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Surface water hydrology considerations in predicting radon releases from water-covered areas of uranium tailings ponds

KIRK K.NIELSON & VERN C.ROGERS
Rogers & Associates Engineering Corp., Salt Lake City, Utah, USA

1 INTRODUCTION

In assessing the releases of radon (Rn-222) from uranium mill sites, the radon escaping from water-covered surfaces of the tailings pond has traditionally been ignored (NRC 1980a, NRC 1980b, NRC 1981). This has been justified by radon diffusion calculations, which suggest that radon cannot penetrate more than a few centimeters of water because of its very low diffusion coefficient (10^{-5} cm^2s^{-1}). The tailings pond is not a motionless body of water, however, and considerable water movement occurs over time periods comparable with the half-life of radon (3.8 days). Therefore, significant advective transport of radon may occur, rendering the pond less effective than previously thought for containing radon gas.

In a recent study for EPA on radon releases from active uranium mills, we examined the potential for advective transport of radon through tailings pond waters along with other radon sources in the mill environment (Rogers et al., 1985). This paper summarizes the parts of the study that dealt with radon releases from the tailings pond area, and discusses the nature and mechanisms of the radon releases from water-covered areas. A reference tailings impoundment is described according to several distinct physical regions, and the conditions affecting radon transport in each are described. Since radon transport through ponded water has not previously been modeled in detail, simple laboratory experiments were conducted to approximate the characteristic transport parameters. The results of these experiments were then used with parameters describing the tailings pond to assess the overall magnitude of radon release expected from the water-covered pond region. The significance of radon releases from the water-covered areas was estimated by comparison to radon fluxes from other, exposed tailings surfaces.

2 REFERENCE TAILINGS IMPOUNDMENT

A reference tailings impoundment that approximates actual impoundments is first defined to illustrate three characteristic regions with distinctly different physical properties that affect radon transport. The reference impoundment also provides a basis to estimate the magnitude of radon releases from tailings ponds. The impoundment contains a central, water-covered pond area, surrounded by a water-saturated beach area, and an unsaturated beach area. Tailings enter the impoundment via a slurry pipeline from the mill, and are depleted in emanated radon for the first few days due to complete radon releases during milling. The total mass of new depleted tailings entering the impoundment is insignificant compared to the total mass in the impoundment, however, so the total radon release rate is relatively constant. Since the slurry pipeline delivers both coarse (sandy) and fine (slime) tailings, the sands tend to accumulate near the pipeline, while the slimes are carried further into the center of the pond. The slurry pipe is typically moved to different positions around the edge of the impoundment, so that the sandy tailings typically comprise most of the saturated and unsaturated beach areas, and the slimes accumulate in the center pond area. The radon source materials and diffusion characteristics in the pond, saturated, and unsaturated areas are thus different, and are described in terms of nominal parameter values to permit estimates of their relative impacts on radon releases.

The unsaturated beach areas are considered to be comprised exclusively of tailings sands, and to be sufficiently above the water level that they are well-drained and similar to surrounding sandy soils in moisture content. Radon originating in these regions is defined in terms of the radium content for the sands, which is typically much lower than that for the slimes. Once radon gas is emanated into the interstitial pore space of the sands, it diffuses according to the characteristics already known and modeled for unsaturated soils and tailings (Rogers et al., 1984a), and is dominated by diffusive transport mechanisms. Advective transport by air or vapor currents in unsaturated regions such as the tailings beaches has been examined and is considered insignificant (Rogers et al., 1983). Accordingly, radon fluxes are computed for the unsaturated beaches as

$$J = 10^4 R \rho E \sqrt{\lambda D} \tag{1}$$

where

J = radon flux from the exposed tailings surface (pCi $m^{-2}s^{-1}$)
R = tailings radium content (pCi g^{-1})
ρ = bulk tailings density (g cm^{-3})
E = radon emanation coefficient for tailings (dimensionless)
λ = radon decay constant (2.1×10^{-6} s^{-1})
D = diffusion coefficient for radon in the tailings pore space (cm^2 s^{-1})

The saturated beach areas are considered to be comprised of approximately 70 percent sands and 30 percent slimes, reflecting the limited mixing of slimes in this part of the tailings mass. Although this area of the impoundment is variable and more difficult to define in terms of physical extent, its diffusion characteristics are more distinct in being saturated by water. Despite wave action over the saturated beach areas, advective transport in the interstitial volume is probably limited to only the top few centimeters, as defined by the wave-pond elevation difference. The radon source term for the saturated beaches is modeled as a weighted average of the respective radium contents of the sands and slimes (70/30 ratio) multiplied by their respective emanation coefficients. Transport of emanated radon to the atmosphere, neglecting liquid advection in the top wave-affected layer, is dominated by diffusion through the saturated

interstitial space, with a typical diffusion coefficient on the order of 10^{-5} $cm^{2}s^{-1}$ (Rogers et al., 1984a).

The tailings beneath the pond area are assumed to be comprised of approximately 50 percent sands and 50 percent slimes. In this region the radon source term is similarly computed as a weighted average of the respective radium contents of the sands and slimes (50/50 ratio) multiplied by their respective emanation coefficients. Movement of emanated radon to the atmosphere includes advective as well as diffusive transport, since considerable water movement occurs within the pond over time periods comparable to the half-life of radon. The movement is partly caused by surface wind currents, thermal gradients, mechanical disturbance from the mill discharge pipe, and biological disturbances (animals, birds, etc.). In addition, radon release from the radium dissolved in the water must now be considered separately, since the water is physically separated by significant distances from the solid tailings material. For analyzing the pond area, radon releases were divided into three components:

1. Radon originating from solid tailings under less than 1 m of water.
2. Radon originating from solid tailings under greater than 1 m of water.
3. Radon from the dissolved radium in the pond water.

The one meter depth is chosen to partition the surface water, where turbulant movement is pronounced and often visible, from deeper layers, where advection is minimal. Although actual advective currents probably decrease continuously with depth, this partitioning conveniently defines a "rapid release" zone for radon and a deeper decay-limited transport zone.

3 LABORATORY MEASUREMENTS AND RESULTS

In order to quantify radon releases from the above pond sources, several parameters were measured in the laboratory, using a sample of slime tailings from the Rifle, Colorado UMTRAP site. The measured parameters included the solubility of the tailings radium and the transport coefficient for radon through "undisturbed" columns of water in the laboratory. In order to interpret the radon transport experiments, radium contents, emanation coefficients, and related tailings parameters were also measured. The key tailings parameters are summarized in Table I.

TABLE I

Characteristics of Tailings used as Radon Sources

R = 4628 pCi/g Ra-226
E = 0.25 pCi Rn-222 released per pCi Ra-226
Porosity = 0.66 in test columns
Solubility = 35 pCi/liter Ra-226 in separated column water.

The slime tailings sample was oven dried, and 200-gram aliquots were weighed into each of four Boyoucos soil test cylinders. Seven hundred ml of water were added to each cylinder, after which they were stirred and allowed to settle and equilibrate for at least 22 days. The settled tailings occupied the bottom 8.9 cm of the 5.9 cm diameter glass cylinders, and the water layer comprised an average height of 19.4 cm above the tailings. Radon flux measurements were then made from the water surfaces after first circulating fresh air over the undisturbed water. After the radon flux measurements, the water was carefully siphoned from the columns without disturbing the tailings layers. Additional radon flux measurements were then made from the bare, saturated tailings. The radon flux measurements utilized both the accumulator can and charcoal canister techniques (Rogers et al., 1984b). The accumulator can measurements gave a ten-minute average flux, and the charcoal canister measurements gave a 24-hour average flux. The results were averaged and reported in terms of a mean and standard deviation.

The results of the laboratory measurements are presented in Table II. The relatively high radon fluxes penetrating 19.4 cm of water indicated clearly that molecular diffusion did not account for the observed radon transport through the columns. Despite precautions to avoid agitation and vibrations, advective transport (probably thermally induced) dominated the observed radon flux, which would have been nearly four orders of magnitude lower with only diffusive transport in undisturbed water. The removal of the water (not disturbing the tailings) allowed measurement of the bare radon flux from the saturated tailings, and gave evidence that the advective forces acting in the water cover were not active in the saturated tailings region. Instead, the low diffusion coefficients typical of water-saturated pore space were found to be typical.

TABLE II

RADON FLUXES MEASURED FROM BARE AND WATER COVERED TAILINGS SURFACES (mean ± S.D.)

Undisturbed water, Accumulator Can	75 ± 19
Undisturbed water, Charcoal Canister	68 ± 7
Bare, saturated tailings, Accumulator Can	84 ± 21

The results of the laboratory flux measurements were compared with values obtained using the RAECOM computer code (Rogers et al., 1984a). Using the radium content, emanation coefficient, and porosity from Table 1, the RAECOM code gave a computed radon flux of 83 pCi $m^{-2}s^{-1}$ using its default (correlation) value for the diffusion coefficient in the saturated tailings. This compares well with the mean measured flux value of 84 pCi $m^{-2}s^{-1}$ and also supports the selection of 4×10^{-5} cm^2s^{-1} for the radon diffusion coefficient in the submerged tailings. Further RAECOM analyses of the 17 percent attenuation provided by the undisturbed water indicated that the effective transport coefficient for the water layer was on the order of 0.003 cm^2s^{-1}. The dissolved radium content measured from the water layers and shown in Table I, gives negligible contribution to the measured fluxes, but gives a nominal solubility parameter for evaluating the reference tailings impoundment.

4 APPLICATIONS AND DISCUSSION

In applying the Laboratory data to estimate radon releases from submerged tailings, the high uncertainties and lack of lab/field experiment correspondence preclude quantitative accuracy from being associated with the conclusions. However, the lab data conclusively show that non-diffusive transport can dominate radon movement, even in visually "undisturbed" water columns. Since surface turbulence is invariably visible in tailings ponds, we infer that greater advective transport occurs in the pond surface layers. In the absence of turbulence data for either the deep or shallow pond water, we qualitatively associate the measured laboratory transport coeffecient with possible transport characteristics of the deep (>1m) impoundment water.

For the shallow (<1m) impoundment water, extrapolations of visual dye movement tests indicate advective velocities may exceed 1-2 mm/minute, resulting in virtually no radon containment by the surface water. If shallow water movement is sufficient to remove radon from the tailings-water interface and transport it to the atmosphere in a short time (several hours), the radon flux from the shallow tailings is nearly as great as that from similar bare saturated tailings, hence no significant radon attenuation is considered.

For tailings at depths greater than one meter, the radon transport properties of the pond water are considered to follow the Laboratory value of 0.003 cm^2s^{-1} up to the 1-meter depth, above which no further attenuation occurs. For dissolved radium, the same water motion that facilitates rapid radon release from the shallow water also allows release of all radon generated in the top meter of the pond. Thus, the applicable flux equation for radon from the top meter of water over the deep fraction of the pond and for the average half-meter of water over the shallow fraction is

$$J_d = 10^6 K_s R \lambda (1 - 0.5\, f_s) \tag{2}$$

where

K_s = ratio of radium in solution to radium in tailings solids (g cm^{-3}).
f_s = fraction of pond area with less than 1 meter depth.

Radon generated from dissolved radium below one meter is transported according to the 0.003 cm^2s^{-1} coefficient up to the one meter depth, where it is rapidly released to the atmosphere. The three radon sources, shallow tailings, deep tailings, and dissolved radium, are added to obtain a simplified estimate for the average flux from the tailings pond,

$$J = 10^4 R \rho E \sqrt{\lambda D}\, [f_s + (1-f_s)A] + 10^6 K_s R \lambda (1 - 0.5 f_s) \tag{3}$$

where

A = attenuation factor for deep water.

The attenuation factor, A_t, is determined from RAECOM calculations, or it can also be approximated by

$$A_t = \exp[-\sqrt{\lambda/D_{tr}}\,(x_p - 100)] \tag{4}$$

where

D_{tr} = effective stagnant water transport coefficient (cm^2s^{-1})
X_p = average pond depth for areas greater than 1 meter deep (cm)

In order to estimate radon releases from a tailings impoundment, numerous site-specific parameters must be defined. Some, such as ore grade, area of the impoundment surfaces, etc. are readily known or measurable, while others, such as diffusion coefficients, are usually unknown without specific measurements. Table III presents nominal values for some of the required parameters for the present estimates. Other values, such as emanation coefficients, moistures and diffusion coefficients for the unsaturated tailings, were based on site-specific data (Rogers et al., 1985).

TABLE III

Tailings Parameters used in Radon Transport Calculations

	Submerged Tailings	Saturated Tailings	Unsaturated Tailings
Sand/slime ratio	50/50	70/30	100/0
Bulk density (g cm^{-3})	1.55	1.57	1.60
Porosity	.40-.42	.39-.41	.38-.40
Moisture Saturation	1.0	1.0	.33-.57
Surface Area (m^2)	4.0E5	2.0E5	9.0E4

For calculating radon emissions from the unsaturated, sandy tailings at the outer edges of the impoundment, equation 1 was used to obtain the normalized radon fluxes in Table IV. The radon release is normalized to account for the typical 4:1 ratio of radium activity in the slimes compared to that in the sands, and also to account for their bulk density difference as defined in Table I. The resulting data in Table IV are thus normalized to the average radium in the original ore not just for the sands alone. It should also be emphasized that the use of specific fluxes presupposes a fixed diffusion coefficient in the source material, and thus does not have general application to areas in which moistures or diffusion coefficients are greatly different.

TABLE IV
Specific Radon Fluxes Computed for Six State Milling Regions for Three Parts of a Uranium Tailings Impoundment
($pCi\ m^{-2}s^{-1}/pCi\ g^{-1}$)

	State						
Tailings	CO	NM	TX	UT	WA	WY	Mean
Unsaturated	0.42	0.76	0.23	0.29	0.29	0.43	0.40
Saturated Beach	0.036	0.062	0.031	0.027	0.031	0.035	0.037
Pond	0.020	0.033	0.019	0.017	0.019	0.021	0.022

For the mixed tailings in the saturated beach areas, Table IV gives the corresponding specific fluxes assuming a 70/30 mass ratio of sands/slimes, and assumes a combined mean density of 1.57 gcm^{-3}. The resulting average specific fluxes are again normalized to the average radium content of the original ore.

For the ponded areas of the tailings, is was assumed that one-fourth of the pond area was less than one meter deep, and that the tailings are 50/50 sands/slimes. The value of K_s (NRC 1980) is 8.92E-4 g/ml. The diffusion coefficient for tailings measured in the laboratory was similar to the predicted value from earlier correlations, and so the correlation value of 4.0E-5 cm^2s^{-1} was used. A lower-bound estimate of the diffusion coefficient for deep water was obtained from the laboratory measurement, D_{tr} = 0.003 cm^2s^{-1}. This value was used in RAECOM calculations (Rogers et al., 1984a) to obtain an average attention factor of A_t = 0.17, which was used in the analysis. The resulting normalized radon fluxes from the water-covered tailings using equation (3), and dividing by R, are shown by state in Table IV.

In order to assess the relative importance of total radon releases for the three tailings regions, a reference uranium mill is defined to process one with an average grade of 0.1 percent U_3O_8. Its tailings impoundment is also defined to have the surface areas shown in Table III. The resulting total radon releases, expressed in Ci/day for each tailings region in the six states are summarized in Table V. The total radon releases vary from 0.9 to 2.3 Ci/day, and are dominated (69%) by the unsaturated sandy tailings, as might be expected. Although the submerged tailings account for only 17% of the total, they are much more important than previously estimated. Although to be regarded qualitatively, this study suggests that rado mitigation by submerging tailings in the pond water may be much less effective than has been previously assumed. From the specific fluxes in Table IV, it is seen that saturating or submerging the tailings is still effective in significantly reducing radon fluxes by an order of magnitude, but that the advantage of additional water over the saturated tailings is proportionately reduced.

TABLE V

Summary of Total Radon Emissions from the Reference Tailings Impoundment in Six States
(Ci/day)

	State						
Tailings	CO	NM	TX	UT	WA	WY	Mean
Unsaturated	0.92	1.66	0.50	0.63	0.63	0.94	0.88
Saturated Beach	0.18	0.30	0.15	0.13	0.15	0.17	0.18
Pond	0.19	0.32	0.19	0.17	0.19	0.20	0.21
Total	1.29	2.28	0.84	0.93	0.97	1.31	1.27

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