Radon Emanation from Disposal of Depleted Uranium at Clive, Utah

1. Introduction, Purpose, and Scope

Large quantities of Depleted Uranium (DU) have been generated in the process of enriching uranium for nuclear reactor fuels and weapons in the US and throughout the world. Like natural uranium, the DU is mostly comprised of ^{238}U, but contains less ^{234}U and ^{235}U than natural uranium. While some DU has been used in fabrication of counterbalances and armor-piercing bullets, much is to be disposed as waste. Several million pounds of DU have been shipped from US Department of Energy operations at the Savannah River Site and other facilities to the EnergySolutions facility in Utah for disposal.

EnergySolutions is licensed to dispose of wastes at its Clive, Utah facility that contain radioactivity ranging from ~ 15 pCi/g ^{226}Ra in slightly contaminated soils to higher-activity wastes on the order of thousands of pCi/g. The ^{226}Ra wastes generate radon (^{222}Rn) gas, which is currently regulated by the US Federal Government to a residual release rate (flux) of ≤20 pCi/m^{2}s after final disposal. Currently, radon release from uranium mill tailings is commonly controlled by earthen covers of sufficient thickness and low diffusivity to delay the escape of the radon, causing most of it to decay within the cover before reaching the top of the cover. The 3.8-day half-life of ^{222}Rn allows a 128-fold reduction in radon flux with a 7-half-life transit time (~27 days) and more than 1,000-fold reduction with a 10-half-life transit time (~38 days).

DU presents unique radon-related regulatory issues because of its unusual properties. The DU does not presently emit significant quantities of radon, and will not do so for tens- to hundreds-of thousands of years into the future. However, calculations project that the very slow decay of DU will generate decay products, including ^{226}Ra, which will increase to a maximum level in about 500,000 to one million years. Radiological equilibrium, governed by the half-lives of parent and product radionuclides, maintains radon activities equal to those of ^{226}Ra (e.g., 1 µCi of ^{222}Rn will be in equilibrium with 1 µCi of ^{226}Ra at any given time, ignoring the fraction of ^{222}Rn that diffuses away from its parent ^{226}Ra). Therefore, the rate of radon generation could increase to levels that would require much thicker radon-barrier covers (or deeper disposal) in tens-of-thousands of years than present regulations require for controlling radon from materials such as uranium mill tailings.

This white paper describes the nature of the radon source term for DU wastes in terms of their two primary controlling factors:

> (1) the ^{226}Ra concentration; and

> (2) the radon Emanation Fraction.

These two factors control the local time-distribution of ^{226}Ra concentrations from the initial concentrations of ^{238}U and ^{234}U and the decay sequence and half-lives of their decay products. This paper then estimates the range of radon Emanation Fractions that could occur for the ^{226}Ra that grows into equilibrium with the DU. Finally, the potential radon source term is estimated as the product of the radium concentration and the emanation fraction to assess its significance for radon control and local concentrations. Although the time intervals projected for significant radon releases occur long after the 1,000-year design period required by present radon regulations, EnergySolutions is exploring methods to assess the safety of DU disposal and methods to render the disposal of DU safe over extended time periods.

2. Radium Concentrations in DU

The quantity of radon gas generated in a uranium-bearing material is the product of the parent ^{226}Ra concentration (R) and the radon emanation fraction (E, the fraction of ^{222}Rn that is emanated or released from solids into the immediate interstitial pore space). This product (R x E) is often called the radon source term, and occurs in generalized radon source equations[1] as well as the simple one-dimensional equation for radon flux (J) from an exposed ore or tailings mass:

$$J = 10^4 R\rho E \sqrt{\lambda D} \tanh\left(x\sqrt{\frac{\lambda}{D}} \right)$$
(1)

where J = radon flux (pCi/m^{2}s);

10^4 = units factor (pCi/cm^{2}s to pCi/m^{2}s);

R = ^{226}Ra concentration (pCi/g);

ρ = bulk density of uranium-bearing material (g/cm^3);

E = ^{222}Rn emanation fraction (dimensionless);

λ = ^{222}Rn decay constant (2.1 x 10^{-6} s^{-1});

D = diffusion coefficient for ^{222}Rn gas in the uranium-material interstitial pore space (cm^2/s);

tanh ( ) = hyperbolic tangent function; and

x = thickness of the radon-barrier cover material above the uranium-bearing material (cm).

The flux of radon released from buried radon-emanating material is attenuated by radioactive decay of radon as it slowly diffuses through the cover soil as well as decay of radon contained in the radon source material. Since the amount of decay increases with the thickness of the cover soil, radon-barrier covers are commonly designed with sufficient thickness and composition to reduce the radon flux to 20 pCi/m^{2}s (i.e., the current design criterion for uranium tailings under 40 CFR 192.32 and related regulations).

Radon source terms for uranium ore, DU, and other radon-generating materials use similar definitions. In the case of initially pure DU, however, the time period when significant radon may be released to the biosphere is far into the future because pure uranium metal of any isotopic composition initially contains virtually no ^{226}Ra that generates radon. Analyses of 22 samples[2] of the DU waste awaiting disposal at EnergySolutions show initial ^{226}Ra levels averaging 0.9 ± 0.3 pCi/g (mean ± s.d.) in 11 of the samples and non-detects (i.e., levels < 0.6 pCi/g) in the other 11 samples. These concentrations are typical of natural ^{226}Ra impurities in soils and many other materials, and do not appear to be related to ^{238}U or ^{234}U decay. The decay sequence by which ^{226}Ra grows into DU is illustrated in Figure 1. The rate of ^{226}Ra in-growth, as well as the rate of in-growth of the other ^{238}U decay products, is governed by their half-lives, which are also shown in Figure 1. Detectable levels of radon are generated only after significant quantities of radioactive daughters of ^{238}U, i.e., ^{230}Th and ^{226}Ra, are formed by ^{238}U and ^{234}U decay.

The refining of uranium from its natural ore removes thorium, radium, and other non-uranium decay products along with the other mineral masses in the original ore. After separation, the uranium decay products begin to "grow" back into the initially-pure uranium at rates governed by their half-lives. Because of the great disparity among half-lives, the rates of decay-product in-growth vary widely. Nevertheless, their in-growth follows well-known and established laws of nuclear physics and can be

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[1]   Multiphase Radon Generation and Transport in Porous Materials, V.C. Rogers & K.K. Nielson, *Health Physics* 60, 807-815, 1991.

[2]   GEL Laboratory report to Mark Ledoux, EnergySolutions, dated Nov 3, 2010.

calculated for any future time (after uranium refinement) using the classical Bateman equations.[3] The in-growth rates are dominated by the yellow-shaded, long-lived radionuclides ^{234}U (240,000 years), ^{230}Th (75,000 years), ^{226}Ra (1,600 years), and ^{210}Pb (22 years).
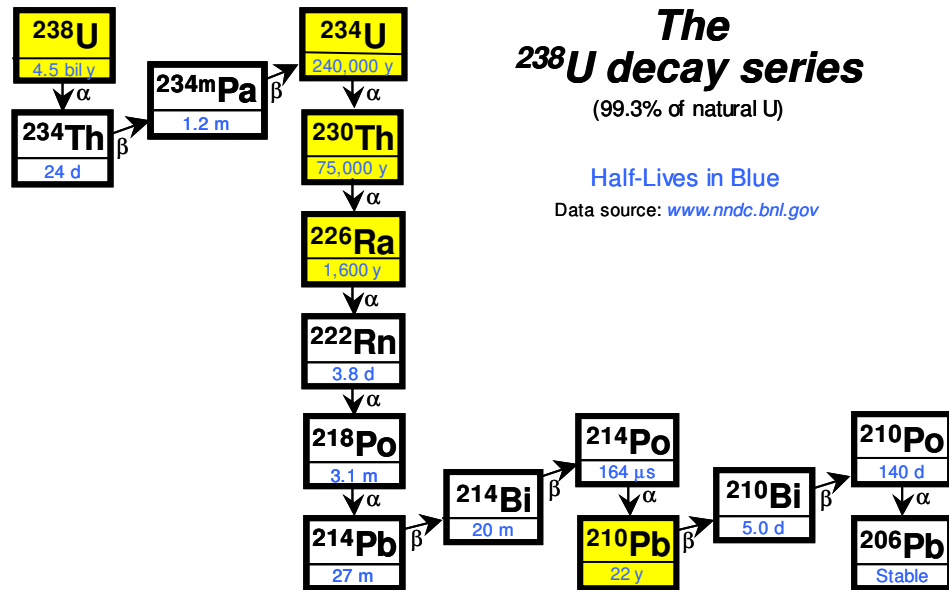
Figure 1. ^{238}U Decay Sequence, Dominated by Long-Lived Radionuclides (shaded yellow).

Figure 2 illustrates the calculated concentrations of ^{238}U and its long-lived decay products that would occur from the first year to 100 billion years after hypothetically-pure ^{238}U is isolated as the starting material. The concentrations are expressed in units of pCi of the radionuclide per gram of uranium. The blue ^{226}Ra curve is mostly overlaid by the brown ^{210}Pb curve as their equilibrium is attained, and they overlay the other three curves at later times as they come into equilibrium with their parent nuclides. Finally, the decrease after about 10 billion years is controlled by ^{238}U decay. The original chemical separation of uranium from its ore included both ^{238}U and ^{234}U, however, so the levels in Figure 2 show only part of the in-growth that would be expected for pure natural uranium.

Figure 3 illustrates the in-growth of long-lived decay products over the same time scale that would result if only ^{234}U were initially present at the same activity as the ^{238}U. In this case the ^{234}U concentration drops in the 100,000 year to few-million-year time frame because it decays and is not supported in this case by new ^{234}U formed from ^{238}U decay. As in Figure 2, the ^{230}Th, ^{226}Ra, and ^{210}Pb again grow from zero to peak together, this time after only about 200,000 years instead of the million-year time frame for the maximum from ^{238}U. Once again, the curves are overlaid by the ^{210}Pb curve as equilibrium is attained with the other radionuclides.

The observation that DU becomes more radioactive with time is simply the fact that DU is approaching its original equilibrium distribution of uranium decay products that existed before the decay products of the natural uranium ore were removed during processing.

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[3]   Principles of Radioisotope Methodology, G.D. Chase and J.S. Rabinowitz, Minneapolis: Burgess Publishing Co., 3rd edition, 1967, p. 177.
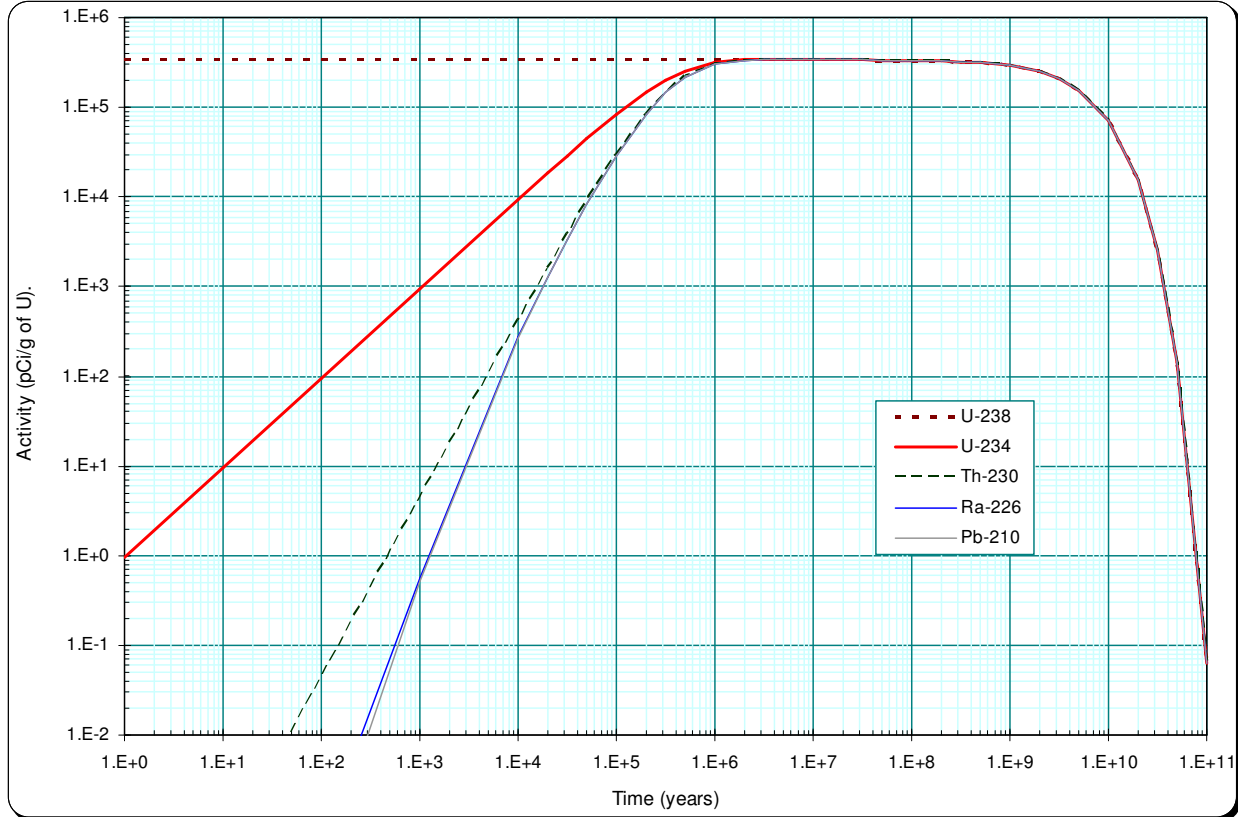
Figure 2. In-growth of ²³⁸U Decay Products from Pure ²³⁸U.
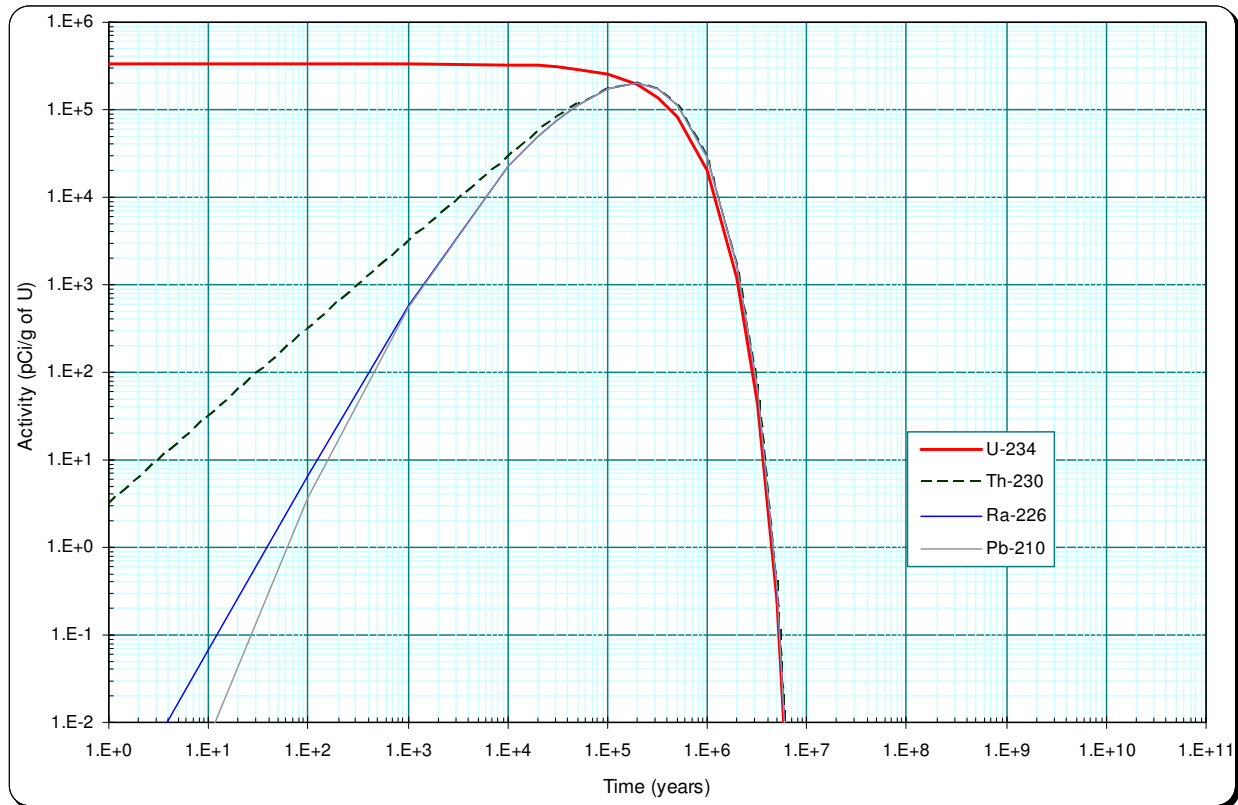
Figure 3. In-growth of ²³⁴U Decay Products from Pure ²³⁴U.

The decay-product in-growth curves expected for natural uranium, which has nearly equal starting activities of ^{234}U and ^{238}U, are approximately represented by the sum of the curves in Figures 2 and 3. In this case, the earlier time for uranium refinement (referenced for Fig. 2) and the time of the isotopic separation of ^{234}U (referenced for Fig. 3) are considered approximately equivalent. The equivalence is justified because any years elapsed between uranium refinement and its isotopic separation (to "deplete" the ^{234}U level) are negligible compared to the tens-of-thousands to millions of years required for ^{226}Ra in-growth.

The combined in-growth curves representing natural uranium (i.e., where ^{234}U and ^{238}U activities are initially equal) are illustrated in Figure 4. In this case and subsequent estimates, the concentrations are expressed as pCi per gram of uranium oxide instead of pure uranium because the chemical form of the DU to be disposed is uranium oxide. The initial DU oxide shipped from the Savannah River Site consisted of DUO_3 (83.2% U), while most future DU oxide to be de-converted from DUF_6 will consist of DU_3O_8 (84.8% U). Because of their similar U contents, all DU oxides are hereafter referred to as DUO_x for purposes of estimating uranium masses. The maximum concentrations of ^{234}U and ^{238}U are therefore slightly lower (2.8×10^5 pCi/g UO_x instead of 3.4×10^5 pCi/g U). In Figure 4 the ^{238}U curve is completely overlaid by the ^{234}U curve up to about 300,000 years, after which the ^{210}Pb curve overlays all of the curves. The ^{226}Ra curve starts (time equal to 1 year) at 7×10^{-4} pCi/g and requires about 124 years to reach 10 pCi/g and 5,380 years to reach 10,000 pCi/g.
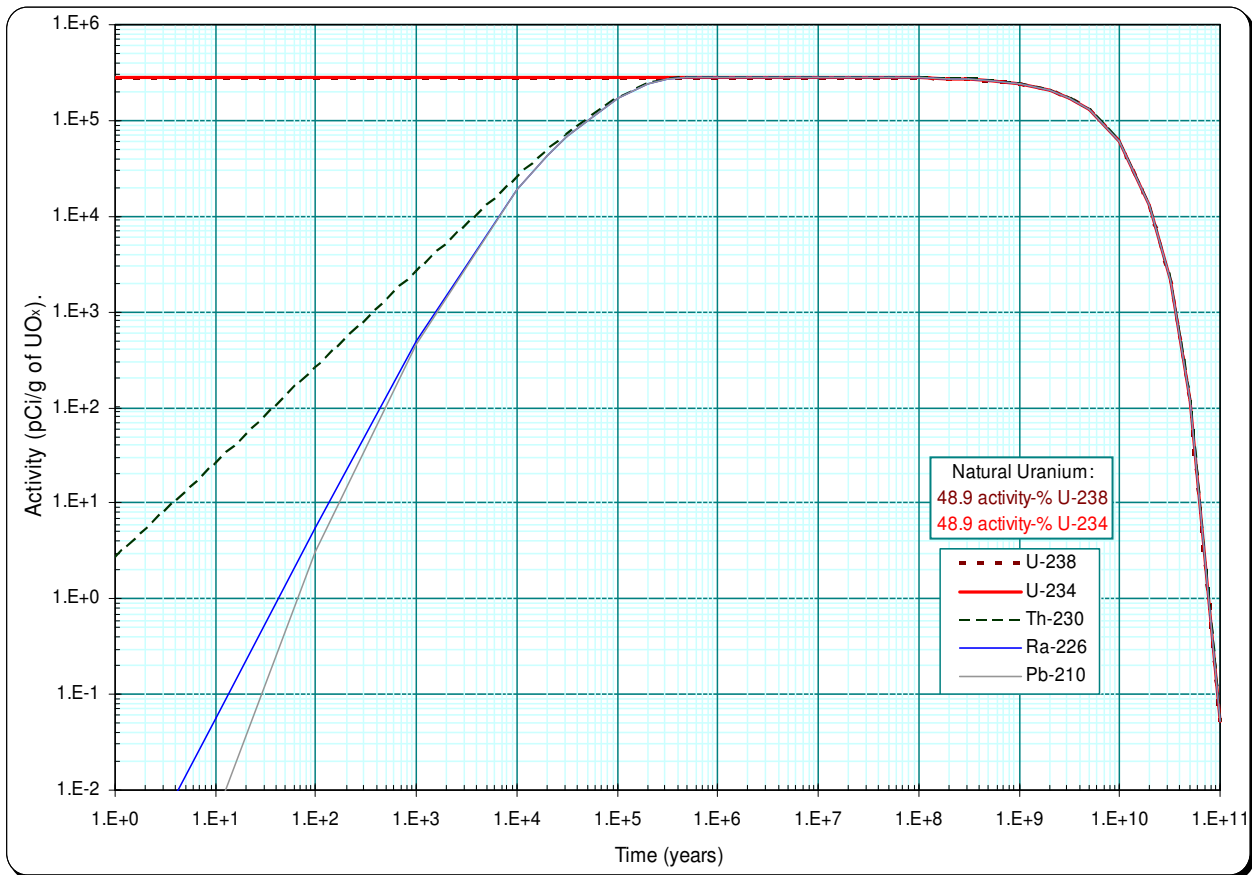

Figure 4. In-growth of Uranium Decay Products from Pure Natural UO$_x$.

3. Characteristics of Depleted Uranium

Data were reviewed for a shipment 52 containers of DUO_3 sent for disposal in December 2009 from the DOE Savannah River Site to the EnergySolutions facility in Clive, Utah.[4] The shipment consisted of 3.58×10^6 kg of DUO_3 that occupied a total container volume of 1,133 m^3. The nominal bulk density of this shipment of DUO_3 was thus about 3.16 g/cm^3. This density is consistent with the range of $1.5 - 4.0$ g/cm^3 estimated for DUO_x in a DOE assessment of preferred DU disposal forms.[5]

The broad range of density values in the DOE assessment stems at least in part from the "fine particulate or powder" nature of the DUO_x in the shipment, which gives it similar compaction properties to many soils that attain increased density as a result of higher compaction forces and optimum moisture conditions. Besides increasing the disposal cost, the increased disposal volume associated with less-than-maximum DUO_x density raises a question of potential future settlement of the powdered waste, leaving void spaces in the disposal containers that could allow future subsidence of the disposal cell and possible damage to the radon barrier cover. One solution to the settlement issue is to apply grout within and around the disposal containers, potentially mixing the DUO_x powder with selected grout to mechanically stabilize the DUO_x waste.

The porosity of DUO_x waste is conservatively estimated using the specific gravity of U_3O_8 (CAS # 1344-59-8) of 8.3 g/cm^3,[6] to obtain a porosity of 0.62, calculated as follows:

$$P = \left(1 - \frac{\rho}{\rho_{sg}}\right) = \left(1 - \frac{3.16}{8.3}\right) = 0.62 .$$

A similar estimate of the waste porosity based on the specific gravity for DUO_3 (7.29 g/cm^3) is 0.57, while that based on the specific gravity of DUO_2 (10.96 g/cm^3) would be 0.71. No data are found for the relative compaction properties of U_3O_8, UO_3, and UO_2. Laboratory analyses of 33 samples of the DUO_3 material shipped to EnergySolutions averaged 90.0 ± 2.0 % (mean ± s.d.) of the total uranium activity from ^{238}U and 8.0 ± 1.8 % of the total uranium activity from ^{234}U. These activity percentages correspond to activity concentrations of 2.8×10^5 pCi ^{238}U per g DUO_x and 2.5×10^4 pCi ^{234}U per g DUO_3. Using these activity concentrations to normalize the decay product in-growth curves from Figures 2 and 3 gives the set of decay product in-growth curves for DUO_x illustrated in Figure 5. The initial ^{226}Ra concentration at 0.9 pCi/g and its initial ^{210}Pb in-growth are based on the residual ^{226}Ra found in the 11 DUO_x samples.

The in-growth curves for the Savannah River Site DUO_3 material in Figure 5 show that the lower initial ^{234}U concentration remains steady for about 2,000 years and then gradually increases as the original ^{234}U decay is slightly over-compensated by in-growth of new ^{234}U produced by ^{238}U decay. The ^{234}U increases by only 29% above its original concentration by 10,000 years, but eventually reaches 95% of the ^{238}U concentration after a million years and is virtually equal to the ^{238}U concentration in two million years.

The ^{230}Th gradually grows in from ^{234}U decay to 9.3 pCi/g in the first hundred years and reaches 93 pCi/g after 1,000 years. The ^{226}Ra concentration begins at 0.9 pCi/g, its initial contaminant level in the DUO_x, and remains steady for nearly a hundred years until significant new ^{226}Ra is generated by the increasing ^{230}Th levels. After about 100 years, ^{226}Ra is dominated by new in-growth. The ^{210}Pb curve shows in-growth from the initial ^{226}Ra level plus DU-related in-growth at later times.

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[4] "100727_SRS_DU_Shipment_Detail.xls" file obtained Nov 3, 2010, from Wayne Johns, Environmental Manager, EnergySolutions, Salt Lake City, UT.

[5] A.G. Croff, J.R. Hightower, D.W. Lee, G.E. Michaels, N.L. Ranek, and J.R. Trabalka, Assessment of Preferred Depleted Uranium Disposal Forms, Oak Ridge National Laboratory report *ORNL/TM-2000/161*, June 2000.

[6] www.ibilabs.com/U3O8.htm
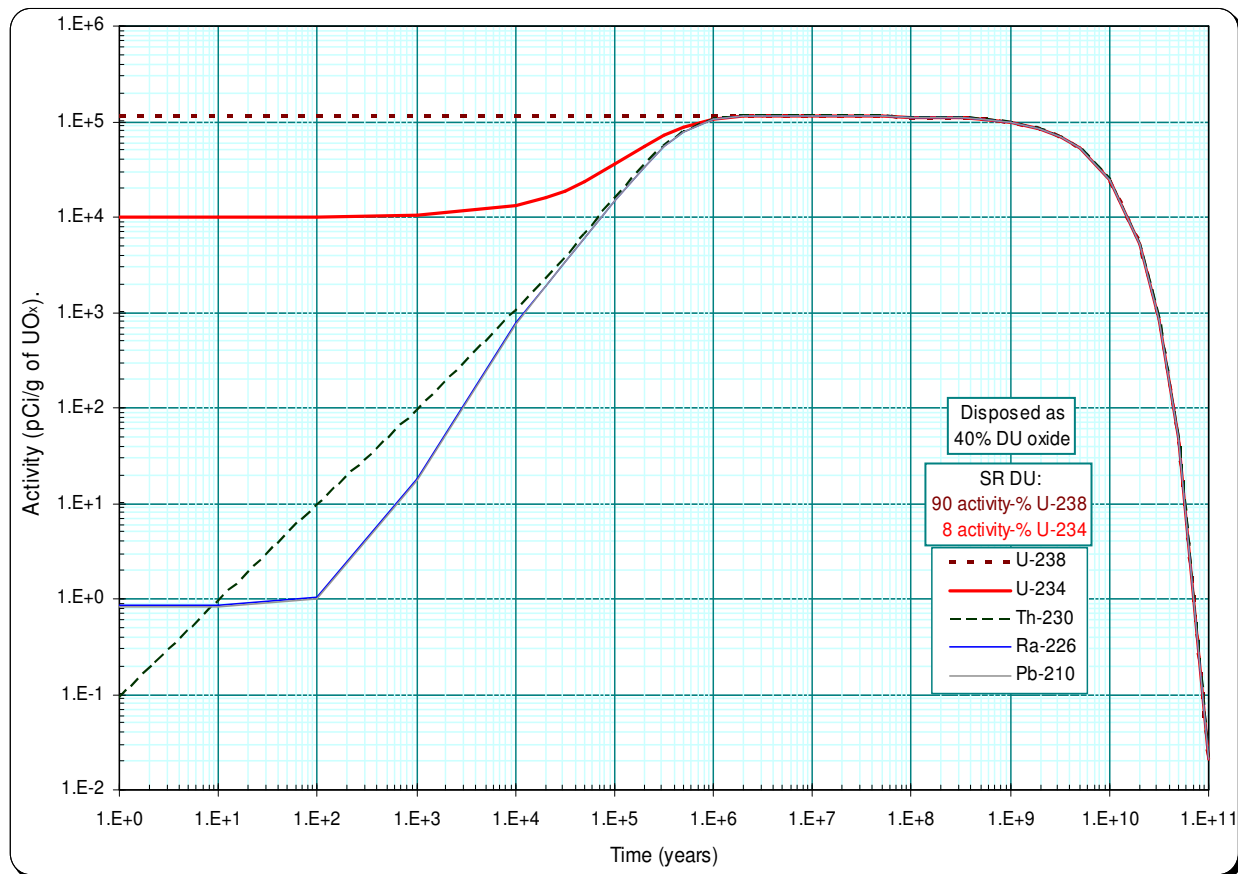
Figure 5. In-growth of DU Decay Products from DUO$_x$ Material from the Savannah River Site.

The ^{226}Ra level increases to 5 pCi/g by 500 years, by which time the initial 0.9 pCi/g of ^{226}Ra is far surpassed by new ^{226}Ra in-growth from ^{230}Th. The ^{226}Ra level reaches 18 pCi/g after 1,000 years, which is the design lifetime presently specified for radon control for uranium mill tailings. The ^{226}Ra level reaches 770 pCi/g at 10,000 years, a level that corresponds to a 0.28% UO$_x$ ore grade (a moderate grade compared to low-grade ores in the 0.01% to 0.25% range and high-grade ore up to 23% mined in Canada). The ^{226}Ra level increases to 15,000 pCi/g at 100,000 years, which is equivalent to 5% UO$_x$, well into the high-grade range for uranium ores. At this time, ^{226}Ra is within 97% of equilibrium with its ^{230}Th parent, but still 2.4 times below its ^{234}U parent and 7.7 times below its ^{238}U parent activity. These concentrations are all maximum values, assuming no dilution by less-concentrated wastes or grout stabilizers, negligible mass added by the disposal containers, and no interstitial space between containers that would be filled by stabilizing fill materials.

The data on the DUO$_3$ received at EnergySolutions indicates a lower ^{234}U/^{238}U activity ratio of 0.09 (uranium is more depleted) than is referenced in an EPA technical brief on DU,[7] which indicates a nominal ^{234}U/^{238}U activity ratio for DU of about 0.19. The higher initial ^{234}U concentration in the DU described by EPA corresponds to intermediate-level initial ^{226}Ra concentrations between those in Figure 5 and those for natural uranium in Figure 4.

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[7] Depleted Uranium Technical Brief, U.S. Environmental Protection Agency, *EPA 402-R-06-011*, 2006.

4. Radon Emanation Fractions

Radon Emanation fractions (also called emanation coefficients, emanating power, escape-to-production ratio, and in percentage units, percent emanation) are defined as the fraction of radon generated in a material that can escape to the pore space of the uranium bearing material and potentially move out of the material. Radon emanation fractions are important here because they represent the fraction of the ^{226}Ra activity concentrations identified in Section 3 that produces the gaseous ^{222}Rn effluents from DU or its oxides. No data were available on emanation fractions for DU or DUO$_x$, primarily because of the extremely long time periods before sufficient ^{222}Rn can be generated for measurement and the very low emanation fractions for DUO$_x$ at ambient temperatures.[8]

Several selected radon emanation measurements give insight into the magnitude of emanation that could be expected from DU in its oxide form. Two independent studies report ambient-temperature emanation fractions for uraninite (UO$_2$) of 0.0053 in one[9] and 0.12 to 0.15 in the other.[10] The large difference in emanation between the two studies was ascribed to differences in the ages of the minerals and in the internal properties (pore distributions) of the samples. Despite the 25-fold difference in the sets of results, both sets are lower than most emanation fractions of common uranium bearing soils and ores, and are probably representative of the more uniform distribution of ^{226}Ra in the uranium oxides than occurs in soils and ores. Although uraninite is a less-oxidized form of uranium than the DUO$_x$ targeted here, it is the only uranium oxide for which radon emanation data were found, and it appears likely to have a relatively uniform ^{226}Ra distribution to approximate that which could grow into DUO$_x$ if the DUO$_x$ could be maintained in its initial oxide waste form without leaching and re-distribution on other mineral surfaces.

The ranges and differences in emanation fractions are best described on the atomic level. When a ^{226}Ra atom undergoes radioactive decay, it emits an energetic (~ 4.8 million electron volt [MeV])[11] alpha particle in one direction and the resulting radon (^{222}Rn) atom recoils in the opposite direction with approximately equal momentum. The energy of the recoil radon atom can propel it about 63 μm in air, 0.1 μm in water, and 0.02 – 0.07 μm in minerals.[12] Depending on the location and trajectory of the recoil radon atom, it can be either emanated to the pore space or retained in the solid mineral phase in several ways that are illustrated in Figure 6.

The recoil radon atom is *emanated to the pore space* when it originates within its recoil range (0.02 to 0.07 μm) from the solid surface (dashed brown line, Fig. 6), is directed toward that surface, and dissipates its residual recoil kinetic energy as follows:

  a. in pore-space air;
  b. in pore-space water;
  c. in a solid material; or
  d. a combination of a), b), and/or c).

The recoil radon atom is *retained in the solid phase* when it:

  e. originates beyond its recoil range from the solid surface;

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[8] J.C. Clayton and S. Aronson, The Emanation of Radon-220 from Sintered UO$_2$ Powders and Plates. Pittsburgh: Bettis Atomic Power Laboratory, *WAPD-276*, 1963.

[9] E. Garver and M. Baskaran, Effects of Heating on the Emanation Rates of Radon-222 from a Suite of Natural Minerals, *Applied Radiation and Isotopes* 61, 1477-1485, 2004.

[10] L. Morawska and C.R. Phillips, Dependence of the Radon Emanation Coefficient of Radium Distribution and Internal Structure of the Material. *Geochim. Cosmochim. Acta.* 57, 1783-1797, 1993.

[11] National Nuclear Data Center, www.nndc.bnl.gov.

[12] A.B. Tanner, Radon Migration in the Ground, A Review, in The Natural Radiation Environment, Adams & Lowder, eds., Univ. of Chicago Press, 1964.

f. is directed away from the solid surface;

g. enters directly into another solid surface; or

h. enters a solid surface with sufficient energy to remain embedded.
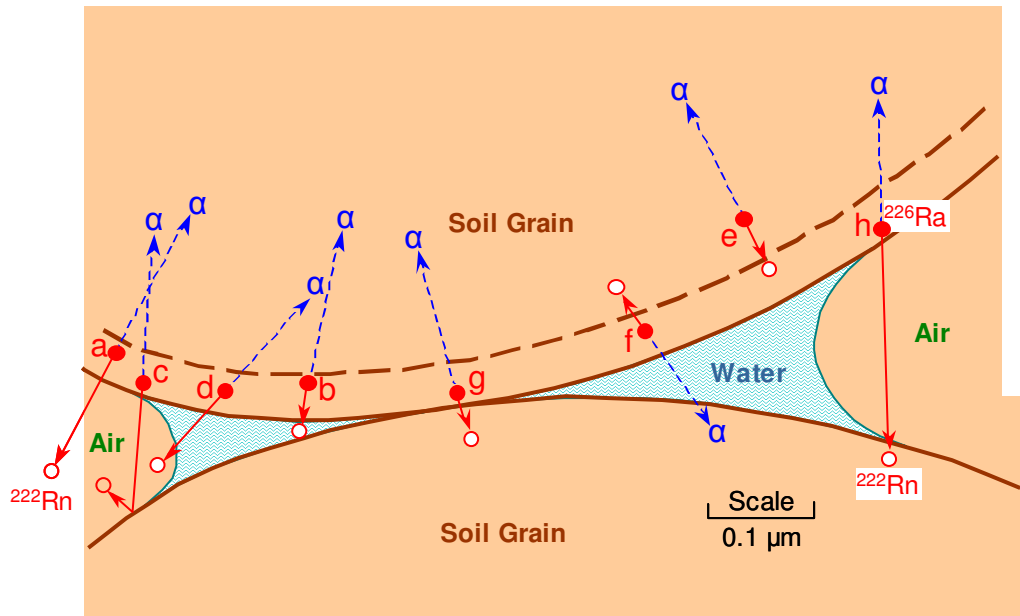
Figure 6. Recoil Radon Atom Emanation and Retention Pathways. [adapted from Tanner[12]]

Moisture in the pore volume can greatly increase emanation fractions by converting most of the Fig.6(h) retention events into Fig.6(b) or Fig.6(d) emanation events. The (h) retention mechanism shown in Fig. 6 involves capture in a mineral pocket that is melted or even vaporized by the kinetic energy of the recoil radon atom. If the impact pocket is not sufficiently deep, the radon is emanated by indirect recoil, Fig.6(c). Once the radon atom is in the contiguous air- or liquid-filled pore space, it is considered emanated because it can then diffuse out of the material before decaying (3.8-day half-life).

Early calculations by Flugge and Zimens,[13] Quet,[14] and others show that emanation fractions should be near zero for most earthen materials because they have grain sizes much greater than several microns and therefore most of their radon corresponds to the trajectory in Fig.6(e) (i.e., too far from the mineral surface to be emanated). Although very low emanation fractions (near 0.01) are typical of lava fields,[15] coal flyash particles,[16] and certain other vitrified materials, most other earthen materials have higher emanation fractions that far exceed the level explained by recoil from uniform material.

For example, western US soils, uranium ores, and uranium tailings emanate about 0.10 – 0.4 of their radon.[17, 18] and this emanation can range from 0.01 to 0.9 in extreme cases.[19] A study of 950 earthen

[13] S. Fluegge and K.E. Zimens, The Determination of Grain Size and Diffusion Constant by the Emanating Power. The Theory of the Emanation Method. *Z. Phys. Chem*. B42, 179-220, 1939.

[14] C. Quet, J. Rousseau-Violet, and P. Bussiere, Recoil Emanating Power of Isolated Particles of Finely Divided Solids, *Radiochem. Radioanal. Lett.* 9, 9-18, 1972.

[15] M.H. Wilkening, Radon-222 from the Island of Hawaii: Deep soils are more important than lava fields or volcanoes, *Science* 183, 413, 1974.

[16] D.R. Kalkwarf, P.O. Jackson, and J.C. Kutt, Emanation Coefficients for Rn in Sized Coal Fly Ash, *Health Phys*. 48, 429-436, 1985.

[17] B.J. Thamer, K.K. Nielson, and K.M. Felthauser, The Effects of Moisture on Radon Emanation, U.S. Bureau of Mines, *OFR-184-82*, 1982.

materials from the western US gave a similar extreme range of 0.02 to 0.95; however the median emanation coefficient of the 950 materials was 0.22, [20] typical of many consolidated ores and rocks. A study of 296 weathered, surface soils throughout Florida also showed a wide emanation range of 0.01 to 0.85, but had a higher median emanation coefficient of 0.39.[21]

Two primary explanations are given for emanation fractions that greatly exceed the low values that would otherwise occur from uniform ^{226}Ra distributions in mineral grains:

1. The ^{226}Ra distributions are non-uniform, distributed on mineral surfaces in organic matter, clay, iron oxide, and accessory minerals in interstitial material between low-radium mineral grains; and

2. Many mineral grains have extensive networks of micro-pores, originating from millions to billions of years of radioactive uranium and thorium decay, and enlarged by weathering and leaching fluids that enter the pores.

Either of these explanations can explain observed emanation phenomena, and both are likely to occur simultaneously in at least some situations.[22] The general incompatibility of uranium with the crystal structures of many major rock-forming minerals contributes to the non-uniformity in explanation 1 above.

Typical radon emanation phenomena include the following:

- Moderate emanation fractions occur from materials at ambient in-situ moistures, but 2 to 3-times lower emanation fractions when the materials are oven-dried (~105 °C).[23]
  Explanation: pore moisture absorbs recoil-radon energy, increasing the probability of emanation into moisture-filled pore space.

- Radon emanation fractions tend to be higher in high-porosity uranium ores (e.g., sandstones and clayey deposits) than in hard-rock crystalline ores of very low porosity.
  Explanation: More ^{226}Ra is within recoil-radon-range proximity of contiguous pore space in interstitial uranium minerals of sedimentary ores than in uniform, un-weathered hard-rock crystalline ores.

- Moderate to high emanation coefficients from ^{226}Ra adsorbed onto clay, iron or manganese oxide coatings, organic matter, and other common mineral surface materials, documented by selective leaching studies.
  Explanation: recoil radon atoms have a lower probability of retention in mineral grains when they originate in lower-density, interstitial pore constituents that are commonly mingled with pore moisture.

- Heating of moderate- to high-emanation materials to about 400 °C to 900 °C irreversibly reduces radon emanation fractions.

[18] K.K. Nielson, V.C. Rogers, M.L. Mauch, J.N. Hartley and H.D. Freeman, Radon Emanation Characteristics of Uranium Mill Tailings, *Uranium Mill Tailings Management - V*, Ft. Collins: Colo. St. Univ., p. 355-367 (1982).

[19] S.R. Austin and R.F. Droullard, Radon Emanation from Domestic Uranium Ores Determined by Modifications of the Closed-Can, Gamma-Only Assay Method, U.S. Bureau of Mines, *RI-8264*, 1978.

[20] P.M.C. Barretto, Radon-222 Emanation Characteristics of Rocks and Minerals, p. 129-150 in *Radon in Uranium Mining*, Vienna, IAEA, 1975.

[21] K.K. Nielson, R.B. Holt, and V.C. Rogers, Statewide Mapping of Florida Soil Radon Potentials, US Environmental Protection Agency, *EPA-600/R-95-142a,b*, 1995.

[22] A.B. Tanner, Radon Migration in the Ground: A Review, p. 161-190 in *The Natural Radiation Environment*, J.A.S. Adams and W.M. Lowder, eds., Univ. of Chicago Press, 1964.

[23] See reference in footnote 17

Explanation: many minerals with large internal micro-pore networks are annealed by heating, effectively reducing the proximity of much of the ^{226}Ra from contiguous pore space. In addition, certain surface minerals with adsorbed ^{226}Ra may fuse with mineral grains, reducing their potential for recoil radon emanation trajectories.

- Radon emanation is near unity (100 % emanation) from aqueous ^{226}Ra. Aqueous radium solutions are commonly used in "bubblers" to deliver known quantities of radon in air bubbled through the solution to collect the dissolved, emanated radon gas.

  Explanation: All radon is emanated because there are no immobilizing solid surfaces to retain the radon.

If disposed DUO_x were to remain unchanged in its powdered oxide form, its emanation could reasonably be represented by the emanation data for uraninite ($E \approx 0.12$). However, given the probable need for grouting or other mixing with backfill materials to stabilize the waste material and the long time before ^{226}Ra in-growth, the DUO_x is likely to be leached and re-deposited on surrounding materials. The processes associated with leaching and re-deposition would likely leave the uranium in the form of surface deposits (secondary minerals) having the chemical form of U_3O_8, which is the most stable form of uranium and the form most commonly found in nature.[24]

Surficial, secondary-mineral deposits of DUO_x are the common form of most natural uranium ores. This suggests representing any DU materials disposed in Utah by emanation fractions similar to those from local or western regional US uranium ores. Table 1 lists moist-material radon emanation coefficients (escape/production ratios) measured in uranium ores from Utah, Colorado, Wyoming, and New Mexico.[25] The Utah ores emanate about 15% of their radon, while Colorado ores emanate about 22% and the median emanation for the 16 western US ores is approximately 25%. The geologic ages of the ores range from about 40 million to 2.5 billion years, similar to the time frame for maximum ^{226}Ra in-growth.

Emanation fractions for pure DU (metal) could be very low ($< \sim 0.001$) because of the uniform distribution of ^{226}Ra that would grow into the metal mass. If a technology were identified to assure that DU remains in a metallic state without leaching and transport over the millions to billions of year timeframes in which ^{226}Ra accumulates, very small emanation fractions could be representative.

Table 1. Radon Emanation Fractions Measured in Western US Uranium Ores.

Origin	Sample	Emanation	± 1sd	n	Mean	Median
Utah	UB	0.130	±.030	18	0.148	0.148
Utah	UL	0.166	±.015	12		
Colorado	CD	0.250	±.020	20		
Colorado	CK	0.189	±.012	21	0.219	0.219
Colorado	CS	0.219	±.007	11		
Wyoming	WG-1	0.095	±.019	20		
Wyoming	WG-2	0.240	±.030	19		
Wyoming	WG-3	0.112	±.019	20	0.271	0.201
Wyoming	WP-1	0.550	±.020	19		
Wyoming	WP-2	0.470	±.020	20		
Wyoming	WC	0.161	±.170	14		
New Mexico	NM-S23-1	0.530	±.020	4		
New Mexico	NM-S23-2	0.370	±.020	13		
New Mexico	NM-S23-3	0.260	±.010	10	0.412	0.440
New Mexico	NM-P10-1	0.460	±.020	17		
New Mexico	NM-P10-2	0.440	±.020	8		
				All	0.29	0.25

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[24] The Encyclopedia of Earth, December 6, 2009, http://www.eoearth.org/article/Uranium.

[25] See reference in footnote 17.

5. Radon Source Term for DUO_x

The long-term projection of a radon source term for disposed DUO_x is highly uncertain due to the uncertainty in projecting terrestrial site conditions over million- to billion-year time scales. However, three scenarios are considered here to have at least some credibility based on the geologic ages of known uranium-bearing materials. These are summarized in Table 2 in terms of the possible ^{238}U and ^{226}Ra concentrations, radon emanation fractions, and the radon source strengths, estimated in terms of the emanating ^{226}Ra concentration (the product of ^{226}Ra concentration and emanation fraction) and the worst-case radon flux from a bare surface of exposed waste at its maximum ^{226}Ra concentration. The radon fluxes can be compared with the present regulatory limit of 20 pCi/m^2s for radon flux from disposed uranium mill tailings.

Table 2. Estimates of Radon Source Strength

Scenario	Likelihood	Maximum Radon Period [a] (years)	Max ^{238}U (pCi/g)	Max ^{226}Ra (pCi/g)	Radon Emanation Fraction	Emanating ^{226}Ra (pCi/g) [b]	Bare-waste Radon Flux (pCi/m²s) [c]
Waste stays in-place as pure DU	Negligible [d]	80,000 – 5 billion	300,000	300,000	0.001 – 0.12	300 – 36,000	980 – 120,000
Grouted waste is minimally dispersed (K_d = 21,000 cm³/g); resembles U ore	Low	300,000 – 60 million	110,000	110,000	0.25	28,000	89,000
Grouted waste is moderately dispersed (K_d = 70 cm³/g); resembles U ore	Moderate	90,000 – 2 million	110,000	19,000	0.25	4,700	15,000
Grouted waste is widely dispersed (K_d = 6 cm³/g); resembles U ore	Moderate	30,000 – 1.6 million	110,000	2,400	0.25	600	2,000

[a]   Years into the future over which radon levels are within 50 % of their maximum level.

[b]   Product of the ^{226}Ra concentration and the radon emanation fraction.

[c]   Calculated for maximum value using equation (1) assuming ρ = 1.6 g/cm³, λ = 2.1 x 10^{-6} s^{-1}, D = 0.02 cm²/s, and x = 300 cm.

[d]   Unlikely to be disposed in metallic form; to be fully protected from leaching and dispersion; and to keep low emanation fractions despite alpha particle damage and helium accumulation.

An additional scenario (disposal as DU metal ingots) was also considered and is listed first in Table 2. However, this scenario was considered as unlikely because of its highly speculative nature (assigning emanation fractions of 0.001 to 0.12 and the difficulty and uncertainties in keeping the DU intact in a metallic form, with low emanation, over extremely long time frames). The emanation fractions are somewhat conservative because they include the range for uraninite rather than metallic uranium. However, even if uranium could be protected from leaching and dispersion for extremely long periods, alpha-particle damage could sufficiently increase the gas diffusivity of the metal to increase its radon emanation, as it apparently does in crystalline minerals and ores. Furthermore, the potential for helium accumulation (from the uranium, thorium, radium, radon, and polonium alpha particles) could also compromise the integrity of the metal ingots, promoting higher gaseous releases.

The second, third and fourth scenarios in Table 2 are all plausible in that they incorporate some interstitial material with the disposed DUO_x; they allow for its leaching and dispersion; and they utilize radon emanation fractions typical of uranium ores. The interstitial material could be grout mixed with the DUO_x to achieve a stable waste form and/or backfill between drums of DUO_x. The quantity of interstitial material is estimated to reduce the DUO_x concentration to 40% of the as-disposed waste mass, consistent with current NRC estimates.[26] The leaching and dispersion of the disposed DUO_x is calculated using the K_d leaching model from the RESRAD code,[27] and the radon emanation fractions were averaged from western US uranium ores.[28] Two of the adsorption (K_d) values for uranium leaching and mobility were taken from Esh,[29] who referenced Sheppard and Thibault[30] for maximum, geometric mean, and minimum K_ds for uranium as 21,000 cm^3/g, 70 cm^3/g, and 2 cm^3/g respectively. The first two of these values were used here, and the last was replaced by a similar, site-specific measured $K_d = 6$ cm^3/g.

Given the broad regions of the western US that contain significant near-surface uranium mineralization (mined from open pits), it is possible that many masses of uranium, similar in quantity to those addressed here, could have existed millions of years ago and been dispersed, leading to the present-day near-surface ore deposits. These have not necessarily caused excessive human risks or exposures, since much of their impact, as well as that of the subject DUO_x mass, depends on land use, life style, and proximity of populations to the mineralized regions.

[26]   D. Esh, Site-Specific Performance Assessment and NRC Depleted Uranium Technical Analysis Overview, Part II: Depleted Uraniuim and NRC Analyses (SECY-08-0147), Public Workshop on Unique Waste Streams including Depleted Uranium, September 2009.

[27]   C. Yu, A.J. Zielen, J.-J. Cheng, D.J. LePoire, E. Gnanapragasam, S. Kamboj, J. Amish, A. Wallo, W.A. Williams, and H. Peterson, User's Manual for RESRAD Version 6, Argonne National Laboratory, *ANL/EAD-4*, July 2001.

[28]   See reference in footnote 17.

[29]   D. Esh, Site-Specific Performance Assessment and NRC Depleted Uranium Technical Analysis Overview, Part I: Performance Assessment and Low-Level Waste Analyses, Public Workshop on Unique Waste Streams including Depleted Uranium, September 2009.

[30]   M.I. Sheppard and D.H. Thibault, Default Soil Solid/Liquid Partition Coefficients, K_ds, for Four Major Soil Types: A Compendium, *Health Physics* 59, 471-482, 1990.