Solving Complex Radioactive Decay Chains for Future Assessment and Cleanup Decisions

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ABSTRACT

There is a need to understand how radionuclide activity changes with time as the activity measured in the past will be different from current and future levels. When a radionuclide decays, its activity decreases exponentially as a function of time transforming into a different atom - a decay product. The atoms keep transforming to new decay products until they reach a stable state and are no longer radioactive. The series of decay products created to reach this balance is called the decay chain. For radionuclide chains, the daughter products can have significant implications in dosimetry and remediation. Thus, risk assessors evaluating sites with radioactive contamination need to plan for future progeny ingrowth, in addition to sampled radionuclides. These are important considerations for risk quantification during the characterization and cleanup plans, particularly when sampling may have occurred years before the remediation cleanup work begins. If a radionuclide's half-life and current activity are known, then hand-calculating the future activity is straightforward. However, calculating the ingrowth of progeny quickly becomes cumbersome for longer chains such as the Thorium-232 decay series. For the more complex chains where many daughters are formed, possibly with multiple branches, this calculation involves solving a complex set of simultaneous differential equations known as the Bateman Equation. The Decay Chain Activity Projection Tool calculates the activity of radionuclides and their progeny as a function of time. This web tool uses a combination of Perl and plot.ly/JS to automatically construct the radionuclide decay chains, solves the resulting Bateman Equation, and provides the user with tabular solution output and plots. The risk assessor may then use the data for exposure assessment and cleanup decisions without further costly sampling.

BACKGROUND

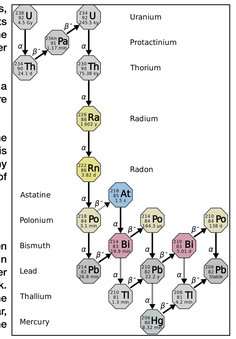
Radioactivity

Radioactivity refers to the amount of ionizing radiation released by a material. Whether it emits alpha or beta particles, gamma rays, x-rays, or neutrons, a quantity of radioactive material is expressed in terms of its radioactivity (or simply its activity). This represents how many atoms in the material decay in a given time period. The units of measurement for radioactivity are the curie (roughly the activity of one gram of Radium-226) and becquerel (amount of a radioactive material that will undergo one transformation per second). The U.S. unit is the Curie (Ci) and the international unit is the Becquerel (Bq).

Radioactive decay is the emission of energy in the form of ionizing radiation. When it decays, a radionuclide transforms into a different atom - a decay product. The atoms continuously transform into new decay products until they reach a stable state and are no longer radioactive. The series of decay products created to reach this balance is called the decay chain.

As a radionuclide decays over time, the activity, or amount of ionizing radiation released, can be quantified for the entire chain if the starting amount of activity for the parent is known. For simple decay chains (one-to-one decay, no branching fractions), this calculation is straight-forward using the derivatives of Lambda, the decay constant. For more complex chains where many daughters are formed with multiple branches, this calculation becomes much more difficult requiring simultaneous equations of derivatives of Lambda and branching fractions.

Figure 1. The Uranium decay series. Source https://en.wikipedia.org/wiki/

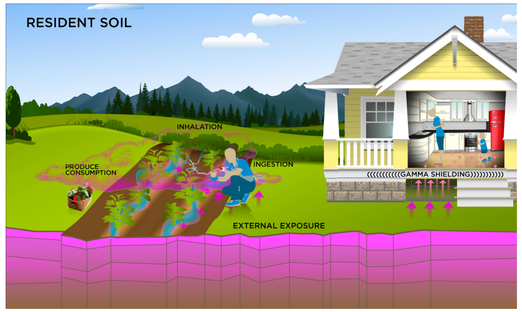


Preliminary Remediation Goals (PRGs)

PRGs are isotope concentrations that correspond to certain levels of risk in air, soil, water and biota. Slope factors (SFs), for a given radionuclide, represent the risk equivalent per unit intake (i.e. ingestion or inhalation) or external exposure of that radionuclide. In risk assessments these SFs are used in calculations with radionuclide concentrations and exposure assumptions to estimate cancer risk from exposure to radioactive contamination. The calculations may be rearranged to generate PRGs for a specified level of risk. SFs may be specified for specific body organs or tissues of interest, or as a weighted sum of individual organ dose, termed the **effective dose equivalent.** These SFs also may be multiplied by the total activity of each radionuclide inhaled or ingested per year, or the external exposure concentration to which a receptor may be exposed, and a chronic daily intake (CDI) term to estimate the risk to the receptor. Cancer slope factors used are provided by the **Center for Radiation Protection Knowledge.**

In general, the radiation dose rate an individual experiences from being near to a radiation source decreases over time. However, this is not always the case. Some radionuclides, such as Cs-137, decay into progeny which are more radiologically hazardous than its parent. For radionuclides such as these, the radiation dose rate actually increases initially as the dangerous progeny activity grows. To account for this phenomenon, PRG values need to be informed by information about how the entire chain activities evolve as a function of time.

Figure 2. This is a representation of the typical residential scenario for exposure to soil encompassing all likely exposure routes.



PRG Scenario Selections

- Residential
- Indoor Worker
- Outdoor Worker
- Composite Worker
- Construction Worker (Site-specific only)
- Recreator (Site-specific only)
- Farmer
- Soil to Groundwater

Media (not all media available for each scenario)

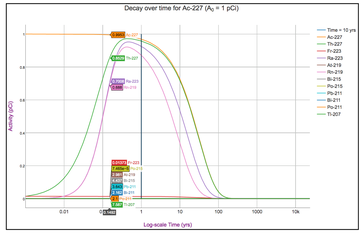
- Soil
- 2-D External Exposure
- Air
- Tap Water
- Fish
- Soil - Unpaved Road Traffic
- Soil - Other Construction Activities
- Surface Water
- Game and Fowl
- Produce (various fruits and vegetables)
- Livestock

RESULTS

Decay information for Ac-227

Radionuclide	λ (yr^{-1})	$T_{1/2}$ (yrs)	Weight (atom-mol)
Ac-227	3.183e-02	2.177e+01	227.027752
Th-227	1.354e+01	5.118e-02	227.027704
Fr-223	1.656e+04	4.186e-05	223.019735
Ra-223	2.213e+01	3.132e-02	223.018502
At-219	3.903e+05	1.776e-06	219.011161
Rn-219	5.519e+06	1.256e-07	219.009480
Bi-215	4.793e+04	1.446e-05	215.001769
Po-215	1.227e+10	5.648e-11	214.999419
Pb-211	1.009e+04	6.868e-05	210.988736
Bi-211	1.702e+05	4.072e-06	210.987269
Po-211	4.235e+07	1.636e-08	210.986653
Tl-207	7.636e+04	9.075e-06	206.977419

Table 1. The radionuclide decay series of Ac-227 based upon the ICRP 107 decay database. Lambda refers to the radiation decay constant in units. $T_{1/2}$ refers to the radionuclide half-life and the final column provides the atomic weights. Note that stable nuclides are not listed in this table.



Summary of activity (A) for Ac-227 and daughters around selected timepoint[1] (10 years)
Activity at Time$_0$ = 1 pCi
(includes last modeled timepoint for which activity is present)

Time (yrs)	Ac-227	Th-227	Fr-223	Ra-223	At-219	Rn-219	Bi-215	Po-215	Pb-211	Bi-211	Po-211	Tl-207
9.999e+00	7.281e-01	7.189e-01	1.004e-02	5.368e-01	7.985e-10	3.378e-01	4.332e-14	1.340e-06	2.300e-10	5.184e-18	1.840e-24	6.640e-22
1.001e+01	7.271e-01	7.177e-01	1.003e-02	5.354e-01	7.917e-10	3.362e-01	4.288e-14	1.328e-06	2.357e-15	5.091e-18	1.799e-24	6.500e-22
2.333e+04	2.964e-323	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00	0.000e+00

Figure 3. This plot shows the activity of Ac-227 and its progeny as a function of time based on the initial condition of a 1 pCi parent activity. The x-axis represents the decay time in log of years and the y-axis represents radionuclide activity in pCi. After ~1.4 years, the activity of Ac-227 drops from 1 pCi to 0.995 pCi. During that time, three progeny activities experienced significant ingrowth (Th-227, Ra-223 and Rn-219) with their activities approaching the theoretical secular equilibrium values.

Image produced with plot.ly.js
https://plot.ly/javascript/

Table 2. The solver output includes a tabular set of results beginning at initial time T_0 through the end of decay chain activity, available as a spreadsheet download. Presented in the browser, however, are the two nearest modeled time points to the user-provided time and the final time point with any activity. If the selected parent is depleted prior to the user time point then only the last active time point is reported.

Highlighted row will be used to generate risk results

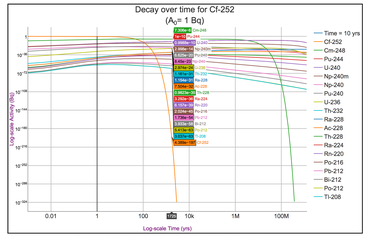


Figure 6. A log-scale plot for difficult chains.

Image produced with plot.ly.js
https://plot.ly/javascript/

PRG Calculations for Parent and Progeny

Isotope	ICRP Lung Absorption Type	Inhalation Slope Factor (risk/pCi)	External Exposure Slope Factor (risk/yr per pCi/g)	Soil Ingestion Slope Factor (risk/pCi)	Particulate Emission Factor (m^3/kg)	Ingestion PRG (pCi/g)	Inhalation PRG (pCi/g)	External Exposure PRG (pCi/g)	Total PRG (pCi/g)	Total PRG (mg/kg)
Ac-227	S	1.49E-07	1.98E-10	2.90E-10	1.36E+09	3.08E+00	5.65E+01	5.95E+02	2.90E+00	4.02E-08
At-219	-	0.00E+00	0.00E+00	0.00E+00	1.36E+09	-	-	-	-	-
Bi-211	-	0.00E+00	1.90E-07	0.00E+00	1.36E+09	-	-	7.15E-01	7.15E-01	1.72E-15
Bi-215	-	0.00E+00	1.08E-06	0.00E+00	1.36E+09	-	-	1.28E-01	1.28E-01	1.11E-15
Fr-223	S	4.07E-11	1.35E-07	1.69E-11	1.36E+09	5.28E+01	2.07E+05	1.05E+00	1.03E+00	2.69E-14
Pb-211	S	4.03E-11	2.91E-07	9.55E-13	1.36E+09	9.35E+02	2.09E+05	4.56E-01	4.56E-01	1.85E-14
Po-211	-	0.00E+00	3.76E-08	0.00E+00	1.36E+09	-	-	3.55E+00	3.55E+00	3.43E-17
Po-215	-	0.00E+00	7.48E-10	0.00E+00	1.36E+09	-	-	1.77E+02	1.77E+02	8.00E-18
Ra-223	S	2.92E-08	4.55E-07	5.99E-10	1.36E+09	1.49E+00	2.89E+02	3.24E-01	2.66E-01	5.20E-12
Rn-219	-	0.00E+00	2.35E-07	0.00E+00	1.36E+09	-	-	5.95E-01	5.95E-01	4.58E-17
Th-227	S	3.50E-08	4.45E-07	1.29E-10	1.36E+09	6.93E+00	2.41E+02	3.28E-01	3.13E-01	1.02E-11
Tl-207	-	0.00E+00	1.59E-08	0.00E+00	1.36E+09	-	-	8.26E+00	8.26E+00	4.34E-14

Table 3. Residential soil PRG output from Preliminary Remediation Goals for Radionuclides (PRG)

Risk Results for Parent and Progeny

Isotope	ICRP Lung Absorption Type	Inhalation Slope Factor (risk/pCi)	External Exposure Slope Factor (risk/yr per pCi/g)	Soil Ingestion Slope Factor (risk/pCi)	Concentration (pCi/g)	Particulate Emission Factor (m^3/kg)	Lambda (1/yr)	Halflife (yr)	2000 m^2 Soil Volume Area Correction Factor	0 cm Soil Volume Gamma Shielding Factor	Ingestion Risk	Inhalation Risk	External Exposure Risk	Total Risk
Ac-227	S	1.49E-07	1.98E-10	2.90E-10	0.7281	1.36E+09	3.18E-02	2.18E+01	9.80E-01	1.00E+00	2.37E-07	1.29E-08	1.22E-09	2.51E-07
At-219	-	0.00E+00	0.00E+00	0.00E+00	7.985E-10	1.36E+09	3.90E+05	1.78E-06	9.00E-01	1.00E+00	-	-	-	-
Bi-211	-	0.00E+00	1.90E-07	0.00E+00	5.184E-18	1.36E+09	1.70E+05	4.07E-06	8.50E-01	1.00E+00	-	-	7.25E-24	7.25E-24
Bi-215	-	0.00E+00	1.08E-06	0.00E+00	4.333E-14	1.36E+09	4.79E+04	1.45E-05	8.36E-01	1.00E+00	-	-	3.39E-19	3.39E-19
Fr-223	S	4.07E-11	1.35E-07	1.69E-11	0.01004	1.36E+09	1.66E+04	4.19E-05	8.13E-01	1.00E+00	1.90E-10	4.84E-14	9.55E-09	9.74E-09
Pb-211	S	4.03E-11	2.91E-07	9.55E-13	2.39E-15	1.36E+09	1.01E+04	6.87E-05	8.73E-01	1.00E+00	2.56E-24	1.14E-26	5.24E-21	5.24E-21
Po-211	-	0.00E+00	3.76E-08	0.00E+00	1.84E-24	1.36E+09	4.24E+07	1.64E-08	8.68E-01	1.00E+00	-	-	5.19E-31	5.19E-31
Po-215	-	0.00E+00	7.48E-10	0.00E+00	1.34E-6	1.36E+09	1.23E+10	5.65E-11	8.76E-01	1.00E+00	-	-	7.59E-15	7.59E-15
Ra-223	S	2.92E-08	4.55E-07	5.99E-10	0.5368	1.36E+09	2.21E+01	3.13E-02	7.84E-01	1.00E+00	3.60E-07	1.86E-09	1.66E-06	2.02E-06
Rn-219	-	0.00E+00	2.35E-07	0.00E+00	0.3378	1.36E+09	5.52E+06	1.26E-07	8.29E-01	1.00E+00	-	-	5.68E-07	5.68E-07
Th-227	S	3.50E-08	4.45E-07	1.29E-10	0.7188	1.36E+09	1.35E+01	5.12E-02	7.93E-01	1.00E+00	1.04E-07	2.98E-09	2.19E-06	2.30E-06
Tl-207	-	0.00E+00	1.59E-08	0.00E+00	6.648E-22	1.36E+09	7.64E+04	9.08E-06	8.83E-01	1.00E+00	-	-	8.05E-29	8.05E-29
Total Risk		-	-	-	-	-	-	-	-	-	*7.01E-07*	*1.77E-08*	*4.43E-06*	*5.14E-06*

Table 4. Residential soil risk output from Preliminary Remediation Goals for Radionuclides (PRG)

As calculated, after 10 years (with no additional source) the excess lifetime risk for an assumed resident exceeds the target of 1E-06.

Preliminary Remediation Goals (PRGs) estimate a concentration to which, if an individual is exposed, has an acceptable excess lifetime cancer risk given the scenario of interest. It is relatively straightforward to calculate individual PRG values for a radionuclide and all of its progeny once the relationship between activity and risk is known. However, the most useful PRG quantity is that which considers the activity of all chain members. In order to accomplish this task, radionuclides from a particular chain need to be added in such a way that exposure to a mixture of radionuclides results in the individual receiving the radiation activity. The method for summing PRG values is displayed in Equation 2. Note that the decay chain calculator is needed to determine the fractional activities of the radionuclides.

$$\text{Composite PRG} = \frac{1}{\frac{f_A}{\text{PRG}_A} + \frac{f_B}{\text{PRG}_B} + \frac{f_C}{\text{PRG}_C} + \frac{f_D}{\text{PRG}_D} + \ldots}$$

Equation 2. Equation required for chain composite PRG

SOLVING THE DECAY SERIES EQUATIONS

In this work, the activities of chain members are solved by a hybrid forward-euler differential equation algorithm published by Leggett et al. This numerical integration method was used in conjunction with the ICRP 107 decay database, which for each radionuclide specifies the progeny, branching ratios, and half-life. The system of linear differential equations is defined by the following equation:

$$\frac{dn_k}{dt} = \sum_{m=1}^{N} \lambda_{m,n} n_m(t) - \lambda_k^e n_k(t)$$

$\lambda_{m,n}$ is the partial decay constant from the mth to the nth nuclide in the decay series

λ_k^e is the effective decay constant

$n_k(t)$ is the number of atoms of the kth member of the chain

Equation 1. The Bateman series of linear, first order differential equations. However, while the solution can be written explicitly for short chains with little effort, the closed form solution is unwieldy for longer chains. It represents the abundances and activities of radionuclides in a decay chain as a function of time. In some chains computational errors arise due to numerical instabilities associated with pairs of similar decay constants. For this reason, numerical integration is often a more practical approach for estimating chain member activities.

SOFTWARE AND WEB INTERFACE

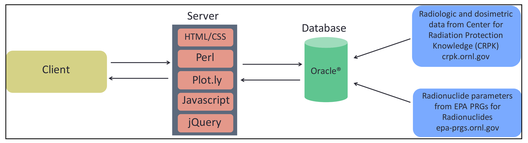


Figure 4. Software and data setup for the Decay Chain Solver

With only 4 required user inputs, an entire series of future activities can be generated for the evaluation of excess lifetime cancer risk. Required selections are:

- The parent radionuclide
- An initial activity (A_0)
- A future time point in years (to allow ingrowth of progeny)
- Preferred units of activity

For more please visit **Preliminary Remediation Goals for Radionuclides (PRG)** at **https://epa-prgs.ornl.gov/cgi-bin/radionuclides/rprg_search**

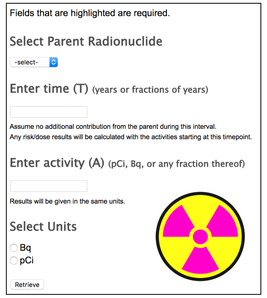


Figure 5. Web User Interface

FUTURE APPLICATIONS AND CONCLUSIONS

A. Plot dose rate as the decay chain evolves
B. Integration of the chain solver routines into:
   - PRG Calculator
   - Building PRG Calculator (BPRG)
   - Surfaces PRG Calculator (SPRG)
   - Dose Concentration Calculator (DCC)
   - Building Dose Concentration Calculator (BDCC)
   - Surfaces Dose Concentration Calculator (SDCC)
C. Peak risk/dose at time period "T"

As more powerful and efficient computational resources become ubiquitous, solving complex problems as described here is not only achievable, but helpful to the risk community and beyond. The Decay Chain Solver Utility will assist in cleanup and assessment decisions as well as alleviate the need for costly re-sampling at future dates.

REFERENCES

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3. Leggett, R., Eckerman, K. and Williams, L., "An elementary Method for implementing complex biokinetic models", Health Physics Society, (1993), 260.