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EFFECTS OF VARIOUS TAILINGS COVERS ON RADON GAS EMANATION FROM PYRITIC URANIUM TAILINGS

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EFFECTS OF VARIOUS TAILINGS COVERS ON RADON GAS EMANATION FROM PYRITIC URANIUM TAILINGS

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Abstract. Radon emanation studies were carried out at an inactive pyritic uranium tailings site in Elliot Lake, Ontario, Canada, to evaluate the effects of various existing dry and wet covers on radon flux rates. Measurements were taken using activated charcoal cartridges for various surface covers consisting of bare, vegetated, acidophilic moss with high degree of water saturation, compacted crushed rock and gravel, and winter snow.

The results showed that at a given site, there was no significant difference in radon emanation rates between various tailings covers and bare tailings. In particular, no increase in radon emanation rates from vegetated areas compared to bare tailings was observed.

Radon emanation rates varied spatially depending on tailings grain size, porosity, moisture content and on pressure and water table variations. The emanation rates were higher for tailings with low water contents compared to those for wet and moss covered tailings.

Introduction

Conventional mining and milling of low grade uranium orebodies produce large amounts of low level radioactive residue or tailings which are usually deposited in extensive tailings impoundments located above grade or in shallow depressions. The residual radioactivity is from uranium decay series radionuclides such as Th-230, Ra-226, Rn-222, Pb-210, etc., which are not recovered in the milling process and are associated with the tailings.

Two of the radionuclide families associated with the parent isotopes, radium (i.e., Ra-226), and their daughters, radon gas (i.e., Rn-222) have been identified as of importance to man, contributing through various environmental pathways. The former through water/food chain pathways, and the latter through air.

Radon (Rn-222 for this study, half-life 3.825 days) is an inert gas, and is produced by the decay of its parent nuclide Ra-226 (half-life 1602 years). Because of its kinetic energy associated with the decay and other diffusional forces, a certain fraction of the radon produced migrates to the tailings pore space. Once in the pore space, radon atoms move through it by molecular diffusion and/or as transport of the interstitial fluid induced by temperature and pressure gradients. There is, thus, a continuous release or emanation of radon from tailings surfaces which is a source of environmental radioactivity in terms of its transport as an airborne gas and its decay products.

The particulate daughter products of radon (i.e., Po-218, Pb-214, Bi-214, etc.) readily attach to aerosols or dust particles and can further be transported by various other environmental pathways. Typical radon release rates from existing tailings piles vary between 35 to 700 atoms/cm^2 s (740 to 14,800 mBq/m^2 s) depending on the nature of the tailings material and cover, moisture contents, etc.[1-4].

The U.S. Nuclear Regulatory Commission (NRC), the U.S. Environmental Protection Agency, and the Canadian Atomic Energy Control Board[5-8] have recommended that appropriate covers be placed on uranium tailings to reduce the surface radon flux to approximately 3 to 18 atoms/cm^2 s (74 to 370 mBq/m^2 s) above that of background levels. Considerable laboratory, field and modelling research has been undertaken to design and evaluate various covers to meet the proposed regulatory guidelines[4,9-16]. The present study was undertaken to evaluate the effects of various existing dry and wet covers on tailings such as compacted crushed rock and gravel, terrestrial vegetation, semi-aquatic moss, sewage sludge, water saturated tailings, and snow on the emanation of radon from inactive uranium tailings.

Method

Site Location and Description

The study was conducted as the Nordic Mine uranium tailings site in Elliot Lake, Ontario, Canada (46° 23'N, 82° 39'W). The site is located about 5 km east of the town of Elliot Lake, Ontario (Figure 1).

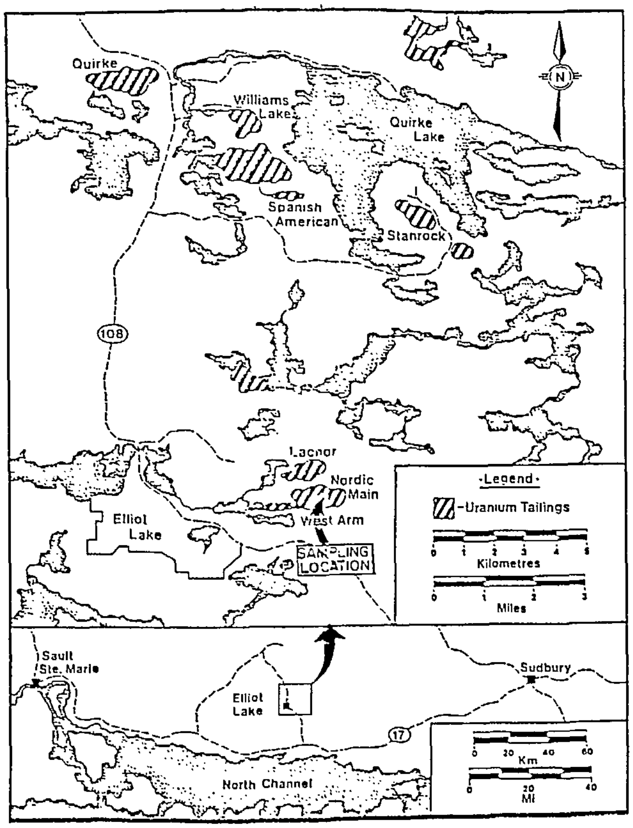

Figure 1. Location of the study site, Elliot Lake, Ontario, Canada.

The Nordic tailings impoundments are shown in Figure 2. The largest impoundment, known as the Nordic Main Tailings, covers an area of 70 hectares and contains approximately 10 million tonnes of tailings with an average thickness of 12 m. The smaller impoundment situated immediately west of the Nordic Main, known as the Nordic West Arm, covers an area of 15 hectares, and contains approximately 2 million tonnes of tailings with an average thickness of 7 m. A peripheral dam of mine waste and overburden, and cross-valley dams complete the impoundment.

The tailings were discharged in the form of neutralized slurry containing approximately 20% solids consisting of a mixture of sand and silt, comprised of quartz, feldspar, approximately 5% pyrite, minor sericite, gypsum and hydroxides formed following lime neutralization.

Deposition of the mill tailings from the Nordic Mine began in 1957 in the Nordic West Arm which was filled to capacity by 1960 and left inactive. The Nordic Main impoundment was then completed and the tailings were deposited there until 1968 when the Nordic Mine and mill ceased operation.

The experimental vegetation program on the tailings surface began in 1973 at the West Arm, and by 1978 most of the area was covered with vegetation except some slimy areas where the water table was high. During 1978-80, a dense vegetation cover was established on the Nordic Main tailings. The revegetation technique consisted of neutralizing the acidified pyritic tailings with agricultural limestone to a depth of approximately 15-20 cm, and direct seeding with mixtures of grasses: Creeping Red Fescue (Festuce rubra L.), and Red Top (Agrostic alba L.), and a legume, Birdsfoot Trefoil (Lotus corniculatus L.). Appropriate fertilizer and reseeding applications were provided for the first five years. Since then the cover has been self-sustaining.

In order to mobilize heavy machinery and equipment during revegetation and subsequent research programs, crushed rock roadways were constructed on tailings as required. For experimental purposes, sewage sludge from the town of Elliot Lake was also placed at designated locations to provide suitable organic matter and microbial environment to promote vegetation growth. To date, most of the tailings areas support a lush green vegetation, except for poorly drained fine-grained tailings sites where acidophilic semi-aquatic moss species have established. A soil like layer, 6 to 8 cm in thichness, consisting of decaying vegetative organic matter has developed on top of the vegetated tailings.

Radon sampling sites were located, as shown in Figure 2, at T-1, for vegetated fine tailings, T-5 and T-7 for vegetated coarse tailings, T-8 for vegetated tailings amended with sewage sludge approximately 10 cm thick, G-5 for compacted crushed rock and gravel cover approximately 0.6 m thick, M-1 and M-2 for semi-aquatic moss covered tailings with high water table, T-9 for bare coarse tailings, and S-5 for winter snow cover approximately 0.5 m thick and frozen tailings.

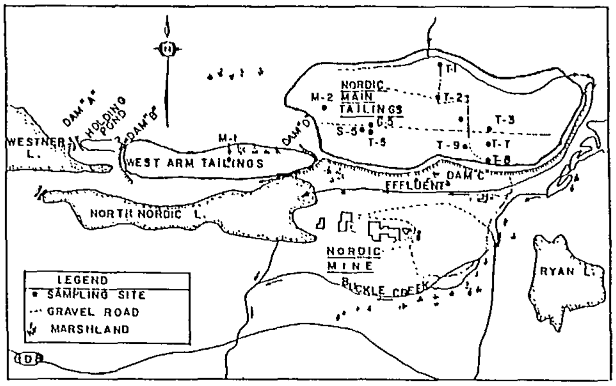

Figure 2. Location of test sites, Nordic tailings impoundment.

Sampling Method

Figure 3 shows the radon emanation sampling arrangement. It consisted of a MSA-GMA gas mask type chemical cartridge (Part No. 459315, Mine Safety Appliance Co., Pittsburgh, PA) containing activated charcoal and measured 7.3 cm in diameter and 2.5 cm in height. The cartridge was attached to the top of a holding ring, 7.4 cm in diameter (I.D), and 25 cm high, with sealing tape. The holding ring was placed over a protective filter (to prevent particulate matter contaminating the cartridge) and pushed slightly into the sampling surface to ensure a good seal. An inverted plastic cup with vent holes on sides near the top covered the sampling assembly as rain and moisture guard. A total of 10 such sampling devices were used, each containing the cartridge from a single batch.

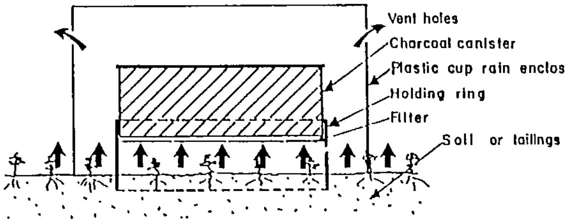

Figure 3. Radon emanation sampling arrangement.

Prior to their placement for sampling, all the cartridges were heated at 125°C for 12 h in a ventilated oven to expel any radon and moisture accumulated by previous usage or storage. The cooled cartridges were sealed in plastic bags until used.

At each sampling site, two adjacent plots of size 1 m x 1 m were selected, one consisted of the appropriate cover, and the other was a bare tailings plot which in some cases was prepared by removing the cover. In each plot, five sampling cartridges were placed, one at each corner and one in the middle to provide a representative coverage of the area sampled. For sampling vegetated plots, the vegetation height was kept small during the sampling period for easy placement and sealing of the sampling cartridge.

The sampling period extended from July to September with the vegetated plots sampled during early summer when the growth was at its peak. For snow covered tailings, the sampling was done in early March when about 0.5 m of snow covered the tailings surface. The sampling time varied from 3 h to 20 h depending on the required sample integration. At the end of sampling each cartridge was again placed in a plastic bag and sealed to prevent any loss of radon. Ambient temperature and pressure were also recorded before and after the sampling period.

As the size of the sampling cartridge was small, to provide an integrated flux rate from the whole sampling area without localized effects, the data from the five sampling cartridges were averaged to obtain a representative measurement of the flux over the entire sampled area.

In order to avoid any effects related to variations in the meteorological parameters, measurements for bare and covered tailings were performed simultaneously.

At site T-9, containing coarse tailings and bare, all the ten sampling cartridges were placed within a 1 m x 1 m sampling area to measure the localized variations within a site.

In a separate study, laboratory emanation tests were also conducted for tailings containing various degrees of moisture saturation.

Analytical Method

The absorbed radon concentration in the charcoal cartridge was measured by gamma spectroscopy where 609 keV decay peak of radon daughter Bi-214 in equilibrium was counted. The experimental arrangement consisted of a Harshaw type 16MB/61B, 10.2 cm x 10.2 cm NaI (Tl) detector, a victoreen model 460401 pre-amplifier, Harshaw model NA-11 RC linear amplifier, Ortec model 446 high voltage power supply, and a Tracor Northern model NS-630 multi channel analyzer and data acquisition system. The detector assembly was mounted in a lead shielded box to shield it from external radiation.

Figure 4 shows a typical gamma spectrum of the absorbed radon and its daughter products in the sample cartridge. Because of the broadening of the low energy 295 and 352 keV peaks of Pb-214 in the presence of absorbed thoron Rn-220 daughters, 609 keV gamma peak of Bi-214 was found most suitable for measuring absorbed radon concentrations.

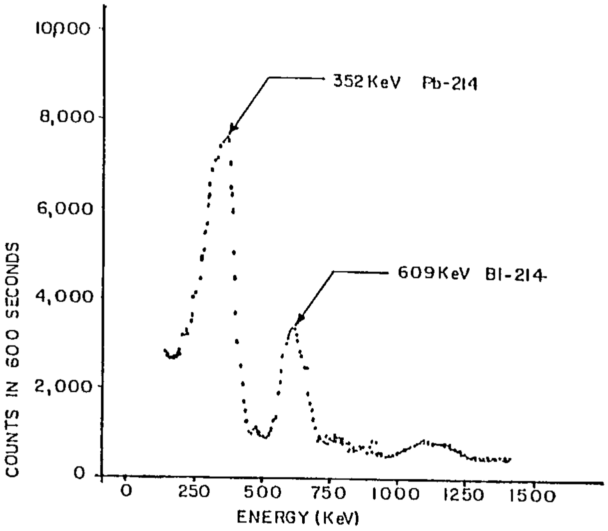

Figure 4. Gamma spectrum of radon and its daughter products using NaI(Tl) detector.

For radon counting, the collected samples were allowed a minimum of 3.5 h equilibrium time to establish secular equilibrium between radon and its daughter products before counting. Typical counting time was for 10 min. The system was calibrated both for energy and counting efficiency, using a sealed tailings source which was prepared by placing 70 g of dried powdered and homogenized tailings containing 36,663 ± 1665 mBq/g (153,986 disintegrations per minute, DPM) Ra-226 in a metal container machined to the same dimensions as those of the charcoal cartridges. The source was sealed airtight and allowed to establish equilibrium between Ra-226 and its daughter products for a minimum of 30 days before counting. It was periodically leak tested for loss of radon gas. For 609 keV Bi-214 peak, the measured calibration efficiency factor ε (counts/disintegration), was 0.0682.

The radon emanation flux J, from a given surface was calculated as:

$$J = \frac{C}{\varepsilon.A.(1 - e^{-\lambda t_1}).e^{-\lambda t_2}} \qquad (1)$$

where, J = radon flux, atoms/cm^2 s
C = net counts (counts-background) for 609 keV Bi-214 peak/s
ε = detector efficiency, counts/disintegration.
A = collection area of the cartridge holding ring, cm^2.
λ = decay constant of radon, h^{-1} (7.5506×10^{-1} h^{-1}).
t_1 = sample collection time, h.
t_2 = time lapse between end of sampling and start of counting, h.

The measured flux in atoms/cm^2 s is converted by multiplying with 20.98 to SI units mBq/m^2 s.

Results and Discussion

Table 1 and Figure 5 show the observed radon emanation fluxes for various covers and bare tailings at different sites. At most of the sites, a localized variation of the order of ±20% in radon emanation was observed within a given site which reflected the heterogeneity of the sampled area in relation to the sampling cartridge. Similar results were obtained for site T-9 which consisted of coarse bare tailings and where all the cartridges were placed within the sampling area of 1 m^2. While comparing results for various sites or

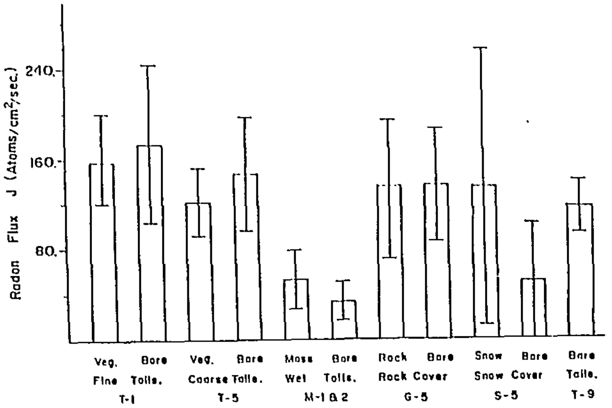

Figure 5. Radon emanation fluxes for various covers and bare tailings at different sites.

Table 1: Radon emanation rates for various covers on uranium tailings surfaces.

Test No.	Sampling Location and Cover Type	Sampling Time, h	Average Radon Flux, J, atoms/cm^2 sec ±1 S.D For Cover	For Bare Tailings	Pressure and Temperature Variation During Sampling ΔT, °C	Δp, Pa
1	Site T-1 Vegetated fine tailings	3.5	157±40	173±71	0	0
2	Site T-1 Vegetated fine tailings	17	305±71	266±71	±19	-800
3	Site T-5 Vegetated coarse tailings	16.6	121±28	136±51	±15	0
4	Site T-7 Vegetated coarse tailings	16.3	236±26	228±49	±12	+533
5	Site T-8 10 cm sewage cover & veg.	6.0	103±33	126±112 (Range 28-254)	-6	+67
6	Site M-1 Moss covered wet tailings	8.0	47±7	34±17	+2	0
7	Site M-2 Moss covered wet tailings	6.8	58±26	32±13	+7	0
8	Site G-5 Crushed rock cover, 0.6 m	17.3	120±122 (Range 46-330)	173±91 (Range 62-264)	-7	600
9	Site G-5 Crushed rock cover, 0.6 m	6.5	150±48	99±31	+1	-133
10	Site S-5 Snow cover, 0.5 m	21.0	134±123 (Range 12-262)	50±52 (Range 17-143)	±10	-1333
11	Site T-9 Bare dry tailings	6.0	-	117±23	1	-133

differences between covered and bare tailings at a given site, the results were, therefore, considered statistically indistinguishable if the difference was less than ±20%.

It is seen from these results that under the present field cover conditions, statistically there was no significant difference in radon emanation rates between a given cover and bare tailings for all sites. The flux rate varied greatly from a low of 32 atoms/cm^2 s for moss covered high water table locations (site M-2) to a high of 300 atoms/cm^2 s for fine grained tailings (site T-1). The values depended on the site and meteorological conditions.

For coarse grained and dry tailings (site T-9), the average radon flux was measured as 117 $\pm$ 23 atoms/cm^2 s under constant conditions of temperature and pressure. Similar results were obtained for vegetated and bare tailings at the coarse grained tailings site T-8, amended with approximately 10 cm of sewage sludge, where the measured fluxes were, respectively, 103 $\pm$ 33 and 126 $\pm$ 112 atoms/cm^2 s.

At the vegetated plot, site T-1, (fine grained tailings covered mostly with Birdsfoot Trefoil), the measured fluxes were, respectively, 157 $\pm$ 40 and 173 $\pm$ 70 atoms/cm^2 s for vegetated and bare tailings, for a short sampling interval of 3.5 h during which the temperature and pressure variations were small. At the same site, however, for a longer sampling period of 17 h, where the temperature and pressure variations were rather large $\Delta T = 19^{\circ}C$ and $\Delta P = -800$ Pa, the measured flux rates were 305 $\pm$ 71 and 266 $\pm$ 71 atoms/cm^2 s, respectively, for vegetated and bare tailings. At the coarse grained tailings site, T-5, with mostly grass cover, the fluxes were, respectively 121 $\pm$ 28 and 131 $\pm$ 51 atoms/ cm^2 s for vegetated and bare tailings with temperature and pressure variations of $\Delta T = \pm 15^{\circ}C$ and $\Delta P = 0$ Pa. At another vegetated coarse grained tailings site, T-7, the values were, respectively, 236 $\pm$ 26 and 228 $\pm$ 49 atoms/cm^2 s for vegetated and bare tailings with $\Delta T = \pm 12^{\circ}C$ and $\Delta P = + 533$ Pa. At this site higher moisture conditions were encountered due to precipitation events before sampling.

Previous studies of Silker[1], Rogers[13] and Hinton[4] have shown that because of the increased porosity and channelling associated with root penetration, radon fluxes for vegetated tailings were significantly higher compared to bare tailings. In Hinton's study[4] the increase was by an order of magnitude. No such effect was observed in our study except increases in radon fluxes during falling barometric conditions and after precipitation events. Both these phenomena are related to increased exhalation of radon gas from zones of higher concentration deeper into the tailings pore space to the surface as the system equilibriates pressure and or water table changes. As the tailings site was vegetated directly without any overburden, clay or top soil capping, no increased effects of root penetration were observed. Similar results were obtained by Scott[19] who measured the same site on a large scale by an aerodynamic method where small changes in radon concentration of air at a given height were measured as the air moved along the study site.

The highest attenuation of radon flux was observed for sites that were characterized by high moisture saturation and where the water table or the capillary fringe were close to the surface. These sites supported acidophillic, semi-aquatic moss (site M-1 and M-2) where the mean radon fluxes for covered and bare tailings were, respectively, 52 $\pm$ 26 and 33 $\pm$ 17 atoms/cm^2 sec. At these locations some crusting of the surface was observed because of upward movement of salts and mineral recrystallization which when broken altered the surface conditions and resulted in wide variation in fluxes within a site.

Figure 6 shows the moisture dependence of radon emanation rates from tailings. The emanation rates increased by a factor of 2-3 as the moisture saturation was increased from a few percent to between 20-70%. For higher moisture contents, it decreased with increased saturation, and at 100% saturation the reduction was by a factor of 7-10. These results are similar to those of Strong[17], Burton[18] and Hinton[4] where the emanation rates increased with modest increase in moisture contents, and decreased at very high moisture values. At lower moisture conditions, increased amount of radon atoms are slowed down in the tailings pore space as the pore moisture content is increased. This leads to high pore radon concentrations resulting in increased emanation rates. At much higher moisture conditions, close to saturation values, radon emanation rates are decreased due to decreased diffusion rate for water compared to that of air.

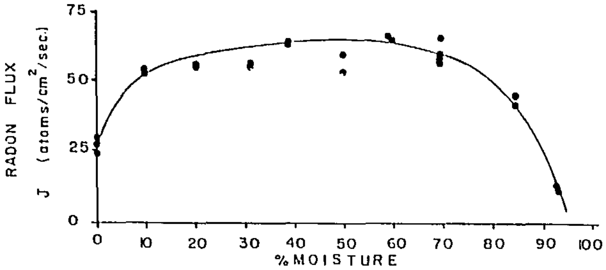

Figure 6. Moisture dependence of radon emanation flux from tailings.

Because of the thinness of the crushed rock and gravel cover (0.6 m) and its coarse nature, no attenuation in radon exhalation rates were observed. Similar results were obtained for winter snow cover (0.5 m thick). As a result of the freezing of the tailings surface (frost penetration depth approximately 0.5 m) a wide variation in flux values, varying between 12 and 262 atoms/cm^2 s with an average of 134 $\pm$ 123 atoms/cm^2 s for snow covered, and 17-143 atoms/ cm^2 s with an average of 52 $\pm$ 52 atoms/cm^2 s for bare frozen tailings, was observed. Channelling is the most probable cause for the observed variation in flux rates.

Conclusions

The results from the present study can be summarized as:

1. For direct vegetated uranium tailings, no effect of the vegetation cover was observed on radon emanation rates compared to that of bare tailings.

2. The highest attenuation in radon fluxes were

obtained for surfaces where the water saturation was high. At lower moisture conditions, radon flux increased with moisture contents.

3. There was no attenuation in radon fluxes for 0.6 m crushed rock and gravel cover or 0.5 m snow cover on tailings, though large variations within a site were observed because of channelling.

4. Radon fluxes increased during falling barometric conditions and after precipitation events by transport related mechanisms.

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