Final Report

REPORT ON THE REVIEW OF METHOD 115 TO MONITOR RADON EMISSIONS FROM URANIUM TAILINGS

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In accordance with the *Quality Assurance Project Plan: Technical and Regulatory Support to Develop a Rulemaking to Modify the NESHAP Subpart W Standard for Radon Emissions from Operating Uranium Mills (40 CFR 61.25)*, this document has been reviewed and approved by the following individuals:

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1.0 INTRODUCTION

EPA promulgated its NESHAP for emissions from uranium tailings in 1989. Section 112(q) of the Clean Air Act (CAA), as amended, requires the EPA to review, and if appropriate, revise or update the Subpart W standard on a timely basis (10-year interval). EPA has determined that it is reasonable and appropriate to revisit this standard at this time, especially in view of renewed interest in uranium exploration and recovery.

Currently, compliance with the existing emission standards for uranium tailings is achieved through the use of Method 115, as prescribed in Appendix B of 40 CFR 61.253.

2.0 OBJECTIVES

The purpose of this report is to evaluate whether Method 115 is still current and applicable to monitor radon emissions from uranium tailings. Secondarily, it evaluates what other methods using current technology may be employed to meet the emissions standard in lieu of dependence upon Method 115.

Furthermore, this report reviews Method 115 as it was first designed and includes consideration of those variations in detection methodologies that could be employed to meet the requirements of Subpart W.

The review also considers various aspects of Subpart W in the context of what is known, or has been learned since its promulgation, regarding radon diffusion through uranium tailings or improvements in radon detection capability.

Subpart W of 40 CFR 61, *National Emission Standards for Radon Emissions from Operating Mill Tailings*, is basically a three-part standard (EPA 1991), summarized as follows:

(1) Radon-222 emissions from an existing uranium mill tailings pile must not exceed 20 pCi/m^2-sec (implicitly this includes background)

(2) Tailings piles after December 15, 1989, have to be designed, constructed, and operated to meet one of two work practices:
- Phased disposal in lined tailings impoundments that have an area of no more than 40 acres with no more than two impoundments in operation (open) at any one time
- Continuous disposal of tailings requires that they are dewatered and immediately disposed of with no more than 10 acres being uncovered at any time

(3) All mill owners and operators must comply with 40 CFR 192.32(a) in the operation of tailing piles (design of impoundments)

The above performance standards depend in part on certain definitions, such as dewatering, which is defined as removing moisture until the water content is less than 30% by weight. This definition can invoke other parameters, which can further influence radon flux from the tailings. For example, the radon flux from tailings with 6% moisture is actually 3.5 times that from tailings with 0.2% moisture (typical of the southwest) and slowly increases to saturation (NRC 1983). Thus, no matter whether the moisture content is 6%, 30%, or even 60%, the radon flux will be about the same (NRC 1983). The 30% moisture definition appears somewhat arbitrary and does not really affect radon flux reduction considerations.

Historically, compliance with the radon flux standard was met through the use of Method 115 of Appendix B, "Test Methods:"

> *§ 61.253 Determining compliance*
> *Compliance with the emission standard in this subpart shall be determined annually through the use of Method 115 of Appendix B. When measurements are to be made over a one year period, EPA shall be provided with a schedule of the measurement frequency to be used.*

Method 115 in Appendix B, entitled "Monitoring for Radon-222 Emissions," consists of numerous sections that discuss the frequency, distribution, number of measurements, and quality assurance from three types of uranium mining or recovery operations:

(1) Radon-222 Emissions from Underground Uranium Mine Vents
(2) Radon-222 Emissions from Uranium Mill Tailings Piles
(3) Radon-222 Emissions from Phosphogypsum Stacks.

The most important elements are measurement frequency and measurement methodologies employed to demonstrate compliance.

3.0 MEASUREMENT FREQUENCY

These were assumed based on reviews of historic tailings piles constructed prior to the standard. In other words, peripheral discharges of tailings from one or more spigots would have eventually resulted in sands depositing close to the spigot (the dam or beach area) and slimes (material less than 200 mesh) depositing further downgrade forming a "pond." According to Sears et al , the slimes typically contained up to 80% of the radioactivity (ORNL 1975) and were often allowed to dry naturally before being covered or mixed with the sands. The size of these piles varied considerably, but 100 acres was common [NRC used 124 acres (50 hectares) in Regulatory Guide 3.59 (NRC 1985)]. On the other hand, the ORNL (1975) report used 100 acres in the adopted model mills.

Section 2.1.3 of Method 115 requires the following measurements:

(1) Water covered area – no measurements required, as radon flux is assumed to be zero

(2) Water saturated beaches – 100 radon flux measurements

(3) Loose and dry top surface – 100 radon flux measurements

(4) Sides – 100 radon flux measurements, except where earthen material is used in dam construction

Thus a minimum of 300 radon flux measurements are required.

The question then becomes whether this required frequency (300 measurements) is necessary under the new standard, which is requiring much smaller ponds. For example, under the new standard, particularly continuous disposal with only 10 acres uncovered at one time, a possible configuration is a reclaimed area and a 10-acre uncovered area with a water saturated beach, and dry top surface and sides. The 300 flux measurements on a 10-acre area translate to one measurement every 1,500 square ft or one every 40 ft. According to NUREG/CR-3166 (NRC 1983), the number of locations a parameter must be measured to determine its average value within a precision of 25% at the 95% confidence level is given by the following:

Number = 45 (coefficient of variation)[1]

Measurements of the coefficient of variation on covered and uncovered tailings piles results in a range for the Number of 25 to 32 radon flux measurements for older ponds (NRC 1983). The recommended minimum number is 30 radon flux measurements for these older and much larger tailings areas (typically 100 acres or more) (NRC 1983). This is an order of magnitude less than the strict interpretation of the current requirements (requiring 300 measurements). It is recommended that the basis for the number of measurements required in Method 115 be provided and perhaps revised. In addition, the definition of water saturated beach and dry top surface should be provided in terms of percent (%) saturation.

The measurement of radon flux has been reviewed and discussed by many authors since the early 1960s (see, for example, Tanner 1964 and Tanner 1978). In the 1970s, the subject was further investigated as regulators began investigating uranium mining and milling (NRC 1983). In all these investigations, the basic technology remained the same. As the direct measurement of radon flux is extremely difficult and very likely could disturb the actual flux with the entry of measuring devices into the flux, an accumulation (can) method was used. Cans, drums, etc., with one end opened are turned over and the open end sealed into the ground to prevent leakage. Radon is allowed to accumulate into the can when, after a period of time, an aliquot is sampled and measured. From knowledge of the can volume, area of the face, time and measured radon concentration, the average radon flux over that time period can be determined. The sampling device can be charcoal in a canister, loose charcoal, a plastic chip (alpha track or track etch systems), or thermoluminescent dosimeters (TLDs). Equivalently if the accumulator can has a valve, a sample can be bled off and measured in a scintillation cell (Lucas cell), or directly measured by solid state devices or other "real time" detectors. The challenge is to determine the optimal measuring system.

The device described in Method 115 is a Large-Area Activated Charcoal Canister (LAACC) discussed in Appendix A of *Radon Flux Measurements on Gardinier and Royster Phosphogypsum Piles near Tampa and Mulberry, Florida* (EPA 1985). The method is basically a large charcoal canister composed of a 10-in diameter PVC end cap with open end, retainer spring, ½-in thick scrubber pad, ½-in thick charcoal support grid, ½-in thick scrubber pad, 1-in

[1] The coefficient of variation (CV) or relative standard deviation (RSD) is a measure of precision calculated as the standard deviation value divided by the average of a set of values. For a population where σ is the standard deviation of a population and μ is the mean, the CV= σ/μ for a population.

thick scrubber pad, ¼-in hole (vent) in the top. A close examination of the appendix indicates a number of potential problems with the design, including the following:

(1) The specifications of the scrubber materials are not defined. For example, is a scrubber pad made of steel wool?

(2) There are two scrubber pads adjacent to each other. Are they made of different materials?

(3) What type of charcoal was used and how was the system calibrated?

The device is homemade and was developed and constructed by Pacific Northwest Laboratory [PNL, now known as Pacific National Northwest Laboratory (PNNL)] whose machine shops, etc., constructed 50 of these LAACC devices. Obviously, the resources of other groups may be much more limited than PNNL.

Variations of the LAACC have also been used. One researcher used the basic technique of the LAACC, but instead of the loose charcoal, a commercial charcoal canister was secured inside the PVC holder (CoPhysics 2008).

A paper by Andreas C. George presented at the Natural Radiation Environment Conference VIII in Buzios, Brazil (George 2007), and also published in the November 2007 Radon Bulletin of the CRCPD, is particularly relevant in terms of radon measuring devices:

> *In the last 20 years, new instruments and methods were developed to measure radon by using grab, integrating, and continuous modes of sampling. The most common are scintillation cell monitors, activated carbon collectors, electrets, ion chambers, alpha track detectors, pulse and current ionization chambers, and solid state alpha detectors.*

The author breaks down radon detection into two large groupings:

I. <u>Passive integrating radon measurements</u>

   (1) Activated carbon collectors of the open face (OF) or diffusion barrier (DB) type. Charcoal canisters often employ a gamma spectrometer to count the radon daughters as surrogates (Bi-214, for example). Liquid scintillation vials also use alpha and beta counting. About 70% of radon measurements in the United States are canister type.

   (2) Electret ion chambers are being used for 2–7 days duration to measure the voltage reduction (drop). The voltage drop on the electrets is proportional to the radon concentration. About 10%–15% of radon measurements use this methodology.

   (3) Alpha track detectors are used for long-term measurements. Alphas from radon penetrate a plastic lattice, which is etched with acid, and the resulting tracks are counted. There is some use in the United States, but this is more popular in Europe.

II. Passive or Active Continuous radon measurements

(1) Scintillation cell monitors mostly include the flow through type

(2) Current and pulse ionization chambers (mostly passive)

(3) Solid State devices are either passive or active if they use a pump to move air through the sensitive volume of the monitor like the RAD 7, which uses a solid state alpha detector (passive implanted planar silicon (PIPS) detector)

He further reviewed the groupings and compiled sensitivities of a number of monitors using data from manufacturers or cited from published literature. He presents his results in three extensive tables, which were modified and summarized below in Table 1. Table 1 presets the sensitivity range for various types of commercially available OF or DB charcoal canisters, followed by the range of sensitivities of various types of continuous radon monitors (CRMs) utilizing solid state detectors or pulse ionization chambers. The last group is the scintillation cells, which are typically cylinders coated with zinc sulphide or other scintillation materials that respond to the alpha decay of radon (also known as Lucas cells). In the first series, the exposed canisters are counted and the radon is determined from the concentration of the Bi-214 or other daughter surrogate.

For the second group, the first set of sensitivities range from 0.18 to 0.36 cpm/4.0 pCi/L). These detectors run continuously, which compensates for their lesser sensitivity. Other solid state monitors use PIPS as the detector or ionization chambers with somewhat higher sensitivities. The last group includes the scintillation cells, which vary in size and sensitivity.

Table 1. Sensitivity of Radon Monitoring Instruments

Instrument	Detection Principle	Sensitivity Range cpm/(4 pCi/L)
DB canister (various types)	gamma	0.80 to 4.2 (range)
OF canister	gamma	250
Db (2g) canister	alpha	54
Continuous radon monitor (CRM)	Solid state	0.18–0.36
Other CRM (e.g., PIPS)	Solid state	2.80
Pulse or continuous ionization	Pulse ion	1.2–3.0
Pylon CRM others	Scintillation Cell	5.7 to 24 (Lucas Cell or equivalent)

The paper also presents the results from various track etch detectors and electret ion chambers with different units for the sensitivity. The sensitivity of various track etch units ranges from 1.70 tracks per cm^2/4 pCi/L-day to 23.4 tracks per cm^2/4 pCi/L, while the sensitivity for the electrets are 8 volts per 4 pCi/L-day for short-term measurements to 0.7 volts per 4 pCi/L-day for long-term measurements.

In summary, it appears that while no one type of detector is "better" than another type, it invariably depends more on the physical circumstances and the time required for the measurement. For radon fluxes over a few days, a series of charcoal canisters can be used or a series of accumulator cans could be deployed, with samples taken periodically over a few days to get an average. It is important to note that there is no inherent advantage to fabricating and using an LAACC device, except for looking at a larger area. The device was first designed to collect radon over a larger area, which, as mentioned, could be accomplished using a similar "shell" and a canister. Note that the amount of charcoal in the LAACC was 400 ml or, assuming a density of 0.4 gm/cc, leads to a mass of 160 gm of activated charcoal. The EPA-recommended 4-in canister used in the home (the size of a tuna can) has a volume of about 66% of the LAACC and likely holds about half as much charcoal. The canisters, however, are easier to use and more standardized, and more importantly, they are commercially available. While these "off the shelf" devices could be used instead of the LAACC, they would have to be evaluated against a specification to assure accuracy, precision, and importantly reproducibility.

It should be noted that the direct measurement of the radon flux from uranium tailings is not required by current guidance from the Nuclear Regulatory Commission (NRC). NRC requirements are presented in Regulatory Guide 4.14, Revision 1, *Radiological Effluent and Environmental Monitoring at Uranium Mills*, April 25, 1980 (NRC 1980) and NUREG-1620 *Standard Review Plan for the Review of a Reclamation Plan for Mill Tailings Sites Under Title II of the Uranium Mill Tailings Radiation Control Act* (NRC 2000). The regulatory guide calls for pre-operational radon flux measurements to establish background flux, but does not require operational flux measurements in this guide. This Regulatory Guide is expected to be updated by December 2009, as it is currently a phase 3 draft guide. The Standard Review Plan indirectly monitors radon emissions through calculations based on measurements or published values for the parameters governing radon diffusion, such as radium-226 concentration, soil type, diffusion coefficient moisture, and so on.

4.0 CONCLUSION

This review of Method 115 demonstrates that its use can still be considered current for use to monitor radon flux from uranium tailings. It is important to note, however, that the specific design protocols were developed with larger tailings impoundments in mind. Alternatively, numerous commercial enhancements to that design are widely available and in use today. Other forms of passive detectors, as well active measurement detectors, are also acceptable alternatives to demonstrate conformance with the standard. In addition, the method as currently written has a number of elements and requirements that should be reviewed and possibly revised, particularly the location and the frequency of measurement. These would be better based on statistical considerations or some other technical basis.

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