Measuring and Modeling Naturally Occurring Radioactive Material: Interpreting the Relationship Between the Natural Radionuclides Present - 8516

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ABSTRACT

The regulatory release of sites and facilities (property) for restricted or unrestricted use has evolved beyond prescribed levels to model-derived dose and risk based limits. Dose models for deriving corresponding soil and structure radionuclide concentration guidelines are necessarily simplified representations of complex processes. A conceptual site model is often developed to present a reasonable and somewhat conservative representation of the physical and chemical properties of the impacted material. Dose modeling software is then used to estimate resulting dose and/or radionuclide specific acceptance criteria (activity concentrations). When the source term includes any or all of the uranium, thorium or actinium natural decay series radionuclides the interpretation of the relationship between the individual radionuclides of the series is critical to a technically correct and complete assessment of risk and/or derivation of radionuclide specific acceptance criteria. Unlike man-made radionuclides, modeling and measuring naturally occurring radioactive material (NORM) and technologically enhanced NORM (TENORM) source terms involves the interpretation of the relationship between the radionuclide present, e.g., secular equilibrium, enrichment, depletion or transient equilibrium.

Isotopes of uranium, radium, and thorium occur in all three natural decay series. Each of the three series also produces a radon gas isotope as one of its progeny. In nature, the radionuclides in the three natural decay series are in a state that is approaching or has achieved secular equilibrium, in which the activities of all radionuclides within each series are nearly equal. However, ores containing the three natural decay series may begin in approximate secular equilibrium, but after processing, equilibrium may be broken and certain elements (and the radioactive isotopes of that element) may be concentrated or removed. Where the original ore may have contained one long chain of natural decay series radionuclides, the resulting TENORM source term may contain several smaller decay chains, each headed by a different longer lived member of the original series. This paper presents the anatomy of common TENORM source terms and the pitfalls of measuring, interpreting and modeling these source terms. Modeling TENORM with common software such as RESRAD is discussed.

INTRODUCTION

The decommissioning process used by the NRC and EPA to evaluate, remediate, and ultimately remove a site from regulatory oversight has flexibility brought about by dose-based acceptance criteria [1, 2, 3, 4, 5, 6, 7 and 8]. The process establishes a radioactivity concentration ‘clean’ criterion based on site specific parameters and the anticipated as-left condition of the site. Remedial decisions are subsequently made based upon the comparison of site characterization data to the cleanup criterion. When the remedial action ends with the residual concentration below the established cleanup criterion, a final status survey is performed and submitted to regulators to demonstrate compliance. The final status surveys designed using the guidance of MARSSIM, Multi-Agency Radiation Survey and Site Investigation Manual, are used to demonstrate compliance with the derived acceptance criteria [9].

Dose assessments are routinely performed to establish derived concentration guideline values (DCGLs) equal to the applicable dose acceptance criteria for the site, for example, the 0.25 mSv (25 mrem) total effective dose equivalent (TEDE) per year for 1,000 years (the 10CFR20 Subpart E radiological criteria for unrestricted use). A dose assessment (pathway analysis) for deriving soil concentration guidelines from a dose limit has four parts:

(1) source analysis – developing the source term,
(2) environmental transport analysis,
(3) dose/exposure analysis, and
(4) scenario analysis.

Source analysis is the part of the dose assessment for which assessments of naturally occurring radioactive material (decay series of radionuclides) differ from assessments of man-made radionuclides. Source analysis addresses the problem of deriving the source term that determine the rate at which residual radioactivity is released into the environment. This rate is determined by several key factors including (a) the geometry of the contaminated zone, (b) the concentrations of the radionuclides present, (c) the in-growth and decay rates of the radionuclides, and (d) the removal rate by erosion and leaching. The “in-growth and decay rates of the radionuclides” factor is applicable primarily to the natural decay series.

Likewise, when an assessment of exposure rate and/or shielding is performed, the development of the appropriate source term is also complicated if naturally occurring radioactive material is present.

Naturally Occurring Radioactive Material

All radioactive material can be classified as either man-made or naturally occurring radioactive materials (NORM). Man-made radionuclides are produced by man, for example, by splitting atoms in nuclear reactors or by bombarding atoms with subatomic particles in accelerators. Common man-made radionuclides include cobalt-60 (Co-60), strontium-90 (Sr-90), and cesium-137 (Cs-137). NORM include primordial radionuclides that are naturally present in the rocks and minerals of the earth's crust and cosmogenic radionuclides produced by interactions of cosmic nucleons with target atoms in the atmosphere and in the earth. NORM consists primarily of material containing potassium-40 (K-40) and radionuclides belonging to the primordial series [10 and 11]. The principal primordial radionuclides are isotopes of heavy elements belonging to the radioactive series headed by the three long-lived parents uranium-238 (U-238) start of the uranium natural decay series, uranium-235 (U-235) start of the actinium natural decay series, and thorium-232 (Th-232) start of the thorium natural decay series. All three of these series have numerous progeny radionuclides in their decay chains, including a radon gas isotope, before reaching a stable end point, a lead isotope.

As stated previously, in nature, the radionuclides in these three series are approximately in a state of secular equilibrium, in which the activities of all radionuclides (parent and progeny) within each series are nearly equal. Two conditions are necessary for secular equilibrium. First, the parent radionuclide must have a half-life much longer than that of any other radionuclide in the series. Second, a sufficiently long period of time must have elapsed, for example ten half-lives of the decay product having the longest half-life, to allow for in-growth of the decay products. For natural decay series in secular equilibrium, the activity of the parent radionuclide undergoes no appreciable changes during many half-lives of its decay products.

Each of the decay series consists of several alpha and beta emitting radionuclides of various half-lives. Some of the members also emit gamma during radioactive decay. The members of each of the three

natural decay series are presented in Table 1 with their half life and mode of decay (alpha or beta emission).

Table I – The Natural Decay Series

Uranium			Thorium			Actinium		
Nuclide	**Decay**	**T1/2**	**Nuclide**	**Decay**	**T1/2**	**Nuclide**	**Decay**	**T1/2**
U-238	alpha	4.5 billion yrs	Th-232	alpha	14 billion yrs	U-235*	alpha	700 million yrs
Th-234	Beta	24 days	Ra-228	beta	5.8 yrs	Th-231	beta	26 hrs
Pa-234m	Beta	1.2 min.	Ac-228*	beta	6.1 hrs	Pa-231*	alpha	33,000 yrs
U-234	alpha	240,000 yrs	Th-228	alpha	1.9 yrs	Ac-227	beta (99%)	22 yrs
Th-230	alpha	77,000 yrs	Ra-224	alpha	3.7 days	Ac-227	alpha (1%)	22 yrs
Ra-226	alpha	1,600 yrs	Rn-220	alpha	56 sec	Th-227*	alpha	19 days
Rn-222	alpha	3.8 days	Po-216	alpha	0.15 sec	Fr-223*	beta	22 min
Po-218	alpha	3.1 min.	Pb-212*	beta	11 hrs	Ra-223*	alpha	11 days
Pb-214*	Beta	27 min.	Bi-212*	beta (64%)	61 min	Rn-219*	alpha	4 sec.
Bi-214*	Beta	20 min.	Bi-212*	alpha (36%)	61 min	Po-215	alpha	1.8 msec
Po-214	alpha	160 μsec	Po-212	alpha	310 nano-sec	Pb-211*	beta	36 min
Pb-210	Beta	22 yrs	Th-208*	beta	3.1 min	Bi-211*	alpha	2.1 min
Bi-210	Beta	5 days	Pb-208	stable	N/A	Th-207	beta	4.8 min
Po-210	alpha	140 days				Pb-207	stable	N/A
Pb-206	Stable	N/A						

*Also emits significant gamma.

Technologically Enhanced Naturally Occurring Radioactive Material

Technologically enhanced NORM (TENORM) is typically defined as any naturally occurring material not subject to regulation under the Atomic Energy Act whose radionuclide concentrations or potential for human exposure has been increased above levels encountered in the natural state by human activities. TENORM is produced when radionuclides that occur naturally in ores, soils, water, or other natural materials are concentrated or exposed to the environment by activities, such as uranium mining, chemical processing or sewage treatment. TENORM is a byproduct of processing mineral ores containing naturally occurring radionuclides. These include uranium, phosphate, aluminum, copper, gold, silver, titanium, zircon and rare earth ores. The ore beneficiation process concentrates the radionuclides above their naturally occurring concentrations. Some TENORM may be found in certain consumer products, as well as fly ash from coal-fired power plants.

The majority of TENORM is waste from industrial processes. Many of the wastes are produced in very large volumes, but have low activity concentration. While some wastes are disposed of, others are put to commercial uses. The industrial sectors producing the majority of TENORM are:

- Uranium Overburden and Mine Spoils

- Phosphate Industry Wastes
- Phosphate Fertilizers and Potash
- Coal Ash
- Oil and Gas Production Scale and Sludge
- Waste Water Treatment Sludge
- Metal Mining and Processing Waste
- Geothermal Energy Production Waste.
- Paper and pulp Industry
- Scrap Metal Release and Recycling

TENORM or Source Material

TENORM is defined as naturally occurring radioactive material, other than source material, whose concentration has been technologically enhanced. Source material is defined in 10 CFR 40.4 as "uranium or thorium, or any combination thereof, in any physical or chemical form; or, ores that contain by weight one-twentieth of one percent (0.05 percent) or more of uranium or thorium, or any combination of uranium or thorium"[12]. Source material (0.05% by weight) is material greater than 2.04 Bq/g (55 pCi/g) of Th-232 or greater than (6.18 Bq/g) 167 pCi/g of U-238.

The definition of TENORM specifically excludes source material and by-product material as both are defined in the Atomic Energy Act of 1954, as amended, as implemented by the Nuclear Regulatory Commission [13]. Some source material by-products and mill tailings defined by 10 CFR 40 are regulated by NRC and Agreement States. This preempts states from regulating these materials as TENORM. Some source material by-products and mill tailings processed prior to 1978 may not be regulated by the NRC and therefore, may be regulated by states as TENORM. Uranium by-product is defined as waste material that has become contaminated from the fuel cycle or uranium recovery operations. However, assessing and modeling source material source terms also requires the same attention to the equilibrium status of the natural decay series radionuclides present as do NORM or TENORM source terms.

MODELING NORM AND TENORM SOURCE TERMS

Almost all NORM and TENORM source terms contain at least nominal activity concentrations of all three decay series (uranium, thorium and actinium). Since the uranium and actinium decay series are parented by uranium isotopes (U-238 and U-235) that constitute the majority of naturally occurring uranium, both series are present together in nature. Natural uranium consists of 99.3% by weight U-238 and 0.7% by weight U-235. Based on the weights, natural uranium contains an approximate activity ratio of 22 to 1 U-238 to U-235. For example a sample impacted with natural uranium (not enriched or depleted radium) containing 22 Bq/g (595 pCi/g) of U-238 also contains 1 Bq/g (27 pCi/g) of U-235. This relationship changes only for enriched uranium (U-235 % increases relative to U-238) and for depleted uranium (U-235 % decreases relative to U-238).

Understanding the state of equilibrium of the NORM or TENORM radionuclides present in your source term is vital to the accuracy of your assessment. The state of secular equilibrium in natural uranium and thorium ores is significantly altered when they are processed to extract specific radionuclides. After processing, radionuclides with half-lives less than one year will reestablish equilibrium conditions with their longer-lived parent radionuclides within several years. For this reason, at processing sites what was once a single, long decay series (for example the natural uranium decay series beginning with U-238) may be present as several smaller decay series headed by the longer-lived decay products of the original series (that is, headed by U-238, U-234, Th-230, Ra-226, and/or Pb-210 in the case of U-238). Each of these sub-series can be considered to represent a new, separate decay series. Understanding the physical

and chemical processes associated with materials containing uranium, thorium, and radium is important when developing your source term for dose assessments.

In addition, since each of the series also contains a radon gas member, equilibrium can be broken when radon gas has the opportunity to escape the solid matrix of the source term material. The analysis of characterization samples, the evaluation of characterization data, the development of the source term and the use of modeling software are key steps in any dose assessment. When the source term includes NORM or TENORM the dose assessor must consider the parent and progeny radionuclides and the status of equilibrium/in-growth during each of these steps.

Sample Analysis

Since each series contain at least 12 radionuclides of which some emit alpha, others beta/gamma, laboratory analysis for each of the members of a decay series is not realistic (time and cost prohibitive). Gamma Spectroscopy (for example EPA "Method 901.1M) is most often used to analyze solid type samples for NORM constituents. Gamma spectroscopy analysis is nondestructive and is capable of identifying almost all gamma emitters associated with NORM radionuclides (Table 1). The gamma emitters listed on a standard laboratory gamma spectroscopy analysis report (present in a NORM sample) vary from laboratory to laboratory. Some laboratories report activity values for Ra-226, Ra-228 and the majority of their gamma emitting progeny. Some laboratories also list other naturally occurring radioactive materials that are not normally associated with TENORM, e.g., K-40, beryllium-7 (Be-7).

Not all of the progeny of the natural decay series emit gamma. Therefore, inferring progeny activity based on the assumption of secular equilibrium and the identification of gamma emitting series progeny further down the series is common. For example, Ra-228 does not emit a gamma ray capable of being quantified through gamma spectroscopy analysis. However, the immediate daughter of Ra-228, Ac-228, emits a gamma ray that is easily quantified using gamma spectroscopy. The assumption of secular equilibrium is valid since secular equilibrium is established relatively quickly between Ra-228 and Ac-228 since the half-life of Ac-228 is relatively short (6.1 hrs).

A similar concept is used when quantifying the amount of Th-228 in a soil sample. Th-228 is a pure alpha emitter and is incapable of being quantified using gamma spectroscopy. Pb-212 is a progeny of Th-228 and is a gamma emitter. By measuring the amount of Pb-212 in a sample, and assuming secular equilibrium between Th-228 and Pb-212 exists, the amount of Th-228 in the sample can be determined. The key to this inference technique is sealing the sample container to allow the radon gas (Rn-220 in this example) to be contained and therefore ensure equilibrium is established from Th-228 through Rn-220 and the remaining short-lived progeny of Rn-220, including Pb-212. Once the sample is sealed and a sufficient amount of time has passed to allow equilibrium to re-establish (24 hours in this case) the sample can be analyzed by gamma spectroscopy and non-gamma emitting progeny can be inferred from gamma emitting progeny within the series.

The sealed sample, followed by a sufficient in-growth time period to elapse prior to gamma spectroscopy analysis, is also used to identify Ra-226 from the gamma emitting, short-lived progeny of Rn-222 (Pb-214 and Bi-214).

Using RESRAD to Model NORM

RESRAD is a computer model designed to estimate radiation doses and risks from RESidual RADioactive materials [14 and 15]. The RESRAD model and computer code was developed as a multifunctional tool to assist in developing cleanup criteria and assessing the dose or risk associated with residual radioactive materials. RESRAD is used to:

- Compute soil guidelines (concentrations that will comply with dose or risk based acceptance criteria),
- Compute potential annual doses or lifetime risks to the critical group (workers or members of the public) resulting from exposures to residual radioactive material in soil,
- Compute concentrations of radionuclides in various media (air, surface water, and groundwater) resulting from residual activity in soil, and
- Support ALARA (as low as reasonably achievable) analysis and/or a cost/benefit analysis that can help in the cleanup decision- making process.

Significant exposure pathways for the critical population group modeled by RESRAD in deriving soil acceptance criteria include the following (as depicted in Figure 1):

- Direct exposure to external radiation from the contaminated soil material;
- Internal dose from inhalation of airborne radionuclides, including radon progeny; and
- Internal dose from ingestion of
- Plant foods grown in the contaminated soil and irrigated with contaminated water,
- Meat and milk from livestock fed with contaminated fodder and water,
- Drinking water from a contaminated well or pond,
- Fish from a contaminated pond, and
- Contaminated soil.

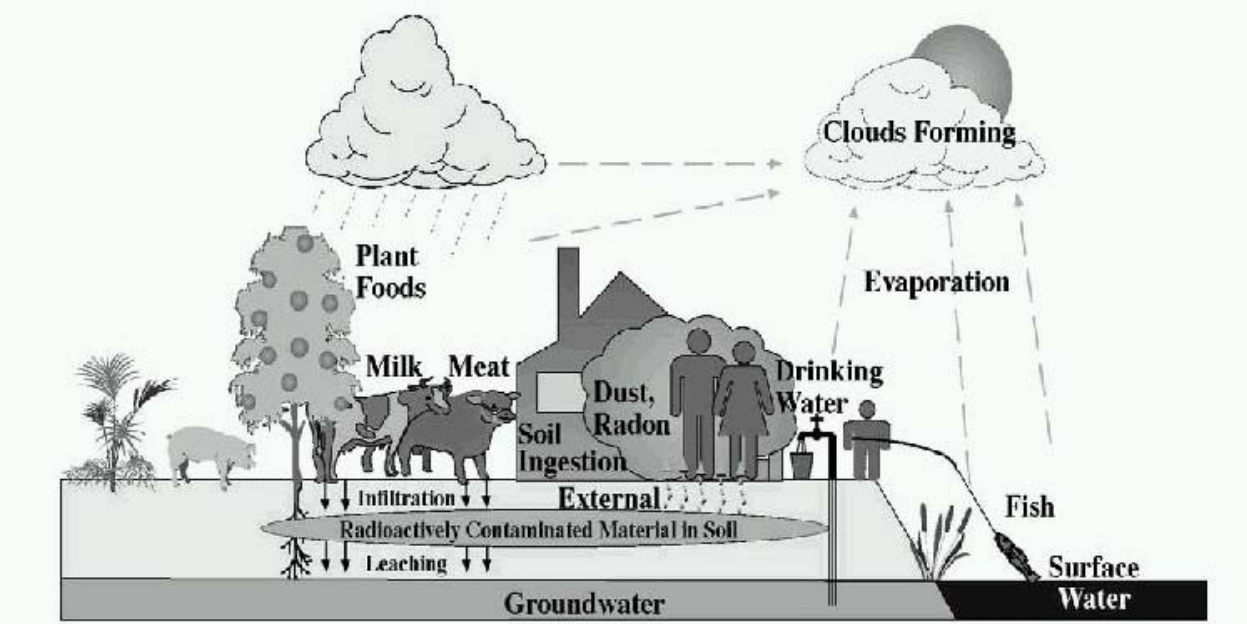

Figure I – RESRAD Environmental Exposure Pathways

When the source term (depicted as Radioactively Contaminated Material in Soil in Figure I) includes any of the natural decay series (NORM or TENORM source term) the additional complexity of in-growth and decay is also modeled. RESRAD can simultaneously estimate risk from exposure to 74 principle radionuclides (half-lives greater than 6 months) and 53 associated radionuclides (short-lived decay products). Primary radionuclide concentrations are entered in pCi/g units and associated radionuclides are entered automatically by RESRAD. For example, Ra-228 is a principle radionuclide with one short-lived decay product (Ac-228). If a risk assessor enters the Ra-228 concentration of 1 Bq/g (27 pCi/g), RESRAD automatically assumes the Ac-228 concentration is 1 Bq/g (27 pCi/g). If the primary radionuclide is part of a multiple radionuclide series, progeny that follow in the series will also be listed,

but the concentration indicated as 0.0. For example, after Ra-228 is entered at 1 Bq/g (27 pCi/g), RESRAD will automatically enter Th-228 at 1 Bq/g (27 pCi/g). The dose assessor can then add the appropriate Th-228 concentration. For example, if the analysis of the characterization data indicates secular equilibrium for the thorium decay series, an equal activity concentration should be entered for each of the short lived progeny of the longer lived progeny in the thorium series. Even if no Th-228 concentration is entered, RESRAD will allow for in-growth over time. Other examples are discussed below.

Assume a site is contaminated with uranium series radionuclides. The site may show the average U-238 activity concentration is 10 Bq/g (270 pCi/g), and this value is entered into RESRAD. The code will automatically add U-234, Th-230, Ra-226, and Pb-210, all at 0.0 Bq/g. The risk assessor should consider the appropriate concentration of each of the progeny radionuclides based on site knowledge, characterization data and/or reasonable rules-of-thumb. If site data characterization data indicates the Uranium series is in secular equilibrium, each progeny should be entered equal to the parent radionuclide (U-238 at 10 Bq/g). If disequilibrium has occurred, for example Th-230 has been extracted as part of a thorium alloy process, the activity concentration indicated by characterization data (the amount resulting from in-growth since thorium removal) should be entered. If data are not available for each radionuclide, concentrations should be estimated to ensure all dose is estimated. The default assumption can be that all radionuclides in a series are present at the same concentration (in equilibrium). This assumption typically would be conservative, but may not be depending on the process history and age of the radiologically contaminated soil. For example, if the contaminant is uranium oxide, it likely contains only trace concentrations of anything besides U-234 and U-238. These uranium isotopes should be entered into RESRAD at the same concentration and the other radionuclides in the series can be left at 0.0 Bq/g. Even after 1,000 years the relative concentrations of non-uranium radionuclides will not change enough to register in dose calculations.

Another site may contain processed uranium ore. The concentration of each individual long-lived radionuclide should be estimated using site characterization data, because equilibrium assumptions may not accurately predict site conditions. For example, Ra-226 may be only 10% of the Th-230 concentration. Additionally, Ra-226 has a half-life that is short enough (1,600 years) so that in-growth and decay will impact risks within the 1,000-year RESRAD default evaluation period. Under this scenario every effort should be used to estimate the concentration of each long-lived radionuclide in the series.

Assume a site is contaminated with thorium decay series radionuclides. The series contains three long lived radionuclides, Th-232, Ra-228, and Th-228. However, the latter two radionuclides have relatively short half-lives at 5.75 years and 1.91 years, respectively. If pure Th-232 is present at time = 0.0 (now), it will only take a few decades (10 half-lives of the progeny) for the Ra-228 and the Th-228 to approach equilibrium conditions. The dose assessor should consider this fact when estimating the source term for thorium series concentrations. In addition, RESRAD has an input parameter titled “time since placement”. For example, if a value of 50 years is entered, the activity concentrations of the long lived members of the decay series, at time equal to 50 years ago, should be entered. RESRAD will make default assumptions about the relative concentrations of natural decay series radionuclides. It is up to the dose assessor to change the default assumptions to match as close as possible to the actual site conditions.

RESRAD Results When Modeling NORM

NORM source terms are usually defined by their large volume and relatively low activity concentration. At least some, if not all of the long lived members, are in equilibrium with their short lived progeny. For example, when modeled over a given time frame, the assessment time frame provided in 10CFR20 Subpart E of 1,000 years, the activity concentration does not decrease appreciably by radioactive decay since the longest lived members of the secular equilibrium series have half lives orders of magnitude

larger than the assessment time frame. The shorter lived progeny, because of secular equilibrium in-growth, also do not decay appreciably over 1,000 years. However, RESRAD output files do not readily represent the phenomena and can lead to a gross misinterpretation of the results.

Once the source term has been established and entered into RESRAD, along with all of the other input parameters, RESRAD will produce both dose results and radionuclide specific, soil activity concentrations equal to the dose limit input. The results are reported for time zero (now), the year of maximum dose (t_{max}) and several other years along the 1,000 year assessment as input by the dose assessment. For NORM radionuclides, the source term activity concentrations are entered for the long lived members of the decay series and RESRAD assumes the same activity concentration for each of the short lived progeny. The RESRAD results are provided for each of the long lived isotopes of the series. The contributions of each of the long lived member's short lived progeny are included in the long lived results as follows:

Uranium Series	Thorium Series	Actinium Series
1. U-238 (long lived)	1. *Th-232 (long lived)*	1. *U-235 (long lived)*
Th-234, Pa-234m	2. *Ra-228 (long lived)*	Th-231
2. U-234 (long lived)	Ac-228	2. *Pr-231 (long lived)*
3. Th-230 (long lived)	*3. Th-228 (long lived)*	*3. Ac-227 (long lived)*
4. Ra-226 (long lived)	Ra-224, Rn-220, Po-216,	Th-227, Fr-223, Ra-223,
Rn-222, Po-218, Pb-214,	Pb-212, Bi-212, Po-212,	Rn-219, Po-215, Pb-211,
Bi-214, Po-214	Th-208	Bi-211, Th-207
5. Pb-210 (long lived)		
Bi-210, Po-210		

For most assessments of NORM or TENORM, the source term includes all three decay series, resulting in a total of 11 long lived radionuclides input and subsequently reported. Figure II provides an example of the input summary provided in the RESRAD output file for a NORM source term.

RESRAD, Version 6.3 T½ Limit = 180 days 05/29/2007 12:55 Page 11
Summary : Subsurface Benchmark Ra-228 Dual Sim 2 DCGLs
File : Subsurface Benchmark Ra-228 Dual Sim 2 DCGLs.RAD

Contaminated Zone Dimensions		Initial Soil Concentrations, pCi/g	
Area:	18400.00 square meters	Ac-227	1.000E+00
Thickness:	1.10 meters	Pa-231	1.000E+00
Cover Depth:	0.15 meters	Pb-210	1.000E+00
		Ra-226	1.000E+00
		Ra-228	1.000E+00
		Th-228	1.000E+00
		Th-230	1.000E+00
		Th-232	1.000E+00
		U-234	1.000E+00
		U-235	1.000E+00
		U-238	1.000E+00

Total Dose TDOSE(t), mrem/yr
Basic Radiation Dose Limit = 2.775E+01 mrem/yr
Total Mixture Sum M(t) = Fraction of Basic Dose Limit Received at Time (t)

t (years):	0.000E+00	1.000E+00	3.000E+00	1.000E+01	3.000E+01	1.000E+02	1.488E+02	3.000E+02	1.000E+03
TDOSE(t):	6.036E-01	6.108E-01	6.249E-01	6.751E-01	8.385E-01	1.670E+00	2.809E+00	2.341E+00	1.849E+00
M(t):	2.175E-02	2.201E-02	2.252E-02	2.433E-02	3.022E-02	6.016E-02	1.012E-01	8.437E-02	6.663E-02

Maximum TDOSE(t): 2.809E+00 mrem/yr at t = 148.8 ± 0.3 years

Figure II – Typical NORM Source Term as Summarized in RESRAD Output File

The example dose assessment included a 6-inch soil cover and standard soil erosion rate of 1 mm/year. These inputs combined with no groundwater pathways results in a maximum dose at the approximate time the soil cover has eroded (148.8 years). Since all 11 long lived decay series radionculides were entered, the RESRAD output file results tables include results for each of the 11 long lived radionuclides, for example, the RESRAD output file table presenting DCGL values for various times.

RESRAD, Version 6.3 T½ Limit = 180 days 05/29/2007 12:55 Page 23
Summary : Subsurface Benchmark Ra-228 Dual Sim 2 DCGLs
File : Subsurface Benchmark Ra-228 Dual Sim 2 DCGLs.RAD

Single Radionuclide Soil Guidelines G(i,t) in pCi/g
Basic Radiation Dose Limit = 2.775E+01 mrem/yr

Nuclide (i)	t= 0.000E+00	1.000E+00	3.000E+00	1.000E+01	3.000E+01	1.000E+02	1.488E+02	3.000E+02	1.000E+03
Ac-227	1.582E+03	1.525E+03	1.442E+03	1.345E+03	1.755E+03	1.532E+04	8.416E+04	1.206E+08	*7.232E+13
Pa-231	1.283E+04	8.477E+03	4.905E+03	1.824E+03	6.132E+02	2.333E+02	1.732E+02	4.637E+02	4.528E+04
Pb-210	2.137E+05	8.189E+04	3.882E+04	1.673E+04	1.151E+04	3.861E+04	1.370E+05	2.455E+07	*7.634E+13
Ra-226	1.257E+02	1.249E+02	1.232E+02	1.173E+02	1.017E+02	6.282E+01	4.339E+01	9.402E+01	3.422E+03
Ra-228	2.007E+02	1.507E+02	1.293E+02	2.024E+02	1.870E+03	5.638E+06	1.411E+09	*2.726E+14	*2.726E+14
Th-228	1.315E+02	1.870E+02	3.783E+02	4.453E+03	5.106E+06	*8.195E+14	*8.195E+14	*8.195E+14	*8.195E+14
Th-230	3.522E+05	1.188E+05	5.056E+04	1.621E+04	4.923E+03	9.076E+02	3.971E+02	2.832E+02	2.434E+02
Th-232	3.512E+03	9.870E+02	3.504E+02	1.115E+02	5.885E+01	2.639E+01	1.466E+01	1.483E+01	1.608E+01
U-234	2.128E+06	7.306E+05	3.193E+05	1.118E+05	4.384E+04	2.079E+04	1.876E+04	4.401E+04	1.715E+05
U-235	7.193E+03	7.049E+03	6.775E+03	5.941E+03	4.257E+03	1.578E+03	7.758E+02	2.038E+03	1.831E+05
U-238	1.411E+04	1.385E+04	1.336E+04	1.192E+04	9.224E+03	5.158E+03	3.435E+03	9.191E+03	*3.361E+05

*At specific activity limit

Figure III – RESRAD Output File Single Radionuclide Soil Guidelines Table

Normally, for a non-NORM source term, the DCGL values reported for the year of maximum dose can be taken from the Single Radionuclide Soil Guideline table and used to develop a MARSSIM based final survey plan. However, if the source term contains NORM the secular equilibrium/in-growth is not accounted for in this table. Specifically, the DCGL values reported for Ra-228 (1.41E+09 pCi/g) and Th-228 (8.20E+14 pCi/g) are obviously too large and will result in doses orders of magnitude greater than the dose limit (in this case 27.75 mrem). The large DCGL values are the result of the method by which RESRAD accounts for activity and resulting dose over time. For each of the radionuclides entered in the source term RESRAD runs the assessment over the time frame (1,000 years) based on the input activity concentration (1 pCi/g) and does not include the additional in-growth activity from the long lived parent of Th-228 or Ra-228. So at time 148.8 years, all of the original 1 pCi/g of Ra-228 and Th-228 has decayed away and the DCGL value calculated on the fraction of the original 1 pCi/g remaining is erroneously large. In reality, a significant amount of activity remains for both Ra-228 and Th-228 from NORM in-growth as revealed in Figure IV. If the summed dose from all in-growth parents of a radionuclide, in the year of max dose (148.8 years) is used to calculate the radionuclide specific DCGL values, the values are correct. Table II summarizes the derivation of DCGLs by dividing the dose limit (27.75 mrem/yr) by the summed dose per year per the 1 pCi/g entered for each radionuclide. Table III presents the RESRAD Single Radionuclide Soil Guideline Table (Figure III) DCGL values and the DCGL values calculated from the RESRAD Individual Nuclide Dose Summed Over All Pathways Table (Figure IV) dose per year per pCi/g factor (Table II).

Individual Nuclide Dose Summed Over All Pathways
Parent Nuclide and Branch Fraction Indicated

Nuclide (j)	Parent (i)	THF(i)	DOSE(j,t), mrem/yr								
			t= 0.000E+00	1.000E+00	3.000E+00	1.000E+01	3.000E+01	1.000E+02	1.488E+02	3.000E+02	1.000E+03
Ac-227	Ac-227	1.000E+00	1.754E-02	1.820E-02	1.925E-02	2.063E-02	1.581E-02	1.812E-03	3.297E-04	2.301E-07	5.537E-22
Ac-227	Pa-231	1.000E+00	2.850E-04	8.997E-04	2.311E-03	8.646E-03	3.090E-02	8.891E-02	1.245E-01	4.653E-02	4.768E-04
Ac-227	U-235	1.000E+00	2.031E-09	1.501E-08	8.835E-08	1.031E-06	1.203E-05	1.469E-04	3.312E-04	2.729E-04	9.952E-06
Ac-227	∑DOSE(j)		1.783E-02	1.910E-02	2.156E-02	2.927E-02	4.672E-02	9.087E-02	1.252E-01	4.680E-02	4.867E-04
Pa-231	Pa-231	1.000E+00	1.879E-03	2.374E-03	3.347E-03	6.568E-03	1.435E-02	3.002E-02	3.566E-02	1.331E-02	1.362E-04
Pa-231	U-235	1.000E+00	2.075E-08	7.621E-08	2.487E-07	1.460E-06	9.266E-06	6.391E-05	1.128E-04	8.489E-05	2.913E-06
Pa-231	∑DOSE(j)		1.879E-03	2.374E-03	3.347E-03	6.569E-03	1.436E-02	3.009E-02	3.577E-02	1.339E-02	1.391E-04
Pb-210	Pb-210	1.000E+00	1.299E-04	3.389E-04	7.148E-04	1.659E-03	2.412E-03	7.187E-04	2.025E-04	1.130E-06	4.077E-17
Pb-210	Ra-226	1.000E+00	2.607E-06	1.671E-05	8.242E-05	6.342E-04	3.692E-03	1.368E-02	1.675E-02	7.888E-03	2.222E-04
Pb-210	Th-230	1.000E+00	4.196E-10	5.865E-09	6.478E-08	1.546E-06	2.957E-05	5.253E-04	1.183E-03	1.949E-03	2.482E-03
Pb-210	U-234	1.000E+00	1.001E-15	2.912E-14	6.977E-13	4.962E-11	2.820E-09	1.708E-07	5.649E-07	1.664E-06	3.388E-06
Pb-210	U-238	9.999E-01	5.895E-22	3.496E-20	1.782E-18	3.740E-16	6.255E-14	1.281E-11	6.298E-11	3.536E-10	1.413E-09
Pb-210	∑DOSE(j)		1.325E-04	3.556E-04	7.973E-04	2.294E-03	6.133E-03	1.492E-02	1.813E-02	9.840E-03	2.707E-03
Ra-226	Ra-226	1.000E+00	2.207E-01	2.222E-01	2.251E-01	2.358E-01	2.692E-01	4.281E-01	6.228E-01	2.873E-01	7.887E-03
Ra-226	Th-230	1.000E+00	4.793E-05	1.450E-04	3.445E-04	1.102E-03	3.845E-03	2.425E-02	6.002E-02	8.740E-02	1.033E-01
Ra-226	U-234	1.000E+00	1.438E-10	1.013E-09	5.439E-09	5.139E-08	5.070E-07	9.605E-06	3.310E-05	7.944E-05	1.415E-04
Ra-226	U-238	9.999E-01	1.019E-16	1.538E-15	1.819E-14	5.072E-13	1.432E-11	8.517E-10	4.209E-09	1.811E-08	5.934E-08
Ra-226	∑DOSE(j)		2.207E-01	2.223E-01	2.255E-01	2.369E-01	2.731E-01	4.523E-01	6.829E-01	3.747E-01	1.114E-01
Ra-228	Ra-228	1.000E+00	9.937E-02	8.881E-02	7.094E-02	3.230E-02	3.403E-03	1.274E-06	5.450E-09	3.262E-17	0.000E+00
Ra-228	Th-232	1.000E+00	6.128E-03	1.761E-02	3.750E-02	8.459E-02	1.457E-01	3.557E-01	6.834E-01	6.759E-01	6.367E-01
Ra-228	∑DOSE(j)		1.055E-01	1.064E-01	1.084E-01	1.169E-01	1.491E-01	3.557E-01	6.834E-01	6.759E-01	6.367E-01
Th-228	Ra-228	1.000E+00	3.891E-02	9.535E-02	1.438E-01	1.048E-01	1.144E-02	3.648E-06	1.422E-08	8.496E-17	0.000E+00
Th-228	Th-228	1.000E+00	2.110E-01	1.484E-01	7.335E-02	6.232E-03	5.435E-06	1.068E-16	3.918E-24	0.000E+00	0.000E+00
Th-228	Th-232	1.000E+00	1.629E-03	1.007E-02	4.069E-02	1.612E-01	3.170E-01	6.667E-01	1.167E+00	1.152E+00	1.048E+00
Th-228	∑DOSE(j)		2.515E-01	2.538E-01	2.578E-01	2.722E-01	3.285E-01	6.667E-01	1.167E+00	1.152E+00	1.048E+00
Th-230	Th-230	1.000E+00	3.086E-05	8.869E-05	2.043E-04	6.089E-04	1.763E-03	5.798E-03	8.673E-03	8.626E-03	8.207E-03
Th-230	U-234	1.000E+00	1.818E-10	1.235E-09	6.407E-09	5.568E-08	4.394E-07	3.858E-06	7.479E-06	1.030E-05	1.142E-05
Th-230	U-238	9.999E-01	1.921E-16	2.805E-15	3.209E-14	8.205E-13	1.838E-11	4.906E-10	1.332E-09	3.049E-09	4.963E-09
Th-230	∑DOSE(j)		3.086E-05	8.869E-05	2.043E-04	6.089E-04	1.764E-03	5.802E-03	8.681E-03	8.637E-03	8.219E-03
Th-232	Th-232	1.000E+00	1.454E-04	4.353E-04	1.015E-03	3.042E-03	8.826E-03	2.896E-02	4.293E-02	4.274E-02	4.091E-02
U-234	U-234	1.000E+00	1.304E-05	3.798E-05	8.688E-05	2.482E-04	6.321E-04	1.321E-03	1.438E-03	5.391E-04	5.574E-06
U-234	U-238	9.999E-01	2.441E-11	1.673E-10	8.677E-10	7.393E-09	5.466E-08	3.764E-07	6.087E-07	4.594E-07	1.583E-08
U-234	∑DOSE(j)		1.304E-05	3.798E-05	8.688E-05	2.482E-04	6.321E-04	1.321E-03	1.438E-03	5.395E-04	5.590E-06
U-235	U-235	1.000E+00	3.858E-03	3.936E-03	4.095E-03	4.669E-03	6.497E-03	1.738E-02	3.532E-02	1.326E-02	1.387E-04
U-238	U-238	5.400E-05	6.086E-10	1.815E-09	4.181E-09	1.199E-08	3.055E-08	6.375E-08	6.903E-08	2.588E-08	2.681E-10
U-238	U-238	9.999E-01	1.966E-03	2.003E-03	2.076E-03	2.327E-03	3.008E-03	5.379E-03	8.077E-03	3.019E-03	3.118E-05
U-238	∑DOSE(j)		1.966E-03	2.003E-03	2.076E-03	2.327E-03	3.008E-03	5.379E-03	8.077E-03	3.019E-03	3.118E-05

THF(i) is the thread fraction of the parent nuclide.

Figure IV – RESRAD Output File Individual Nuclide Dose Summed Over All Pathways, Parent Nuclide and Branch Fraction Indicated Table

Table II – DCGL Calculation Summary

Radio-nuclide	mrem/yr / pCi/g	DCGL (pCi/g)	Radio-nuclide	mrem/yr / pCi/g	DCGL (pCi/g)	Radio-nuclide	mrem/yr / pCi/g	DCGL (pCi/g)
Ac-227	1.25E-01	2.22E+02	Ra-228	6.83E-01	4.06E+01	U-234	1.44E-03	1.93E+04
Pa-231	3.58E-02	7.76E+02	Th-228	1.17E+00	2.38E+01	U-235	3.53E-02	7.86E+02
Pb-210	1.81E-02	1.53E+03	Th-230	8.68E-03	3.20E+03	U-238	8.08E-03	3.44E+03
Ra-226	6.83E-01	4.06E+01	Th-232	4.29E-02	6.46E+02			

Table III – DCGL Comparison

Radio-nuclide	RESRAD DCGL (pCi/g)	Calculated DCGL (pCi/g)
Ac-227	8.42E+04	2.22E+02
Pa-231	1.73E+02	7.76E+02
Pb-210	1.37E+05	1.53E+03
Ra-226	4.34E+01	4.06E+01
Ra-228	1.41E+09	4.06E+01
Th-228	8.20E+14	2.38E+01
Th-230	3.97E+02	3.20E+03
Th-232	1.47E+01	6.46E+02
U-234	1.88E+04	1.93E+04
U-235	7.76E+02	7.86E+02
U-238	3.44E+03	3.44E+03

Assuming each of the three decay series are in secular equilibrium, a decay series specific sum of fractions can be performed to calculate the value for each series that will result in the dose limit (27.75 mrem) from each series. This value can then be used as a surrogate for the series when final status survey samples are analyzed and only one of the series is identified by gamma spectroscopy. The sum of fractions calculation is summarized for both sets of DCGL (Table III) values in Table IV.

Table IV – Sum of Fractions for Each Decay Series

	Radio-nuclide	RESRAD DCGL (pCi/g)	Activity (pCi/g)	Fraction (-)	Calculated DCGL (pCi/g)	Activity (pCi/g)	Fraction (-)
Thorium Series	Th-232	1.47E+01	**14.7**	1.0027	6.46E+02	**14.7**	0.0227
	Ra-228	1.41E+09	14.7	0.0000	4.06E+01	14.7	0.3620
	Th-228	8.20E+14	14.7	0.0000	2.38E+01	14.7	0.6182
		Sum of Fractions:		1.00	Sum of Fractions:		1.00
Uranium Series	U-238	3.44E+03	**38.6**	0.0112	3.44E+03	**38.6**	0.0112
	U-234	1.88E+04	38.6	0.0021	1.93E+04	38.6	0.0020
	Th-230	3.97E+02	38.6	0.0972	3.20E+03	38.6	0.0121
	Ra-226	4.34E+01	38.6	0.8896	4.06E+01	38.6	0.9499
	Pb-210	1.37E+05	38.6	0.0003	1.53E+03	38.6	0.0252
		Sum of Fractions:		1.00			1.00
Actinium Series	U-235	7.76E+02	**142**	0.1830	7.86E+02	**142**	0.1807
	Pa-231	1.73E+02	142	0.8199	7.76E+02	142	0.1830
	Ac-227	8.42E+04	142	0.0017	2.22E+02	142	0.6407
		Sum of Fractions:		1.00	Sum of Fractions:		1.00

SUMMARY

RESRAD modeling (dose assessments) to derive single radionuclide, dose based acceptance criteria, requires a good understanding of the physical, chemical and biological factors/input parameters applicable to the selected exposure scenario(s). When NORM or TENORM source terms are modeled, an additional understanding of the status of equilibrium, is necessary to accurately perform a dose assessment in support of dose based acceptance criteria. Historical information about the site processes/ores, selection of appropriate analytical analyses to identify key decay series radionuclide and a comprehensive review of the characterization data are needed to understand the equilibrium status of the decay series present. Once the source term has been characterized (in regards to relative activities of the radionuclides within a decay series) the source term must be input into RESRAD to reflect that status of equilibrium at time zero, or at the time since placement, if the characterization data reflects the equilibrium status of dated material. When the RESRAD output file is reviewed, depending on the time of maximum dose, DCGL values may be artificially high in value. Sum of fraction calculations, based on the status of equilibrium of each decay series, can also be used to assess the RESRAD results and develop an appropriate MARSSIM final status survey protocol.

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