# Radon Fluxes from an Earthen Barrier Over Uranium Mill Tailings After Two Decades of Service - 17234

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## ABSTRACT

Radon (Rn) fluxes were measured at a disposal facility for uranium mill tailings after two decades of service. The facility was closed with an earthen cover vegetated with grasses. Measurements were made on the surface of the Rn barrier and directly on the surface of the tailings. Fluxes were measured using extra small (area = 0.018 $m^2$), small (0.071 $m^2$), medium (0.59 $m^2$), and large (2.32 $m^2$) flux chambers to evaluate the impact of measurement scale on Rn flux. Activated carbon (AC) passive collectors and electric radon detectors (RAD7) were used to measure Rn concentrations. Tests were conducted at various locations on each cover representing conditions that can lead to different levels of soil structural development and different water content. Rn fluxes at the surface of the Rn barrier were much lower than Rn fluxes measured at the surface of the tailings, indicating the barrier remained effective for Rn containment. Geometric mean Rn fluxes measured at the surface of the Rn barrier in each test pit were below the regulatory requirement (0.74 $Bq/m^2$-s). Rn concentrations measured using AC samplers in the flux chambers were 60% of concentrations measured using the RAD7, on average, and Rn fluxes computed with the AC data were 9% of those computed with the RAD7 data, on average. Size of the flux chamber had no systematic effect on Rn flux, indicating that 20 y of soil forming processes had not created a pore network causing scale-dependent Rn flux. Geometric mean Rn fluxes in the test pits were similar regardless of differences in surface conditions (vegetation, thickness of soil cover) known to influence soil forming processes and soil structure, but were lower in areas where surface conditions promoted higher water content in the Rn barrier.

## INTRODUCTION

Disposal facilities for uranium mill tailings generated by current and historic uranium beneficiation operations have been constructed at locations throughout the United States as required by the Uranium Mill Tailings Radiation Control Act (UMTRCA). Nearly all UMTRCA disposal facilities rely on a surface cover to control the rate at which contaminants migrate in the gas and water phases from the tailings and into the surrounding environment.

A schematic profile of a typical surface cover is shown in Fig. 1. The lowermost layer, commonly referred to as the "low-permeability radon barrier" (or "Rn barrier") is the primary component that controls fluxes in the gas and water phases. Rn barriers with low saturated hydraulic conductivity and low gaseous diffusivity can be very effective in controlling the emission of radon into the atmosphere and ingress of precipitation into the underlying waste. The Rn barrier is overlain by protection and erosion control layers intended to stabilize the cover and provide protection against physical and biological processes that can increase the saturated hydraulic conductivity and gaseous diffusivity of the Rn barrier. The erosion control layer may be riprap or a vegetated soil layer depending on site-specific conditions.

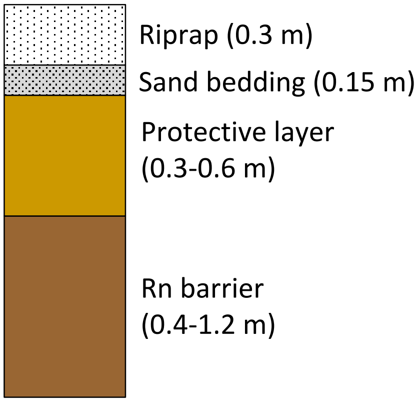

Fig. 1. Typical profile for a UMTRCA surface cover.

Field research at disposal facilities has demonstrated that abiotic and biotic surface processes accelerate natural soil-forming processes within cover profiles [1,2]. Root intrusion, insect and animal intrusion, wet-dry cycling, and freeze-thaw cycling induce volume change, cracking, and translocation of materials and soil aggregates. These processes, which are inevitable and ubiquitous at the near surface, can create macro-structure in the Rn barrier, causing the hydraulic conductivity and diffusivity to increase, and potentially resulting in greater radon emissions and seepage of contaminants to groundwater [3-7]. Soil drying caused by evapotranspiration may also reduce the water saturation of the Rn barrier, resulting in a higher gaseous Rn diffusion coefficient and higher fluxes.

Understanding how these abiotic and biotic processes affect radon emissions is critical for designing and predicting the performance of future surface barriers. Although the mechanisms and impacts associated with these processes have been established, their significance in terms of water and gas transport into and out of uranium mill tailings disposal facilities is unknown. We are currently conducting a study to evaluate how these ecological and structure-forming processes may affect gaseous and water fluxes in UMTRCA disposal facilities. Part of this study involves measuring Rn fluxes from Rn barriers at existing UMTRCA disposal facilities that have been in service for an extended period. This paper describes Rn fluxes measured at the UMTRCA disposal facility in Falls City, Texas that has been in service for 20 y, and provides a comparison with regulatory limits.

## FIELD SITE

The cover profile at the Falls City, Texas disposal facility is shown in Fig. 2. The top deck area of the cover consists of (bottom to top) a clay Rn barrier (0.91 m), a growth medium/protective layer (0.76 m) constructed of similar soil as the Rn barrier (but not compacted), and top soil (0.15 m). The top deck is vegetated with indigenous and introduced grasses with occasional mesquite brush. The side slopes have a thinner Rn barrier (0.61 m) overlain by a sand bedding layer (0.15 m) and rip rap (0.61 m). An apron along the edge of the top deck provides a transition between the top deck and the side slope. In the apron, the Rn barrier is 0.91-m thick, the bedding layer is 0.30-m thick, and the rip rap is 0.61-m thick. Construction of the cover was completed in 1996 over a 50-ha area. The disposal cell contains 6,480,020 dry Mg of waste and 47.2 TBq of $^{226}$Ra.

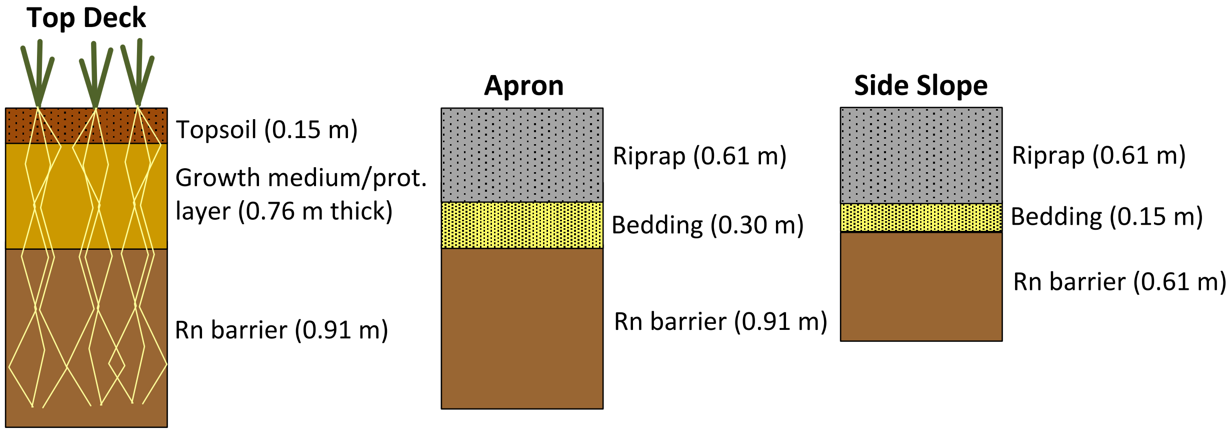

Fig. 2. Schematic cover profiles for disposal facility at Falls City, Texas.

Falls City is in southeastern Texas, approximately 75 km southeast of San Antonio and 320 km southwest of Houston. The climate is warm and humid, with an average annual precipitation of 740 mm distributed relatively uniformly throughout the year. The average annual air temperature is 20.4 $^{o}$C. Subfreezing air temperatures are uncommon and subfreezing temperatures at the depth of the Rn barrier are not expected.

## METHODS

### Test Pits

Test pits were excavated in groups of two at three locations on the cover. Each pit was approximately 4 m x 4 m and was excavated down to the surface of the Rn barrier using a backhoe and hand tools. After Rn flux measurements were made on the surface of the Rn barrier, a smaller pit was excavated through the Rn barrier to the surface of the tailings so that flux from the tailings could be measured. After the measurements were made, the entire cover profile was reinstated to as-built specifications.

Two sets of pits were on the top deck and one set was on the apron and side slope. On the top deck, one pit in each group was excavated in an area vegetated with grasses and the other in an adjacent area with mesquite. The expectation was that the deeper woody root system of the mesquite would have a different impact on the Rn barrier relative to the shallower root systems associated with grasses. The two different top deck locations were selected corresponding to different levels of activity anticipated in the tailings (lower vs. higher). The side slope test pits were selected to contrast conditions in the apron vs. the actual side slope and to assess whether Rn fluxes are different in areas covered by rip rap vs. areas covered with grasses.

## Radon Flux Measurements

Radon fluxes were measured using flux chambers equipped with a RAD7 continuous electronic radon flux detector (Durridge Company, Inc. Billerica, MA) and a 100-mm-diameter open-faced activated carbon (AC) canister (Radon Testing Corporation of America, RTCA, Elmsford, NY) (Fig. 3). Four chamber sizes with different cross-sectional area were used to evaluate potential scale effects in the Rn measurement: large (surface area, A = 2.32 m$^2$; volume, V = 0.35 m$^3$), medium (A = 0.59 m$^2$, V = 0.20 m$^3$), small (A = 0.071 m$^2$, V = 0.011 m$^3$), and extra small (A = 0.018 m$^2$, V = 0.002 m$^3$). The small chamber is similar to chambers used historically to measure Rn fluxes for construction documentation and certification [8, 9]. Each chamber was fitted with gas-tight ports and tubing to connect to a RAD7 detector [9] (Fig. 3). The perimeter of the chamber was sealed with bentonite paste.

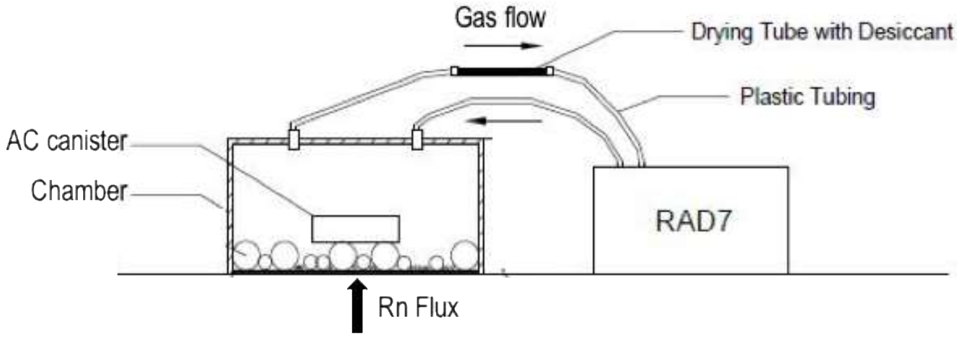

Fig. 3. Schematic of Rn flux chamber used in study.

The RAD7 detector is a solid-state $\alpha$ detector that measures disintegration of $\alpha$ progenies over a designated time period [9, 10] in gas cycled through the detector. A pump flow rate of 800 mL/min and a cycle time of one hour were used for all tests. The AC canisters (National Radon Safety Board device code 10331) contained 90 g of AC. Canisters were sealed in packing envelopes provided by RTCA and shipped to RTCA for analysis immediately after measurements were made.

A typical Rn build-up curve measured with the RAD7 is shown in Fig. 4. The Rn concentration builds linearly during the early phase of the test, and then the rate of increase diminishes as the concentration gradient diminishes. In the example in Fig. 4, the linear portion occurs for approximately the first 7 h.

Radon flux (J) from the RAD7 data was obtained by fitting the analytical solution in Chao et al. [10] to the data for each build-up curve. The analytical solution is:

$$C = \left( C_i - \frac{JA}{V(\lambda + D)} \right) \left( 1 - e^{-(\lambda + D)t} \right) + \frac{JA}{V(\lambda + D)} \qquad \text{(Eq. 1)}$$

where C is Rn concentration in the chamber at time t, $C_i$ is the initial Rn concentration, λ is the decay rate, D is the back-diffusion coefficient, and V is the volume of the chamber. The decay term is necessary because of the short half-life of Rn (3.8 d). The back-diffusion term accounts for diffusion of Rn back into the source material as the concentration builds inside the chamber. The flux J in Eq. 1 corresponds to the Rn flux into the chamber at time zero. Eq. 1 was fit to the Rn build-up data using non-linear least-squares regression (example fit shown in Fig. 4). Fluxes computed by fitting Eq. 1 were checked by comparison with fluxes computed from the initial linear portion of the build-up curve using linear regression.

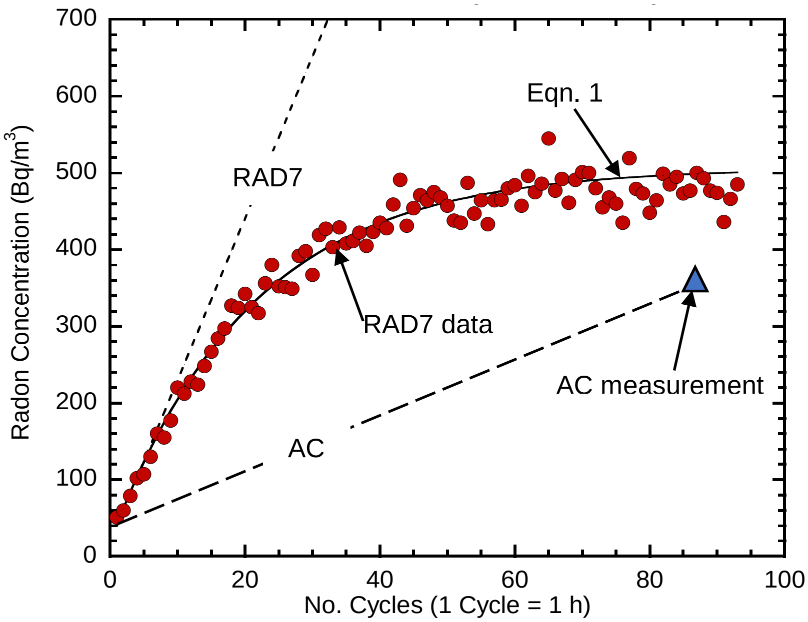

Fig. 4. Example of Rn build-up curve (from laboratory calibration test) measured with RAD7 and fit with Eq. 1 along with Rn concentration from AC measurement.

Flux was calculated from the AC data assuming the build-up of Rn in the chamber is linear and that the AC is in equilibrium with the gas in the chamber. When build up occurs non-linearly, the flux computed from AC data is lower than the actual radon flux, as illustrated by comparing the RAD7 and AC slopes shown in Fig. 4.

## RESULTS

### Rn Build-Up at Surface of Tailings and Radon Barrier

Typical Rn concentrations measured on the surface of the Rn barrier and on the surface of tailings are shown in Fig. 5 for a test pit excavated in the top deck in an area vegetated with grasses. Similar graphs were obtained for each test pit. The Rn concentrations are much higher and build more rapidly on the surface of the tailings than on the surface of the radon barrier, indicating that the Rn barrier is effective in reducing release of Rn from the tailings.

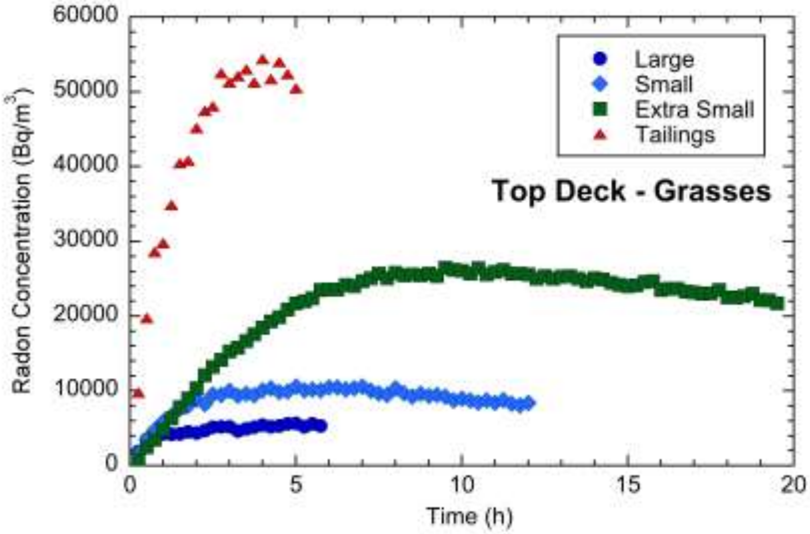

Fig. 5. Flux buildup curves measured with flux chambers and RAD7 radon detectors on surface of tailings and on surface of Rn barrier.

The initial slope is similar for each of the buildup curves measured on the surface of the Rn barrier, indicating that the Rn flux from the Rn barrier in this test pit does not depend significantly on the scale of the measurement. The buildup curves level off at different concentrations due to differences in volume of the chambers.

### Fluxes from RAD7

Rn fluxes measured with the different size chambers in the six test pits are compared in Fig. 6. There is no systematic effect of size of the flux chamber for any of the test pits. This suggests that soil-forming processes that introduce structure into engineered barriers did not affect the network of pores controlling gas-phase diffusion in the radon barrier at this site. Similar geometric mean Rn fluxes were measured for Test Pit (TP) 1 (top deck with mesquite) and Test Pit 2 (top deck with grasses), even though different root structures were observed in the Rn barriers beneath the areas with mesquite relative to areas with grasses. Similarly, geometric mean fluxes in the apron and side slope areas for Tests Pits 5 and 6 were comparable, even though the thickness of material over the Rn barrier in these locations differed (Fig. 2), providing different levels of protection to the Rn barrier. The activity of the underlying waste also differed substantially between the apron and side slope (see fluxes on waste surface for Test Pits 5 and 6 in Fig. 6).

The geometric mean Rn flux is nearly 10x higher for Test Pit 3 (top deck with mesquite) relative to the Rn flux for Test Pit 4 (top deck with grasses). This difference cannot be attributed to different attributes of the Rn barrier, however, as the tailings activity beneath the Rn barrier differed significantly between Test Pits 3 and 4, despite their close proximity. The Rn flux measured on the surface of the tailings beneath Test Pit 3 was 5-15x higher than beneath Test Pit 4, which most likely contributed significantly to the difference in the geometric mean Rn fluxes.

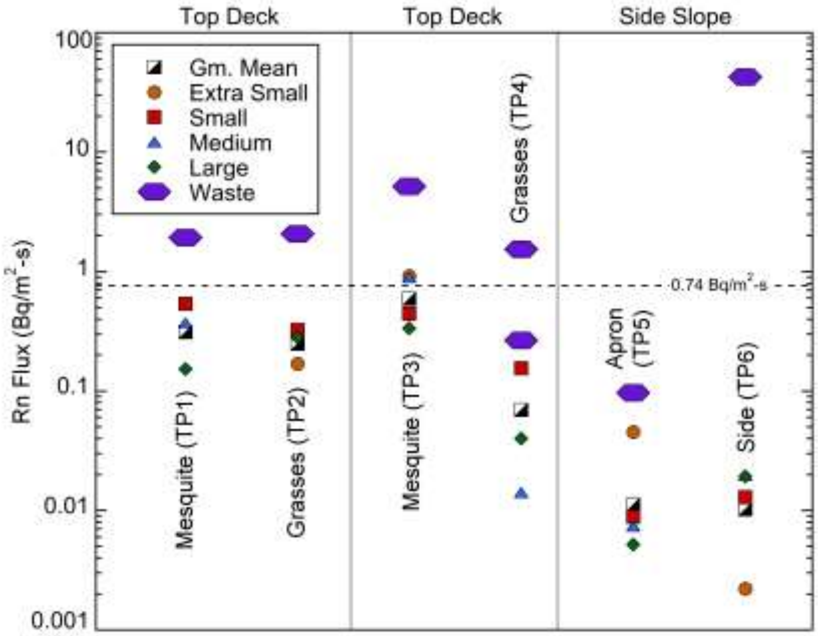

Fig. 6. Fluxes measured on the surface of the radon barrier and on surface of waste with flux chambers using RAD7 radon detectors. TPX = Test Pit X. Dashed horizontal line is regulatory threshold for Rn flux (0.74 Bq/m$^2$-s).

Rn fluxes on the side slope (Test Pit 6) are substantially lower than those from the top deck, even though the Rn flux from the surface of the tailings beneath Test Pit 6 was substantially higher than in the other pits. These lower fluxes are attributed in part to higher water contents in the Rn barrier on the side slope relative to the top deck. On the side slope, the water content of the Rn barrier was 9-13% higher than in the top deck, which results in lower gas diffusivity and therefore lower Rn flux. The rock cover with little vegetation apparently promoted ingress of precipitation and limited evaporation, keeping the Rn barrier wetter.

All but two of the fluxes from the surface of the Rn barrier in Fig. 6 are below the regulatory threshold (0.74 Bq/m$^2$-s), and for each test pit the geometric mean Rn flux is below the threshold. Thus, after 20 y of service, the Rn barrier at Falls City, Texas appears to be functioning as intended in terms of Rn flux. Other engineering properties of the barrier important to containment are currently being studied.

### Comparison of Fluxes from RAD7 and AC Data

Ratios of Rn concentrations and fluxes measured with the AC canisters relative to those measured with the RAD7 are shown in Fig. 7. Rn concentrations measured with AC canisters range from 49 to 69% of concentrations measured with RAD7, with a geometric mean of 60%, even after corrections were applied to account for humidity in the AC. Rn fluxes measured with the AC canisters range between 2 and 28% of the fluxes measured with the RAD7, with a geometric mean of 9%.

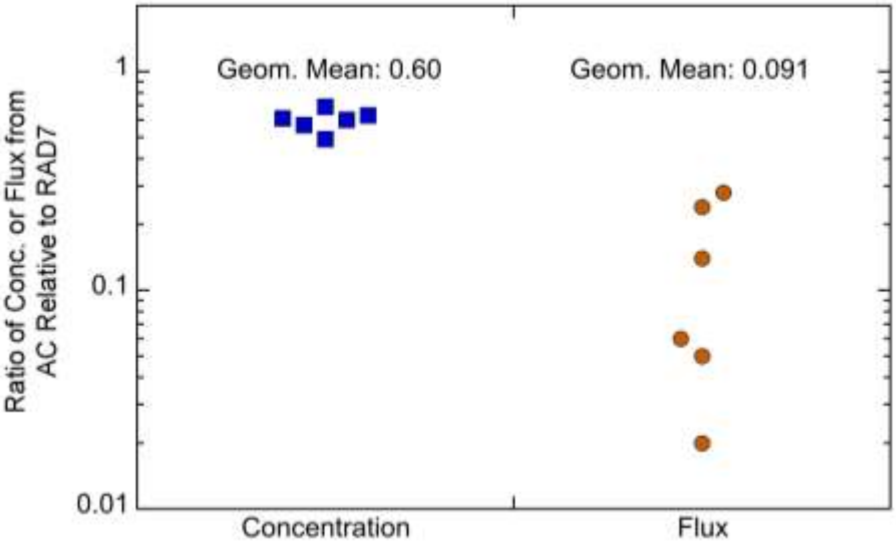

Fig. 7. Ratio of Rn concentration or flux measured with AC relative to RAD7.

Factors contributing to the lower Rn concentrations measured with AC are still being explored, and likely include mass transfer limitations affecting diffusion of Rn into or out of the AC. The lower fluxes measured with the AC data are due to the combined effects of the under-estimate of the Rn concentration along with ignoring the non-linearity in the Rn buildup curve when making computations using a single-point (linear) measurement with AC.

## SUMMARY, CONCLUSIONS, AND PRACTICAL IMPLICATIONS

Rn concentrations and fluxes were measured on the surface of the Rn barrier in six test pits excavated in a final cover over a uranium mill tailings disposal facility in Falls City, Texas. The final cover had been exposed to soil forming processes combined with wetting and drying while in service for two decades. Measurements were made with flux chambers having four different sizes to determine if soil structure in the Rn barrier would contribute to scale-dependent fluxes. Measurements were also made on the surface of the tailings in each test pit. Rn concentrations and fluxes were measured with activated carbon (AC) canisters and RAD7 electronic radon detectors. The AC canister provides a single-point measurement of Rn concentration, whereas the RAD7 provides a continuous Rn buildup curve.

Comparison of Rn concentrations and Rn fluxes measured on the surface of the Rn barrier and the surface of the tailings indicates that the Rn barrier is effective in reducing Rn concentrations and Rn flux. Rn concentrations and Rn fluxes were substantially higher on the surface of the tailings relative to the surface of the Rn barrier, and the geometric mean Rn flux from the top surface of the Rn barrier was beneath the regulatory threshold (0.74 Bq/m$^2$-s) in each test pit after 20 y of service.

No systematic differences were found between Rn fluxes measured at different scales or in Rn barriers exposed to different soil forming processes. Similar geometric mean Rn fluxes were measured beneath areas vegetated with grasses and areas with mesquite, even though different root structures in the Rn barriers were observed. Similar geometric mean Rn fluxes were also obtained in areas with rip rap on the surface even though the thickness of soil over the Rn barrier differed. This suggests that, at least at this disposal facility, soil forming processes were not creating structure in the Rn barrier that affected Rn transport.

Lower fluxes from the Rn barrier were measured in the apron and slide slope areas that were covered with rip rap relative to the top deck, which was covered with fine-textured soils vegetated with grasses and mesquite. Water content of the Rn barrier was also substantially higher in the areas covered with rip rap, which probably resulted in a lower gaseous diffusivity and therefore lower fluxes. Higher water content of the Rn barrier in the areas covered with rip rap is attributed to greater infiltration of precipitation and lower evapotranspiration due to the rip rap cover.

Rn concentrations measured with AC canisters were 60% of Rn concentrations measured with the RAD7, on average, and fluxes computed from the AC canister concentrations were 9% of those measured with the RAD7, on average. The downward bias in concentrations from the AC canisters is being explored. The downward bias in Rn flux computed with data from the AC canister is believed to be due to the lower concentration and ignoring non-linearity in the Rn buildup curve in the flux chamber. Conducting both measurements simultaneously may have also affected the measurements and is being explored. Electronic radon detectors like the RAD7 are recommended for measuring Rn concentrations and fluxes from Rn barriers.

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