressure on radon flux was evaluated in two ways. First, measurements were collected with the Model 1029 radon flux monitor placed on the larger exhalation bed. Measurements were collected daily from July 26, 2012 to September 19, 2012. The monitor was placed on the bed and was allowed to accumulate radon for at least one hour. The first three data points—corresponding to one hour of accumulation time—were used to calculate the rate of change in radon concentration inside the accumulator with respect to time (dC/dt), as discussed in Appendix A.

For measurements performed with the atmospheric chamber, the large exhalation bed and the Model 1029 were placed inside the chamber, and the lid was sealed. The vacuum pump was turned on until the desired pressure was reached. The radon concentration was measured over time and the exhalation rate was calculated using the same method as described in Appendix A—the same as for calculating radon flux with the accumulator. For measurements made with the atmospheric chamber, the 55-gallon steel drum served as the radon accumulator.

4.2.2 Investigation of Soil Moisture Effects

The fill dirt layer (Figure 4.1) was first dried overnight in an oven at 100 °C. Deionized water was added to the fill dirt of the smaller exhalation bed to assess the affect of soil moisture on radon flux and to determine if the Model 1029 would be able to differentate between varying levels of water content. Enough water was added so that the fill dirt contained 0%, 5%, 10%, and 15% water by weight. The wet dirt was placed on top of the uranium ore and the radon exhalation was measured with the atmospheric chamber. Soil with a water content of 0% by weight will abosrb moisture from the ambient air. However, with the short measurement time, it was assumed that the soil moisture remained at 0% during the measurement.

4.2.3 Inter-comparison of Flux Monitors

Radon flux measurements were collected using four different monitors to assess inter-comparability. The four monitors used in the inter-comparison were the three monitors discussed in Section 4.2 and the atmospheric chamber discussed in Section 4.3. The small exhalation bed was used for all inter-comparison measurements. The uranium ore contents were varied from 0.2 kg to 10.7 kg. Measurements with all four monitors were collected at each quantity of uranium ore. Radon was allowed to equilibrate in the cover layer before any measurements were made. Additionally, measurements with the LAACC, E-PERM, and Model 1029 were made on soil to assess background inter-comparability.

Repeated measurements were made with the LAACC to assess measurement variability. A total of four 24-hour measurements were made on the small exhalation bed. The charcoal was then analyzed by gamma spectroscopy and the radon-222 activity collected per hour was calculated.

5. RESULTS AND DISCUSSION

The results of the atmospheric pressure, soil moisture, and inter-comparison measurements are summarized below.

5.1 ATMOSPHERIC PRESSURE

Radon exhalation measurements collected under naturally varied atmospheric conditions are presented in Figure 5.1. Atmospheric pressure during the measurement periods ranged from 989 to

1,001 hPa and the associated radon exhalation values varied from 2.18 to 11.7 $pCi/m^2 s^1$. The average width of the 95% confidence interval for all measurements was 18% of the observed mean range in radon exhalation. The radon exhalation measurement data in Figure 5.1 have a slight negative correlation with respect to atmospheric pressure.

For the ORAU study, a linear regression was applied to the data, represented by the red line in Figure 5.1. The resulting R^2 value was negative at -0.04, indicating that the data do not fit the linear regression model. A negative R^2 value is generated by the graphing software when the variance of the regression is worse than the variance resulting from fitting a horizontal line. As shown in Figure 5.1, two data points are significantly higher than the rest. However, nothing suggests that that the two data points are questionable and, therefore, cannot be excluded from the regression model. Other environmental factors for those two data points recorded by the Model 1029, temperature and relative humidity, were compared to the rest of the population. As seen in Table B-1, temperature was not appreciably different for those two data points compared to the rest of the population. Relative humidity for those two points was slightly higher than the other measurements but is likely not the cause. An increase in relative humidity would decrease the sensitivity of the radon monitor causing an underestimation of the radon concentration.

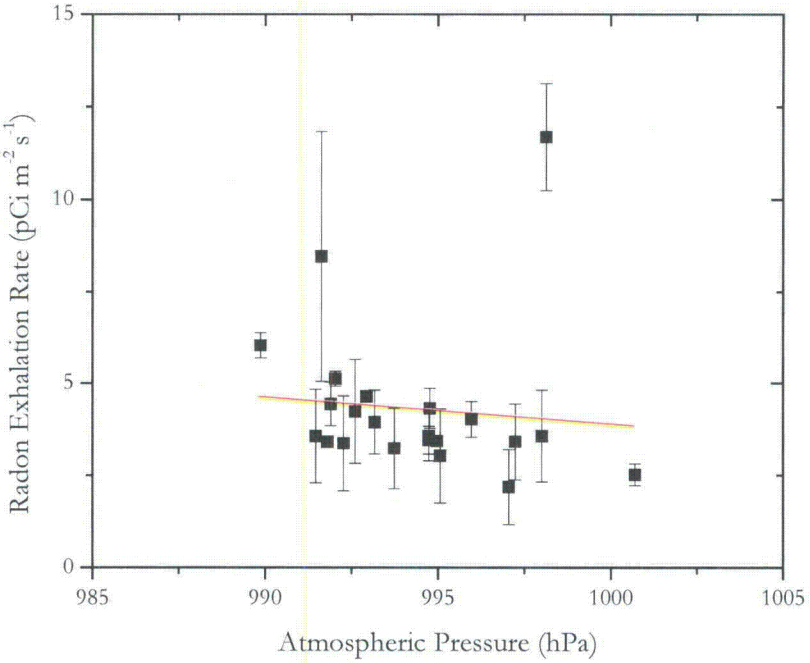

Figure 5.1. Radon Exhalation vs. Normal Atmospheric Pressure

Figure 5.2 represents radon exhalation measurements made while the pressure was varied with the atmospheric chamber. Ambient pressure inside the chamber ranged from 751 to 1,170 hPa and the resulting radon exhalation ranged from 4.51 to 7.90 pCi/m^2 s. The pressure range represents a much wider range than what would be observed under normal conditions. Comparing Figures 5.1 and 5.2, the pressure range in Figure 5.1 is only 3% of the range in Figure 5.2. The pressure was varied far outside of the normal range in order to achieve a better fit of the data.

A linear regression was applied to the data and the resulting R^2 value was 0.17, which represents a better fit than the previous data set. The resulting R^2 value means that only 17% of the variation in radon exhalation can be explained by the variation in the ambient pressure. The small R^2 value indicates that the regression performed on the data in Figure 5.2 is not useful for application in the field. A possible cause for the inadequacy of the model is that the Model 1029 may not be sensitive enough to resolve the change in radon exhalation due to atmospheric pressure alone. The average width of the 95% confidence interval of all measurements is 53% of the observed range in radon exhalation. A trend may be more pronounced, resulting in a higher R^2 value, if the measurement error is significantly decreased.

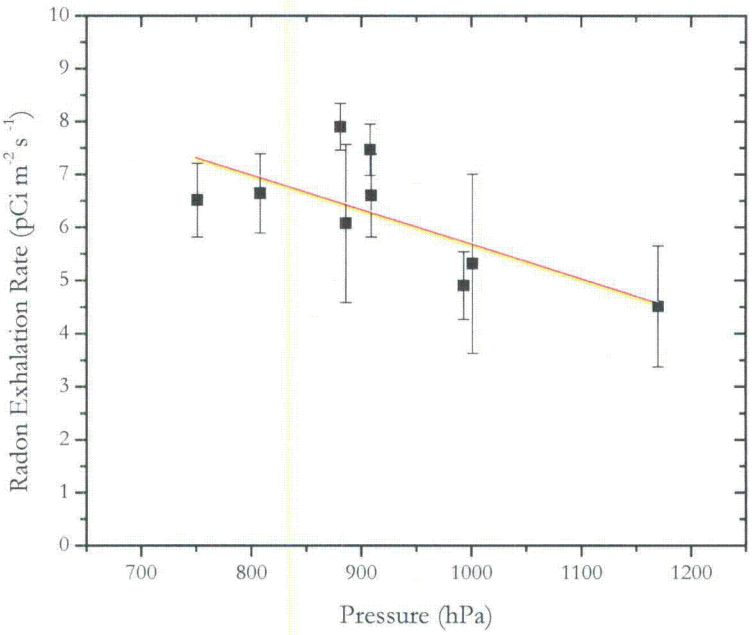

Figure 5.2. Radon Exhalation vs. Ambient Pressure Outside of Normal Range

5.2 SOIL MOISTURE

Radon exhalation was found to remain relatively constant with water content in the fill soil between 0% and 5% (% by weight). When the water content was between 5% and 10%, radon exhalation began increasing with increasing soil moisture. Figure 5.3 shows the relationship between radon exhalation and the fill soil percent moisture. The barrier between the uranium ore and fill dirt was permeable to water. As the moisture content in the fill layer increased, more water was available to penetrate into the ore layer. The moistened layer of ore would have a much greater emanation factor according to one study (Straden 1984). Under dry soil conditions, the range of the recoil radon-222 atoms is greater than the distance between soil particles. As water content in the soil increases, the air in the soil pores is replaced by water, which stops the recoil atom inside the soil pore. The radon is then free to diffuse through the soil. The soil moisture was not expected to infiltrate the ore layer; this effect is the cause of the exhalation rate increasing with water content.

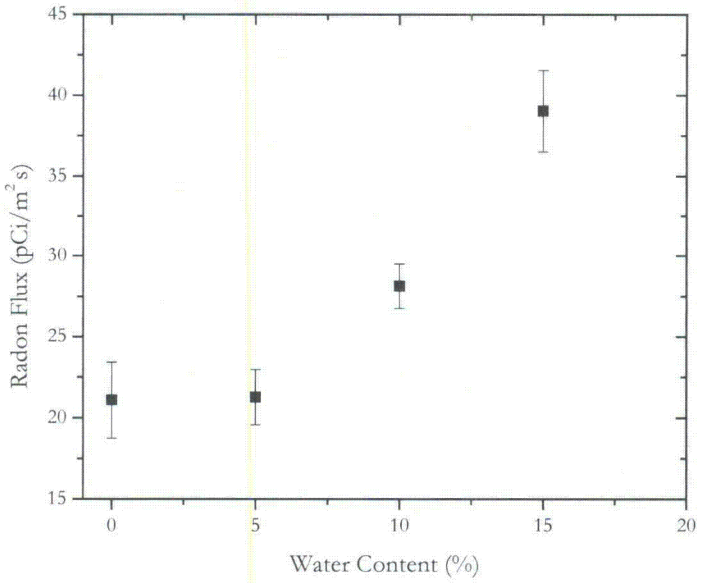

Figure 5.3 Radon Exhalation Rate as a Function of Water Content in the Fill Dirt

5.3 MEASUREMENT SYSTEM INTER-COMPARISON

Radon exhalation rate vs uranium ore quantity for each of the measurement systems is presented in Figure 5.4. At a relatively high uranium ore weight, the LAACC and the atmospheric chamber are in good agreement. However, the Model 1029 and E-PERM under-responded relative to the atmospheric chamber and LAACC. As the quantity of uranium ore decreased, the responses of the Model 1029 and E-PERM began to converge to the responses of the atmospheric chamber and LAACC. For measurements made on background soil, the difference between the E-PERM and the LAACC were less substantial. The radon exhalation rate for the background soil was 0.15 ± 0.01 pCi/m^2 s as measured by the LAACC which is in agreement with the E-PERM's result of 0.14 pCi/m^2 s. The model 1029 malfunctioned during the background level inter-comparison and returned a result of approximately 0 pCi/m^2 s. Due to project deadlines the issue was not able to be resolved.

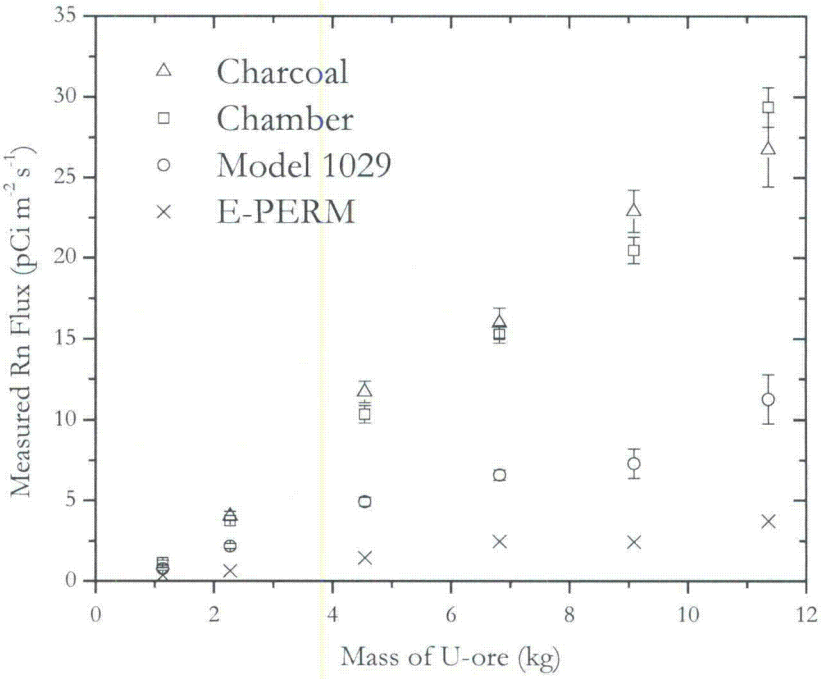

Figure 5.4. Results of the Inter-comparison Exercise

The results presented in Figure 5.4 prompted an investigation to determine the cause of the under–response of the Model 1029 and E-PERM relative to the LAACC and atmospheric chamber. For a fixed accumulation volume and measurement period, the magnitude of back-diffusion is proportional to radon exhalation. This means that at a higher radon exhalation, the effect of back-diffusion will be more pronounced. The data presented in Figure 5.4 indicate that the Model 1029 and E-PERM exhalation monitors suffer from significant back-diffusion effects, causing an under-response relative to the LAACC and atmospheric chamber. Accumulator volume can also impact the degree of back-diffusion. At a given radon exhalation rate, decreasing the accumulator volume will increase susceptibility to back-diffusion. The accumulator volume of the E-PERM is 1×10^{-4} m^3, which is significantly less than that of the Model 1029 at 1×10^{-2} m^3. This difference in volume explains why the back-diffusion effect on the E-PERM is more pronounced. The atmospheric chamber and LAACC do not suffer from back-diffusion due to their specific designs. The accumulator volume in the atmospheric chamber is very large relative to the soil gas volume in the exhalation bed and for the LAACC back-diffusion is not an issue due to the flow-through

design. The data also suggest that even a short accumulation time, as identified in the literature, does not significantly reduce the effect of back-diffusion.

Because the LAACC is the widely accepted standard for radon exhalation measurements, the results of the Model 1029 and the E-PERM were compared directly with the results of the LAACC. The difference in results was dependent on the quantity of uranium ore present in the bed (i.e., dependent on the magnitude of back-diffusion). It was found that the negative bias of the responses, relative to the LAACC, ranged from 12% to 68% for the Model 1029 and 58% to 89% for the E-PERM. Figure 5.4 shows how the back-diffusion is dependent on radon exhalation for the E-PERM and Model 1029. The data presented in Figure 5.4 are consistent with the theory of back-diffusion. That is, with increasing uranium ore mass, the concentration inside the accumulator will increase. With the increasing concentrations within small, non-flow through accumulator design, the effects of back diffusion on the exhalation measurement negative bias also increases.

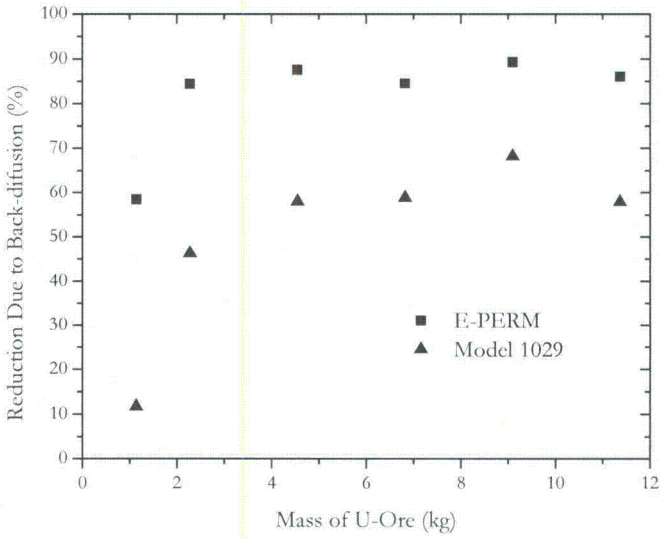

Figure 5.4. Reduction in Monitor Response Relative to the LAACC

Several articles have presented the results of inter-comparison tests with a number of radon exhalation monitors (Grossi et al. 2011; Hutter and Knutson 1998). Grossi et al. reported a fairly good agreement between the monitors with a coefficient of variation (CV) from 10% to 23% while Hutter Knutson reported a CV of 34%. However, the data presented in these articles were collected

from areas where the radon exhalation level was very small compared to the levels in the small exhalation bed used in this study. The maximum recorded radon exhalation value from the aforementioned articles was 1.2 pCi/m^2 s compared to a maximum value of 27 pCi/m^2 s from the exhalation bed.

Figure 5.5 shows the activity collected per hour by each individual measurement; the measurements have an average relative error. The variability measurements with the LAACC were not performed under identical atmospheric condition. However, based on the data presented in Section 5.1, the variability introduced by atmospheric pressure changes is small relative to the measurement uncertainty.

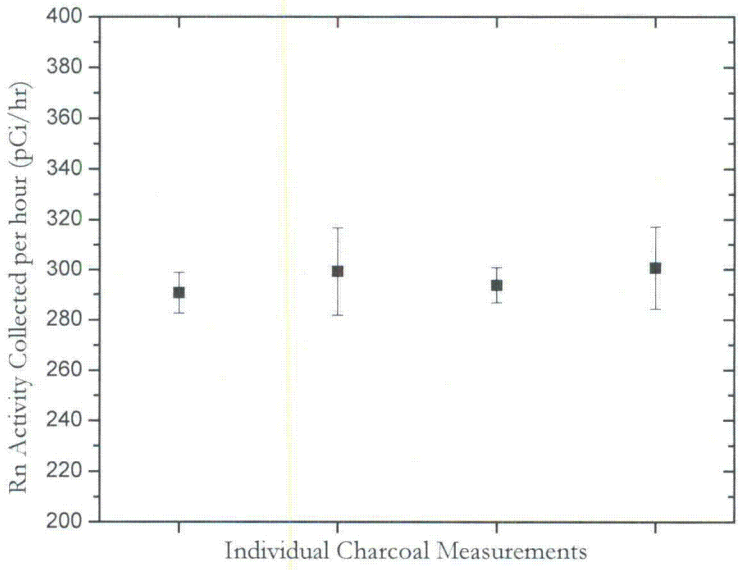

Figure 5.5. Repeated Measurements Using the LAACC

6. SUMMARY AND CONCLUSIONS

6.1 ATMOSPHERIC PRESSURE

Radon exhalation rate was found to have a slight negative correlation with atmospheric pressure (the exhalation rate decreased with increasing atmospheric pressure), which was expected based on the literature review. However, the linear regression model applied to the normal atmospheric data was

not valid, as indicated by the negative R^2 value. The data set for normal atmospheric pressure had two suspect data points present. These suspect data were not removed from the regression model as there was no evidence to question the data quality.

When atmospheric pressure was varied outside of the range of naturally occurring pressure differentials—the lowest and highest recorded pressures were 751 and 1,170 hPa, respectively—the relationship between radon exhalation and atmospheric pressure was stronger. The linear regression model applied to the second set of atmospheric data yielded an R^2 value of 0.17, which is better than the model applied to the normal atmospheric data set.

The measurement uncertainty of the Model 1029 was much larger than the variation in radon exhalation due to atmospheric pressure. The average width of the 95 % confidence interval of all measurements was 18% and 53% of the observed range in exhalation for the normal and artificially varied atmospheric pressure, respectively. This would make it difficult to quantify small changes in radon exhalation due to normal environmental variation in atmospheric pressure. If it were possible to reduce the measurement error, a better relationship may be established.

6.2 SOIL MOISTURE

Radon exhalation was found to increase significantly with increasing soil moisture.. The moisture in the soil layer infiltrated the ore layer which greatly increased the emanation power of the ore. The increase in emanation from the ore was enough to overcome the decrease in exhalation caused by moisture in the soil layer, thus resulting in a net increase in radon exhalation. Radon exhalation increased from 20 pCi/m^2s to 38 pCi/m^2s when percent moisture in the fill dirt layer increased from 0 to 15%. Thus, a valid relationship between soil moisture and radon exhalation could not be established with the current experimental setup. Further efforts were not pursued due to the results of the inter-comparison, discussed in Section 6.3, as a valid relationship between radon exhalation and soil moisture would not impact the overall outcome.

6.3 INTER-COMPARISON

Based on the results of the inter-comparison study, radon back-diffusion was causing the under response for both the Model 1029 and E-PERM. Previous inter-comparison exercises, as mentioned in the Section 5.3, were performed at radon exhalation levels close to background. However, the

concentrations of radon associated with uranium mill tailings are orders of magnitude higher than the concentrations encountered in background. Therefore, a major recommendation from the study is to ensure that the selected monitoring system has been evaluated for performance with the anticipated exhalation measurement conditions.

Therefore, the inter-comparison study resulted in the following conclusions for the measurement systems. The Model 1029 radon exhalation monitor would not be very useful for radon exhalation measurements at a uranium mill tailing site. This monitor is significantly affected by back-diffusion when performing measurements at an area with a high radon exhalation background. Therefore, because of back-diffusion issues, the Model 1029 is not considered a viable system for assessing radon exhalation across uranium mill tailing piles. The findings of the inter-comparison exercise result in the selection of option 3 from the list of AAs in Table 3.1. Therefore, a more accepted measurement method, such as activated charcoal, must be relied upon to assess radon exhalation over time.

7. PATH FORWARD

As a result of the data collected during the pilot study, the DQOs were reevaluated. As stated in Section 3.5, if the problem statement could not be adequately addressed, then ORAU would determine the next best solution. The following subsections provide a general overview of an alternate method to assess radon exhalation at a uranium mill tailings site over time. Specific procedures would be detailed in the project-specific-plan. The proposed method involves collecting periodic radon exhalation measurements at a mill tailings site and evaluating the trend in measurements over time, if any. Previous radon exhalation data would only be viable if the measurements were collected from the same locations periodically.

7.1 TREND TESTING

There are several statistical methods for evaluating trends over time (Helsel and Hirsch 2002; Meals et. al. 2011).Two trend tests are recommended. First, the Mann-Kendall test could be performed on the radon exhalation data set collected over time. The Mann-Kendall test is a nonparametric test and the values being tested do not require an underlying distribution assumption. The effects of

covariates such as atmospheric pressure, soil moisture, and temperature may mask any apparent trend. If a reasonable linear regression can be obtained between radon exhalation and any of the covariates, then the covariate influence on flux can be accounted for. Based on the results of the literature review and this study, soil moisture is expected to dominate the impact on flux over that of all other covariates. If the covariates could be successfully removed, then a second trend test can be performed on the residual (a residual is the difference between the actual measurement and the predicted value) from the regression model. Should flux residuals increase over time then it can be concluded that radon exhalation is increasing. If the covariates cannot be removed, then a simple trend test, such as the Mann-Kendall, could still be used on the data set. The sensitivity of the trend test is dependent on the rate of change of radon exhalation over time, if any, and thus cannot be predicted before mobilization to the site. A small, gradual increase over time will require more measurements (time) to detect than a large increase over a short period of time.

Exhalation measurements should be performed periodically; other covariates such as atmospheric pressure, soil moisture, and temperature should be recorded as well. The covariates should be averaged over the measurement period. The trend analysis should be performed at each measurement location. A test on the average exhalation rate is not performed as the average could hide small changes in individual locations. The null hypothesis (H_0), or assumed base condition, is that there is no trend in the radon exhalation rates. The alternative hypothesis (H_A) is that there is a trend present in the radon exhalation rates. The confidence level of the test should be set at 95%.

7.2 RADON EXHALATION DATA SET

There are two potential data sources for the trend test, one being previous radon exhalation measurements to satisfy regulatory requirements. The other potential data set would be generated by performing radon exhalation measurements for the sole purpose of trend testing.

7.2.1 Historical Data

A site that has numerous years of radon exhalation data could be selected for a retrospective trend analysis. A simple trend test could be performed on the mean exhalation rate. The mean rate would need to be used as it is unlikely that the measurement locations would be the same for each data set. Using previous data has the advantage that the data analysis cost is relatively small compared to

collecting new data. However, a major disadvantage is that specific locations could not be evaluated over time.

7.2.2 Collected Data

A large enough number of measurement locations must be selected so that the results can be representative of the tailings impoundment. The measurement results should be representative of the tailing site so they could be extrapolated to the entire site, and thus decisions made about the study boundary would be representative of the entire impoundment. Therefore, a ranked-set sampling (RSS) scheme is proposed to select the measurement locations. Ranking locations would be assigned a low, medium, or high value dependent upon a static NaI measurement. The number and placement of RSS measurement locations would be determined using Visual Sample Plan (VSP). A large number of measurements is expected based on the results of national emission standards for hazardous air pollutants monitoring at the Canon City facility in Colorado where radon exhalation values varied by up to a factor of 100 (Cotter 2009). Measurements locations would be recorded with global positioning system (GPS) equipment.

Two of the underlying assumptions of the proposed trend testing process are that 1) radon exhalation at measurement locations would be expected to correlate proportionately with the relative concentration of radon decay products at a given location, and 2) radon exhalation at measurement locations increase the closer the material is to the surface. In other words, tailing pile areas with high concentrations close to the surface and with a thinner cover layer would be expected to have both the highest exhalation rates and should also show an increasing gamma radiation signature. These assumptions must be true in order to rank measurement locations with a NaI detector. Therefore, gamma walkover scans would be used to select judgmental radon exhalation measurement locations, both high and low. The judgmental locations would then be selected for routine monitoring. Gamma walkover scans would be performed during each site visit to ensure that areas of the study boundary are not changing relative to other locations (i.e., locations with the highest gamma radiation levels should remain high and locations with a low gamma radiation detector response should remain low relative to each other). NaI detectors would be coupled to GPS equipment that enable real-time gamma count rate and spatial data capture. The gamma data collected from each campaign could also be evaluated over time to identify trends in the gamma

signature from the pile. These data would provide a secondary standard for evaluating cover performance, especially when posted together with the flux measurement data.

7.2.2.1 Radon Exhalation Monitor

Measurements are proposed to be collected using the LAACC method as specified in Method 115b, Appendix B to 40 CFR 61. Additionally, the LACCs will be shielded from direct sunlight to minimize heating of the charcoal. Duplicate measurements are recommended by co-locating LAACCs at 10% of the measurement locations. The duplicate measurements would be evaluated using the duplicate error ratio (DER). A DER less than 3 indicates a 99% confidence that the two measurements are in agreement. Measurement times will be kept shorter than 24 hours to minimize the effects of temperature and humidity on radon adsorption onto activated charcoal.

$$DER = \frac{|S-D|}{\sqrt{(Us^2)+(Ud^2)}} = \leq 3$$

Where :
S = Measurement result
D = Duplicate result
Us = Measurement uncertainty (one-sigma level)
Ud = Duplicate uncertainty (one-sigma level)

Charcoal samples would be analyzed using solid-state gamma spectroscopy. Analyzing samples by gamma spectroscopy would provide a lower relative error when compared with systems using a NaI detector.

7.4 LOCATION OF SITE AND FREQUENCY OF MEASUREMENT EVENTS

A mill tailings site for this study has yet to be selected. One possible site is the Gas Hills site located approximately 60 miles east of Riverton, Wyoming. This site is proposed because of the potential support from site personnel. ORAU provides support to the state of Pennsylvania. It is possible an arrangement could be made to perform the proposed study at the abandoned tailing pile in Canonsburg, Pennsylvania. One factor that would be considered when selecting the site would be whether or not the tailing impoundment was covered by rip-rap. The rip-rap would need to be removed at each measurement location, which may not be feasible.

The frequency of measurements is yet to be determined. The site geographic location may seasonally constrain measurement events.

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APPENDIX A
EXHALATION RATE CALCULATIONS

The following sections describe how radon exhalation rate was calculated using the Model 1029 monitor. The minimum detectable flux (MDF) is also presented.

A.1 CALCULATION OF RADON EXHALATION RATE

When measuring radon exhalation with the closed can technique, radon concentration inside of the accumulator will increase with respect to time. The radon concentration profile over time will follow an exponential ingrowth, as shown in Figure A.1. Eventually equilibrium will be reached inside of the accumulator where the radon concentration is not changing over time. The rate of change of radon concentration with respect to time (dC/dt) remains constant initially then slowly decreases as the steady state is reached. However, when performing closed can measurements environmentally there are other factors that affect the buildup of radon inside the accumulator.

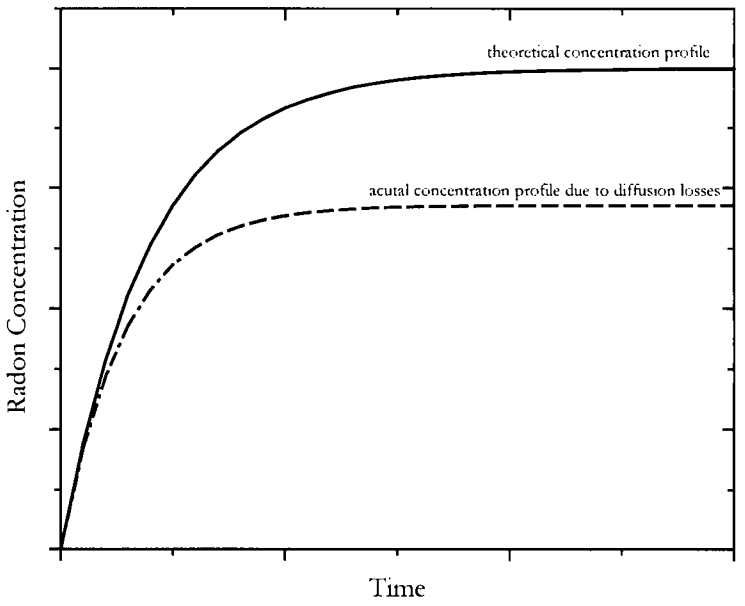

Figure A.1. Theoretical vs Actual Radon Concentration Profile for Closed Can Measurements

As the radon concentration inside of the accumulator increases the probability increases for radon atoms to diffuse back into the soil, where they are then free to diffuse outside of the accumulator.

This process of back-diffusion suppresses the steady state radon concentration inside of the accumulator, as shown by the dashed line in Figure A.1. The magnitude of the suppression is dependent on the accumulator volume and the radon exhalation rate, both of which determine how fast the radon concentration increases.

Radon concentration at a given time (t) can be calculated by,

$$C(t) = \frac{FA}{V\lambda_e}[1 - exp(-\lambda_e t)] \qquad \text{(Equation 1)}$$

where F is the undisturbed radon exhalation rate, λ_e is the effective time constant for radon buildup, A is the surface area of the accumulator, and V is the accumulator volume (Mayya 2004).

The effective time constant takes into account radon losses from both decay and leakage (i.e., back-diffusion). The rate of change of radon concentration inside the accumulator is determined by taking the derivative of with respect to time. When the measurement time is short, dC/dt for the theoretical and actual curves are equal, meaning that losses due to back-diffusion are insignificant. At t = 0 the increase in radon concentration is linear and given by (Mayya 2004)

$$\frac{dC(t)}{dt} = \frac{FA}{V} \qquad \text{(Equation 2)}$$

Thus, for a short accumulation time, the undisturbed radon exhalation rate can be calculated by Equation 2. The time at which dC/dt for the theoretical curve does not equal that of the actual curve will be dependent on the magnitude of the exhalation rate.

The Sun Nuclear Model 1029 radon monitor will measure radon concentration every half-hour. An example accumulation curve for a 24 hour measurement period is shown in Figure A.2. Based on the curve in Figure A.2, an accumulation time of three hours or less would provide certainty that back-diffusion losses are not significant. The data points collected within the first three hours show a linear relationship and can be fitted using linear regression. Origin Pro 8 was used to develop a linear regression model of the increase in radon concentration inside of the accumulator; up to the first six data points were used as inputs of the model. The model had the form of y = m*x + b, where m is equivalent to the rate of change of radon concentration.

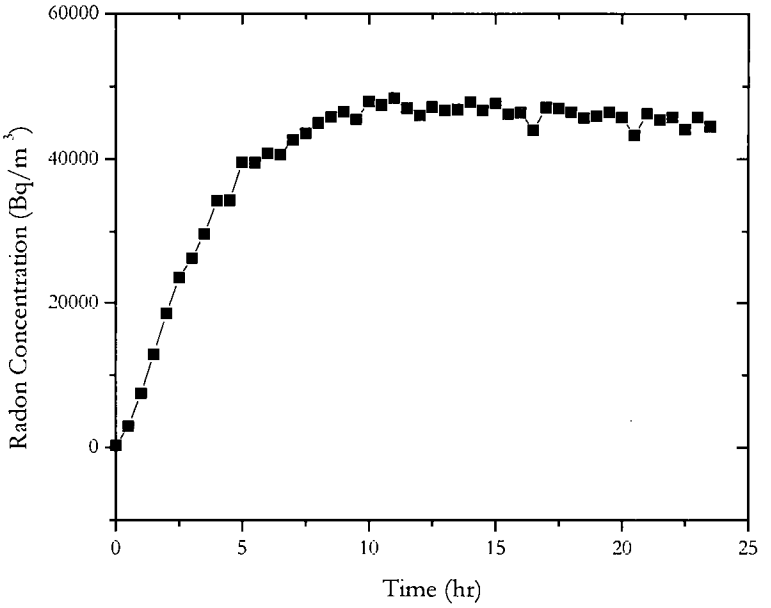

Figure A.2. Radon Concentration Inside the Model 1029 Accumulator

APPENDIX B
TABLES

Table B-1. Radon Exhalation Measurements Made With the Model 1029 Under Normal Environmental Conditions					
Measurement Date	**Temperature (°C)**	**Pressure (hPa)**	**Relative Humidity (%)**	**dC/dt[a] (pCi/s)**	**Exhalation Rate (pCi/m² s)**
07/26/2012	28.5	991.6	66.7	2.39 ± 0.49	8.44 ± 3.39
07/27/2012	29.3	992.6	61.5	1.20 ± 0.20	4.24 ± 1.41
07/31/2012	26.9	992.0	64.0	1.45 ± 0.03	5.12 ± 0.21
08/01/2012	26.0	992.9	62.2	1.31 ± 0.00	4.65 ± 0.02
08/06/2012	24.5	998.1	66.2	3.31 ± 0.21	11.7 ± 1.4
08/07/2012	25.3	994.8	66.3	1.22 ± 0.08	4.32 ± 0.54
08/08/2012	26.4	994.7	64.6	0.979 ± 0.06	3.46 ± 0.38
08/09/2012	24.4	992.3	64.5	0.954 ± 0.19	3.37 ± 1.29
08/13/2012	23.5	995.0	56.0	0.971 ± 0.00	3.43 ± 0.00
08/15/2012	21.9	991.8	62.8	0.966 ± 0.02	3.41 ± 0.13
08/16/2012	21.6	995.1	61.5	0.858 ± 0.19	3.03 ± 1.28
08/20/2012	20.9	991.9	59.8	1.26 ± 0.09	4.44 ± 0.59
08/21/2012	19.7	994.7	55.2	1.01 ± 0.10	3.57 ± 0.67
08/22/2012	20.4	997.2	55.8	0.97 ± 0.15	3.42 ± 1.03
08/23/2012	20.6	998.0	56.1	1.01 ± 0.18	3.57 ± 1.25
09/05/2012	24.5	991.5	64.6	1.01 ± 0.18	3.57 ± 1.27
09/06/2012	21.9	993.2	62.8	1.12 ± 0.13	3.95 ± 0.87
09/10/2012	26.4	997.1	48.6	0.62 ± 0.15	2.18 ± 1.02
09/11/2012	18.9	1000.7	55.6	0.71 ± 0.04	2.52 ± 0.29
09/17/2012	21.6	993.7	60.0	0.92 ± 0.16	3.24 ± 1.10
09/17/2012	21.4	989.9	63.9	1.71 ± 0.05	6.04 ± 0.35
09/19/2012	16.5	996.0	56.4	1.14 ± 0.07	4.03 ± 0.48

[a]dC/dt = rate of change of radon concentration inside the accumulator with respect to time