

Measurement of ^{222}Rn Flux, ^{222}Rn Emanation, and $^{226,228}\text{Ra}$ Concentration from Injection Well Pipe Scale

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Abstract

Radon-222 flux (Bq s^{-1}) was measured from the ends of twenty sections of produced water injection tubing (pipe) containing barite scale contaminated with naturally occurring radioactive material (NORM). Exposure measurements near the pipes were as high as $300 \mu\text{R h}^{-1}$. Flux measurements were accomplished by first purging the pipes with dry nitrogen and then collecting the outflow (nitrogen and radon) on charcoal columns affixed to the end of the pipe for 66 hours. As determined in this manner, ^{222}Rn flux from the ends of the pipe ranged from 0.017 to 0.10 Bq s^{-1} (0.46 to 2.7 pCi s^{-1})

Following the radon flux measurements, pipe scale was removed and a representative sample was taken for ^{226}Ra and ^{228}Ra concentration measurements and determination of ^{222}Rn emanation fractions (the fraction of the total radon contained in a material that is released from the material and free to migrate). The samples were also analyzed for gross mineral content. Emanation fraction measurements for ^{222}Rn ranged from 0.020 to 0.063, while ^{226}Ra concentrations ranged from 15.7 to 102 Bq g^{-1} (424 to 2760 pCi g^{-1}). Barite was the predominate mineral in 17 of the 20 scale samples collected. Much of the previous work dealing with radon emanation fraction measurements has involved uranium mill tailings. Compared to mill tailings and natural soils which have emanation fractions that typically range from 0.1 to 0.3,

the emanation fractions measured for these NORM scales are substantially lower.

Introduction

Most minerals in the earth's crust contain small, but measurable concentrations of naturally occurring radioactive materials (NORM). Geochemical properties associated with petroleum formation can result in locally elevated concentrations of ^{238}U and ^{232}Th within source rocks (Snaveley 1989). Isotopes of radium and radon are key components of both the ^{238}U and ^{232}Th decay series. Under certain physical and chemical conditions, brines or formation waters present within the petroleum producing geologic formation may leach radium from clays. Although the initial production of oil and gas from a reservoir is typically dry, as the natural pressure within the petroleum bearing formation falls, the formation water may be produced along with the oil and gas (Smith 1987). Radium may be a dissolved constituent in these waters. Isotopes of radium are estimated to comprise over 90% of the total radioactivity (excluding ^{222}Rn) found in produced waters (Snaveley 1989, EPA 1991). In contrast, only trace amounts of ^{238}U , ^{232}Th , ^{228}Th , ^{210}Pb , ^{210}Po , and other NORM radionuclides are typically transported to the surface in produced water. The high proportion of radium in produced waters is due to the increased solubility of radium relative to other NORM elements.

Decreases in temperature and pressure allow solutes contained within the produced waters to precipitate as they are brought to the surface (Smith 1987). These precipitation processes can result in the formation of hard, extremely insoluble scale deposits on the interior of piping and other production equipment, eventually resulting in sufficient flow resistance to require replacement of the scaled pipe segments (e.g. Raabe 1996, Bernhardt et al. 1996). Soluble radium co-precipitates along with barium and calcium as complex sulfates or carbonates (EPA 1991). The formation of these hard scales is also related to factors such as flow rate, reservoir delivery time, and geologic structure (Bassignani et al. 1991). The trend is for the relative quantity of NORM to increase as the well ages and water production increases. Details of the

characteristics of scale formation in well pipes are discussed by Wilson and Scott (1992).

The concentrations of radium in these scales can vary from background to highly elevated levels, depending on the concentration of uranium and thorium within the geologic formation and other factors. Surveys of oil and gas wells sampled in Oklahoma, Texas and Louisiana indicated that from 50% to 75% of the facilities had produced water containing ^{226}Ra concentrations of over 50 pCi L^{-1} (Snavely 1989). The U.S. Environmental Protection Agency (EPA) has estimated that as many as one-third of all domestic oil and gas wells produce some radium contaminated scale (EPA 1991). By way of contrast, a study conducted in Italy involving 544 facilities, indicated that scale formation was small, with quantity of scale produced rarely exceeding 10 kg per plant per year (Bassignani et al. 1991). In a study involving over 36,000 external gamma measurements at American Petroleum Institute (API) member facilities in 20 states and three offshore areas, a total of 14.9% of the leases examined were found to have external gamma readings above background with a maximum reading of 3.5 mR h^{-1} (Otto 1989). As with most NORM surveys conducted to date, however, this did not provide a statistical representation of the industry because many of the wells investigated were known to be associated with elevated radioactivity.

Although the presence of elevated NORM concentrations in petroleum reservoirs and in oil and gas production and processing facilities has been recognized since the early 1930s (EPA 1991), concerns have been recently expressed regarding the potential for radiation exposure from uncontrolled releases of NORM to the public. However, large uncertainties remain in estimates of the extent and potential ramifications of NORM contamination in the petroleum industry. The focus of health concerns related to NORM associated with petroleum production involves the generation and release of chemically inert radon gas. Radioactive decay of ^{226}Ra (from the uranium decay series) results in formation of ^{222}Rn (3.82 d half life). Decay of ^{228}Ra also yields an isotope of radon (^{220}Rn), but is much shorter-lived (55 s half life) than ^{222}Rn , and therefore does

not pose as serious a problem from a transport and exposure standpoint. Radon present in pore space of the material may be transported to the surrounding atmosphere via advection or diffusion. The risks associated with the handling and disposal of NORM contaminated material are dependent on the overall rate at which Rn is released from the material matrix.

Radon emanation fraction is commonly defined as that fraction of the total radon contained in a material that is released from the material and free to migrate from the material in the gas phase. The physical properties of the radium bearing material largely determine the radon emanation fraction of the material. These physical properties include: (1) the distribution of radium within the material; (2) whether the material is massive or granular; (3) the type and magnitude of porosity of the material; (4) the moisture content of the material; and (5) the effective radon diffusion coefficient in the material. These various properties can interact in complex ways that are often counter-intuitive. However, a general understanding of the processes responsible for the release of radon atoms from a material is useful at this point. The following information regarding emanation mechanisms is presented in detail in Tanner (1980).

Special concerns have been raised regarding radon release from pipe at pipe cleaning facilities. A common method for cleaning scale accumulation in pipes is under-reaming, a process whereby a high-speed rotating device is inserted into the pipe to break-up and loosen the scale, which is then blown out of the pipe with compressed air. This process generates considerable volumes of waste which may contain NORM. Furthermore, dust generated in the process may contain ^{226}Ra and attached ^{222}Rn daughters, thereby providing pathways for exposure via inhalation and ingestion.

The Conference of Radiation Control Program Directors (CRCPD) has developed Model Regulations to cover the distribution and disposal of NORM generated by various industries (CRCPD 1988). Several states including Texas, Louisiana, and Mississippi have moved to adopt NORM regulations based to some degree on the CRCPD Model Regulations, and in several

instances, the focus of these regulations is on the oil and gas industry (Cameron 1996). Although radon is the primary concern, these regulations are generally based on activity concentrations of radium in the material. However, the relationship between radon concentration and radiation dose to humans is highly variable, depending in part on the radon emanation fraction. Because of the complex way that the physical properties of the scale materials may interact, direct measurement of the emanation fraction are needed to better estimate the exposure potential of oil field NORM.

The objectives of this investigation were (1) to characterize the radon release rates from NORM-scale contaminated production tubing being stored above ground; (2) to determine the radon emanation fraction of the bulk scale material when removed from the tubing; and (3) to quantify the radium isotope concentration in the pipe scale.

Materials and Methods

Twenty sections of 2 3/8-inch production tubing (pipe) were selected for evaluation from among the pipes showing the highest surface gamma exposure rate readings at a pipe yard in Texas. These pipe sections ranged in length from 8.92 m (29.25 ft) to 10.00 m (32.80 ft). Radon flux (Bq s^{-1}) was measured by purging the volume of each pipe with a carrier gas (commercially available dry nitrogen) thereby entraining any radon emanating from the scale. The purge gas stream was passed through an activated carbon bed to collect the radon. Following the collection interval, the activated carbon was returned to the laboratory for gamma analysis to determine the radon activity collected. The pipe sections were purged of any accumulated radon prior to the start of the sample collection interval so that the measurement would reflect the true radon release rate from the pipe. Carrier gas flow rates of approximately $2.5 \text{ liter min}^{-1}$ were employed. A sample collection interval of approximately 66 hours was used to ensure that sufficient radon was collected for an accurate gamma measurement. Sample decay times from end of sample collection to start of counting ranged from 26 to 37 hours. The activated carbon bed was fabricated from a length of 1-inch, schedule-40, PVC pipe, packed with approximately 180 g of

coconut-husk-type activated carbon. Gamma analysis of the radon-laden activated carbon samples was carried out using a 3 × 3-inch sodium iodide scintillation detector and support electronics. Equation 1, after Hartley and Freeman (1986) was used to reduce the raw count data to a radon flux in Bq s⁻¹.

$$J = \frac{C \lambda^2}{K(1 - e^{-\lambda T1})(e^{-\lambda(T2-T1)} - e^{-\lambda(T3-T1)})} \quad (1)$$

Where:

- J = measured radon flux (Bq s⁻¹)
- C = net counts for the region of interest (counts)
- λ = decay constant for ²²²Rn (s⁻¹)
- K = calibration factor for the region of interest (counts s⁻¹ Bq⁻¹)
- $T1$ = sample collection time (s)
- $T2 = \Delta t$, start of sample collection to start of counting (s)
- $T3 = \Delta t$, start of sample collection to end of counting (s)

The calibration factor K was determined from repeated analysis of two different ²²⁶Ra-on-activated-carbon standards of the same counting geometry as the samples.

A sample of the scale from each pipe was obtained for ²²⁶Ra analysis and ²²²Rn emanation fraction measurements following completion of the radon flux measurements. Pipe scale samples were obtained in a somewhat crude manner. Each pipe section was hoisted about two meters off the ground and tipped to one side. Scale samples were obtained by striking the pipe with sledge hammers to dislodge the scale, and “rattling” the scale fragments down the pipe to a collection bucket placed at the end of the pipe. This procedure was repeated by tipping the opposite end of the pipe down and collecting sample from the other end of the pipe. About 0.006 m³ (1.6 gallons) of dislodged scale was collected from each pipe. The broken-up scale was then mixed with a trowel and a one liter sample was extracted and placed in a polyethylene container. The sample collection bucket and trowel were rinsed with water and wiped clean with a paper towel between samples to avoid cross contamination.

Gamma exposure measurements were made of the each sample and ²²⁶Ra concentrations

were estimated based on calculations using the Microshied[®] code and assuming 16 percent equilibrium (1 day ingrowth) between ²²⁶Ra and daughter products. This was done to ensure Department of Transportation activity limits for shipping limited quantity radioactive material (0.05 C for ²²⁶Ra) were not exceeded. An aliquot of approximately 130 g was forwarded to the University of Texas at Austin for gross mineralogical identification.

The radon emanation fraction was measured using an aliquot of the 1 L scale sample recovered from each tubing section. The general methodology employed in these measurements was to flush all free gaseous Rn from the sample aliquot and measure the ingrowth of the emanating fraction of the total radon in the sample by gamma spectroscopy. Spectral gamma measurements of the 0.352 MeV peak from ²¹⁴Pb and the 0.609 MeV and 1.764 MeV peaks from ²¹⁴Bi were obtained at two different times during the ingrowth period. The emanation fraction was determined based on the theoretical ingrowth curve. Fig. 1 shows the theoretical ²²²Rn ingrowth curve. The sample activities at times t_1 and t_2 (I_1 and I_2) are described by Equations 2 and 3 below.

$$I_1 = I_o + N(1 - e^{-\lambda t_1}) \quad (2)$$

$$I_2 = I_o + N(1 - e^{-\lambda t_2}) \quad (3)$$

Where:

- I_1 = activity at time t_1
- I_2 = activity at time t_2
- I_o = bound radon activity at time t_o
- N = free, or emanating radon activity at radioactive equilibrium
- λ = radon decay constant (s^{-1})

These two equations can be solved simultaneously for the unknown quantities I_o and N .

Substituting A for $(I - e^{-\lambda t_1})$ and B for $(I - e^{-\lambda t_2})$ simplifies the equations. The solutions are given in Equations 4 and 5, and the emanation fraction F is computed from Equation 6.

$$N = \frac{(I_1 - I_2)}{A - B}, \quad (4)$$

$$I_o = \frac{I_2 A - I_1 B}{A - B} \quad (5)$$

$$F = \frac{N}{N + I_o} \quad (6)$$

It is not necessary to determine the actual sample activities (I_1 and I_2) at times t_1 and t_2 . The net count rates (C_1 and C_2) observed at t_1 and t_2 are proportional to the activities (I_1 and I_2) through a calibration constant, which cancels out when the final ratio is taken in Equation 6. Thus C_1 and C_2 may be substituted for I_1 and I_2 .

The radon emanation fraction of most granular materials is dependent upon the grain size (surface to volume ratio) as well as the moisture content of the sample (Colle' et al. 1981, Tanner 1980). Because of this concern, the samples prepared for radon emanation fraction measurement were not crushed or otherwise further reduced in size beyond that which occurred during the field sampling until the emanation fraction measurements were completed. It was believed that the method of obtaining the scale samples used in this investigation would produce a grain size distribution representative of scale materials removed from tubing and pipe by the "rattling" process commonly used in the past. The emanation fractions reported here are not intended to be representative of those belonging to scale materials removed by the under-reaming process, which is the process currently used at most pipe cleaning facilities.

Representative 0.4 L aliquots of the 1 L scale samples were obtained by the cone-and-quarter method. Each aliquot was placed as a thin layer in a flushing chamber constructed from a 70 cm length of 4-inch PVC pipe. A small laboratory air pump was used to draw conditioned air through the flushing chambers at a flow rate of approximately 1 L min⁻¹. Previous experience with uranium mill tailings materials indicated that using room air for flushing results in significant

drying of the sample. In order to avoid this problem, a humidifying system was employed to condition the air stream drawn into the flushing chamber to 98% relative humidity, and maintain it at the same temperature as the flushing chamber. By maintaining the air stream close to the saturation moisture content, it was anticipated that the intergranular moisture would remain constant in the sample.

Weight gains were, however, observed in all of the samples following removal from the flushing chambers. The increases were probably the result of iron oxide minerals taking up hydration waters from the saturated atmosphere. There is no question that the measurement process affected the moisture content of the samples, relative to the "as-collected" condition. It can be argued, however, that the saturated air of the measurement condition closely approximates the moisture conditions that exist in most soils.

The spectral gamma measurements were obtained with a high purity, planer germanium detector used with a personal computer based multichannel analyzer. Two spectra were collected for each sample at two different times, (t_1 , t_2) during the ingrowth period. Samples were counted for 1500 s. Net peak areas were obtained for three different regions of interest comprised of the 0.352, 0.609, and 1.764 MeV lines for each count. The sum of the net peak areas from these three regions of interest are proportional to the sample activity at times t_1 and t_2 . Substitution of these values into Equations 4, 5, and 6 yields the radon emanation fraction.

The same 0.4 L aliquot used in the emanation fraction measurements was used as the starting sample material for the ^{226}Ra and ^{228}Ra analyses. The 0.4 L aliquots were first dried at 110°C and then crushed to a sieve size of 80 mesh and thoroughly blended. A 100 gram aliquot was then extracted and placed in a metal container, sealed and allowed to sit 21 days for radon progeny ingrowth. Each sample was counted on a high purity germanium detector (HPGE) using the 0.609 MeV ^{214}Bi photopeak for ^{226}Ra and 2.614 MeV ^{208}Tl photopeak for ^{228}Ra . Samples were corrected for density differences between the sample matrix and the calibration standards.

Results and Discussion

Concentrations of ^{226}Ra in the 20 pipe scale samples ranged from 15.7 to 102 Bq g^{-1} (424 to 2760 pCi g^{-1}) with a mean of 62 Bq g^{-1} (1670 pCi g^{-1}) (Table 1). In comparison, natural soils typically have ^{226}Ra concentrations that range from 0.019 to 0.074 Bq g^{-1} (0.5 to 2.0 pCi g^{-1}). Concentrations of ^{228}Ra ranged from 47.9 to 150 Bq g^{-1} (1300 to 4050 pCi g^{-1}), with a mean of 96 Bq g^{-1} (2600 pCi g^{-1}). Based on these concentrations, disposal of these scaled pipes would be regulated in those states that have enacted NORM regulations.

Radon flux from the tubing ends (Table 1) ranged from 1.7×10^{-2} to 0.1 Bq s^{-1} (0.46 to 2.7 pCi s^{-1}) with a mean of 4.0×10^{-2} Bq s^{-1} (1.1 pCi s^{-1}) and a standard deviation of 2.3×10^{-2} Bq s^{-1} (0.62 pCi s^{-1}). By way of comparison, federal regulations limit Rn flux from uranium mill tailings to 0.74 $\text{Bq m}^2 \text{ s}^{-1}$ (20 $\text{pCi m}^2 \text{ s}^{-1}$). This suggests that risks associated with storage of above ground NORM contaminated piping are not substantially greater than a properly remediated uranium mill tailings pile. It is interesting also to compare the radon flux values measured from the tubing segments with the range of radon-flux density values observed in a variety of soils. Wilkening, et al. (1972) give a worldwide average of 1.6×10^{-2} $\text{Bq m}^{-2} \text{ s}^{-1}$ (0.43 $\text{pCi m}^{-2} \text{ s}^{-1}$) for radon-flux density from natural soils, with a range from 2.2×10^{-4} to 5.2 $\text{Bq m}^{-2} \text{ s}^{-1}$ (0.006 to 140 $\text{pCi m}^{-2} \text{ s}^{-1}$). Thus, the average section of NORM contaminated pipe from this site is comparable to approximately 2.5 m^2 of average ground surface, in terms of radon production potential.

Radon emanation fractions ranged from 0.020 to 0.063 with a mean of 0.04 and standard deviation of 0.01. These values are substantially lower than emanation fractions measured for uranium mill tailings of comparable ^{226}Ra content, which typically range from 0.1 to 0.3. Thus, health impact assessments for disposal of uranium mill tailings cannot be extrapolated to NORM scale materials because the radon emanation fraction of the two materials differs substantially.

In order for a radon atom to be released from a material in which it is contained, it must first be transported to the surface of that material. In granular materials, such as uranium mill tailings, the surface of interest is the grain surface, and as a practical matter the grain is usually bounded by a void or pore space in the bulk material. An important mechanism for the transport of radon atoms is the so-called direct recoil mechanism. When a ^{226}Ra atom decays by the emission of a 5.3 MeV alpha particle, approximately 86 keV of energy is imparted to the parent nucleus. This recoil energy drives the newly formed radon atom in the opposite direction as the alpha particle. As the radon atom travels along its recoil path, it continues to lose energy until it finally stops. The distance traveled is dependent upon the material through which it travels. For minerals with common densities this range is 20-70 nm (Tanner 1980). If the medium is water the range is approximately 0.1 μm , whereas in air the range is about 60 μm (Tanner 1980). Thus, if the parent radium atom is located substantially greater than 70 nm from the grain surface, recoil of the radon atom will not result in its escape from the grain, and it will not contribute to the emanated fraction. If, on the other hand, the original radium atom lies within the recoil range of the grain boundary, and the recoil path terminates within the pore volume, then the atom is free to migrate away as gaseous radon.

Another possibility for the recoil path is for it to cross the grain boundary, but terminate in an adjacent grain. In traveling into the adjacent grain, the atom produced an ionization damage trail. Because of the ionization damage, the radon atom is able to diffuse back out of the grain much more readily than if there were no damage. This is referred to as the indirect-recoil mechanism.

Because the surface area to volume ratio is dependent upon grain size, the size and distribution of grains within granular materials can have a large impact on the radon emanation fraction. The distribution of the radium may also play a significant role in determining the emanation fraction. For many types of uranium ores deposited in sandstone, the uranium, and

consequently the radium is located in a relatively thin coating of secondary minerals on the clastic grains. This is one reason for the relatively high emanation fractions exhibited by many uranium mill tailings.

In contrast to granular materials, NORM pipe scales are commonly deposited as a massive form of the scale minerals, typically barite. Although these scale deposits exhibit some porosity (at crystal boundaries and as fracture porosity), it is significantly lower than for a granular material such as uranium mill tailings. Additionally, the radium distribution tends to be more uniform throughout the bulk volume of the scale, which significantly reduces the fraction of parent radium atoms that are within the recoil range of pore features or surfaces of the scale material. Thus estimates of radon emanation fractions for NORM scale materials are relatively low. Nielson et al. (1988) and Baird et al. (1990) estimate approximately 5% for emanation fractions for scale materials. In contrast, estimates for the emanation fraction of NORM contaminated sludges are four to five times higher because their physical properties more closely approximate a granular material than the massive scales.

The emanation fractions presented here represent a more appropriate value to use in an assessment of NORM scale. However, these data are of limited scope in that they represent only one physical location. More measurements from a wide variety of locations and operating conditions are needed to develop distributions of emanation fractions, that are appropriate for a risk assessment of NORM contaminated scale material.

Mineralogical analyses indicated that barite (BaSO_4) was the predominate mineral in 17 of the 20 scale samples collected, and was the second most predominate mineral in the remaining three samples. Akaganeite [$\beta\text{-FeO(OH)}$] and Halite (NaCl) were the predominate minerals found in the other three samples. This is consistent with earlier reports that NORM contamination tends to be associated with barite scales where the radium ion replaces the barium ion in the crystal lattice. Other minerals found in the scale samples include the iron oxide dimorphs, Lepidocrocite

and Goethite [α -FeO(OH)]. It was not determined if the iron-bearing minerals were authigenic or corrosion products from tubing.

Conclusions

These data show that while ^{226}Ra and ^{228}Ra concentrations may be of the same magnitude as uranium mill tailings and other materials regulated for their radon release potential, the radon emanation fraction is significantly lower in scales than for typical mill tailings and soils. The radon flux rates measured from pipes containing this scale material are comparable to a few square meters of average soil. These results suggest a lower radon related risk for NORM scales than for other materials currently regulated for their radium content. The generality of these conclusions is severely limited because these results represent one production facility, from a single geologic setting, and there is no evidence to suggest that these results are typical. Clearly more measurement results are needed from a variety of locations and operating conditions if the radon related health risks of oil field NORM are to be accurately characterized.

References

Baird, R.D.; Merrell, G.B.; Klein, R.B.; Rogers, V.C.; Nielson, K.K. Management and disposal alternatives for NORM wastes in oil production and gas plant equipment. Rogers and Associates Engineering Corporation, RAE-8837/2-2; 1990.

Bassignani, A.; DiLuise, G.; Fenzi, A. Radioactive scales in oils and gas production centers. In: Proceedings of the Society of Petroleum Engineers First International Conference on Health, Safety, and Environment. The Hague, Netherlands, November 10–14; 1991.

Bernhardt, D.E.; Owen, D.H.; Rogers, V.C. Assessment of NORM in pipe from oil and gas production. In: Proceedings of the 29th Midyear Topical Meeting of the Health Physics Society. Scottsdale, Arizona January 7–10; 1996.

Cameron, F.X. The odyssey of the good ship NORM: The search for a regulator safe harbor. In: Proceedings of the 29th Midyear Topical Meeting of the Health Physics Society. Scottsdale, Arizona January 7–10; 1996.

Colle', R.; Rubin, R.J.; Knab, L.I.; Hutchinson, J.M.R. Radon transport through and exhalation from building materials: a review and assessment. Washington DC; U.S. Department of Commerce, National Bureau of Standards; NBS Technical Note 1139: 1–97; 1981.

CRCPD. Model state regulations for the control of radiation, proposed rules for Part N, Regulation and licensing of naturally occurring radioactive material; 1988.

EPA (U.S. Environmental Protection Agency). Draft diffuse NORM waste characterization and preliminary risk assessment. Air and Radiation Report ANR-460; 1991.

Hartley, J.N.; Freeman, H.D. Radon flux measurements on Gardinier and Royster phosphogypsum piles near Tampa and Mulberry, Florida. U.S. Environmental Protection

Agency Report EPA 520/5-85-029; 1986.

Nielson, K.K.; Rogers, V.C., Bollenbacher, K.K. Safety analysis for the safe disposal of naturally occurring radioactive materials in Texas. Rogers and Associates Engineering Corporation Report RAE-8818-1; 1988.

Otto, G.H. A national survey on naturally occurring radioactive materials (NORM) in petroleum producing and gas processing facilities. Report to the American Petroleum Institute: 1-265; 1989.

Raabe, O.G. Studies of the solubility of naturally-occurring radionuclides in petroleum pipe scale. In: Proceedings of the 29th Midyear Topical Meeting of the Health Physics Society. Scottsdale, Arizona January 7-10; 1996.

Smith, A.L. Radioactive scale formation. Journal of Petroleum Technology 39:697-706; 1987.

Snaveley, E.S. Radionuclides in produce water - a literature review. American Petroleum Institute Publication No. 5404: 1-85; 1989.

Tanner, A.B. Radon migration in the ground: A supplementary review, In: T.F. Gesell and W.M. Lowder (eds.), Natural radiation environment III, Volume 1. U.S. Department of Energy Report CONF-780422: 5-56; 1980.

Wilkening, M.H.; Clements, W.E.; Stanley, D. Radon-222 flux measurements in widely separated

regions, In: J.A.S. Adams, W.M. Lowder, and T.F. Gesell (eds.) The natural radiation environment II. U.S. Energy Research and Development Administration Report CONF-720805-P2: 717-730; 1972.

White, G.J. Naturally-occurring radioactive materials (NORM) in oil and gas industry and equipment and wastes. U.S. Department of Energy, Bartlesville Project Office, Bartlesville Oklahoma. DOE/ID/01570-T158; 1992.

Wilson, A.J.; Scott, L.M. Characterization of radioactive petroleum piping scale and evaluation of subsequent land contamination. Health Phys. 63:681-685; 1992.

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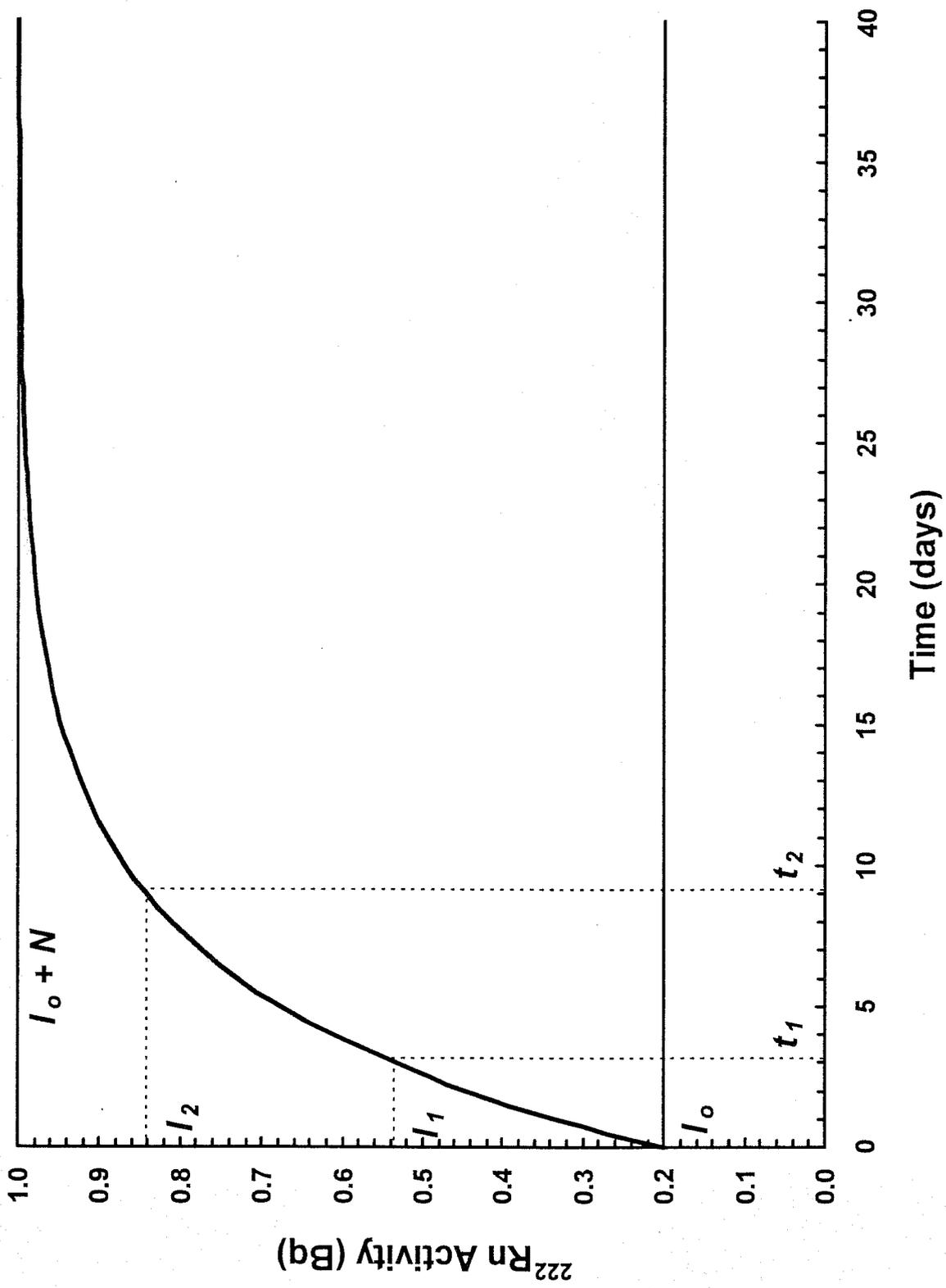


Figure Captions:

Fig. 1. Radon-222 ingrowth curve for a 1 Bq ^{226}Ra source and a 0.2 Bq initial ^{222}Rn activity (I_0). Radon activity at times t_1 and t_2 are the activities, I_1 and I_2 shown in the figure. The ^{222}Rn emanation fraction is the ratio of N (the total activity at equilibrium minus the initial radon activity in the sample) to $N + I_0$.

Table 1. Measurement results for ^{226}Ra and ^{228}Ra concentrations, ^{222}Rn flux from pipes, and ^{222}Rn emanation fractions. The weight gain ratio relates the weight of the sample before and after the radon emanation measurement.

Average

Pipe No.	scale thickness (cm)	Radon flux (Bq s ⁻¹)	Radon emanation fraction	Weight gain ratio	²²⁶ Ra (Bq g ⁻¹)	²²⁸ Ra (Bq g ⁻¹)	Loss on drying (%)
1	0.25	0.030	0.063	1.04	33.1	55.8	6.7
2	0.49	0.061	0.040	1.03	79.6	109	10.7
3	0.21	0.017	0.027	1.03	73.2	111	5.5
4	0.23	0.030	0.026	1.03	71.8	103	5.3
5	1.08	0.104	0.030	1.00	79.8	111	20.7
6	0.54	0.038	0.054	1.03	14.7	19.0	9.0
7	0.53	0.033	0.052	1.04	15.7	19.8	9.4
8	0.20	0.040	0.030	1.03	102	150	4.9
9	0.26	0.019	0.032	1.03	38.6	60.0	6.2
10	0.24	0.039	0.020	1.03	85.9	134	5.0
11	0.28	0.089	0.039	1.04	82.3	139	5.8
12	0.23	0.034	0.027	1.03	97.3	149.4	4.1
13	0.28	0.024	0.037	1.04	26.5	48.6	7.3
14	0.28	0.034	0.031	1.04	86.5	133	5.5
15	0.33	0.073	0.038	1.04	102	150	5.2
16	0.33	0.025	0.035	1.04	26.2	47.9	8.8
17	0.25	0.038	0.030	1.03	93.6	150	4.2
18	0.32	0.020	0.034	1.04	24.3	50.5	6.5
19	0.26	0.033	0.033	1.03	83.5	124	4.6
20	0.25	0.024	0.057	1.04	21.9	55.6	6.5

Footnotes

* Microshield Version 4.0 Grove Engineering Inc. 15215 Shady Grove Rd., Suite 202 Rockville, MD 20850