Radon Emissions from Tailings and Evaporation Ponds

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Regulatory Context

- **40 CFR 61, Subpart W NESHAP limits radon emissions from operating mill tailings (EPA 1989)**
- **Defines the Regulatory limit in Subpart W for tailings impoundments in existence as of December 15, 1989**
  - Flux of 20 pCi m^{-2} s^{-1}
- **After that date, two work practice requirements:**
  - Total area in operation* at any time 2 X 40 acres
  - Limited to 10 acres uncovered at any time

* Operated per § 192.32(a) as determined by NRC

Why Regulatory History is of Current Importance

- Per Settlement Agreement with Plaintiffs of November 2009, EPA agreed to review and potentially revise 40 CFR 61
- Subsequently, EPA has conducted series of public meetings and solicited public input
- EPA has indicated they are considering applying work practice requirements of Sub W to water ponds (e.g., conventional U mill and ISR evaporation ponds)

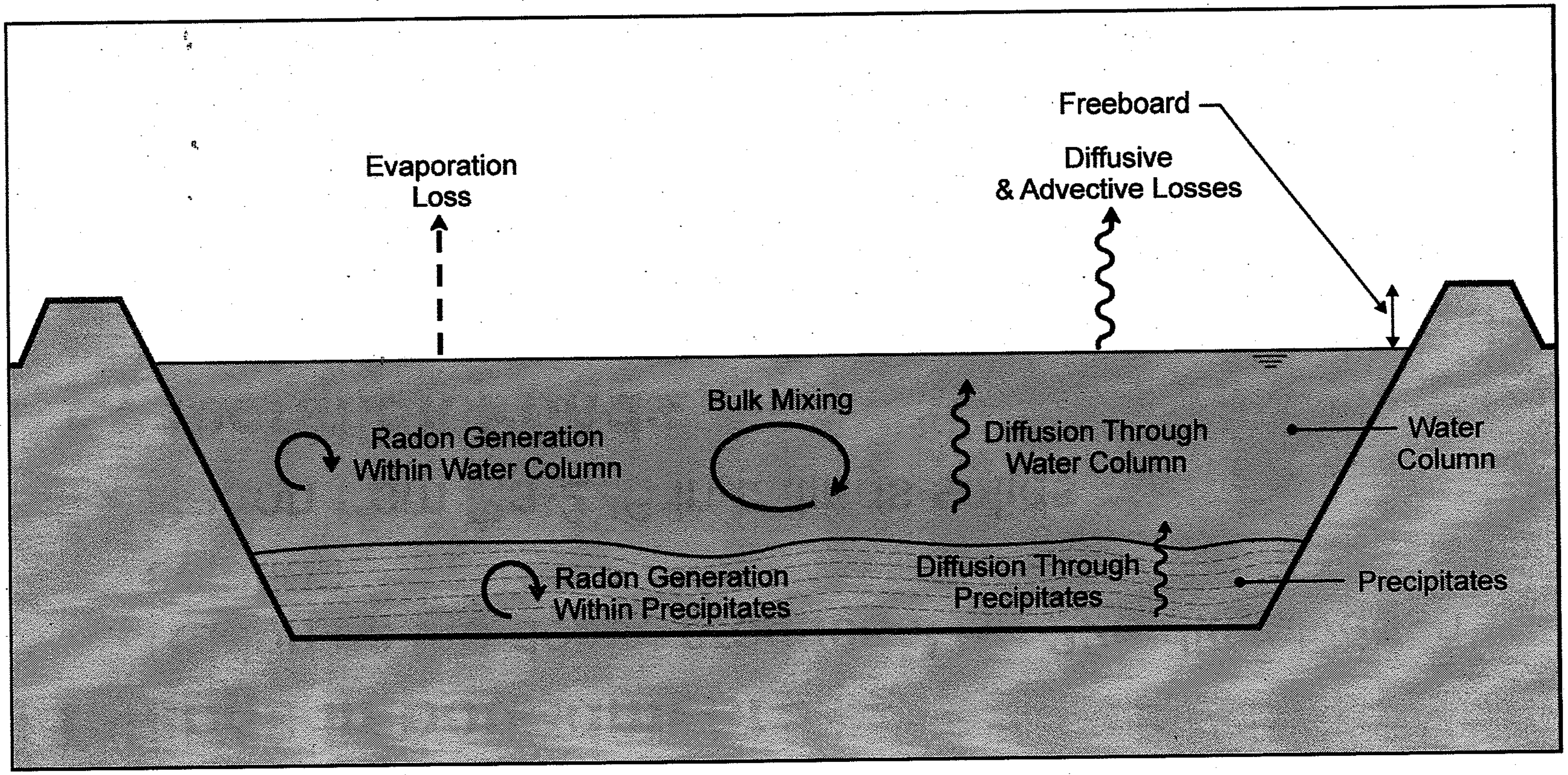
Issues and Concerns

- Direct measurement of radon flux over water surfaces via accepted methods (e.g., EPA # 115) is problematic
- Application of Work practice acreage limitations to include water ponds at new uranium recovery facilities could severely limit production and could make operation in some cases "impossible"
- The basic physics, historical and recent studies of radon emission from water surfaces suggests it is "trivial" and is the subject of this presentation

Sources of Radon

- **Tailings Impoundment**
  - Radon as the decay product of Ra-226 dissolved in solution
  - Radon from Ra-226 in tailings solids
- **Evaporation Ponds**
  - Radon as the decay product of the Ra-226 dissolved in solution
  - Radon from Ra-226 in the precipitates
- **ISRs**
  - Dissolved Radon from formation

Radon Release Mechanisms



Diffusion Length

$$L = \sqrt{\frac{D}{\lambda P}}$$

Where:

L = diffusion length

= distance to which concentration decreases by factor of e (= 2.718)

D = bulk diffusion coefficient (cm^2/s)

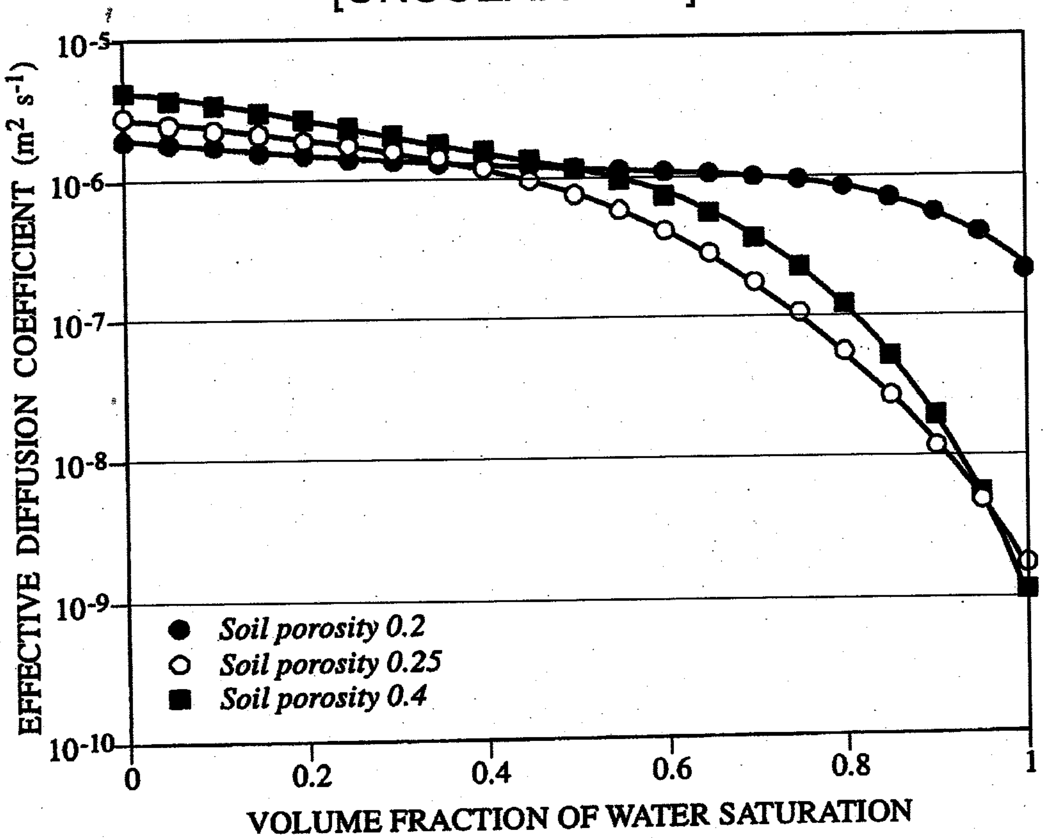
λ = radon decay constant

= 2.1 ´ 10^{-6}/s

P = porosity (void volume/total volume)

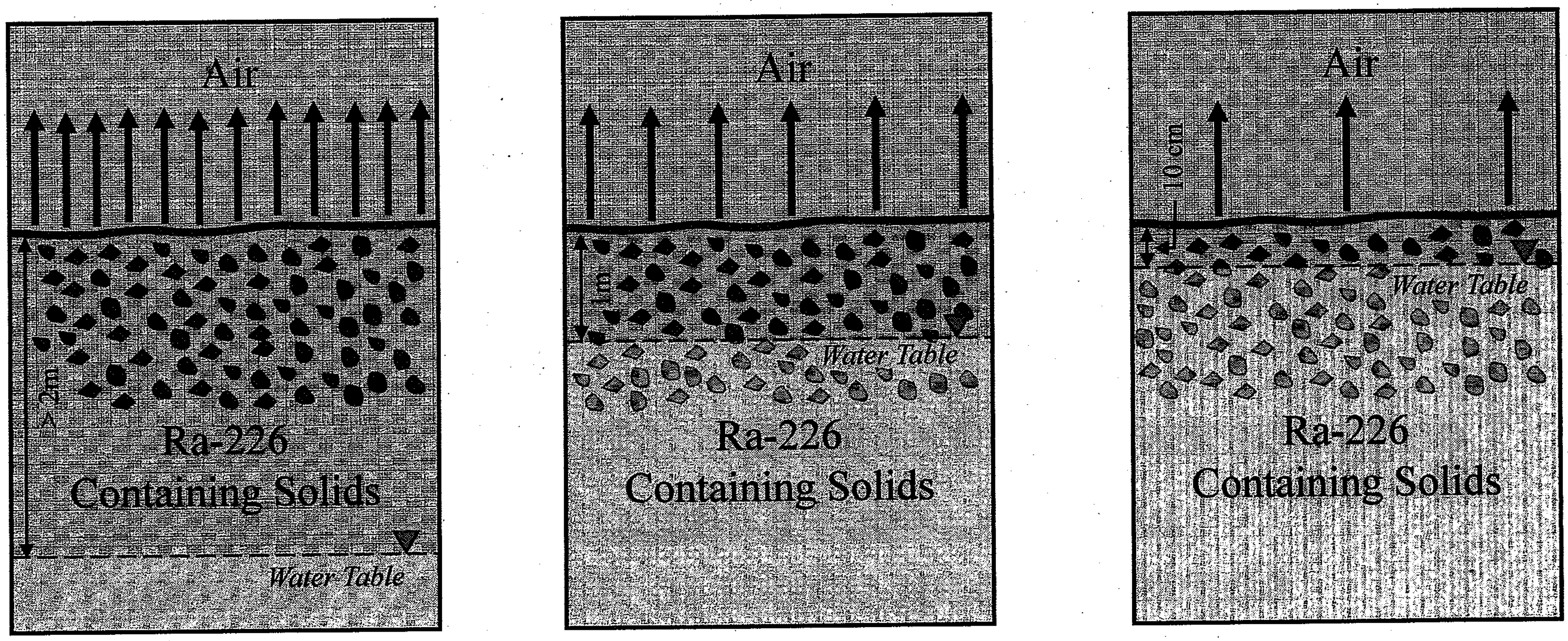
Experimental Diffusion Coefficients

[UNSCEAR 2000]



SOURCE: After UNSCEAR 2000

Effects of Water Saturation



Radon from Water Covered Tailings Solids and Precipitates

- Diffusion coefficient of radon through solids decreases with increasing water content
- Tailings solids and precipitates under water will be saturated
- For practical purposes, "zero" radon flux as radon produced within tailings solids/precipitates decays before migrating to water/solids interface

Radon Release Mechanisms

- Diffusion
- Diffusion enhanced by mechanisms such as natural convection in the water column and wind action
- Evaporation

Release via Diffusion

- **Radon in still water diffuses toward the air-water interface where it is released to the ambient air**
- **The diffusion of radon is described by Fick's Law: the flux density of the diffusing radon is linearly proportional to:**
  - its concentration gradient
  - its diffusion coefficient in water

Release via Diffusion

- **Diffusion Coefficient of Radon in Water and Air at 20°C** [Drago, 1998]

Medium	Value	Unit
water	1.2×10^{-5}	cm^2/s
air	1.4×10^{-1}	cm^2/s

- **The diffusion coefficient of radon in air is approximately 10,000 times larger than its diffusion coefficient in water**

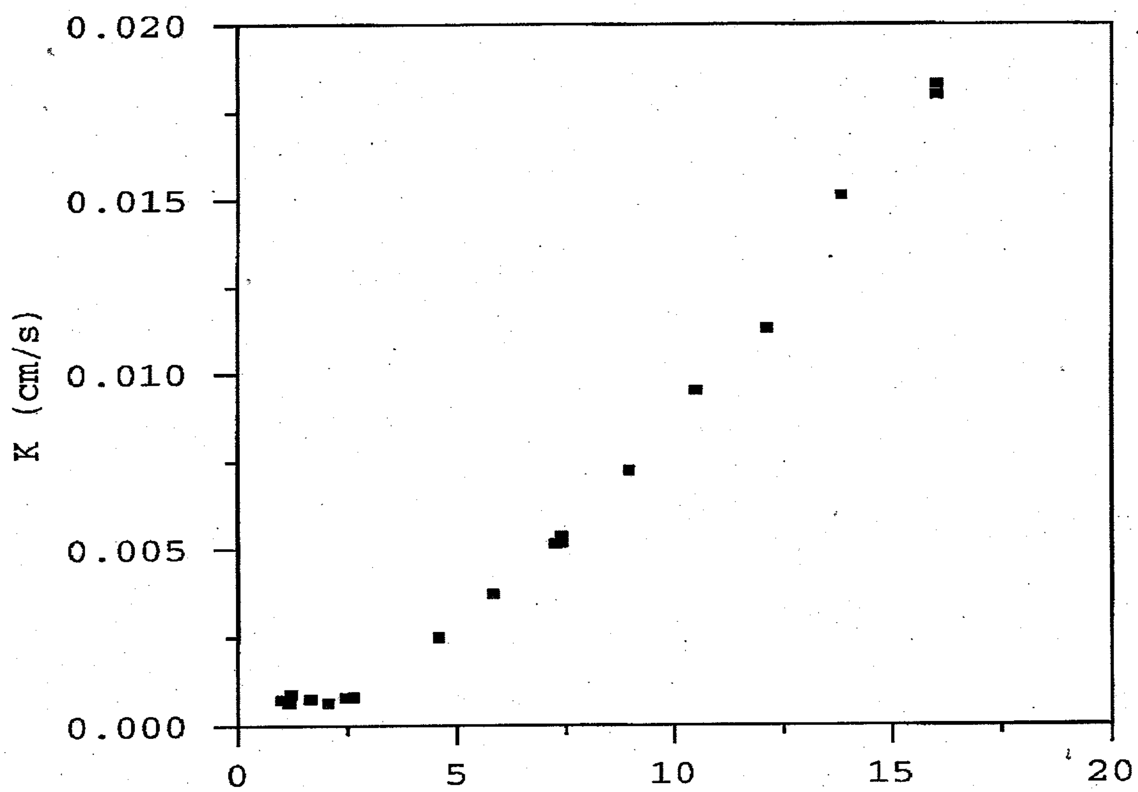
General Release Mechanism - 1

- In reality, gaseous diffusion is enhanced by various mechanisms such as natural convection in the water column and wind action
- Hence, Fick's Law is expressed as an effective diffusion coefficient
- In addition, the radon flux across the air/water interface is expressed as overall mass transfer coefficient – a compound factor of diffusion in water, air and the effect of convections in both water column (wave action) and air (wind action)

General Release Mechanism - 2

Typical Plot of Mass Transfer Coefficient K versus Wind Speed (m/s)

[Source: Saylor and Handler, 1997]



General Release Mechanism - 3

- At wind speed of 8 miles/hour (3 m/s) or less, no significant wave action expected. At these wind speeds, release rates are typically independent of wind speed (diffusion controlled)
- The wave dimensions versus wind speed depend on the geometry and size of ponds
- For typical tailings impoundments (40acres) and evaporation ponds, the maximum wave depth would be less than 1 ft for the wind speed of 23.4 miles/hour (8.1 m/s)

Radon From Wave Action

- Rn-222 is produced at the rate of 2.1 x 10^{-6}/s from Ra-226
- Wave action induced turbulence assumed to release radon at air/water interface as it is produced from Ra-226 within "turbulent" layer

Solution Ra-226 (pCi/L)	Depth of Turbulent Mixing (cm)	Rn-222 (pCi/m^2 • s)
10	10	0.002
	50	0.01
100	10	0.02
	50	0.1
1000	10	0.2
	50	1

NRC Models

- NRC uses the Rogers et. al. models (based on the Fick's Law of diffusion) in NRC 1984 handbook for uranium tailings cover design
- Nielson and Rogers work is also basis for NRC's Regulatory Guide 3.64 on radon attenuation by earthen tailings covers (NRC 1989)
- The "NRC" model considered that mixing leading to non-diffusive radon emissions could take place in the top 1 m of water cover within tailings impoundments
- Over evaporation ponds, the magnitude of the wave dimensions may not be large enough to induce complete mixing in the top 1 m of the water column

Evaporative Emission

- **A 2010 analysis by SENES (conventional mill currently under licensing review) showed that evaporative loss of ponds is extremely insignificant compared to diffusional release of radon**
  - Continuous evaporation from thin film at interface based on Henry's Law constant for radon and concentration of radon in ambient air
  - Assumes 75% radon removal efficiency [Rost, 1981] for radon produced from radium in solution and used for spray evaporation
- **The SENES calculations indicated that the total radon emission from evaporation is insignificant vs. estimated value based on Neilson and Rogers model (diffusion and wave action from the evaporation ponds).**

Radon and ISRs

- **Two mechanisms of release:**
  - Dynamic release of Rn dissolved in lixiviant when initially exposed to atmospheric pressure (small leaks in well fields, IX - elution interface, surge ponds, restoration)
  - From decay of Ra 226 dissolved in water
- **Since sources are many and diffuse, cannot measure directly**
- **Accordingly, historical approach for demonstrating compliance to, e.g. 10 CFR 40.65 semi annual effluent reporting and public dose limits is via calculations and environmental monitoring**

Estimating Radon Releases – 10 CFR 40.65 Reporting Requirements for ISRs

- NRC regulatory Guide 3.59* used to estimate Radon source terms and/or
- Results of environmental monitoring (Rn passive detectors, air particulate sampling) compared to 10 CFR 20, Appendix B, Table 2 unrestricted area concentration limits
- Offsite doses estimated via ratios of environmental monitoring results to 10 CFR 20, App. B limits X 100 mrem / yr or via RG 3.59 + MILDOS – AREA computer code (Argonne National Lab, 1997 – See Appendix D, NUREG 1569)

* RG 3.59, 1987 - *Methods for Estimating Radioactive and Toxic Airborne Source Terms for Uranium Milling Operations,* Section 2.6: Radon Release During In Situ Operations

Rn from ISRs – Example Results

- Estimates of Rn source terms via RG 3.59 and/or MILDOS – AREA (NUREG 1569) in recent applications and in 40.65 reports = several hundred to several thousand Ci/yr. *
- Rn concentrations in unrestricted areas via environmental monitoring by licensees consistently << 10 CFR 20, App. B, Table 2 limits and offsite doses consistently < 10 CFR 20.1301 public dose limits
- From NRC NUREG 1910 (ISR GEIS), Table 4.2-2: "Calculated doses are solely for radon releases**.. these sites have no yellowcake emissions since they use vacuum dryer technology.. All doses reported are well within the 10 CFR Part 20 annual radiation dose limit for the public of 1 mSv [100 mrem/yr)"

* As a frame of reference, at a typical almost anywhere, natural background radon flux of 1 - 2 pCi/m^2-sec, a square mile of earth has a " Rn source term" of 50 – 100 Ci/yr.

** Dose (TEDE) as calculated by MILDOS is actually from the radon progeny since radon, as an inert gas, is dosimetrically insignificant

Radon , ISRs and 40 CFR 61 Subpart W

- Accordingly, application of Sub W Rn emission limit and/or work practices should not be necessary for ISRs since:
  - Adequate public protection and standards of care are provided under the AEA (e.g., 10 CFR 20; 10 CFR 40 App A)
  - Operating experience consistently demonstrates unrestricted area concentrations and public dose limits are achieved

QUESTIONS ?

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