Calculation of ^{222}Rn Concentrations at Allied Chemical and Dye Corporation, North Claymont, Delaware

prepared by

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June 30, 2015

At the June 24, 2015, meeting of the Advisory Board's Subcommittee on Dose Reconstruction, a question arose regarding the ^{222}Rn concentrations at the Allied Chemical and Dye Corporation plant in North Claymont, Delaware. Allied Chemical is known to have performed a pilot study to extract uranium from phosphoric acid. In three previous documents (Behling 2014, Farver and Mauro 2014, Mauro 2015), SC&A staff presented results of dose estimates based on exposures to assumed radon concentrations of 4 pCi/L. During the June 24 meeting, the subcommittee agreed with SC&A's proposal to apply the radon model previously developed for estimating radon concentrations in Building 40 at the Blockson Chemical Company in Joliet, Illinois (SC&A 2008) to answer two questions:

1. What is the likely radon concentration in the indoor air, given the estimated production of uranium at Allied Chemical?

2. What throughput of phosphate ore would have been required to produce a radon concentration of 4 pCi/L?

1 Methodology

Radon could have been released into the building through two mechanisms:

1. Emanation from ore within the building, before, during, and after the grinding of the ore

2. Evolution from the hot sulfuric acid after the crushed ore is dissolved

Radon emanates from the solid matrix of the phosphate ore by recoil of the radon atom following the emission of an alpha particle by its parent nuclide, ^{226}Ra. According to UNSCEAR 2000: "The range of recoil distance for ^{222}Rn is 20–70 nm in common minerals." This distance is less than 0.1% of the diameter of the crushed ore particles, which is less than 74 µm. Consequently, the crushing of the ore would have little effect on the radon emanation.

In the second mechanism, radon that has remained embedded in the solid matrix of the rock would enter into the acid solution after the rock has dissolved. There are no readily available data on the rate of evolution of radon from aqueous solutions. However, since the ~30% sulfuric acid is heated and agitated to promote the dissolution of the ore, such evolution cannot be ruled out.

The rate of release of radon into the building atmosphere, as well as its removal by air exchange and radioactive decay, is given by the following differential equation:

$$\frac{dB}{dt} = A_{226\text{Ra}} R \left(t_r \varepsilon_d + \frac{f\left[\left(1 - \varepsilon_d\right)\left(1 - e^{-\lambda t_r}\right) + \left(1 - \varepsilon_w\right) e^{-\lambda t_r}\right]}{\lambda} \right) - B(r + \lambda) \qquad (1)$$

$$= 0$$

B = atoms of daughter product (^{222}Rn)

$A_{226\text{Ra}}$ = specific activity of ^{226}Ra in phosphate rock (Bq/kg)

R = processing rate of ore

$$= \frac{R_w}{t_w}$$

= 0.006159 kg/s

R_w = weekly processing rate

$$= \frac{a_{238\text{U}} M_{\text{U}}}{A_{238\text{U}} t_y}$$

= 887 kg/week

$a_{238\text{U}}$ = specific activity of ^{238}U in natural uranium (Bq/kg)

M_{U} = assumed annual production of uranium metal (kg)

= 10 lb

$A_{238\text{U}}$ = specific activity of ^{238}U in phosphate rock from Central Florida (Bq/kg)

t_w = weekly operating time (s)
= 40 h

t_r = residence time of phosphate rock in processing building (s)

ε_d = emanation coefficient of radon from dry phosphate rock

f = fraction of radon evolving from sulfuric acid

ε_w = emanation coefficient of radon from wet phosphate rock

λ = decay rate of ^{222}Rn

r = outside air exchange rate inside processing building (s^{-1})

The first term inside the large parentheses in the first line of equation 1 represents the rate at

which radon emanates from the phosphate ore during its passage through the processing building. The second term (the algebraic fraction with λ in the denominator) represents the evolution of radon after the rock is dissolved in sulfuric acid. The first term in the numerator represents the evolution of radon resulting from the decay of ^{226}Ra in the dry rock following calcining, while the second term represents the evolution of radon that was in secular equilibrium with ^{226}Ra in the wet rock prior to calcining. The final term of equation 1 represents the removal of radon due to ventilation and radioactive decay. The radioactive decay and ingrowth of radon during the calcining step is not accounted for. The ingrowth of radon during calcining would result in a slightly elevated concentration in the crystalline matrix because the partially dried rock has a lower emanation coefficient than the wet rock. However, since the duration of the calcining step is most likely short as compared to the mean radioactive life of radon, this process would not significantly affect the results of our model.

The second line of equation 1 denotes the steady-state solution to the first equation: the rate of change equals zero. This condition allows a direct solution of the equation:

$$B = \frac{A_{226\mathrm{Ra}} R \left(t_r \varepsilon_d + \frac{f\left[\left(1 - \varepsilon_d\right)\left(1 - e^{-\lambda t_r}\right) + \left(1 - \varepsilon_w\right) e^{-\lambda t_r}\right]}{\lambda} \right)}{r + \lambda} \qquad (2)$$

The activity concentration of radon in the ambient air is given by

$$C = \frac{A_{226\mathrm{Ra}} R \left(t_r \varepsilon_d \lambda + f\left[\left(1 - \varepsilon_d\right)\left(1 - e^{-\lambda t_r}\right) + \left(1 - \varepsilon_w\right) e^{-\lambda t_r}\right] \right)}{(r + \lambda) V} \qquad (3)$$

C = activity concentration of radon at Allied Chemical (Bq/m^3)

V = volume of processing building

1.1 Method of Calculating ^{222}Rn Concentration

Unlike the Monte Carlo analysis used in the Blockson model, the present calculation of the radon concentration was deterministic, and used the mean value of each parameter. The complete list of parameters is presented in Table 1.

Table 1. Parameters Used in Analysis

Symbol	Description	Value	Units
A_{226Ra}	Specific activity of ^{226}Ra in phosphate rock from Central Florida	1,460	Bq/kg
a_{238U}	Specific activity of ^{238}U in natural uranium	12.347E+06	Bq/kg
A_{238U}	Specific activity of ^{238}U in phosphate rock from Central Florida	1,263	Bq/kg
f	Evolution fraction of Rn from sulfuric acid	0.35	—
M_U	Assumed annual production of uranium metal	4.536	kg
r	Air exchange rate	6.98E-04	s^{-1}
t_r	Residence time of ore in processing plant	14,400	s
t_w	Weekly operating time	144,000	s
t_y	Annual operating time	50	weeks
V	Volume of building	20,652	m^3
ε_d	Dry emanation coefficient	0.113	—
ε_w	Wet emanation coefficient	0.299	—
λ	Decay constant of ^{222}Rn	2.0979E-6	s^{-1}

1.1.1 Fraction of Radon Evolving from Sulfuric Acid

In the absence of available data on the evolution of radon from dilute, hot sulfuric acid, we used data from Hopke (2006), who had measured the emanation of radon from water in a household shower at temperatures of 21 ºC–32 ºC. The average emanation fraction was 0.7—there appears to be little correlation with temperature. The small water droplets produced in a shower would tend to maximize the emanation. Since it is unlikely that the emanation from the acid would be higher, this value can be used as the upper bound of the distribution of the fraction of radon evolving from the acid. We found no basis for assigning a lower bound that was significantly higher than zero. In the current analysis, this fraction is therefore assigned the mean of a uniform distribution ranging from 0 to 0.7, which has a value of 0.35.

1.1.2 Air Exchange Rate in Processing Building

The most applicable data on the air exchange rate in the processing building was presented by Parker (1985), who measured the air exchange rates in two industrial buildings, using SF_6 as a tracer. One building was a machine/wood shop, with a floor area of 20,000 ft^2 (1,858 m^2). This is smaller than the total area of Building 40 at Blockson, which is estimated to be 2,459 m^2.[1]

General ventilation of the shop is through 12-ft × 25-ft bay doors, which were opened

[1] This is the area of the building's footprint, as scaled from Wynveen et al. 1983, Figure 10, and corresponds to the upper bound of the volume of the building.

periodically during the 2-h period of measurement. The shop does not have a central HVAC system—local ventilation is provided for welding or metal stripping. The measured rate was 5.5 air changes per hour.

A second building studied by Parker was a single-story warehouse of approximately 100,000 ft^2 (~9,300 m^2), over five times the size of Blockson Building 40. According to Parker:

> The warehouse is heated by overhead forced air electric heaters, but has no central HVAC system. Cooling is via several roof-mounted mechanically opened vents. The warehouse contains several 12-ft × 25-ft bay doors at either end of the building. During a working day, these doors are normally open and one or two receiving doors on the west side of the building are opened periodically.

Measured rates during a working day were 0.05 air changes per hour in the morning and 0.2 in the afternoon, which yield an average rate of 0.125 per hour.

It is reasonable to assume that the ventilation rate of a building is proportional to the area of the walls, barring any infiltration through the roof:

$$R_v = A_w \, c$$

R_v = ventilation rate (m^3/s)

A_w = area of walls (m^2)

c = proportionality constant (m/s)

For a rectangular building,

$$A_f = L \, W$$

$$= k \, W^2$$

A_f = area of floor (m^2)

L = length (m)

W = width (m)

k = ratio of length to width = $\frac{L}{W}$

and

$$A_w = 2H(L + W)$$

$$= 2H(1 + k)W$$

$$= 2H(1 + k)\sqrt{\frac{A_f}{k}}$$

H = height (m)

Combining the above equations, we obtain the following expression for the air exchange rate:

$$r = \frac{R_v}{V}$$

$$= \frac{R_v}{A_f H}$$

$$= \frac{2c(1 + k)}{\sqrt{A_f k}}$$

V = volume of building (m^3)

= A_f H

Thus, for a given shape building (i.e., *k* is constant) and similar air flows through walls, doors, and windows in units of cubic meters of air per second per unit wall area (*c* is constant), the air exchange rate, *r*, is inversely proportional to the square root of the floor area. Multiplying the air exchange rate of each of the two buildings, as cited above, by the square root of the ratio of the floor area of the building to the floor area of Building 40 yields values of 4.78 and 0.243 air changes per hour, respectively. The mean of these values was adopted as the air exchange rate in the present analysis.

The result of this calculation yields a ^{222}Rn activity concentration of 0.156 Bq/m^3, or 0.0042 pCi/L.

1.2 Throughput of Phosphate Ore corresponding to Assumed ^{222}Rn Concentration

We can use equation 3 to solve for *R*(*C*), the throughput of phosphate ore corresponding to a given ^{222}Rn concentration, *C*. Setting *C* = 148 Bq/m^3 (4 pCi/L), we obtain R = 5.85 kg/s (46,408 tons/y), assuming the same parameters as before.

2 Conclusion

Based on this deterministic calculation, using the mean values of the probabilistic distributions of input parameters in the Blockson model, and site-specific values of operating durations, we find that the assumed production of 10 lb of uranium in one year leads to a ^{222}Rn concentration that is much lower than the 4 pCi/L previously assumed by SC&A. We also find that the assumed ^{222}Rn concentration of 4 pCi/L would imply an unrealistically high throughput of phosphate ore.

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