Solving Complex Radioactive Decay Chains for Future Assessment and Cleanup Decisions – 17046

Fred Dolislager *, Stuart Walker **, Leslie Galloway *, Michael Bellamy ***,
Kathryn Bolus ***
* University of Tennessee
** US EPA
*** Oak Ridge National Laboratory

ABSTRACT

In support of U.S. Environmental Protection Agency's on-line calculators for risk and dose assessment, a decay chain tool was developed to address the 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 tool currently uses a combination of Perl, JavaScript, Plot.ly, and jQuery to automatically construct the radionuclide decay chains, solve the resulting Bateman Equation, and provide the user with tabular solution output and plots. Additional software may be used as development advances. The risk assessor can then use the data for exposure assessment and cleanup decisions without further costly sampling. The benefits of providing this calculator are to prevent overestimation of risk due to decay of the parent isotope and to avoid underestimation of risk due to ingrowth of progeny more toxic than the parent. Graphs are provided to show the simultaneous decay rates and ingrowth, as well as tabular output at time-point T and all the decay half-life intervals.

INTRODUCTION

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). (U.S. EPA 2017a)

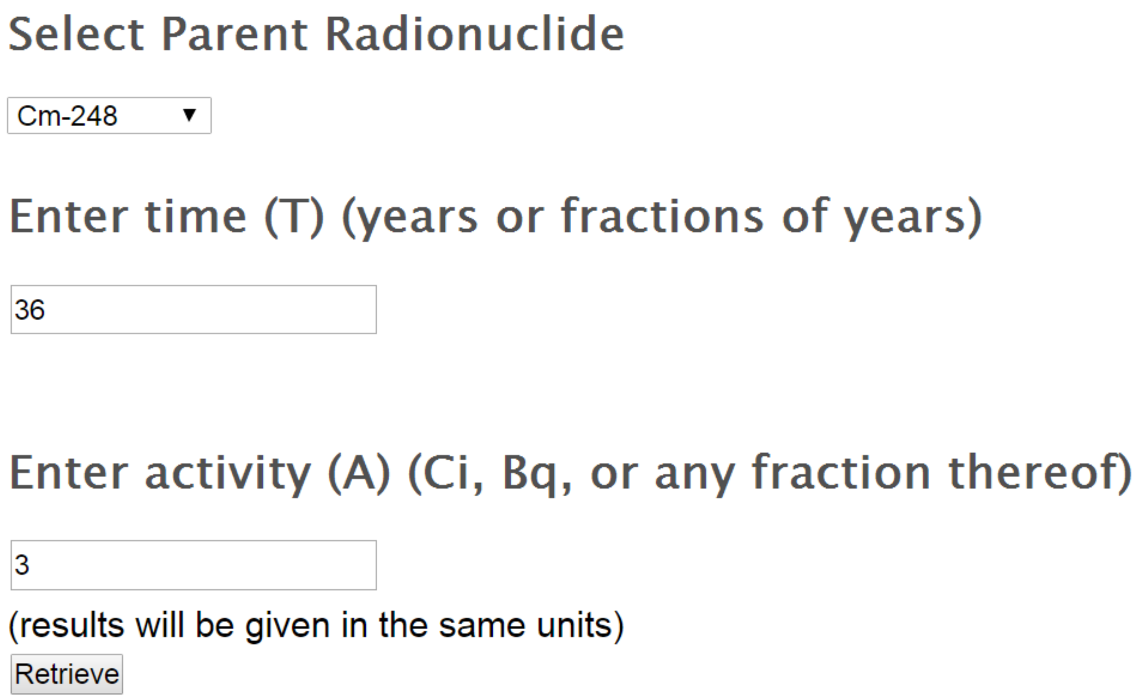
Radioactive decay is the emission of energy in the form of ionizing radiation. When it decays, a radionuclide transforms into a different atom - a decay product. The atoms keep transforming to new decay products until they reach a stable state and are no longer radioactive. The series of decay products created to reach this balance is called the decay chain. (U.S. EPA 2017b)

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.

Using a hybrid forward-euler differential equation algorithm published by Leggett et al., this tool can predict the activity after a period of time (T) given a measurement of activity (A) for the chain parent. (Leggett et al. 1993) Graphs are provided to show the simultaneous decay rates and ingrowth, as well as tabular output at time-point T and all the decay half-life intervals.

CALCULATOR OPERATION

The calculator only requires three inputs from the user: nuclide selection, time at which to solve the chain, and the initial activity of the nuclide. See Figure 1.



Select Parent Radionuclide

Cm-248

Enter time (T) (years or fractions of years)

36

Enter activity (A) (Ci, Bq, or any fraction thereof)

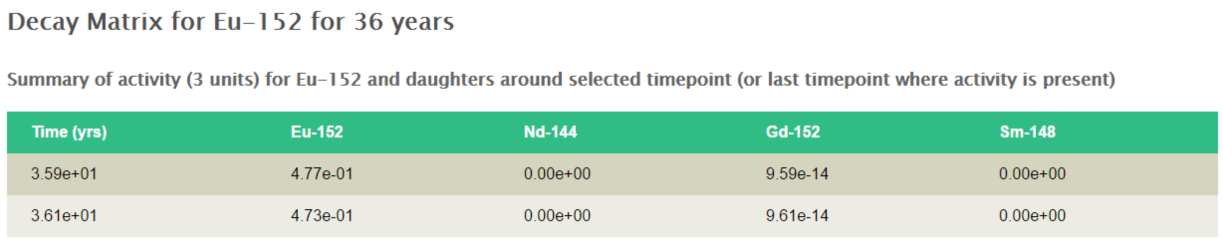
3

(results will be given in the same units)

Figure 1. User interface for data entry.

INTERPRETATION OF RESULTS

After the user selects the “Retrieve” button, the results are displayed. The first section on the results page is a tabular display of the parent and progeny activities at the time requested. See figure 2 for an example of Eu-152. In this example, it may be inferred that two of the progeny are completely gone; however, examination of the graphical display gives evidence that the progeny have not yet grown in. See Figure 3. The graphical display is interactive. Figure 4 presents the menu bar that can be used for taking a screenshot, saving plot to a file, zooming, panning, scaling, axis control, and attaching data labels to the cursor. See figure 5 for the activated data labels. The data labels give the activity for each progeny in the order of decreasing activity. Figure 6 presents a complicated chain for U-238.



Decay Matrix for Eu–152 for 36 years

Summary of activity (3 units) for Eu–152 and daughters around selected timepoint (or last timepoint where activity is present)

Time (yrs)	Eu-152	Nd-144	Gd-152	Sm-148
3.59e+01	4.77e-01	0.00e+00	9.59e-14	0.00e+00
3.61e+01	4.73e-01	0.00e+00	9.61e-14	0.00e+00

Figure 2. Tabular presentation of decay chain activity projection tool.

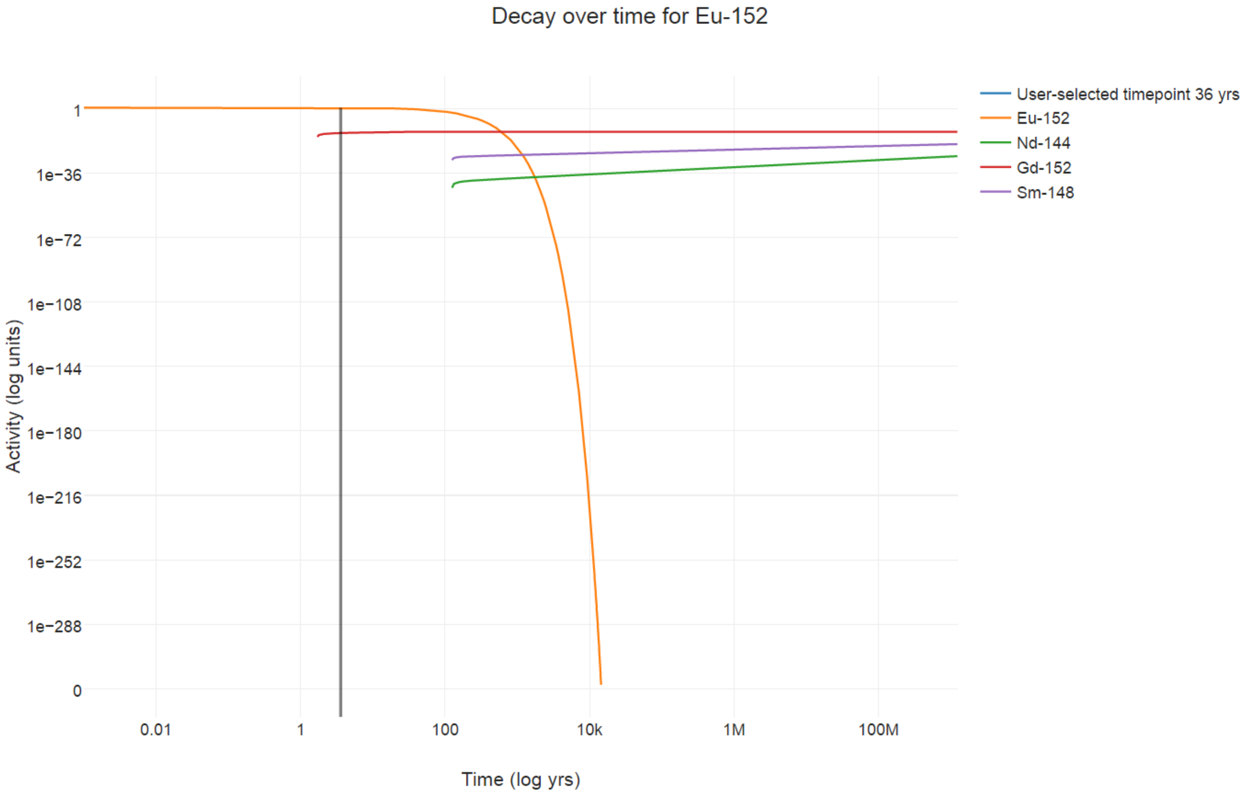


Figure 3. State of the chain over time for Eu-152.

Figure 4. User interface controls for the active plot.

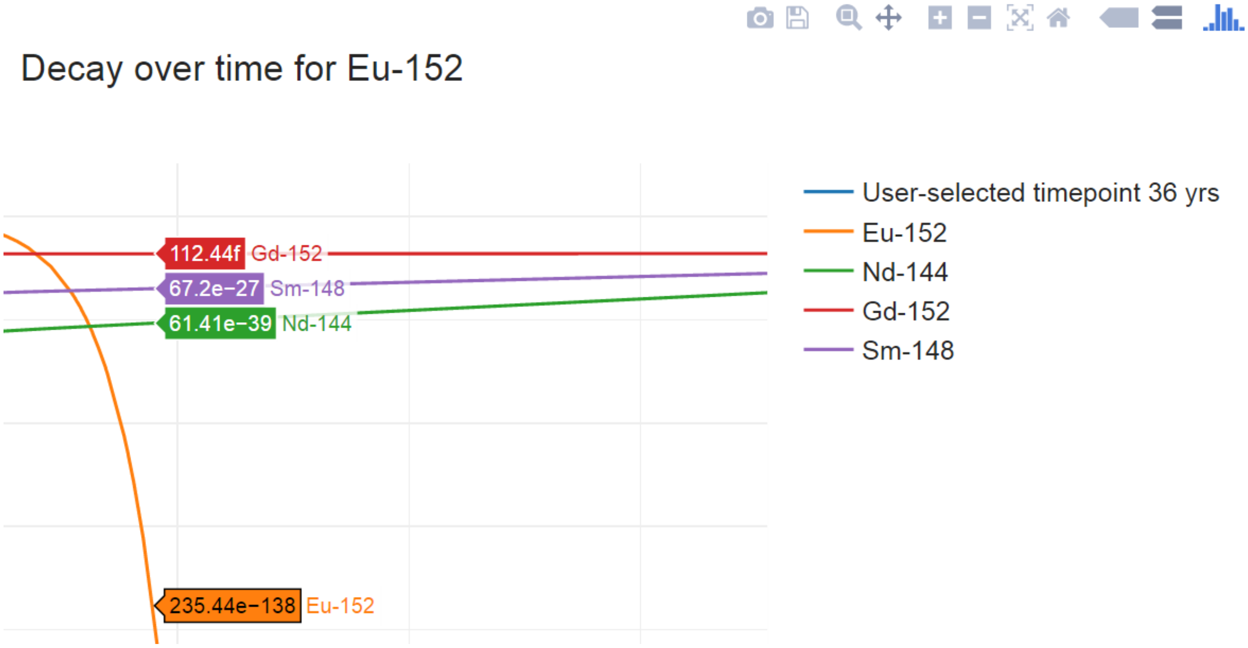


Figure 5. Close up detail of the data labels attached to cursor movement.

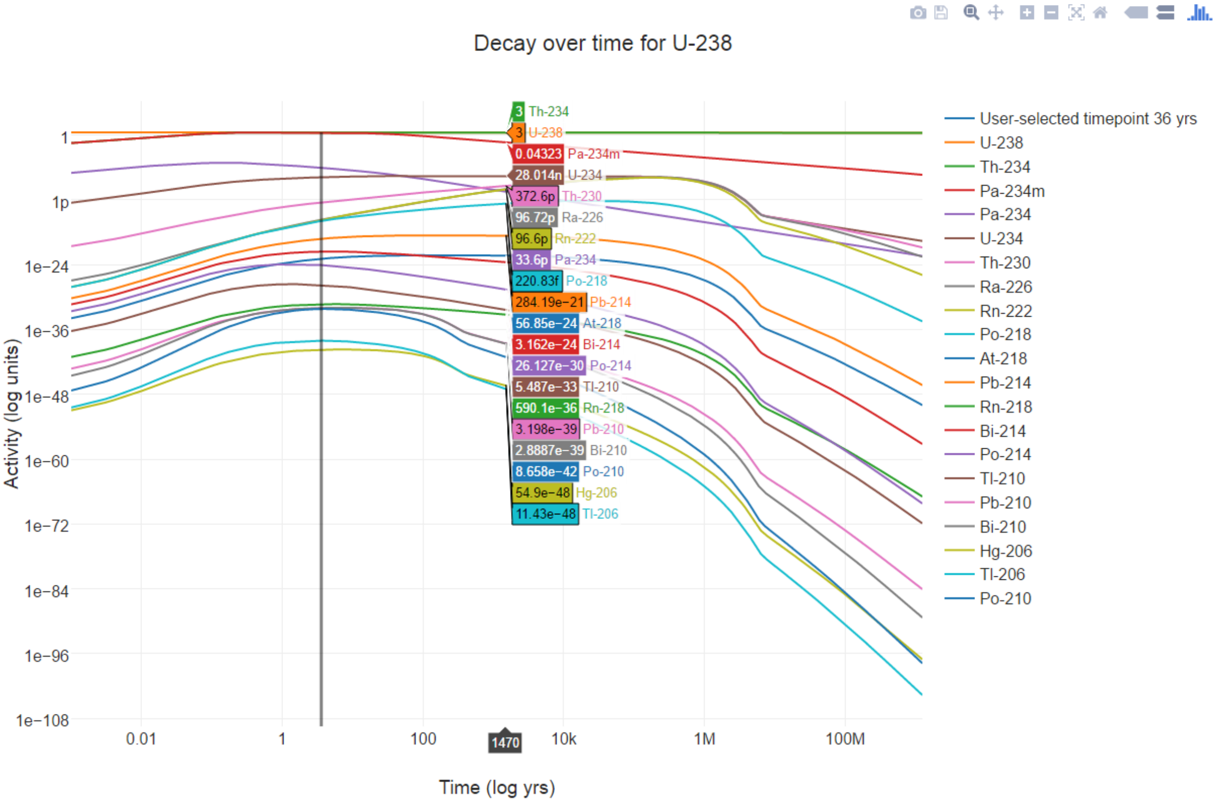


Figure 6. U-238 chain state at 1470 log years.

APPLICATION

The activities presented by this calculator can be used in dose and risk assessments. Often, only the parent nuclide is analyzed and the dose or risk contribution of the progeny are not always assessed. The dose or risk contribution of progeny can be more significant than the parent over time.

CONCLSIONS AND FUTURE DEVELOPMENT

The release of this website will enhance the assessment of cancer risk and dose assessments performed at contaminated sites by predicting the activity of progeny ingrowth. Knowledge of future activity can help predict when a site may again be used for various land uses. Integration of the decay solutions into risk and dose-based screening levels will ensure protective site assessments.

Most dose and risk assessments deal with an exposure duration (ED) in units of years. With the state of the chain known at any given time, it is planned to allow the user to enter the date of the analysis, the date when exposure is expected to begin and end. With this information the activity for each progeny can be calculated by determining the area under the curve.

With more advanced code development it is expected that the year, or span of years (ED), of peak dose or risk can be calculated. This development would be integrated into the existing screening level and risk calculators for the US EPA.

REFERENCES

U.S. EPA 2017a Radiation Basics Website. Accessed January 2017.
https://www.epa.gov/radiation/radiation-basics#tab-3

U.S. EPA 2017b Radioactive Decay Website. Accessed January 2017.
https://www.epa.gov/radiation/radioactive-decay

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