Risk Assessment Revision for
40 CFR Part 61 Subpart W –
Radon Emissions from Operating Mill Tailings

Task 5 – Radon Emission from Evaporation Ponds

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EXECUTIVE SUMMARY

The National Emission Standards for Hazardous Air Pollutants (NESHAPs) includes radon emissions for uranium mill tailings (40 CFR Part 61 Subpart W – National Emission Standards for Radon Emissions from Operating Mill Tailings – December 15, 1989). At the time of promulgation, the overwhelming number of uranium processing facilities were conventional acid or alkaline leach mills. Radon emissions from these facilities were primarily from the dried out portions of large (greater than 100-acre) tailings ponds. With the promulgation of Subpart W, this large area source was reduced by the requirements to limit the size of new tailings areas to either 40 acres for phased disposal or 10 acres for continuous disposal (40 CFR 61 Subpart W). New facilities had to abide by these new tailings pond area limits. Additionally, and more importantly, economic and other considerations have lead commercial uranium recovery companies to submit license applications/amendments to develop, upgrade or restart a significant number of in-situ leach (ISL) facilities over conventional facilities.

Unlike conventional processes, ISL facilities do not produce tailings or other solid waste products. They do, however, generate significant amounts of liquid wastes during uranium extraction and aquifer restoration. During extraction, extraction solution (lixiviant), composed of groundwater enhanced by an oxidant and carbonate/bicarbonate, is injected through wells into the ore zone. This lixiviant moves through pores in the ore body and mobilizes the uranium. The resulting “pregnant” lixiviant is withdrawn by production wells and pumped to the processing plant, which recovers the uranium. To prevent leakage of the lixiviant outside the production zone, it is necessary to maintain a hydraulic cone of depression around the well field. This is accomplished by bleeding off a portion of the process flow. Other liquid waste streams are from sand filter backwash, resin transfer wash, and plant wash down. One method to dispose of these liquid wastes is to evaporate them from ponds.

The water in these evaporation ponds can contain significant amounts of radon and radium, which will generate radon gas. This radon gas could escape from the pond, and result in exposures to individuals living nearby. The goal of this task is to develop a model that can be used to conservatively estimate the amount of radon that could be released from evaporation ponds to determine whether the ponds are a significant source of radon exposure to nearby individuals.

A review of the various models used for estimating radon flux from the surface of water bodies indicates that the stagnant film model (also known as the two bottleneck model) coupled with a wind correction equation, can be used to estimate the radon flux based on the concentration of radium in the pond’s water and the assumption that radon is in secular equilibrium with the radium. Using this model, the radon flux from the surface of an evaporation pond, as a function of the wind speed (for winds less than 24 mph), can be estimated with the following equation.

$$J = \frac{1.48\times10^{-4}}{e^{-0.351V}} C_w \qquad \text{(ES-1)}$$

Where J = Radon flux (pCi/m^2-sec)
C_w = Concentration of radium in the water (pCi/L)

V = Wind speed (m/sec)

Implicit in this model is the fact that in pond water, the radon diffusion coefficient is 10^{-5} cm^2/sec and that the thickness of the stagnant film layer can be estimated by an exponential relationship with wind speed.

Measurements conducted on the Homestake evaporation ponds agree with the stagnant film model estimates. However, the Homestake measurement method did not allow the measurement of wind-generated radon fluxes, as the collar used to float the canister makes the wind speed zero above the area being measured. No data was found for measurements of the radon flux on evaporation ponds versus wind speed.

The model should not be used for wind speeds above 10 m/sec (24 mph). However, this is not expected to be a major limitation for estimating normal operational evaporation pond radon releases and impacts.

Using actual radium pond concentrations and wind speed data, Equation ES-1 was used to calculate the radon pond flux from several existing ISL sites. It was determined that the radon flux ranged from 0.07 to 13.8 pCi/m^2-sec. From this, it can be seen that the radon flux above some evaporation ponds can be significant (e.g., may exceed 20 pCi/m^2-sec). If this were to occur, there are methods for reducing the radium concentration in the ponds; the most straightforward being dilution. This, however, is temporary, as eventually evaporation will increase the concentration. A second method is to use barium chloride ($BaCl_2$) to co-precipitate the radium to the bottom of the pond. The radon generated at the depths of the bottom sediments will decay prior to reaching the pond surface.

Again using actual ISL site data, the total annual radon release from the evaporation ponds was calculated and compared to the reported total radon release from three sites. The evaporation pond contribution to the site's total radon release was small (i.e., <1%).

Two additional sources of radon release were investigated: 1) the discharge pipe, and 2) evaporation sprays. The discharge pipe is used to discharge bleed lixiviant to the evaporation pond. Radon releases occur when the bleed lixiviant exits the pipe and enters the pond. It was found that these radon releases are normally calculated using the NUREG-1569, Appendix D methodology, thus, this source is currently included in the total radon releases reported for an ISL site. Nonetheless, a simple scoping calculation showed that the discharge pipe radon release is larger (in some cases, much larger) than the radon releases once the waste water is in the evaporation pond.

Spray systems are sometimes used to enhance evaporation from the ponds. A model to calculate radon releases during spray operation was developed. Also, data from ISL ponds were used to estimate this source of radon release. The radon releases from spray operation were calculated to range from <0.01 to < 3 pCi/m^2-sec. Furthermore, operation of the sprays would reduce the radon concentration within the pond, hence, the normal radon release would be depressed once the sprays are turned off (until the radon has had an opportunity to re-equilibrate with the radium).

1.0 INTRODUCTION AND BACKGROUND

The National Emission Standards for Hazardous Air Pollutants (NESHAPs) includes radon emissions for uranium mill tailings (40 CFR Part 61 Subpart W – National Emission Standards for Radon Emissions from Operating Mill Tailings – December 15, 1989). At the time of promulgation, the overwhelming number of uranium processing facilities were conventional acid or alkaline leach mills. Radon emissions from these facilities were primarily from the dried out portions of large (greater than 100-acre) tailings ponds. With the promulgation of Subpart W, this large area source was reduced by the requirements to limit the size of new tailings areas to either 40 acres for phased disposal or 10 acres for continuous disposal (40 CFR 61 Subpart W). New facilities had to abide by these new tailings pond area limits. Additionally, and more importantly, economic and other considerations have lead commercial uranium recovery companies to submit license applications/amendments to develop, upgrade or restart a significant number of in-situ leach (ISL) facilities (NRC 2009) over conventional facilities.

Unlike conventional processes, ISL facilities do not produce tailings or other solid waste products. They do, however, generate significant amounts of liquid wastes during uranium extraction and aquifer restoration. During extraction, extraction solution (lixiviant), composed of groundwater enhanced by an oxidant and carbonate/bicarbonate, is injected through wells into the ore zone. This lixiviant moves through pores in the ore body and mobilizes uranium (and other elements). The resulting “pregnant” lixiviant is withdrawn by production wells and pumped to the processing plant, which recovers the uranium. To prevent leakage of the lixiviant outside the production zone, it is necessary to maintain a hydraulic cone of depression around the well field (NRC 2003, Appendix D). This is done by bleeding off a portion of the process flow, typically one to three percent of the process flow or a few tens of gallons per minute (gpm) to 100 gpm or more (NRC 2001, NRC 2003). Other liquid waste streams are from sand filter backwash, resin transfer wash, and plant wash down. All of these liquid wastes may be disposed in an evaporation pond via permitted deep injection or by treatment to unrestricted release levels and disposed of by irrigation. This report focuses on disposal in an evaporation pond.

The water in these evaporation ponds can contain significant amounts of radon and radium, which will generate radon gas. In addition, some ponds may contain sediment which can also generate radon. This radon gas could escape from the pond and result in exposures to individuals living nearby. The goal of this task is to develop a model that can be used to conservatively estimate the amount of radon that could be released from the evaporation ponds, to determine whether the ponds are a significant source of radon exposure to nearby individuals.

To develop the evaporation pond radon release scenario, the following tasks were conducted:

- A review as to the important parameters governing radon release from water, including the general behavior of radon in water (Section 2.1);
- A review of measurements or estimates from models of radon and radon flux from the surface of various water bodies, including, but not limited to, tailings piles/ponds, lakes and oceans, and evaporation or settling ponds (Section 2.2);

- A review of the various models used for estimating radon flux from the surface of water bodies (Section 2.2);
- Selection of the model (Section 2.2.3) and parameters (Section 3.1) suited to evaporation ponds including modeling turbulent influence from the wind (Section 2.2.4);
- Review of current and past evaporation ponds/tailings, including using the joint frequency wind distribution to bound the problem (Section 3.2);
- Estimates of radon flux from selected evaporation pond configurations, including estimates from other sources such as discharge pipes (Section 3.3) and enhanced evaporation systems (Section 3.4).

Two measures were used to evaluate whether evaporation ponds are a significant source of radon at ISL sites:

- The estimated pond radon flux was compared to 20 pCi/m^2-sec, which is the Uranium Mill Tailings Radiation Control Act (UMTRCA, 40 CFR 192.32) standard for radon release when averaged over the entire tailing pile or impoundment. It is used in this report not as a radon release standard, per se, but as a guide to indicate whether the evaporation pond radon flux that has been calculated is significant.

- Using actual ISL site pond dimensions, pond radium concentration, and wind speed distribution, the model developed in this report was used to calculate the annual evaporation pond radon release. The calculated annual pond radon release was then compared to the total annual radon release for each site, as reported in site specific documents.

2.0 POND RADON EMISSION MODEL

For this task, SC&A has developed a model to bound radon emission from evaporation, holding, or settling ponds. In the discussion that follows, the term “evaporation ponds” includes holding and settling ponds, as well as evaporation ponds.

2.1 Sources of Radon in Pond Water

The following presents a basic review of some of the sources and properties of radon in water that influence the radon in evaporation ponds. Sources of radon in the water of an evaporation pond can include:

- Radon dissolved in the water (lixiviant) which passed through the ore body;
- Radon generated from radium containing sediment deposits on the bottom of the evaporation or holding pond;
- Radon from the decay of dissolved or suspended radium in the water; and
- Radon diffusing back into the water from the atmosphere.

The last of these four sources is considered to be negligible, while the first three sources are discussed further in the following sections.

2.1.1 Radon Dissolved from the Ore Body

The first source (radon in the water from an ISL facility) comes from the decay of radium in a solid matrix, likely the ore body, in addition to that dissolved in the water. The recoil range of a radon atom after radium disintegration is 30 to 50 nm in solids, 95 nm in water, and 64,000 nm in air (Lawrence 2005). The diffusion coefficient of radon in a solid matrix is 10-20 cm^2/sec, therefore, only those radon atoms produced in a layer 30 to 50 nm thick on the surface of solids will potentially enter the air or water pore spaces of sediments or soils. This limits the radon from the ore body. In the ore body, the ratio of the amount of radon in the pore space to the total amount of radon produced in a solid is the emanation coefficient. The emanation coefficient ("E") varies depending on the material (ranging from 1% to 80% and averaging 20% depending on the soil type), pore space, and water content (NRC 2001).

This source is also time dependent given the radon half life of 3.8 days. Should the water discharged to the evaporation pond be held up or delayed for any length of time, a fraction of the initial radon concentration would decay. However, in an ISL production well field, the movement of process water allows the radon to circulate and discharge before any significant decay reduction can occur.

Measurement of radon in groundwater at some ISL facilities ranges from 1.3×10^5 to 1.93×10^7 pCi/L (NRC 2001). The NRC assumed a concentration of 8×10^5 pCi/L in pregnant lixiviant for their risk analysis which approximates "the highest value reported inside a uranium recovery facility" (NRC 2001).

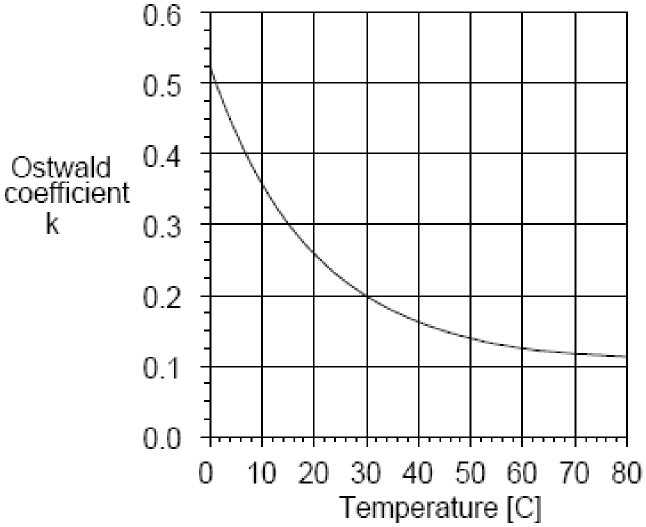
Investigations of radon in drinking water showed that over small distances there is often no consistent relationship between measured radon levels in groundwater and radium levels in the groundwater or in the parent bedrock (FR 64(38):9569). Radon's volatility is rather high compared to its solubility. Radon rapidly volatizes from surface water, therefore measured radon in surface water is often much lower than in groundwater. This is also the basis for the use of radon as an atmospheric tracer.

2.1.2 Radon Generated from Sediment Deposits

Radon generated from radium containing sediment deposits on the bottom of evaporation or holding ponds is also dependent on the emanation coefficient and the diffusion coefficient. A portion of the radon generated in the sediment, dependent on the emanation coefficient, will diffuse through the sediment layer. The thickness of the sediment layer, along with its diffusion coefficient, governs how much radon will be released to the overlying water layer. The radon diffusion coefficient in a fully saturated material is estimated at about 10^{-5} cm^2/sec. (ANL 1993)

2.1.3 Radon from Radium in the Water

Radon from the decay of dissolved or suspended radium in the water is also released to the atmosphere. While soluble in water, radon's solubility decreases with increasing temperature (Surbeck 1996). The Ostwald coefficient "k", defined as the ratio of the radon concentration in water to the radon concentration in air, decreases by over 250 percent from 0 °C to 40 °C, which is the range of temperatures expected in a uranium processing facility (see **Figure 1**).



Source: Surbeck 1996

Figure 1: Radon Water Solubility Versus Temperature (Ostwald Coefficient)

At constant temperature however, Henry's Law influences the release of radon from the water surface. That is, at constant temperature, the amount of radon released is proportional to the partial pressure of radon in equilibrium with the water. This is the basis for the stagnant film model discussed later.

2.2 Modeling Radon Emission from Water Bodies

2.2.1 Tailings and Tailings-Type Pond Models

A review of radon and radon flux from the surface of various water bodies, including, but not limited to, tailings piles/ponds, lakes and oceans, and evaporation or settling ponds was conducted to determine the probable range of radon and the potential for bounding the radon from an evaporation pond. These systems, which are similar to a lined evaporation pond, provide an overview of radon releases and should encompass the radon release from an evaporation pond.

Sears, Blanco, Dahiman, Hill, Ryan, and Witherspoon

One of the earliest published results was provided by Sears, et al. (ORNL 1975) from Oak Ridge National Laboratories as part of a series of reports documenting the cost of waste from the nuclear fuel cycle. Her group compiled and analyzed data from 15 operating mills during 1973 and 1974 and, using the data, developed two model mills: an acid leach mill and an alkaline leach mill. Each mill was assumed to have a capacity of 2000 tons per day ore processing and was assumed to run for 20 years, 365 days per year. The mills were located at two hypothetical sites, one in Wyoming and one in New Mexico. The Sears report presents a series of source terms for radon emanation from active tailings areas for the model mills at the end of their 20-year life. Radon releases from both ponds and dry active areas were considered. The pond source term included contributions from the tailings under the water cover, as well as radium dissolved in the pond water. They assumed that all of the radon diffusing from the tailings into the pond water plus all the radon from the decay of radium dissolved in the pond water was released to the atmosphere (i.e., the stirred-pond model). As Sears states, a "quiet-pond model, where some of the radon decays as it diffuses through the pond, would yield somewhat lower radon releases." (ORNL 1975, page 61) A portion of the ORNL 1975 results are presented in **Table 1**. As **Table 1** indicates, the pond radon release is about 0.0046 pCi/m^2-sec per pCi/L of radium dissolved in the pond's water. Since evaporation ponds are not expected to have large inventories of radium containing sludge on their bottoms, the contribution to the radon release from submerged tailings shown in **Table 1** is not expected to be applicable to evaporation ponds.

Table 1: Radon Release from Tailings Ponds

Case No.	Pond Type	Radium Pond Conc. (pCi/ml)	Radon Release (pCi/m^2-sec)[1]		
			Radium Decay	Submerged Tailings	Total
New Mexico Acid (Solvent) Extraction Mill					
1	Pond	1.7	7.8	9.0	16.8
2	Pond	1.7	7.8	9.0	16.8
3	Pond	2.3	10.5	9.0	19.6
4	Precip. Line pond	0.25	1.2	9.0	10.2
5	Precip. Line pond	0.25	1.2	9.0	10.2
Wyoming Acid (Solvent) Extraction Mill					
1	Pond	1.7	7.8	9.0	16.8
2	Pond	1.7	7.8	9.0	16.8
3	Pond	2.3	10.5	9.0	19.6
4	Precip. Line pond	0.25	1.2	9.0	10.2
5	Precip. Line pond	0.25	1.2	9.0	10.2
New Mexico Alkaline Leach Mill					
1	Pond	0.26	1.2	9.0	10.2
2	Pond	0.26	1.2	9.0	10.2
3	Pond	0.33	1.5	9.0	10.5
4	Precip. Line pond	0.088	0.4	9.0	9.4
5	Precip. Line pond	0.088	0.4	9.0	9.4

Table 1: Radon Release from Tailings Ponds

Case No.	Pond Type	Radium Pond Conc. (pCi/ml)	Radon Release (pCi/m^2-sec)[1]		
			Radium Decay	Submerged Tailings	Total
Wyoming Acid (Solvent) Extraction Mill					
1	Pond	0.26	1.2	9.0	10.2
2	Pond	0.26	1.2	9.0	10.2
3	Pond	0.33	1.5	9.0	10.5
4	Precip. Line pond	0.088	0.4	9.0	9.4
5	Precip. Line pond	0.088	0.4	9.0	9.4

Tailing in cases 4, 5 and 6 were chemically treated such as lime neutralization (acid leach) or copperas (alkaline leach) reducing the Ra-226 concentrations.

Source: ORNL 1975, Table 4.20

1. Units converted from pCi/day-cm^2 to pCi/m^2-sec for ease in comparing with other data.

Nielson and Rogers

A similar study was published in 1986 by K. K. Nielson and V.C. Rogers entitled “Surface water hydrology considerations in predicting radon releases from water-covered areas of uranium tailings ponds” (Nielson 1986). In this study, the authors conducted a series of experiments using dried slime tailings from a mill (Rifle, Colorado UMTRAP site) placed into glass cylinders and covered with various depths of water. The radium-226 content of the slimes was about 4600 pCi/g. The dissolved component was 35 pCi/L Ra-226. The radon flux was measured from the water surface after circulating fresh air over the undisturbed water. Then the water was carefully siphoned off and additional flux measurements were taken over the saturated tailings. The results showed that diffusion did not account for all the radon transported from the water surface, and advective transport, likely thermally induced, dominated the observed radon flux. The authors state that the radon flux was four orders of magnitude greater than a radon flux governed by diffusion alone through the water. The measurement of flux with the water removed (saturated tailings) indicated no advective forces in the saturated material, only diffusion. In comparing their measured fluxes to radon fluxes calculated with a computer model, Nielson and Rogers found similar results.

The authors further analyzed a hypothetical tailings pile and separated the less than one meter of water-covered pond area from regions with greater than one meter of water cover. For the shallow-depth regions (less than one meter cover), visual dye tests indicate advective velocities result in almost no containment by the surface water. Thus, the radon flux from shallow water-covered tailings is nearly as great as that from bare saturated tailings.

For areas with water cover greater than one meter, they define an effective transport coefficient, which attenuates the radon generated in the covered tailings and from radon generated by radium in the water. These are “attenuated” or effectively transported up to the one-meter depth, where, as noted previously, there is almost no reduction in radon due to local advection.

In summary, the authors propose three contributors to the radon on the water surface: shallow, unsaturated tailings; deep, water saturated tailings and slimes, and radium dissolved in pond water. The applicable equation from Nielson and Rogers 1986 has been reproduced in Appendix A.

To use the equation, the authors selected nominal values for many of the required parameters, including bulk density, porosity, moisture saturations, surface area, and so on. Other parameters, such as emanation coefficient and diffusion coefficients for the unsaturated tailings, were based on site-specific data. The calculations presented are for the three contributors of tailings pond radon previously identified. **Table 2** presents their results for various states with uranium milling regions and were normalized to the radium content of the original ore.

Table 2: Specific Radon Fluxes from Six State Milling Regions for Three Components of a Uranium Tailings Impoundment
(pCi/m^2-sec) / (pCi/g)

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

Source: Nielson and Rogers 1986, Table IV

The table was used to further calculate the total daily radon emissions from each of these hypothetical ponds. Importantly, while the mean pond emissions are less than 5% of the total, they are not insignificant. Also, assuming an average ore grade of 0.1% U_3O_8 — or about 290 pCi/g Ra-226 — leads to a mean pond radon flux of 6.4 pCi/m^2-sec, which is comparable to the prior estimate by Sears, et al. (ORNL 1975) in **Table 1**.

World Information Service on Energy and Mudd

The Nielson and Rogers equation (see Appendix A) were put on the World Information Service on Energy (WISE) Uranium Project Internet site in 2002 (WISE 2002). Also, G.M. Mudd used the Nielson and Rogers model to estimate the radon flux from the Australian Ranger mill tailings pile (IAEA 2003). He estimated the radon flux as 20.25 pCi/m^2-sec from the above-ground dam (water depth 1.3 m), and 2.2 pCi/m^2-sec from the tailings repository in Pit # 1 (water depth greater than 10 m). These values are again comparable to the prior work.

Usman and Spitz

The role of advection in the transport of radon to the surface of ponds was also pointed out in work conducted by S. Usman and H. Spitz (IRPA 2004) who were investigating the long-term storage of radium-bearing sand at the Department of Energy's Fernald Ohio Site (also known as the K-65 tank waste) using a water barrier. The K-65 waste had extremely high concentrations of radium-226, with an average concentration of 391,000 pCi/g in silo 1, and 195,000 pCi/g in silo 2, which are orders of magnitude greater than a typical tailings pond. They conducted a series of laboratory experiments (with lesser concentrations of radium in the source material) and determined that advection played a role in the transport of radon through a water column. As stated in their paper, conventional values for molecular diffusion of radon in water range from 1.14×10^{-5} to 1.56×10^{-5} cm^2-sec^{-1}. Values observed in their study were 30 to 50 times greater: 5.14×10^{-4} to 8.14×10^{-4} cm^2-sec^{-1}. They attribute this to "radio-turbulence" from the energy released in the decay of radium to radon. Of importance to this work, however, is again the evidence that advection plays a role in radon from the surface of a "tailings type" pond.

2.2.2 Models for Other Water Bodies

The prior section focused on water bodies associated with uranium or uranium extraction, e.g., tailings piles and pits. As such, they have material on the bottom that contains radium which contributes to the surface flux. In larger bodies of water (oceans, lakes, etc.), the depth negates any such contribution. This is equivalent to an evaporation pond with no sediment on the bottom and is the focus of this section.

During the same period as the above tailings pond studies, a series of investigations into radon transport in water bodies was being conducted by oceanographers using radon as a tracer. The primary interest of these investigators was the exchange of gases over water bodies, particularly the exchange of carbon dioxide between the atmosphere and the ocean or lakes. B. Bolin was one of the first to investigate this gas transfer using the C^{14}/C^{12} ratio in the atmosphere and ocean (Bolin 1960). Bolin's model assumed a hypothetical boundary of water at the sea surface through which gas exchange is only by molecular diffusion. Building on Bolin's work, in 1971, W.S. Broecker and T.H. Peng proposed using radon to determine gas exchange rates on the open ocean (Broecker and Peng 1975). In this model, the atmosphere and water are viewed as two turbulent bodies separated by a thin unstirred, or stagnant, layer through which gases pass by molecular diffusion. As the water becomes more turbulent, the stagnant layer becomes thinner, and thus the gas exchange increases. Other researchers also measured radon from the ocean surface; for example, C. Duenas measured the radon flux directly at a location off the coast of Malaga Bay on the Spanish Mediterranean Coast (Duenas C. 1983). He also determined quantitative relations between the radon flux and the radon concentrations in the surface water with temperature and wind speed. The effect of wind speed is discussed in Section 2.2.4.

The basic model used by the above investigators is known as the classical stagnant film model, whose principal parameter is the transfer coefficient across the air-sea interface (Schwarzenbach, R.P., et al. 2003). The stagnant film model assumes the rate of transfer of gas between water and air is controlled by the thickness of a thin layer of water through which gas is transferred by molecular diffusion, that is, through a hypothetical film of stagnant water. The air above this film is assumed to be very well mixed. Similarly, the water below the film is assumed to be well mixed. The rate of gas exchange is dependent on the thickness of the film, which is in turn dependent on the degree of agitation of the surface. It follows that the greater the agitation due to wind on the surface, the thinner the thickness of the stagnant film layer. This concept is discussed further in Section 2.2.4.

At the same time as the stagnant film model was being applied to the transfer of gases between the ocean and the atmosphere, it was also being applied to smaller bodies of water, such as lakes. Emersin (1975) studied the radon exchange rate in three Canadian lakes that ranged in size from 3.5 to 5.5 hectares (35,000 to 55,000 m^2), with depths ranging from 7 to 10 m. Emerson shows that with the appropriate film thickness, the stagnant flow model radon exchange rates “conform reasonably well with the predictions made by wind tunnel experiments.” Emerson’s results are discussed further in Section 2.2.4. Broecker and Peng (1975) also applied the stagnant film model in their study of CO_2 exchange rates at three lakes in the western United States, ranging in size from 1.8×10^8 to 5.2×10^8 m^2. Finally, Chambers (2009) presents a pond radon flux model consisting of two release mechanisms: diffusion and turbulence (wave action). For the diffusion portion of his model, Chambers uses the stagnant film model, with a fixed film thickness. These studies demonstrate that the stagnant film model can be applied to water bodies other than the oceans.

2.2.3 Evaporation Pond Model Development

The concepts developed above were recently applied to current evaporation ponds by Baker and Cox (2010) and by Chambers (2009). The model used by both was the same as that discussed above, i.e., the stagnant film model, also known as the two bottleneck model (Schwarzenbach 2003). Using the model, Baker and Cox calculated the radon flux at the surface of a pond with dissolved radium and compared the results with charcoal canister measurements which were floated on the surface of the pond. They concluded that the 1.65 pCi/m^2-sec predicted by the model compares well with the mean measured flux of 1.13 pCi/m^2-sec. Following the Baker and Cox approach, Appendix B presents a derivation of the radon flux equation based on the bottleneck model and continuity of flux across the boundary. The basic equations and background may be found in Schwarzenbach (Chapter 19 – Bottleneck Boundaries). As noted therein, the flux across a two bottleneck boundary may be expressed as:

$$J = V_{tot}\left(\frac{C_a}{K_{aw}} - C_w\right)10 \qquad (1)$$

Where:			
J	=	Radon flux	(pCi/m^2-sec)
V_{tot}	=	Gas exchange velocity	(cm/sec)
K_{aw}	=	Air-water partition coefficient	(dimensionless)
C_a	=	Concentration of radon in the air	(pCi/L air)
C_w	=	Concentration of radon in the water	(pCi/L water)
10	=	Units conversion factor	(L/m^3) (m/cm)

The air-water partition coefficient (K_{aw}) estimated from the Henry’s Law constant from NIST (NIST 2008) and conversion factors is:

$$K_{aw} = 4.35$$

From the previous discussion, the thickness of the stagnant air film and the stagnant water film can vary considerably. The selected diffusion coefficients for molecular diffusion of radon are about 10^{-5} cm^2/sec in water and 0.11 cm^2/sec in air. Baker estimated these film thicknesses as 100 microns for water and 1000 microns for air (0.01 cm and 0.1 cm). Substituting into the definitions for transfer velocities for water and air (see Appendix A):

$$V_w = \frac{D_w}{Z_w} = \frac{10^{-5}}{0.01} = 10^{-3}$$

And (2)

$$V_a = \frac{D_a}{Z_a} = \frac{0.11}{0.1} = 1.1$$

Where:	V_w	=	Transfer velocity for water	(cm/sec)
	V_a	=	Transfer velocity for air	(cm/sec)
	D_w	=	Diffusion coefficient for water	(cm^2/sec)
	D_a	=	Diffusion coefficient for air	(cm^2/sec)
	Z_w	=	Water film thickness	(cm)
	Z_a	=	Air film thickness	(cm)

However, the total transfer velocity is defined as:

$$V_{tot} = \frac{V_w \ V_a \ K_{aw}}{V_w + V_a \ K_{aw}} \tag{3}$$

Noting that $V_a K_{aw} >> V_w$, means that the denominator can be approximated by $V_a K_{aw}$, which leads to:

$$V_{tot} \approx \frac{V_w \ V_a \ K_{aw}}{V_a \ K_{aw}} \approx V_w \tag{4}$$

It is assumed that the atmospheric radon concentration is much less than the water radon concentration, or:

$$\frac{C_a}{K_{aw}} << C_w \tag{5}$$

Then the flux (Equation 1) reduces to:

$$J = V_w \ C_w \ 10$$

$$= \frac{D_w}{Z_w} \ C_w \ 10 \tag{6}$$

$$= 10^{-2} \ C_w \tag{7}$$

Where	J	=	Radon flux	(pCi/m^2-sec)
	C_w	=	Concentration of radon in the water	(pCi/L)
	V_w	=	Transfer velocity for water	(cm/sec)
	D_w	=	Diffusion coefficient for water	(cm^2/sec)

= 10^{-5} (cm^2/sec)
Z_w = Water film thickness (see Section 2.2.4) (cm)
= 0.01 (cm)
10 = Units conversion factor (L/m^3) (m/cm)

Given a deep evaporation pond, for any sediment less than 2 meters below the surface, it is assumed the radon in the water is in secular equilibrium with the radium in the water. For a normalized concentration of radium in the water of 1 pCi/L, the flux is 0.01 pCi/m^2-sec. This flux matches the calculated radium normalized radon flux from Baker and Cox (2010), and compares favorably with the Baker and Cox (2010) measured normalized flux of 0.0068 pCi/m^2-sec and the 0.0058 pCi/m^2-sec radon flux due to diffusion given by Chambers (2009)[1]. This indicates that while the flux from an evaporation pond may be limited, it is not negligible.

Baker measured the radium concentration in an evaporation pond at the Homestake ISL facility at 165 pCi/L. Assuming a direct conversion to Rn-222 (165 pCi/L), the flux is estimated from Equation 7 at 1.65 pCi/m^2-sec. This is comparable to later measurements of the flux carried out by Baker, et al. using charcoal canisters. The average measured flux was 1.13 pCi/m^2-sec. This flux is also comparable to the flux calculated by Nielson and Rogers in the pond portion of a tailings pond (see **Table 2**), assuming an average ore grade of 0.1% U_3O_8 —or about 290 pCi/g Ra-226. The radon flux is 6.4 pCi/m^2-sec. This is also similar to the radon flux reported by Mudd (Section 2.2.1) of 2.2 pCi/m^2-sec from the tailings repository in Pit # 1 (water depth greater than 10 m) (IAEA 2003).

These values are the same order of magnitude in the Sears (ORNL 1975) Table (**Table 1**) for approximately the same order of magnitude of radium in the water.

Lastly, it is noted that the work conducted by Baker and Cox (2010) did not attempt to take wind speed into account. In fact, using the floating charcoal canisters on the pond surface insures that the wind on the surface being measured is zero, since the wind is blocked by the edge of the canisters flotation collar (Baker and Cox 2010). This can bias the radon flux results, as will be shown in the next section.

2.2.4 Modeling the Effects of Wind Turbulence

A number of researchers have investigated wind speed in conjunction with reduction in the film thickness and subsequent increase in the flux of gases from the surface (carbon dioxide, radon and so on). Schwarzenbach (2003) found that the thickness of the film layer is proportional to the inverse of the square of the average wind speed for winds greater than 2 m/sec in the open ocean. Broecker and Peng (1975) noted that at an average wind speed of 7 m/sec (12 knots or 13.8 mph), the film thickness was measured at 64 microns (64 µm), while when the average wind velocity was 12 m/sec (22 knots or 25.3 mph), the measured film thickness decreased to 20 microns, which follows the inverse squares prediction [i.e., 64 µm/20 µm ≈

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[1] The flux reported by Chambers (2009) is actually 5.8 × 10^{-6} pCi/m^2-sec per pCi/L radium. However, he forgot to convert the radium concentration from pCi/L to pCi/m^3 before calculating the flux, so his reported flux is three orders of magnitude too low.

$(12\ \text{m/sec})^2/(7\ \text{m/sec})^2$]. This section presents a mathematical expression for estimating the influence of wind speed on radon flux.

Besides the work of Broecker and Peng (1975) and also Duenas (1983) previously mentioned, further investigations regarding wind speed and film thickness were published including a paper by Wanninkhof (1992). In this paper, Wanninkhof discusses and summarizes some of the questions of wind speed and gas exchange over the ocean. In a later paper, he and J. Crusius consider the question of gas transfer velocities measured at low wind speed over a lake (Crusius and Wanninkhof 2001). Again, the gas transfer velocity is the diffusion coefficient divided by the film thickness. Crusius and Wanninkhof (2001) present various published relationships between transfer velocities and wind speed. However, these relationships probably do not hold for bodies of water with surface areas as small as evaporation ponds.

Much of the prior research was conducted on the open ocean. An important paper for limited-area water bodies, like evaporation ponds, was the work of S. Emerson (Emerson 1975). He considered the influence of wind speed on the film thickness in Canadian Shield lakes. In the lake, Emerson used a limnocorral for his experiments; that is, a polyethylene curtain (a limnocorral) deployed in a lake such that it enclosed and “isolated” a triangular area in the top-most layer of a thermally stratified lake (the epilimnion). Thus a small area, rather than the entire lake surface, was investigated. This smaller area can be taken as analogous to an evaporation pond. He determined a ”boundary layer” (film thickness) of about 600 microns from a series of three independent experiments conducted from1971 through 1972. Emerson noted that the relationship with wind speed breaks down for wind velocities below 2 m/sec (3.9 knots or 4.5 mph) and presented a plot compiling various researchers’ data of film thickness versus wind speed. Figure 4 of his paper (reproduced as **Figure 2**) presents a compilation of his and other author’s results, which includes stagnant boundary layer values from tank and wind tunnel experiments along with other measurements (Emerson 1975). Note that the wind speeds are all adjusted to the standard 10-meter height wind speed; see for example the measurements of Crusius (Crusius 2003) whose anemometer was at one meter and who used the conversion that the speed at 10 meters is equal to 1.22 times the speed at one meter.

To develop a mathematical expression for the influence of wind speed on the radon flux, a straight line was drawn through the semi-log plot of points on Emerson’s Figure 4, as shown on **Figure 2**.

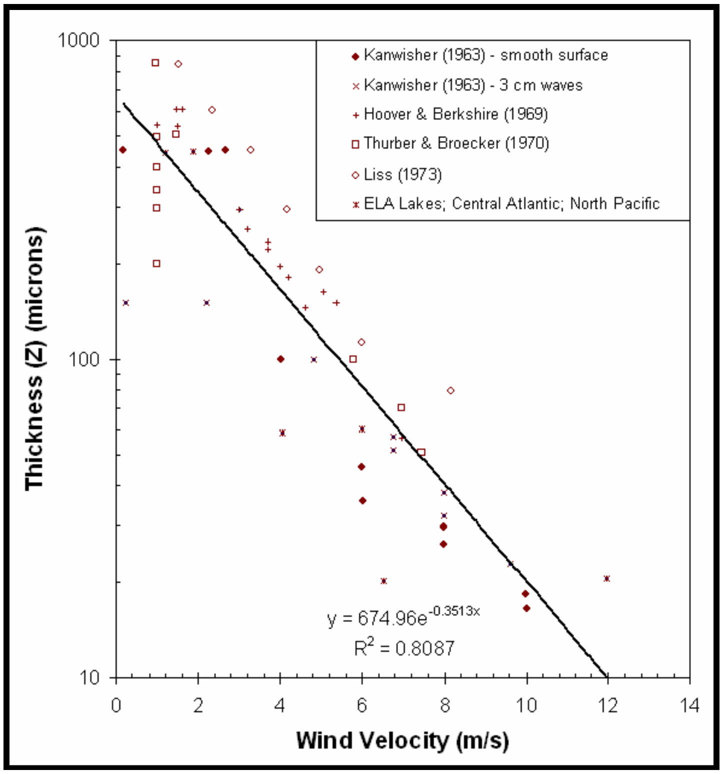

Figure 2: Boundary Layer Thickness Versus Wind Speed
(Based on Emerson 1975, Figure 4)

There is a fair amount of scatter in the points on this semi-log plot, which can result in considerable error. Both polynomial (square and cube) and exponential curves were fitted to the straight line on the figure. The exponential was selected, as polynomials were negative at high wind speed. It is important to note that this functional relationship is not part of a model or function having a physical basis. Rather, it is a mathematical construct. The exponential function was:

$$Z_w = 674.9 \exp(-0.351 * V) \qquad (8)$$

Where: Z_w = film thickness (microns)
V = wind speed (m/sec)

As shown in **Figure 2**, Equation 8 has a coefficient of determination (R^2) equal to 0.8087, which indicates that about 81% of the variation in data points can be explained by Equation 8. A coefficient of determination of 81% is generally considered a good fit to the data.

Using Equation 8, the film thickness was determined, which was then entered into Equation 6. The resulting normalized radon fluxes (i.e., pCi/m^2-sec per pCi/L radium) are shown in **Table 3** for wind speeds ranging from 2 to 10 m/sec (4.5 to 22.4 MPH). **Table 3** indicates that the radon flux would increase by over an order of magnitude when the wind speed increases by a factor of five.

Table 3: Effect of Wind Speed on Pond Radon Release

Wind Speed @ 10 m		Film Thickness (µm)	Normalized Radon Flux* (pCi/m^2-sec)
(m/sec)	(MPH)		
2	4.5	335	0.0030
4	8.9	166	0.0060
6	13.4	82	0.012
8	17.9	41	0.025
10	22.4	20	0.050

* Per pCi/L radium.

Equation 8 was used to calculate the film thickness for conventional uranium mill and ISL facilities. Wind speeds for both conventional mill and ISL facilities are provided and discussed in Section 3.2.

As stated in Section 2.2.2, Chambers (2009) included turbulence (wave action) in his model of radon release from evaporation ponds. Chambers assumed that all of the radon produced by the decay of radium in the turbulent layer was released, and presented calculated radon fluxes of 0.0002 and 0.001 pCi/m^2-sec per 1 pCi/L of radium for turbulent layer thicknesses of 10 and 50 cm, respectively. There are two problems with this approach: 1) there is no guidance on how to select the turbulent layer thickness, and 2) compared to the **Table 3** results, the radon flux is low, probably due to limiting the radon generation to only the turbulent layer.

Although the stagnant film model that has been developed is limited to wind speeds that are less than about 10 m/sec, since most sites experience wind speeds that are this high or higher only a small portion of the time, this should not be a serious constraint for using the model to evaluate radon releases from evaporation ponds. For example, **Table 5** shows that for 11 existing or potential pond sites, the amount of time that the wind speed exceeds 10 m/sec ranges from <1% to <6%. Also, even if there was a high-speed wind event, the radon that could be released would be limited by the total amount that could be dissolved in the pond water (e.g., <1 Ci), after which it would require a period of time (one to two weeks) for the radon to re-equilibrate with the radium, during which time the pond's radon release would be depressed.

3.0 ESTIMATE OF EVAPORATION POND RADON RELEASE

This section presents estimates for three sources of radon: (1) from the surface of evaporation ponds based on the material in the prior sections, (2) from the direct discharge of "process bleed" lixiviant or purge water into the evaporation pond, and (3) from the spray of purge water into the evaporation pond. These are generic estimates. It is expected that the limits for each ISL evaporation pond will be determined on a case-by-case basis using site specific parameters. For each site, the number of ponds, and the flux from each pond, will be superimposed and used as the source term for the site and, ultimately, the dose. As discussed in Section 2.2.4, the wind speed plays a prominent role in determining the flux from an evaporation pond, which is dependent on the location of the facility.

3.1 Evaporation Pond Descriptions

This section presents an overview of evaporation ponds, their size, and the quantity of radium and radon in the water. According to the NRC (NRC 2009), typical evaporation ponds at ISL recovery operations have surface areas ranging from 0.1 to 6.2 acres (4,356 to 270,000 ft^2). However, based on the review presented below, the latter, larger area seems more typical of ISL operations.

For example, the original Crow Butte design report (WWC 1988) had five (5) evaporation ponds in an area adjacent to the ore body. Ponds 1, 2, and 5 had bottom dimensions of 850 × 200 ft = 170,000 ft^2, while ponds 3 and 4 were 700 × 250 ft^2, or 175,000 ft^2. Each pond was 15 feet deep, giving a total capacity of some 114 acre-feet, or about 37×10^6 gallons.

Twenty years later, at the same upgraded facility, Crow Butte (2009 NRC Inspection Report), during the period October 2008 through September 2009, there were three commercial ponds and two Research and Development (R&D) ponds. The commercial ponds (numbers 1, 3, and 4) had a 2:1 slope and were nominally 900 feet by 300 feet by 17 feet deep (area 270,000 ft^2 by 17 feet deep). The storage capacity of the pond system was 122.4 acre-feet or about 40×10^6 gallons, which is similar to the original design.

The ponds at the Christensen Ranch and the Irigaray Site are not as deep. At the Christensen Ranch, there are four ponds 100 feet by 400 feet by 9 feet with an operating capacity of 5.51 acre-feet each, or approximately 22 acre-feet altogether (7.1×10^6 gallons).

At the Irigaray Site, there are five lined evaporation ponds (see **Table 4**) and two lined restoration ponds (Areva 2008). The five lined evaporation ponds have a total capacity of 27.9 acre-feet, and were constructed in 1978 and 1979 under WDEQ Permit to Mine No. 478 and Source Material License SUA-1341. The two lined restoration ponds were constructed in 1979.

Table 4: Irigaray ISL Evaporation Ponds

ID	Size (ft)	Depth (ft)	Capacity (acre-feet)	Evaporation (acre-feet/yr)
A	100 × 390	6	6.3	6.12
B	250 × 250	6	6.3	6.03
C	100 × 390	6	6.3	6.12
D	250 × 250	6	6.3	6.02
E	100 × 250	6	2.7	2.73

Currently (May 2008), five of the ponds (Ponds A, C, D, E, and RA) have been decommissioned. The liners, leak detection system, and all contaminated materials have been removed and disposed of at the licensed Shirley Basin facility. The berms and supporting earthworks have been maintained intact. It is anticipated that a combination of ponds A, C, D, and RA will be re-installed as necessary to support the evaporative disposal of process water, up to 25 gpm, resulting from resumption of uranium recovery operations. Note that the depth of the ponds (about two meters) is still greater than the one-meter cutoff investigated by Neilson for influence of any bottom sediment.

At the Smith Ranch-Highlands facility, there are two small, lined solar evaporation ponds in operation. The capacity of each pond is 0.78 acre-feet of water. Each pond is 100 ft by 100 ft and 8 feet deep. During operations, a 3-feet freeboard is maintained in each pond to protect the berms from wave action due to winds. (PRI 2003)

3.2 Evaporation Pond Radon Release

To determine the average flux over the year, the joint frequency wind speed distributions were considered for three conventional uranium facilities and eight ISL facilities. **Table 5** presents the wind speed distribution for these sites. Each of the bins represents the wind speed summed over the sixteen compass directions. To calculate the flux, the center of each wind bin was determined and converted to meters/second. As is evident, the "windiest" location appears to be the Smith Ranch – Highland, with winds greater than 15.5 MPH for more than 45% of the time.

Table 5: Uranium Sites – Wind Speed Data Percentage in Each Wind Bin

		Wind Speed @ 10 meters					
	Wind "Bin" MPH	< 4	4 to 7	8 to 12	13 to 18	19 to 24	> 24
	Center of Bin MPH	1.5	5.5	10	15.5	21.5	28
	Center of Bin m/sec	0.7	2.5	4.5	6.9	9.6	12.5
Mill Type	**Site**	**Percentage of Wind Speed in Each Wind Bin**					
Conventional Uranium Mills	Sweetwater	19.5	21.6	22.3	20.7	10.9	5.1
	White Mesa	16.5	42.8	27.6	10.6	2.1	0.5
	Canon City	0.6	48.8	30.5	20.2	0.0	0.0
In-Situ Leach	Smith Ranch–Highland	5.7	21.1	28.2	26.1	13.2	5.7
	Crow Butte	10.3	29.0	30.2	22.0	7.4	1.9
	Crown Point	44.2	26.4	23.6	4.4	1.0	0.4
	Christenson/Irigaray	26.8	28.7	20.9	16.9	5.2	1.2
	Church Rock	44.2	26.4	23.6	4.4	1.0	0.4
	Alta Mesa 1,2,3	27.4	24.7	26.3	18.5	2.8	0.4
	Kingsville Dome 1,3	27.4	24.7	26.3	18.5	2.8	0.4
	Vasquez 1,2	27.4	24.7	26.3	18.5	2.8	0.4

Source: SCA 2010

From the above table and the exponential expression for film thickness versus wind speed, the film thickness (Z_w) was calculated for each wind speed bin. A normalized radon flux was determine for each wind speed (pCi/m^2-sec per pCi/L) using Equation 6, the diffusion coefficient for water ($D_w = 10^{-5}$ cm^2/sec), the aforementioned Z_w, and a radium concentration of 1 pCi/L.

As is evident, the normalize flux varies over a factor of two dependent on the wind speed and the percentage of time for each wind speed. For two wind speed bins, 0.7 m/sec and >12.5 m/sec, the film thickness versus wind speed functionality does not hold (Emerson 1975), so the film thicknesses for the adjacent bin was used in the flux calculation.

Figure 3 shows the results of the calculation for the three conventional mills and the eight ISL facilities. The facility with the greatest average wind speed also has the highest flux. Also, the ratio of the highest to lowest flux is somewhat less than a factor of three.

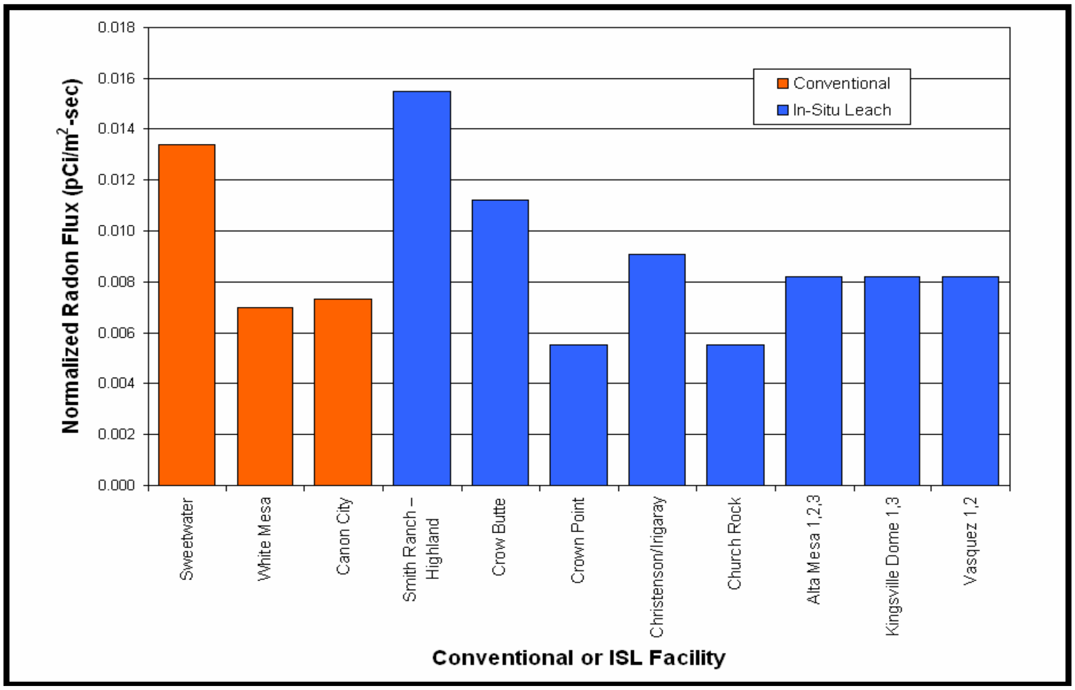

Figure 3: Normalized Radon Flux for Three Conventional and Eight ISL Uranium Facilities

Table 6 presents the radon flux for the aforementioned facilities for three radium in water concentrations, 1 normalized, 100 pCi/L and 1000 pCi/L. The fluxes at the largest concentration, while below the criteria, are not negligible.

Table 6: Radon Flux for Various Radium Concentrations

Mill Type	Radium Concentration (pCi/L)	1	100	1,000
	Site	Radon Flux (pCi/m^2-sec)		
Conventional Uranium Mills	Sweetwater	0.0134	1.34	13.4
	White Mesa	0.0070	0.70	7.0
	Canon City	0.0073	0.73	7.3
In-Situ Leach	Smith Ranch – Highland	0.0155	1.55	15.5
	Crow Butte	0.0112	1.12	11.2
	Crown Point	0.0055	0.55	5.5
	Christenson/Irigaray	0.0091	0.91	9.1
	Church Rock	0.0055	0.55	5.5
	Alta Mesa 1,2,3	0.0082	0.82	8.2
	Kingsville Dome 1,3	0.0082	0.82	8.2
	Vasquez 1,2	0.0082	0.82	8.2

Consider the flux from the Smith Ranch – Highland, which is about three times that from the Church Rock ISL. For the same radium concentration, say 1,500 pCi/L, the radon flux from a Smith Ranch – Highland evaporation pond would be 23.3 pCi/m^2-sec (which exceeds 20 pCi/m^2-sec), while the same radium concentration at Church Rock gives a radon flux of 8.3 pCi/m^2-sec (well below 20 pCi/m^2-sec). Thus, the radon flux is very dependent on wind environment, which is, in turn, dependent on the geographic location. Evaporation ponds located in calm areas can have higher concentrations of radium than those in windy areas. This is somewhat analogous to the situation of radon from soils. Soils with low porosity and tortuosity, such as clays, have low bulk diffusion coefficients versus sandy soils (7×10^{-3}cm^2/sec varved clay versus 6.8×10^{-2} cm^2/sec fine quartz, ORNL 1975). Thus, radium concentrations in different soils can be considerably elevated in these types of materials and still meet the 20 pCi/m^2-sec UMTRCA criteria.

To determine radon fluxes at various sites, the radium in the evaporation pond water was compiled from different sources for three sites. **Table 7**, **Table 8**, and **Table 9** present the reported radium-226 concentrations at the Christenson/Irigaray, Smith Ranch – Highland and the Crow Butte facility, respectively. Only the Christenson/Irigaray concentrations (**Table 7**) were measured directly in the ponds. The Smith Ranch – Highland concentrations (**Table 8**) were measured at the discharge from the radium treatment system, which would also be the concentration of the water entering the evaporation ponds. For the Crow Butte site, it was assumed that the radium concentration entering the ponds would be the same as the radium concentration for water injected into the deep disposal wells (**Table 9**).

Table 7: Christenson/Irigaray Evaporation Pond Radium Concentration

Pond ID #	Radium (pCi/L)				
	04/19/09	05/21/09	08/18/09	11/11/09	Average
IR-B	141	191	131	152	154
IR-RB	3.5	2.4	N/D	21	9
CR-1	87.3	182	205	104	145
CR-2	63.8	75.7	60.2	46	61
CR-3	69.7	47.3	57.7	87	65
CR-4	152	101.7	133.3	136	131

Source: COGEMA 2010, Table 1

Table 8: Smith Ranch – Highland Evaporation Pond Radium Concentration

Date	Radium (pCi/L)		Date	Radium (pCi/L)	
	No. 2	No. 3		No. 2	No. 3
Jan-09	7.5	3.3	Jan-10	1.1	65
Feb-09	3.2	32	Feb-10	3.8	1.5
Mar-09	18	8.5	Mar-10	1.5	48
Apr-09	0.95	4.8	Apr-10	1.5	1.8
May-09	2.5	32	May-10	11	23
Jun-09	6	3.4	2010 Ave	4	28
Jul-09	3.4	3.4	2009 and 2010 Average	4	20
Aug-09	5	27			
Sep-09	3.9	6			
Oct-09	2.7	0.29			
Nov-09	0.77	N/D			
Dec-09	1.2	3.3			
2009 Ave	5	11			

Source: PRI 2010, Tables 7-11 and 7-12

Table 9: Crow Butte Evaporation Pond Radium Concentration

Date	Radium (pCi/L)	Date	Radium (pCi/L)
Jan-07	1,060	Jul-07	856
Feb-07	1,200	Aug-07	782
Mar-07	1,030	Sep-07	974
Apr-07	1,090	Oct-07	788
May-07	918	Nov-07	820
Jun-07	1,000	Dec-07	1,230
Average (pCi/L)			980

Source: CBR 2007a, Appendix D and CBR 2008, Appendix D

These radium concentrations vary from 4 pCi/L to 980 pCi/L, indicating the large range of lixiviant and/or the potential of dilution from other contributors of water to the evaporation ponds. Smith Ranch – Highlands is at the low end of the range, likely due to its use of a barium chloride treatment system to reduce the radium concentrations (see Section 3.5). Smith Ranch – Highlands notwithstanding, it is expected that the radium content would be in the 100's to 1000's pCi/L range at most ISL sites. These values for radium in the water are less than or comparable to those used to calculate risk from ISL facilities in a 2001 NRC review (NRC 2001). The assumed radium-226 activity in the pregnant lixiviant in that study was 3,400 pCi/L of radium-226.

Using the normalized fluxes previously calculated, the radon flux for each of the evaporation ponds are presented in **Table 10**. The fluxes for the most part are low, however, they are not negligible. For Crow Butte, the flux is a factor of ten higher than the others, reflecting the elevated radium content in the evaporation pond. On the other hand, for Smith Ranch – Highlands, the radium content is low due to the radium treatment system.

Table 10: Radon Flux at the Christenson/Irigaray, Smith Ranch – Highland and Crow Butte Facilities

Facility	Pond ID	Radon Flux (pCi/m^2-sec)	
		Average	Maximum
Crow Butte	1, 3, or 4	11.0	13.8
Christenson/Irigaray	IR-B	1.40	1.74
	IR-RB	0.08	0.19
	CR-1	1.31	1.86
	CR-2	0.56	0.69
	CR-3	0.59	0.79
	CR-4	1.19	1.38
Smith Ranch-Highland	No. 2	0.07	0.28
	No.3	0.25	1.01

As a last comparison, the calculated annual radon released from the evaporation ponds was compared to total site reported radon releases. For Crow Butte, the facility total radon releases to the environment for 1995 through 2006 are given in the license renewal application (CBR 2007b, Table 5.8-8). For Christenson/Irigaray, the total facility radon releases for 1995 through 2000 are given in license renewal application (COGEMA 2008, Table 5.13). For Smith Ranch – Highlands, the facility total radon release was taken from Section 4.1.2 of the source material license application (PRI 2003). As shown by **Table 11**, the calculated evaporation pond radon release is less than 1% of the facility total radon releases for each of the three sites analyzed.

Table 11: Comparison of Calculated Evaporation Pond Radon Emissions to Reported Facility Total Radon Emissions

Facility	Radon Release (Ci/yr)		Pond Fraction of Facility Total
	Facility Total*	Pond	
Crow Butte	3,537 to 4,760	26.2	0.59%
Christenson/Irigaray	649 to 1,454	0.70	0.07%
Smith Ranch—Highlands	6,738	0.009	<0.001%

* Source: Crow Butte: CBR 2007b, Table 5.8-8
Christenson/Irigaray: COGEMA 2008, Table 5.13
Smith Ranch-Highlands: PRI 2003, Section 4.1.2

3.3 Direct Discharge Radon Release

Another source or radon is the point source from radon in the water being transferred to the surface of the pond, called the bleed lixiviant, process bleed, or purge water. As previously stated, the bleed lixiviant is necessary to maintain a hydraulic cone of depression around each well field to prevent leakage of mining solutions outside the production zone. The radon in the bleed lixiviant is assumed to be the same as that in the pregnant lixiviant. The radon in this lixiviant ranges in various studies from 1.3×10^5 pCi/L to 1.93×10^5 pCi/L (NRC, 2001). The transferred amount, that is the amount that goes into the atmosphere during the discharge process, can be estimated from studies of the transfer of radon in the home from water containing dissolved radon. The transfer coefficient is defined as the ratio of the average incremental concentration throughout a house to the average concentration in water. The transfer coefficient is really of value only to the release of radon in a closed area within the processing facility. It does not hold for an open system such as the discharge pipe. However, the transfer efficiency, which is the fraction of radon in the water that is released to the air during activities such as showering, clothes washing or other water-moving activities, is similar to discharge. The weighted average of measurements by various researchers of this radon transfer efficiency was 0.52 (NAS 1999).

NRC has also considered this source in NUREG-1569 Appendix D (NRC, 2003). Their hypothesis is that almost all the radon in the purge lixiviant is readily released to the atmosphere as versus the radon transfer efficiency of 52%. The amount of radon available for release is dependent on the water volume purge rate and the concentration of radon in the water. For a "typical" ISL, NRC assumes the radon concentration in the purge water is 3.2×10^5 pCi/L, which is in the range discussed previously. The purge rate was taken as 5.5×10^5L/d or about 100 gpm. This purge rate is very conservative. For example, the purge rate at the Irigaray Site is about 25 gpm. The radon released per year can be calculated from

$$Rn= 3.65 \times 10^{-10}\ (Ci/pCi)(d/y) \times (3.2 \times 10^5\ pCi/L) \times 5.5 \times 10^5 L/d \qquad (9)$$

or

$$Rn= 64\ Ci/year$$

It is noted that continual (365 days per year) processing is assumed to occur. While this release can be used to estimate source terms for dose calculations, it cannot be used for flux estimates, as the area is unknown and any assumed area (such as the evaporation pond area) is arbitrary with no physical basis.

3.4 Enhance Evaporation Sprays Radon Release

In order to reduce the size of the evaporation ponds, some ISL facilities utilize enhanced evaporation systems, see **Figure 4**.

Figure 4: Sprays Used to Enhance Evaporation

It is expected that any radon contained within the pond water that is sprayed into the air would be released.

At equilibrium, with the sprays functioning, the radon activity within the evaporation ponds can be calculated by Equation 10.

$$\frac{dA'_{Rn}}{dt} = \lambda_{Ra} A'_{Ra} - \lambda_{Rn} A'_{Rn} - f_s A'_{Rn} = 0 \qquad (10)$$

Where: A'_{Rn} = Radon-222 activity in the pond (atoms)

A'_{Ra} = Radium-226 activity in the pond (atoms)

λ_{Ra} = Radium-226 decay constant (sec^{-1})

λ_{Rn} = Radon-222 decay constant (sec^{-1})

f_s = Fractional radon release rate (sec^{-1})

$= \frac{F_s}{V_p} \varepsilon$

F_s = Spray flow rate (L/sec)

V_p = Pond volume (L)

ε = Spray radon removal efficiency (dimensionless)

Solving Equation 10 and converting from atoms to Curies gives:

$$A_{Rn} = \frac{\lambda_{Rn} A_{Ra}}{\lambda_{Rn} + f_s} = \frac{\lambda_{Rn} C_{Ra} V_p}{\lambda_{Rn} + f_s} \qquad (11)$$

Where: A_{Rn} = Radon-222 activity in the pond (Ci)

A_{Ra} = Radium-226 activity in the pond (Ci)

C_{Ra} = Radium-226 concentration in the pond (Ci/L)

All others terms have been previously defined.

Notice that when the spray system is turned off (i.e., $f_s = 0$), Equation 2 results in the radon activity (or concentration) (A_{Rn} or C_{Rn}) being equal to the radium activity (or concentration) (A_{Ra} or C_{Ra}), as would be expected.

The radon release (R_{Rn}) due to the sprays may be calculated from Equation 12.

$$R_{Rn} = A_{Rn} f_s = \frac{\lambda_{Rn} C_{Ra} V_p f_s}{\lambda_{Rn} + f_s} = \frac{\lambda_{Rn} C_{Ra} F_s \varepsilon}{\lambda_{Rn} + f_s} \qquad (12)$$

All terms have been previously defined.

To demonstrate the effect of using a spray system to enhance evaporation on radon release, the ponds shown in **Table 12** were evaluated using Equation 12. Note, while the dimensions shown on **Table 12** are for actual ISL evaporation ponds, the actual ponds do not necessarily use, or contain, spray systems.

Table 12: Evaporation Pond Dimensions

Site	Pond ID	Length (ft)	Width (ft)	Depth (ft)	Volume (V_p) (L)
Irigaray	I-1	100	250	6	4.2E+06
Irigaray	I-2	100	390	6	6.6E+06
Christensen Ranch	CR-1	100	400	9	1.0E+07
Irigaray	I-3	250	250	6	1.1E+07
Crow Butte	CB-1	850	200	15	7.2E+07
Crow Butte	CB-2	700	250	15	7.4E+07
Christensen Ranch	CR-2	900	300	17	1.3E+08

The values assumed for the other parameters needed to solve for the amount of radon released due to the operation of the sprays are shown in **Table 13**.

Table 13: Parameters Used to Estimate Spray System Radon Release

Parameter	Case 1	Case 2	Case 3	Case 4	Units
Radon-222 half-life	3.823				days
Radon-222 decay constant (λ_{Rn})	2.1E-06				sec^{-1}
Spray radon removal efficiency (ε)	1	1	1	0.8	—
Radium-226 concentration in the pond (C_{Ra})	600	600	300	600	pCi/L
Spray flow rate	100	1,000	100	100	gpm
Spray flow rate (F_s)	6.31	63.1	6.31	6.31	L/sec

For the spray removal efficiency, Rost (1981) demonstrated the ability of spray aeration to remove radon from water. Depending upon the spray arrangement used, spray aeration achieved between 76% and 93% radon removal. Because of these high removal rates, and because it is conservative, a 100% radon removal has been assumed for three of the four cases analyzed. For the fourth case, a nominal 80% radon removal has been assumed. The second case shows the effect of increasing the spray flow rate by an order of magnitude, while the third case shows the effect of increasing the Ra-226 concentration in the water entering the pond. Note, for this analysis credit was not taken for either the buildup of radium in the pond nor for the removal of radium due to the addition of barium chloride to the pond water.

The results for the four cases analyzed are shown in **Table 14**.

Table 14: Calculated Radon Releases Due to Enhanced Evaporation Sprays

Pond ID	Case 1				Case 2			
	f_s	C_{Rn}	R_{Rn}		f_s	C_{Rn}	R_{Rn}	
	(sec^{-1})	(pCi/L)	(pCi/sec)	(pCi/m^2-sec)	(sec^{-1})	(pCi/L)	(pCi/sec)	(pCi/m^2-sec)
I-1	1.5E-06	351	2,217	0.95	1.5E-05	74	4,686	2.02
I-2	9.5E-07	413	2,604	0.72	9.5E-06	108	6,836	1.89
CR-1	6.2E-07	463	2,923	0.79	6.2E-06	152	9,585	2.58
I-3	5.9E-07	468	2,950	0.51	5.9E-06	157	9,880	1.70
CB-1	8.7E-08	576	3,634	0.23	8.7E-07	424	26,726	1.69
CB-2	8.5E-08	577	3,638	0.22	8.5E-07	427	26,953	1.66
CR-2	4.9E-08	586	3,700	0.15	4.9E-07	487	30,743	1.23

Table 14: Calculated Radon Releases Due to Enhanced Evaporation Sprays

Pond ID	Case 3				Case 4			
	f_s	C_{Rn}	R_{Rn}		f_s	C_{Rn}	R_{Rn}	
	(sec^{-1})	(pCi/L)	(pCi/sec)	(pCi/m^2-sec)	(sec^{-1})	(pCi/L)	(pCi/sec)	(pCi/m^2-sec)
I-1	1.5E-06	176	1,108	0.48	1.2E-06	383	1,933	0.83
I-2	9.5E-07	206	1,302	0.36	7.6E-07	440	2,222	0.61
CR-1	6.2E-07	232	1,462	0.39	5.0E-07	485	2,450	0.66
I-3	5.9E-07	234	1,475	0.25	4.8E-07	489	2,469	0.43
CB-1	8.7E-08	288	1,817	0.12	7.0E-08	581	2,931	0.19
CB-2	8.5E-08	288	1,819	0.11	6.8E-08	581	2,933	0.18
CR-2	4.9E-08	293	1,850	0.074	3.9E-08	589	2,973	0.12

Table 14 shows that even with a relatively large spray flow rate (1,000 gpm), the radon release rate is small (≤2.58 pCi/m^2-sec), or ≤0.0043 pCi/m^2-sec per pCi/L of radium in the pond water. For the largest and smallest ponds analyzed, **Figure 5** shows the effect of varying the spray flow rate on the radon release rate. As shown, the radon release from the large and small ponds becomes asymptotic 0.011 and 0.0038 pCi/m^2-sec per pCi/L of radium, respectively. For a pond radium concentration of 1,000 pCi/L, the large and small pond radon releases become asymptotically 11 and 3.8 pCi/m^2-sec, respectively. These fluxes are similar in order of magnitude to those from high-wind areas previously discussed.

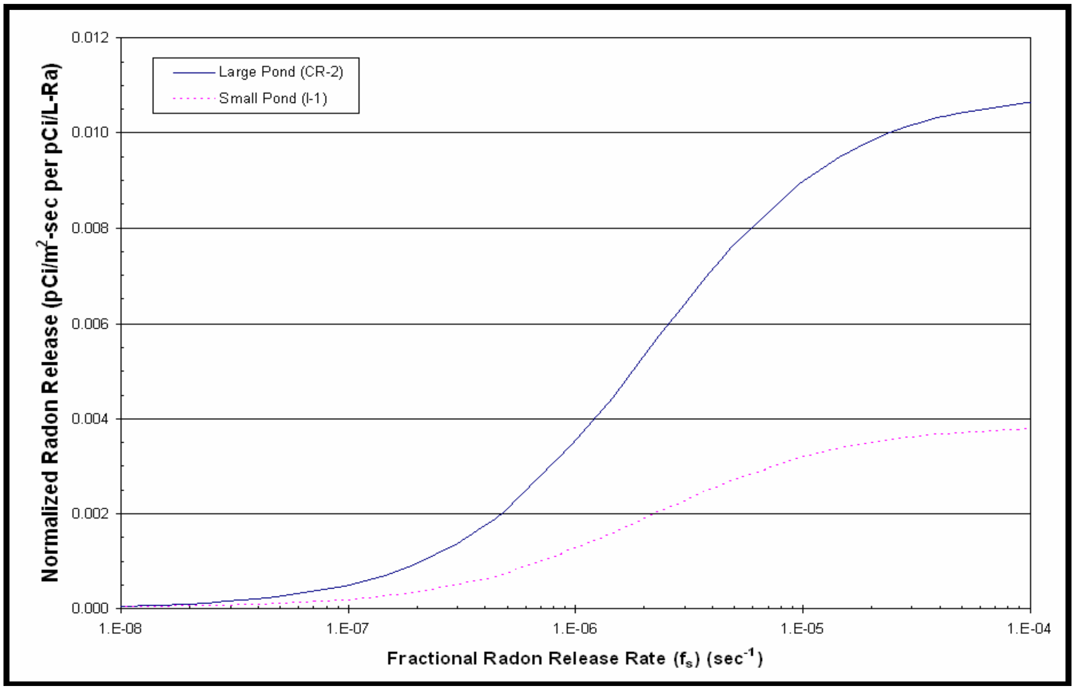

Figure 5: Effect of Spray Flow on Radon Release

3.5 Barium Chloride Treatment to Control Radium

Water entering the evaporation ponds exits the ponds via evaporation, however, radium does not evaporate, and thus builds up to concentration levels that can exceed the concentration of radium in the water entering the ponds. This buildup of radium would result in higher radon releases due to water to air transfer, as well as sprays used to enhance evaporation. In order to reduce radium pond concentrations, barium chloride ($BaCl_2$) is often added to the water. Co-precipitation of radium occurs when natural sulfate (SO_4) in the water combines with radium (Ra) and barium (Ba) to form $RaBaSO_4$ (Powertech 2009).

> Barium chloride ($BaCl_2$) treatment is widely used in the uranium industry to remove radium at mining sites. Radium concentrations below 8.1 pCi/L (0.3 Bq/L) can easily be achieved for wastewater containing sulfate ions. At pH values between six and eight, barium sulfate ($BaSO_4$) has a low solubility and readily precipitates out, co-precipitating radium at the same time. Only 0.00007–0.00013 lb (30–60 mg) of barium chloride per liter of wastewater will achieve 95–99 percent removal of radium. (EPA 2006)

Table 15 shows the effectiveness of barium chloride at removing radium from the Church Rock/Crown Point sample mine water. This reflects the 95% to 99% radium removal efficiency for barium chloride given by the EPA (EPA 2006).

Table 15: Hydro Resources, Inc. Data on Barium Chloride Treatment for Removing Radium from Wastewater

Test	$BaCl_2$ Concentration (mg/L)	Final Radium Concentration (pCi/L)
Test 1	0.0 (waste stream)	74.0
	10.5	0.21
	14.0	0.24
	17.5	0.24
Test 2	0.0 (waste stream)	73.4
	10.5	0.66
	14.0	0.28
	17.5	0.40
Test 3	0.0 (waste stream)	73.4
	14.0	0.20
	17.5	0.64

Source: NUREG- 1508, Table 2.3

Once the radium has precipitated out of the water, it will settle on the pond bottom. Because of the depth of evaporation ponds, radon produced from the precipitated radium would decay before it could be released from the ponds surface. This would only become problematic at the end life of the pond when the water is evaporated out and the pond sediment removed.

4.0 CONCLUSIONS

A review of the various models used for estimating radon flux from the surface of water bodies indicates that the stagnant film model (also known as the two bottleneck model), coupled with a wind correction equation, can be used to estimate the radon flux based on the concentration of radium in the pond's water and the assumption that radon is in secular equilibrium with the radium. Using this model, the radon flux from the surface of an evaporation pond, as a function of the wind speed, can be estimated with the following equation.

$$J = \frac{1.48 \times 10^{-4}}{e^{-0.351V}} C_w \qquad (13)$$

Where
J = Radon flux (pCi/m^2-sec)
C_w = Concentration of radium in the water (pCi/L)
V = Wind speed (m/sec)

Implicit in this model is the fact that in pond water, the radon diffusion coefficient is 10^{-5} cm^2/sec and the thickness of the stagnant film layer can be estimated from Equation 8.

Measurements conducted on the Homestake evaporation ponds by Baker and Cox (2010) agree with the stagnant film model estimates. However, as pointed out previously, the Baker and Cox (2010) measurement method does not allow the measurement of wind-generated radon fluxes, as the collar used to float the canister makes the wind speed zero above the area being measured. No data was found for measurements of the radon flux on evaporation ponds with the wind blowing.

The model should not be used for wind speeds above 10 m/sec. However, for the reasons given in Section 2.2.4, this is not expected to be a major limitation for estimating normal operational evaporation pond radon releases and impacts.

Using actual radium pond concentrations and wind speed data, Equation 13 was used to calculate the radon pond flux from several existing ISL sites. It was determined that the radon flux ranged from 0.07 to 13.8 pCi/m^2-sec (see **Table 10**). From this, it can be seen that the radon flux above some evaporation ponds can be significant (e.g., may exceed 20 pCi/m^2-sec). If this were to occur, there are methods for reducing the radium concentration in the ponds, the most straightforward being dilution. This, however, is temporary, as eventually evaporation will increase the concentration. A second method is barium chloride precipitate discussed in Section 3.5. The barium chloride will co-precipitate the radium to the bottom of the pond. As shown previously, radon generated at these pond depths will decay prior to reaching the surface.

Again, using actual ISL site data, the total annual radon release from the evaporation ponds was calculated and compared to the reported total radon release from the site. As **Table 11** shows, the evaporation pond contribution to the site's total radon release is small (i.e., <1%).

Two additional sources of radon release were investigated: 1) the discharge pipe, and 2) evaporation sprays. The discharge pipe discharges bleed lixiviant and liquid waste from the processing plant to the evaporation pond. Radon releases occur when the waste water exits the pipe and enters the pond. These radon releases can be estimated using the NUREG-1569, Appendix D methodology, thus, this source is currently included in the total radon releases reported for an ISL site. Nonetheless, a simple scoping calculation in Section 3.3 shows that the discharge pipe radon release is typically larger than the radon releases once the waste water is in the evaporation pond.

Spray systems are sometimes used to enhance evaporation from the ponds. In Section 3.4 a model to calculate radon releases during spray operation was developed. Section 3.4 also used data from ISL ponds to estimate this source of radon release. The radon releases from spray operation were calculated to range from <0.01 to < 3 pCi/m^2-sec (see **Table 14**). Furthermore, operation of the sprays would reduce the radon concentration within the pond, so that the normal radon release would be depressed once the sprays are turned off (until the radon has had an opportunity to re-equilibrate with the radium).

5.0 REFERENCES

Argonne National Laboratory (ANL), 1993, Data Collection Handbook to Support Modeling Impacts of Radioactive Material in Soil, Argonne National Laboratory, Argonne, Illinois, April 1993.

Argonne National Laboratory (ANL), 1997, MILDOS-AREA: An Update with Incorporation of In Situ Leach Uranium Recovery Technology, Argonne National Laboratory, Argonne, Illinois May 1997.

Areva Cogema Mining, Inc., 2008, Irigaray and Christensen Ranch Projects U.S. NRC License Renewal Application Source Material License SUA-1341, May 2008.

Baker, K.R. and A.D. Cox, 2010, "Radon Flux from Evaporation Ponds," presented at the National Mining Association/Nuclear Regulatory Commission Uranium Recovery Workshop, May 26-27, Denver, Colorado, http://www.nma.org/uranium_recovery_workshop.asp.

Bolin B., 1960, "On the Exchange of Carbon Dioxide between the Atmosphere and the Sea," Tellus 12, 274-281.

Broecker H.C. and T.H. Peng, 1974, "Gas Exchange Rates Between Air and Sea," Tellus, 24, 21-35.

Broecker H.C. and T.H. Peng, 1980, "Gas Exchange Rates for Three Closed-Basin Lakes," Limnom. Oceanog., V. 25(5), 789-796.

Burnett, W.C., J. Chanton, D.R. Corbett and K Dillon, "The Role of Groundwater in the Nutrient Budget of Florida Bay," Part I Final Report, NOAA Project # NA96OP0234, July 14, 2000.

Chambers, Douglas B., 2009, "Radon Emissions From Tailings Ponds," presented at the National Mining Association/Nuclear Regulatory Commission Uranium Recovery Workshop, July 2, Denver, Colorado.

COGEMA Mining Inc., 2010, "Irigaray and Christensen Ranch Projects, 2009 Annual Effluent and Monitoring Report, January 1 through December 31, 2009," February.

Crow Butte Resources, Inc. (CBR), 2007a, "Crow Butte Uranium Project, Radiological Effluent and Environmental Monitoring Report for First and Second Quarters, 2007."

Crow Butte Resources, Inc. (CBR), 2007b, "SUA - 1534 License Renewal Application," November.

Crow Butte Resources, Inc. (CBR), 2008, "Crow Butte Uranium Project, Radiological Effluent and Environmental Monitoring Report for Third and Fourth Quarters, 2007."

Crusius J., and R. Wanninkhof, 2003, "Gas transfer velocities measured at low wind speed over a lake," Limnom. Oceanog., 48(3), 1010-1017.

Duenas, C., M. C. Fernandez and M. Perez Martinez, 1983, "Radon-222 from the Ocean Surface," Journal of Geophysical Research, 85(C13), 8613-8616.

Emerson, S., 1975, "Gas Exchange Rates in Small Canadian Shield Lakes," Limnom. Oceanog., V. 20(5), 754-761.

Federal Register 1999, Federal Register Vol 64, No 38, "Radon in Drinking Water Health Risk Reduction and Cost Analysis Notice" pp. 9560-9599, page 9569.

Lawrence, C., 2005, *Measurement of* ^{222}*Rn Exhalation Rates and* ^{210}*Pb Deposition Rates in a Tropical Environment,* Thesis Queensland University of Technology, School of Physical and Chemical Sciences, Queensland , Australia, March.

Mudd, G.M., 2002, Uranium Mining in Australia: Environmental impact, radiation releases and rehabilitation. *Protection of the Environment from Ionising Radiation-Proceedings of the Third International Symposium*, Darwin, Australia, 22-26 July, Vienna, International Atomic Energy Agency.

Nielson K. K., V.C. Rogers, 1986, "Surface water hydrology considerations in predicting radon releases from water-covered areas of uranium tailings ponds," Geotechnical & Geohydrological Aspects of Waste Management / Fort Collins, 215-222

Oak Ridge National Laboratory (ORNL), 1975, "Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing 'as Low as Practicable' Guides-Milling of Uranium Ore," ORNL-TM-4903 Volume 1, Sears, M.B., R.E. Blanco, R.C. Dahiman, G.S. Hill, A.D. Ryan, and J.P. Witherspoon, May.

Power Resources, Inc., (PRI), 2003, "Smith Ranch-Highland Uranium Project, Source Material License Application," NRC License No.: SUA-1548, Docket No.: 40-8964, March 12.

Power Resources, Inc., dba Cameco Resources, (PRI), 2010, "NRC License SUA-1548, Docket No. 40-8964, Semi-Annual Effluent and Environmental Monitoring Report, January 1 through June 30, 2010," June 30.

Powertech (USA) Inc., 2009, "Dewey-Burdock Project, Supplement to Application for NRC Uranium Recovery License, Dated February 2009," August.

Rogers, V.C., K.K. Nielson, 1991, "Multiphase Radon Generation and Transport in Porous Minerals," Health Physics Vol. 60, No. 6, 807-813.

Rost, K.L., 1981, "Report on Spray Aeration for Division of Health Engineering," Maine Department of Human Services.

S. Cohen and Associates (SCA), 2010, “Risk Assessment Revision for 40 CPR Part 61 Subpart W – Radon Emissions from Operating Mill Tailings, Task 4 – Detailed Risk Estimates,” to be Published.

Schwarzenbach, Rene, P. Philip, M. Gschwend, and Dieter M. Imboden, 2003, *Environmental Organic Chemistry* 2nd edition, Chapter 19, Wiley-Interscience, Hoboken New Jersey, IBSN-0-471-35750-2.

Surbeck H., 1996, *A Radon-in-Water Monitor Based on Fast Gas Transfer Membranes,* International Conference on Technologically Enhanced Natural Radioactivity (TENR) Cause by Non-Uranium Mining, October 16-19, Szczyrk, Poland.

Usman, S.S., H.Spitz, and J Weisman, 2004, *Transport of Radon in Still Water under Steady State and Transient Conditions*, 11th International Congress of the International Radiation Protection Association, Madrid, Spain, May.

U.S. Environmental Protection Agency (EPA), 2006, “Technical Report on Technologically Enhanced Naturally Occurring Radioactive Materials from Uranium Mining,” Previously published on-line and printed as Vol. 1 of EPA 402-R-05-007, January 2006, Updated June 2007 and printed April 2008 as EPA 402-R-08-005.

U.S. Nuclear Regulatory Commission (NRC), 2009, *Generic Environmental Impact Statement for In-Situ Leach Uranium Milling Facilities-Final Report*, NUREG-1910 June.

U.S. Nuclear Regulatory Commission (NRC), 2003, *Standard Review Plan for In Situ Leach Uranium Extraction License Application – Final Report*, NUREG-1569.

U.S. Nuclear Regulatory Commission (NRC), 2001, *A Baseline Risk-Informed, Performance-Based Approach for In Situ Leach Uranium Extraction Licensees”,* NUREG/CR-6733 September.

U.S. Nuclear Regulatory Commission (NRC), 1997, “Final Environmental Impact to Construct and Operate the Crownpoint Uranium Solution Mining Project, Crownpoint, New Mexico,” NUREG-1508, February.

Western Water Consultants, Inc. (WWC), 1988, *Crow Butte Project, Solar Evaporation Pond Engineering Design Report*, April 27.

Winninkhof, R., 1992, “Relationship Between Wind Speed and Gas Exchange Over the Ocean,” Journal of Geophysical Research, 97(c5), 7373-7382.

WISE, 2002, WISE Uranium Project, Uranium Mill Tailings Radon Flux Calculator. Researched by Peter Diehl, Germany (2002) http://www.wise-uranium.org/ctb.html accessed September 2010.

APPENDIX A: NIELSON AND ROGERS RADON FLUX MODEL

Equation 3 from Nielson and Rogers 1986 can be used to calculate the radon flux from a pond due to three sources: shallow tailings, deep tailings, and dissolved radium. Nielson and Rogers 1986, Equation 3 has been reproduced as Equation A-1.

$$J = 10^4\, R\, \rho\, E \sqrt{\lambda D}\, \left[f_S + \left(1 - f_S\right) A_t\right] + 10^6\, R_w\, \lambda\, S_d \left(1 - 0.5\, f_S\right) \qquad \text{(A-1)}$$

Where:	J	=	Radon flux from the exposed pond surface	(pCi/m^2-sec)
	R	=	Solids radium content	(pCi/g)
	ρ	=	Bulk solid density	(g/cm^3)
	E	=	Radon emanation coefficient for solids	(dimensionless)
	λ	=	Radon decay constant	(sec^{-1})
	D	=	Radon diffusion coefficient in pore water	(cm^2/sec)
	f_S	=	Fraction of pond area with less than 1 m deep	(dimensionless)
	A_t	=	Attenuation factor for deep water	(dimensionless)
		=	$e^{\sqrt{\lambda / D_{tr}}\left(X_p - 100\right)}$	
	D_{tr}	=	Effective stagnant water transport coefficient	(cm^2/sec)
		=	0.003	(cm^2/sec)
	X_p	=	Average pond depth for areas greater than 1 m	(cm)
	R_w	=	Water radium content	(pCi/cm^3)
	S_d	=	Depth of surface layer from which all radon is assumed to be released	(m)
		=	1	(m)
	10^4	=	Conversion factor	(cm^2/m^2)
	100	=	Depth above which attenuation does not occur	(cm)
	10^6	=	Conversion factor	(cm^3/m^3)

APPENDIX B: FLUX FROM AN EVAPORATION POND

This appendix provides the derivation of the flux from the two bottleneck boundaries (also called the bottleneck boundary between two different media) for the model used by Baker as presented in (Schwarzenbach, R.P., et al. 2003).

Figure B-1 presents a diagrammatic sketch of the system.

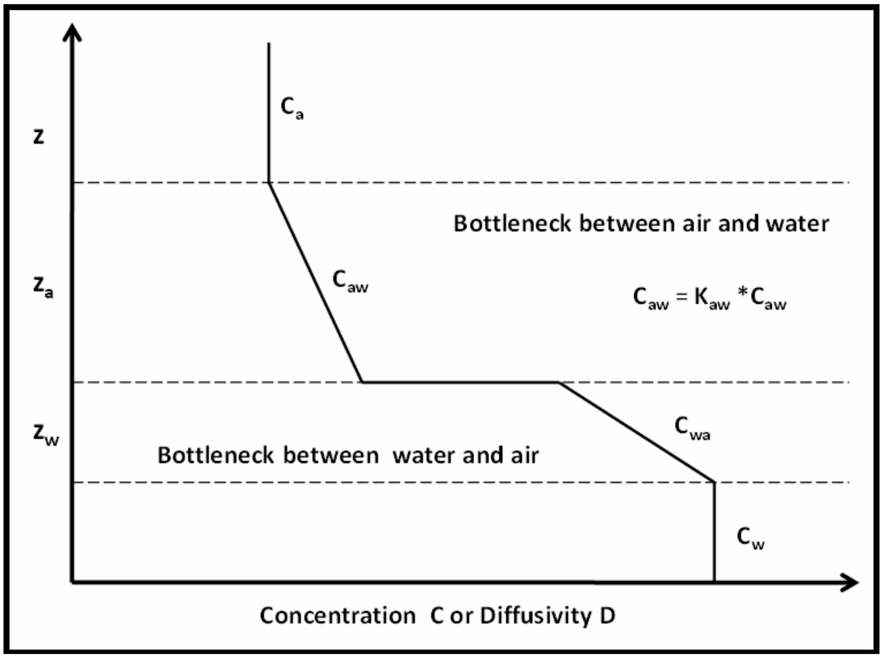

Figure B-1: Diagrammatic Sketch of a Two Bottleneck System

As presented by Schwarzenbach and noted by Baker:

Step 1 - Radon transfers from the water body to the stagnant water zone (bottleneck between water and air). This transfer occurs via the turbulent nature of the water and is dependent on water movement.

Step 2 - Radon transfers from the stagnant water zone to the stagnant air zone. This is a diffusion process. The diffusion gradient, and therefore the diffusion rate, are dependent on the depth of each zone (z_w and z_a) and the radon concentration.

Step 3 - Transfer of radon from the stagnant air zone to the air. Similar to Step 1 radon transfer due to the turbulent nature of the air and wind dispersion of radon into the air.

From Henry's law, the equilibrium partition coefficient can be defined as:

$$K_{aw} = \frac{C_a}{C_w} \qquad \text{(B-1)}$$

Where: K_{aw} = Air-water partition coefficient (dimensionless)
C_a = Concentration of radon in the air (pCi/L air)
C_w = Concentration of radon in the water (pCi/L water)

At equilibrium, the water concentration (C_w) is “imprinted” by the atmospheric concentration (or partial pressure) of radon.

Note from Figure B-1 that at the interface separating the two phases, the concentration jumps from C_{wa} on the water side to C_{aw} on the air side, so using Henry’s law:

$$K_{aw} = \frac{C_{aw}}{C_{wa}} \tag{B-2}$$

Where: K_{aw} = Air-water partition coefficient (dimensionless)
C_{aw} = Air/water bottleneck radon concentration (pCi/L)
C_{wa} = Water/air bottleneck radon concentration (pCi/L)

The unknown quantity is the “contact” concentration C_{aw}. This can be solved from the flux equations as the flux must be continuous across the boundary. The general term for the flux J is:

$$J = D\frac{\delta(C)}{\delta(z)} \tag{B-3}$$

Where: J = Radon flux (pCi/cm^2-sec)
D = Diffusion coefficient (cm^2/sec)
$\delta(C)$ = Change in radon concentration (pCi)
$\delta(z)$ = Distance (cm)

The above is sufficient to determine the contact concentration C_{wa}. The flux across the interface is continuous, that is, equal on either side of the interface (J_{aw}=J_{wa}). Substituting in Equation B-3 and setting the fluxes equal gives:

$$\frac{D_w}{Z_w}\left(C_{wa} - C_w\right) = \frac{D_a}{Z_a}\left(C_{aw} - C_a\right) \tag{B-4}$$

Solving for the water/air bottleneck concentration (C_{wa}) gives:

$$C_{wa} = \frac{V_a C_a + V_w C_w}{V_w + V_a K_{aw}} \tag{B-5}$$

Where: $V_w = \frac{D_w}{Z_w}$ and $V_a = \frac{D_a}{Z_a}$

From Equation A-4, substituting for C_{wa}, the flux (Equation B-3) becomes:

$$J = V_w \left[\frac{V_a C_a + V_w C_w}{V_w + V_a K_{aw}} - C_w \right] \qquad \text{(B-6)}$$

If the inverse of the total transfer velocity (V_{tot}) is defined as:

$$\frac{1}{V_{tot}} = \frac{1}{V_w} + \frac{1}{V_a K_{aw}} \qquad \text{(B-7)}$$

then by rearranging the terms of Equation B-7, the total transfer velocity (V_{tot}) is given by:

$$V_{tot} = \frac{V_w V_a K_{aw}}{V_w + V_a K_{aw}} \qquad \text{(B-8)}$$

Finally, rearranging the terms of Equation B-6 gives:

$$\begin{aligned} J &= \left[\frac{V_w V_a K_{aw}}{V_w + V_a K_{aw}} \right] \left(\frac{C_a}{K_{aw}} - C_w \right) \\ &= V_{tot} \left(\frac{C_a}{K_{aw}} - C_w \right) \qquad \text{(B-9)} \end{aligned}$$

Henry's law constant gives the distribution, or partitioning, of a compound between air and water. From NIST (2008), Henry's law constant for radon is:

$$\begin{aligned} K_H^{\circ} &= 9.3 \times 10^{-3} \text{ (mol/kg-bar) / 0.987 (atm/bar)} \qquad \text{(B-10)} \\ &= 9.42 \times 10^{-3} \text{ (mol/kg-atm)} \end{aligned}$$

$$\begin{aligned} K_H^{CP} &= 9.42 \times 10^{-3} \text{ (mol/kg-atm)} \times 997 \text{ (kg/m}^3\text{)} \qquad \text{(B-11)} \\ &= 9.39 \times 10^{-3} \text{ (mol/L-atm)} \end{aligned}$$

The air-water partition coefficient is defined as the inverse of Henry's law constant:

$$K'_{aw} = \frac{1}{K_H^{CP}} = 106.4 \text{ (L–atm/mol)} \qquad \text{(B-12)}$$

A dimensionless form of Henry's law constant can be computed by using the ideal gas law and multiplying K_H^{CP} by the universal gas constant times temperature. Thus, to obtain a dimensionless air-water partition coefficient (K_{aw}), K'_{aw} is divided by the universal gas constant times temperature:

$$K_{aw} = \frac{K'_{aw}}{R\,T} = 4.35 \qquad \text{(B-13)}$$

Where: K_{aw} = Air-water partition coefficient (dimensionless)
K'_{aw} = Air-water partition coefficient (L-atm/mol)
R = Universal gas constant (L-atm)/(K^o-mol)
= 0.0821 (L-atm)/(K^o-mol)
T = Temperature (K^o)
= 298.15 (K^o)

The diffusion coefficients for molecular diffusion of radon are about 10^{-5} cm^2/sec in water and 0.11 cm^2/sec in air. The assumed film thicknesses are 100 microns for water and 1000 microns for air (0.01 cm and 0.1 cm). Substituting into Equation B-5 gives 10^{-3} cm/sec and 1.1 cm/sec for the transfer velocities for water (V_w) and air (V_a), respectively, and into Equation B-8 gives ~10^{-3} cm/sec for the total transfer velocity (V_{tot}). Thus, for a 1 pCi/L unit concentration of radon in the water, the flux (J) can be approximated as:

$$\begin{aligned} J &= 10^{-3}\ \text{(cm/sec)}\ 1\ \text{(pCi/L)}\ 10^{-3}\ (\text{L/cm}^3)\ 100^2\ (\text{cm}^2/\text{m}^2) \\ &= 0.01\ (\text{pCi/m}^2\text{-sec})\ \text{per (pCi/L of dissolved radon)} \end{aligned}$$