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SUBJECT: FINAL - TECHNICAL BASES AND GUIDANCE FOR RADON FLUX MONITORING AT URANIUM MILL TAILING SITES (RFTA 11-010) DCN 2042-TR-01-0

Dear Mr. McLaughlin:

Enclosed is a final version of the report mentioned in the subject heading. Please direct any questions or comments you may have to me via my information below or Tim Vitkus at 865.576.5073.

Sincerely,

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TECHNICAL BASES AND GUIDANCE FOR RADON FLUX MONITORING AT URANIUM MILL TAILING SITES

1.0 INTRODUCTION

Uranium-bearing ores are mined in order to extract the uranium for use in the nuclear fuel cycle. These ores contain naturally concentrated uranium in natural isotopic abundances (U-238, U-235, and U-234) as well as the complete radioactive decay chain in secular equilibrium. The process of extracting the uranium from the ore yields large volumes of residual wastes, referred to as uranium mill tailings, that are placed in large capped and covered long-term storage piles. The tailings contain the progeny radionuclides, beginning with Th-230 through the stable Pb-206. The immediate daughter of Th-230, Ra-226, decays into an inert radioactive gas, radon-222 (radon). Radon is considered a significant human health concern as it has been classified as a group 1 human carcinogen by the International Agency for Research on Cancer (IARC 1988). The radon concentrations present in mill tailings can be as much as three orders of magnitude higher than the concentration in natural soils (Ferry et al. 2002). Radon has a relatively short half-life of 3.8 days, however, there is time for radon to diffuse to the soil surface, where it becomes available for human breathing before sufficient decay can occur.

Radon flux[1] is the activity of radon that passes through an area per unit time, usually expressed as Becquerel (Bq) or picocuries per square-meter per second (pCi m^{-2} s^{-1}). Radon flux is measured differently than radon concentration, but there are similarities between both measurement techniques. Both techniques measure radon concentration. However, radon flux monitors measure both the radon concentration and account for the surface area from which the radon emanated; whereas concentration monitors simply measure ambient radon concentration.

Radon monitors can be categorized in two ways: active and passive. Active monitors draw in sample air. Passive monitors do not actively draw in air but rather allow it to accumulate inside the detector. The collected radon can then be measured (e.g., by gamma spectroscopy, ion chamber). In general, active sampling instruments, such as those that employ the two-filter method, are more sensitive and have a lower limit of detection than their passive counterparts. These factors make them most useful

[1] Flux is defined by ICRU 60 as dN/dt, where dN is the increment of particle number in time dt. Flux density or fluence rate is the number of particles crossing an area per unit time. In the literature these are used almost interchangeably.

for measuring background radon levels. However, active monitors are not appropriate for measuring radon flux because one is unable to determine over what soil surface area the sample volume emanates.

The first part of this paper will focus on passive radon monitors, since active monitors cannot be used to make radon flux measurements. Guidance developed by the U.S. Environmental Protection Agency (EPA) in order to meet United States Code of Federal Regulations (CFR) Title 40 Part 61.222(a) will also be reviewed. The second part of this paper will investigate how atmospheric conditions affect radon exhalation from the earth's surface. Finally, recommendations for radon flux monitoring at uranium mill tailing sites will be discussed.

2.0 REGULATORY REQUIREMENTS

40 CFR Part 61 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, respectively. Radon flux emissions are limited to an average of 20 pCi $m^{-2} s^{-1}$ in both subparts. Method 115 in Appendix B describes monitoring methods to demonstrate compliance with Subparts T and W. A set of radon flux measurements must be made over the course of a year, but may be made more frequently. Method 115 further states that a measurement set should be made for each region—a clearly defined area that has a homogenous make up—in order to determine an accurate measurement of radon flux. The number of sample locations necessary to accurately determine the mean radon flux will be dependent on the homogeneity of the pile measured (Hartley and Freeman 1986). That is, fewer samples are necessary where the expected radium concentration is homogenous over the areal extent of the tailings pile with more samples required to estimate the average flux with decreasing homogeneity. Method 115 takes somewhat of a conservative approach and requires 100 sample locations for each region (water covered areas require no monitoring because the radon flux is assumed to be zero).

3.0 GENERAL RADON MEASURMENT ISSUES

Other isotopes of radon are present naturally, radon-220 (historically, thoron) and radon-219 (historically, actinon) that can potentially interfere with radon-222 measurements. These radon isotopes (Rn-220 and Rn-219) have significantly less radiological impact due to their relatively low abundances and short half-lives (Budnitz 1974). Because of a short half-life (4 seconds), actinon is

unable to diffuse out of the earth's crust. Moreover, actinon originates from U-235, which is in reduced concentration in mill tailing piles. Therefore, actinon is of little concern when measuring radon-222. Thoron, and some of its daughters, can be collected and measured if thoron is allowed to enter the active volume of the detector. Active monitors usually have a delay volume that allows thoron (half-life of 55 seconds) to decay before entering into the active volume of the monitor. Passive monitors used for radon flux measurements, however, are placed directly on the radon emanating surface. Therefore, some passive monitors may record thoron counts along with radon.

4.0 PASSIVE RADON FLUX MONITORS

Passive radon monitors have two basic design types: closed and flow-through accumulators (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 radon concentration in the detector, from which radon flux can be deduced. There is a wide range of detectors used in passive flux monitors. Depending on the type of detector, passive radon monitors can provide real-time radon flux monitoring. Real-time monitoring better allows for measurement of the diurnal variations of radon flux.

One might assume that placing an accumulator on the ground does not perturb the radon exhalation rate. However, this may not be the case as 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). There are sampling methods that have been used for eliminating back diffusion. These methods include using either activated charcoal to absorb radon, flow-through accumulator designs, or short accumulation times (Mayya 2004). However, while these methods reduce back diffusion, they also limit radon concentration in the accumulator which results in an increased detection limit. The net result is limited ability to quantify low concentration radon fluxes and to differentiate the variability. Because

the radon flux at uranium mill tailing sites are potentially higher than those expected from natural soil, the methods for reducing back diffusion mentioned previously may still prove valuable.

4.1 ACTIVATED CHARCOAL RADON FLUX MONITORS

There are few published, peer-reviewed journal articles that investigate radon flux monitoring at uranium mill tailing sites. An internet search did reveal a report prepared by Tellco Environmental for Cotter Corporation to demonstrate compliance with 40 CFR 61.252(a). The report discussed the use of activated charcoal containers to measure radon flux emissions from the Canon City milling facility (Cotter Corporation 2009). Activated charcoal containers are one of the simplest radon flux measuring devices (Countess 1976). A canister, containing activated charcoal, is inverted, placed on the soil to be measured, and sealed to prevent the sample volume from escaping from the ambient air. The canister is left undisturbed for a predetermined time period during which radon exhaled from the soil into the canister volume is absorbed onto the activated charcoal. The charcoal is removed and analyzed by gamma spectroscopy. Radon flux is calculated based on the canister surface area, sample time, and radon daughter activity measured in the charcoal. The detection limit of activated charcoal canisters is dependent on the gamma spectroscopy system used for counting radon daughters. The detection limit is dependent only on the counting system when the flux sampling time is short, because the activated charcoal collects close to 100% of the radon for sample times less than 24 hours (Hartley and Freeman 1986).

4.2 E-PERM RADON FLUX MONITORS

Another radon flux monitor, with similar capabilities as charcoal canisters, is the Electric Passive Environmental Radon Monitor (E-PERM). E-PERM radon detectors consist of a charged Teflon disk that will begin to discharge when exposed to radiation. Discharge of the Teflon disk is a measure of radon exposure. In order to distinguish between discharge from radon and other radiation sources, a control detector is placed near the sample. The control is isolated from radon exposure to measure the amount of discharge from other radiation sources. There are commercially available E-PERM based radon flux monitors (Rad Elec Inc[2]). It has been reported that these radon flux instruments have a lower limit of detection of 9 and 3 pCi $m^{-2} s^{-1}$ for 8 and 24 hour sample time,

[2] Product information may be found at:
http://radelec.com//index.php?option=com_content&task=section&id=18&Itemid=77

respectively, with a 10% error (Stieff et al. 1996). However, the actual measurement error is likely higher as the 10% error reported assumes that the only source of error comes from the ability to measure the voltage of the Teflon disk. The measurement error would increase if the discharge were not entirely due to radon. Gervino et al. made comparisons between E-PERM radon detectors and activated charcoal (2004). E-PERMs showed a lower deviation between a group of measurements than activated charcoal. However, overall accuracy was not studied.

4.3 REAL-TIME RADON FLUX MONITORS

Other types of radiation detectors are commercially available and can be attached to the accumulator to perform *in situ* radon concentration measurements; such as a specialized ionization chamber, semiconductor detectors, and scintillation cells. AlphaGUARD (Genitron Instruments, Germany) is a relatively small, specialized ionization chamber designed to measure radon concentration. The radon concentration build up inside of the accumulator is given by

$$C(t) = \frac{J\,V}{\lambda_e\,A}[1 - exp(-\lambda_e t)], \quad (1)$$

where J is the unperturbed radon flux,
V is the volume of the accumulator,
A is the face area of the accumulator,
and λ_e is the effective time constant.

The effective time constant corrects for back diffusion as well as radioactive decay. Concentration data taken at various times can be fitted to Equation 1 in order to determine λ_e and J. The AlphaGUARD has been used in conjunction with an accumulator to measure radon flux as well as to evaluate experimental diffusion models at uranium mill tailing sites (Sahoo et al. 2010, Ferry et al. 2000a, Ferry et al. 2000b, Ferry et al. 2002). The system developed by Ferry et al. features an AlphaGUARD detector fitted inside an accumulation chamber (2000a). The chamber is fitted to an actuator that automatically removes the chamber from the sample surface. A one hour accumulation time is used to minimize back diffusion. For short accumulation times, Equation 1 can be differentiated to obtain:

$$J = \frac{V}{A}\,\frac{dC(t)}{dt}, \quad (2)$$

where the time derivative of radon concentration in the accumulation chamber with respect to time is taken at the origin. Shorter accumulation times can be used at mill tailing sites because the expected equilibrium radon concentration in the accumulator is much higher than that of natural soil. The AlphaGUARD is also able to record air temperature and pressure inside of the accumulation chamber.

Alpha-spectroscopy can also be used to measure radon concentration inside an accumulator. The energy resolution inherent to semiconductor detectors enables the analysis of the decay energy spectrum of radon daughters. These detectors are based on the well known electrostatic method (Wada et al. 2010, Ui et al. 1996, Iida et al. 1996). The electrostatic collection method works by negatively biasing the semiconductor detector. Because Po-218 is left positively charged about 88% of the time after radon decay, the Po-218 atoms will migrate and collect on the detectors (Hopke 1989). Concentration can be deduced from counting Po-218 decays. Similar to measurements made with the AlphaGUARD, there is an optimal measurement time before back diffusion becomes significant. When accumulation time remains short, radon flux can be calculated from Equation 2. As with the AlphaGUARD detector, this method is able to differentiate decays caused by radon and thoron. Humidity does have an effect on the electrostatic collection process. High humidity can cause neutralization of the Po-218 atoms leading to a decreased count rate (Hopke 1989). Carbon dioxide can also increase Po-218 neutralization in the chamber (Tuccimei and Soligo 2008, Hopke 1989). Both the effects of CO_2 and humidity can be accounted for by determining the collection efficiency of the detector under operational conditions.

Radon can also be detected by scintillation, typically by using ZnS(Ag) as the scintillating material. A scintillation detector based on the flow-through accumulator was developed by Zahorowski and Whittlestone (1996). This system features two scintillation cells, each one liter in volume, and an accumulator. Air from the accumulator is drawn into the scintillation cell where it is counted. There is a delay between the two scintillation cells such that all of the thoron is allowed to decay. Zahorowski and Whittlestone found that back diffusion can be significant for this system (1996). In order to combat back diffusion, the ventilation rate of the accumulator is kept much higher than the rate of loss by back diffusion. Radon flux is determined by a different mathematical procedure than those for the semiconductor and AlphaGUARD based instruments (for a detailed description, see references). This system has the capability to measure radon flux long-term and is able to measure

radon flux variability with seasonal changes (Whittlestone et al. 1998). Detection limits are 4 mBq m^{-2} s^{-1} and 1 mBq m^{-2} s^{-1} for 24 minute and 1 hour count time, respectively.

5.0 FACTORS AFFECTING RADON EXHALATION

There are numerous interrelated factors that affect radon exhalation from soil surface including soil type, atmospheric pressure, soil moisture (i.e. rainfall), and soil temperature. It is difficult to quantify the change in radon exhalation because of these factors because of their interrelation (i.e. a precipitation event is associated with a drop in pressure). The importance of these factors is necessary to consider when determining the optimum flux measurement system.

5.1 SOIL

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, radon soil and concentration. Sandy soil leads to a greater amount of radon exhalation than soil with large amounts of 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 dryer, which generally leads to an increase in radon exhalation.

5.2 ATMOSPHERIC PRESSURE

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. Pressure effects on radon flux in the short term are still debated in the literature (Schubert and Schulz 2002).

5.3 MOISTURE

Soil moisture, largely a function of precipitation, impacts the diurnal radon flux from the air-soil interface. Periods of precipitation results in decreased radon exhalation rates (Ferry et al. 2000a, 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 a slight increase in moisture content led to an increase in radon emanation (1984). Only three materials were tested in their study, one was soil (shale bearing) the others were concrete and shale. The increase in soil moisture was less pronounced for the soil tested than for the other materials

5.4 OTHER FACTORS

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. Intuitively, one might posit that radon flux would increase with increasing temperature because 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.

6.0 RECOMMENDATIONS

Table 1 summarizes the advantages and disadvantages of the radon flux monitors reviewed in this paper. Monitors chosen to make flux measurements will ultimately depend on the study goals. For a large number of measurements, such as those to comply with 40 CFR 61, E-PERM flux

TABLE 1: COMPARISON OF RADON FLUX MONITORS

MONITOR TYPE	ADVANTAGES	DISADVANTAGES
Activated Charcoal	Ease of operation Relatively inexpensive	Lacks ability to measure diurnal variations in radon flux Post processing cost (man-hours/equipment).
E-PERM	Ease of operation Relatively inexpensive Smaller variations in measurements when compared to activated charcoal	Lack ability to measure diurnal variations in radon flux Teflon disks must be replaced after each use
AlphaGUARD	Able to measure diurnal variations in radon flux Measures atmospheric pressure and temperature Can be deployed long-term	Expensive
Semiconductor	Same as above	Not commercially available
Emanator	Same as above	Not commercially available

monitors would be best. E-PERM flux monitors are suggested over activated charcoal because flux values can be calculated almost instantly after a collection time of about 24 hours. The E-PERM flux monitors should be deployed with an appropriate number of controls to measure background radiation. If atmospheric conditions are to be assessed along with radon flux, other instruments, not discussed in this paper, will need to be used. In order to measure diurnal variations in radon flux an monitor containing an AlphaGUARD detector is recommended. This type of instrument can make hourly measurements of radon flux as well as record temperature and atmospheric pressure. Another benefit shared by both the E-PERM flux monitor and AlphaGUARD is that they are commercially available and their performance is well documented. The use of scintillation and solid state detectors is discouraged because of the increased complexity with no added value.

In terms of sample amount and locations, the study region should be divided into areas of uniform radon exhalation. This is accomplished by first deploying a large number of flux monitors to characterize radon exhalation. The total number of measurements is dependent upon the allowable error and should be selected to satisfy the student-t distribution. The number of samples are acceptable when:

$$\frac{\tau(n)\,\sigma}{n} < (\varepsilon)\overline{x}, \tag{3}$$

where τ (n) is the student-t distribution,
n is the sample size,
ε is the allowable error,
and x is the sample mean (Hartley and Freeman, 1986).

Equation 3 is not able to predict the number of samples needed *a priori*. The samples should be placed at random locations across the study region. The random locations may be geo-referenced using GPS technology to enable repetitive measurements over time at the same locations. The total number of measurements need not be collected all at once. For example, if 200 measurements were needed, 100 flux monitors could be used to make two measurements over a specified period of time. These measurements would have to be collected under similar atmospheric conditions. The measurements should not be made within 24 hours of rainfall, per Method 115 B. Once the tailings pile is well characterized, an AlphaGUARD based monitor could be deployed in an area that was representative of the entire pile for obtaining information on the diurnal variability and other environmental variables.

If the goal of radon flux monitoring is to assess the cap effectiveness and integrity, then using a single real-time monitor in an area that is deemed representative of the pile may not be sufficient. This would only be acceptable if the cap were completely uniform with no fractures or varying erosion rates. To assess the integrity, it may be more advantageous to take biannual or even quarterly measurements at the same locations using a large number of E-PERM flux monitors. Even with flux variations due to season and weather conditions, it should be possible to detect a rise in radon flux due to cap degradation, if the measurements were taken over a period of a few years.

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