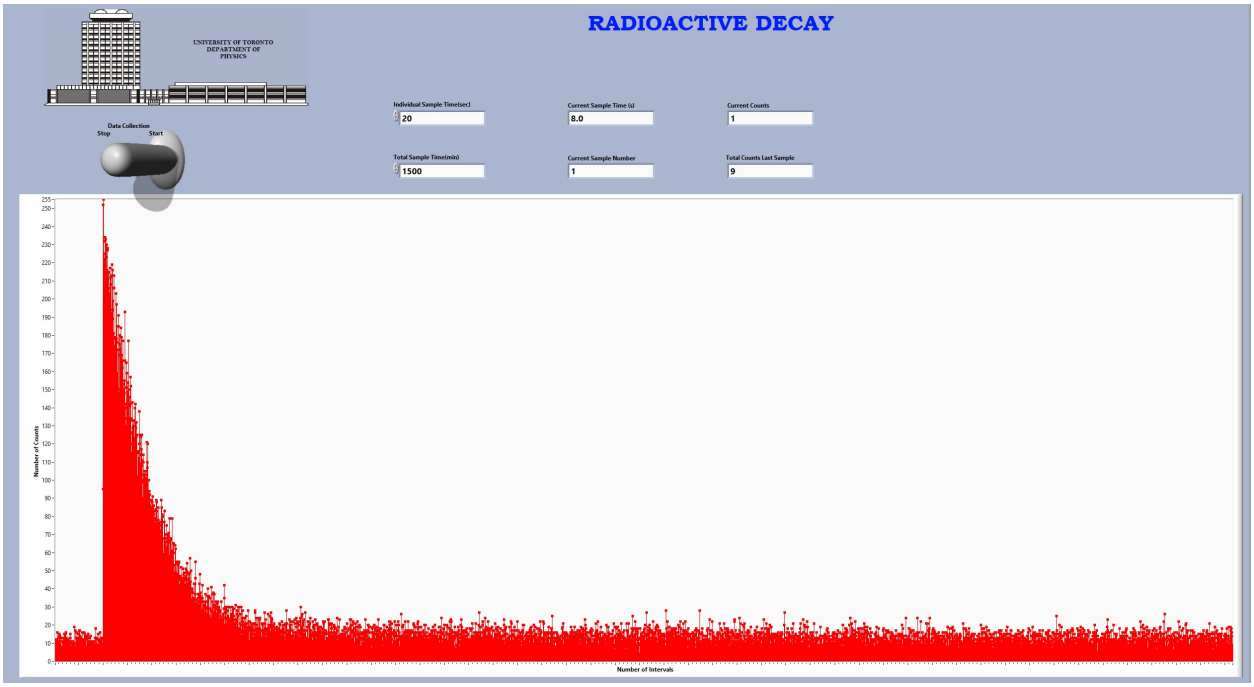
Physics
UNIVERSITY OF TORONTO

Radioactivity in the Air



Revisions

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2020 T. Vahabi and A. Harlick
2017 R. M. Serbanescu, A. Liblong
2013 R. M. Serbanescu, B. Scaunasu, M. Korelek

current revision: 3fcb776
date: March 17, 2025

Introduction

Radon is a radioactive gas which is naturally present in the atmosphere as a decay product of the primordial radionuclides ^{232}Th, ^{238}U, and ^{235}U. Each of these nuclides decays through a long chain of daughters, the members of which can be determined from a chart of the nuclides or found in any number of texts, as well as in graphical format below (Figure 1). The decay products of radon isotopes will adhere to dust particles in the air, which can be collected on filter paper and measured. The amount of activity and proportions of nuclides present in the sample generally depend on the filtration duration, the weather, and potential emissions from nearby industrial sources.

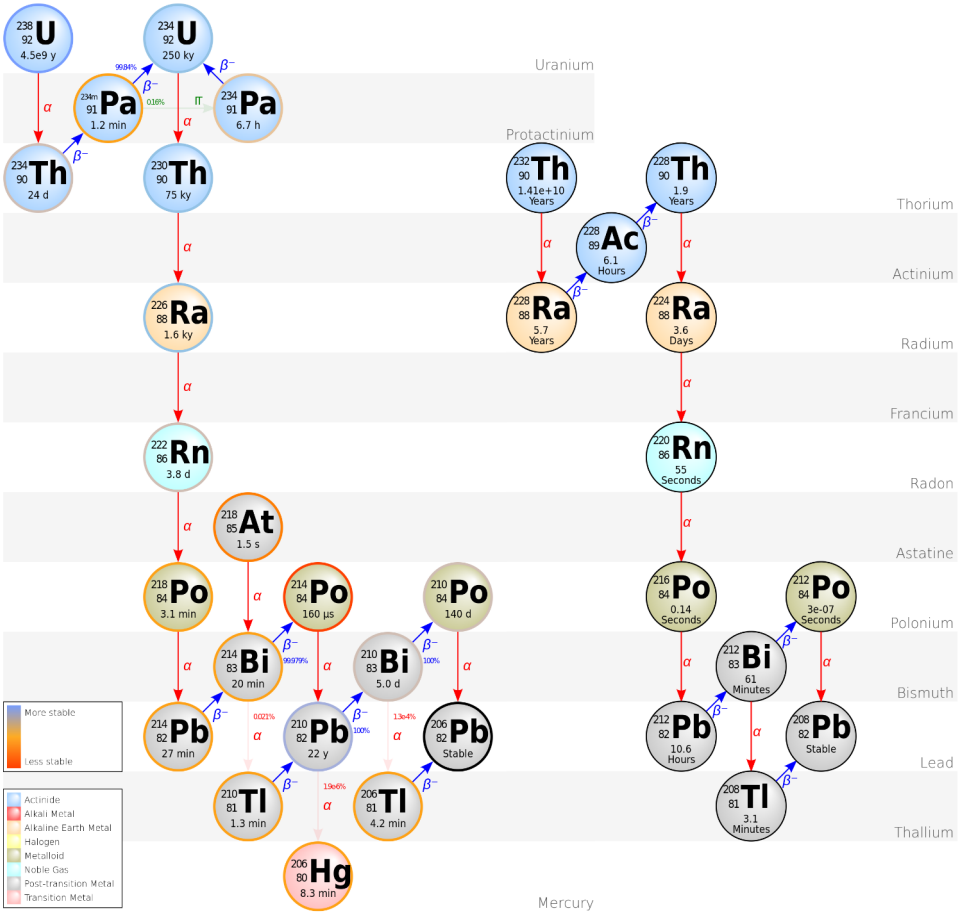

Figure 1: Decay chains of ^{238}U (left Liblong [2017b]) and ^{232}Th (right Liblong [2017a])

In this experiment, you will count β decays from an air filter sample using a Geiger-Müller (G-M) counter, isolate activity from two particular short-lived radionuclides by subtracting background, fit the resulting curve with a sophisticated model to determine the initial concentrations of these two nuclides, then compute the atmospheric concentration of radon.

The most abundant atmospheric radon isotope is ^{222}Rn, originating from ^{238}U. Referring to the ^{238}U decay series, we can expect observable β activity from this chain to come solely from ^{214}Pb and ^{214}Bi , hereafter referred to by their respective historical names, RaB and

RaC (based on their positions in the decay chain of ^{226}Ra, itself a daughter of ^{238}U); ^{210}Pb has a half-life on the order of years, so its activity will be negligible.

Somewhat less abundant in the atmosphere is ^{220}Rn, originating from ^{232}Th. ^{220}Rn is also called *thoron*; henceforth, mentions of "radon" will refer specifically to ^{222}Rn. From the thoron series, we can expect to observe activity from ^{212}Pb, ^{212}Bi, and ^{208}Tl. This subseries will be in *secular equilibrium*. Secular equilibrium is a phenomenon occuring when a radioactive parent has a much longer half-life than its radioactive daughter. As the parent decays, the quantity of the daughter increases, eventually reaching an equilibrium point where it's decaying at the same rate it's being produced. Essentially, the parent's decay is the rate-limiting step in the series. The combined activity of this thoron subseries in secular equilibrium is an exponential function with the same decay constant as the parent, ^{212}Pb (half-life 10 hours). Therefore, if we count for long enough that the RaB and RaC have been effectively depleted, we can fit the curve with a simple exponential, then extrapolate the fit back to time 0 and subtract it as a background.

The rate at which RaA (^{218}Po), RaB, and RaC will accumulate on dust particles varies according to the atmospheric abundance of dust or smog. In very clear conditions one might expect most of the RaA to α decay to RaB without first becoming attached to dust since the half-life is only a few minutes. There will therefore be relatively more accretion of RaB and RaC onto atmospheric dust. Conversely, under smoggy conditions there will be much more RaA accretion on dust, and therefore a greater initial activity from its first decay product, RaB.

1 Equipment

1.1 Geiger-Müller tube

This apparatus is a gas-filled pair of concentric cylinders acting respectively as anode and cathode. A high voltage is applied across the electrode so that when ionizing radiation passes through the gas, an ionization event triggers an avalanche of ion-electron pair production — a phenomenon called Townsend discharge. The electrons and ions drift to the anode and cathode respectively, and the accumulated negative charge on the anode creates a standard voltage negative pulse over the connected resistor.

1.2 Picker Scaler

This is both an analog counter and a high voltage power supply for the G-M tube.

2 Experiment

This experiment makes full use of two lab periods, though most of this is passive in letting the apparatus count. In the first session, you will determine the optimal operating voltage of the G-M tube by counting a ^{137}Cs source at different voltages, then plotting the voltage vs. count rate and finding the plateau point (the point at which the count rate stops increasing appreciably). The remainder of the period will be spent counting background radiation. In

the second session, you will use the air sampler in room 126 (speak to the lab technologist about this) to collect an air sample for one hour, while simultaneously counting background radiation. Then, you will count the air sample for the remainder of the period.

2.1 Using the counter

You will use the "Radioactivity In Air" (RIA) program to digitally count radiation events rather than using the analog counter in the Picker scaler.

Observe the construction of the G-M tube. Note the pair of sliding metal plates. The bottom one is a platform serving as the sample bay; the top one has a sheet of thin metal foil. **Be careful not to damage the foil**. It has an area density of between 1.4 and 2.0 mg/cm^2 which is thin enough to transmit beta particles, but thick enough to stop alpha particles of energy less than about $2MeV$. The gas is not efficiently ionized by γ rays, so this counter is most useful for β particle detection.

2.2 Using the oscilloscope

There are no quantitative measurements to be taken from the oscilloscope in this experiment, but you should use it to **make qualitative observations**.

- Do not operate the oscilloscope with voltage above $1000V$. Disconnect or switch off the oscilloscope when running at voltages higher than $1000V$.
- Do not change the high voltage setting while the osilloscope is connected.
- Connect the oscilloscope to the "pulse input" terminal only *after* the Picker scaler has been set into operation.

Use the following settings:

- Set the "Intensity" dial to maximum (so you can observe the pulses easily).
- Set the vertical and horizontal position dials to equilibrium.
- Set the "VOLTS/DIV" slider to so that the line in the pulse terminal appears clear and sharp.
- If there is a "FOCUS" dial on the oscilloscope, set it to equilibrium.
- If there is a "SOURCE" switch on the oscilloscope, set it to "LINE" so that it doesn't flash every time there is a pulse.

Procedure

Period 1

Pulse observation

Open the "Radioactivity in the Air" program on the computer. Place a ^{137}Cs source in the sample bay of the G-M tube. This tube should be connected to the Picker Scaler "detector input" terminal. Switch on the Picker Scaler (the main power, NOT the high voltage). Set the voltage dial on the Picker Scaler to the minimum value. Switch on the voltage. Set the voltage somewhere between $600V$ and $1000V$. **Reminder:** do not surpass $1000V$ with the oscilloscope connected. Connect the oscilloscope to the "pulse input" terminal. Note the following:

- shape and length of the pulse
- effect of moving the source further or closer to the detector
- effect of varying the voltage
  Reminder: disconnect the oscilloscope each time before switching the voltage.
- deadtime of the detector

Disconnect the oscilloscope, then connect it to the "output pulse" terminal and observe the standardized pulse that the discriminator produces.

Determination of ideal operating voltage

Disconnect the oscilloscope. For a range of voltages between $700V$ and $1100V$, count the ^{137}Cs source for a fixed period of time. Choose the counting duration based on how much time remains in the period and how many voltage values you want to use; be sure to leave time at the end of the session (ideally one hour) to count background. Consider also the effect of the counting time on the variance of the total count; recall that for a Poisson-statistical counting experiment with N counts, the standard deviation is $\sqrt{N}$.

Requirement: plot the count rate against the voltage, and determine qualitatively the plateau point. Include error bars with this plot.

Switch to the operating voltage you determine and count background for the remainder of the lab period.

Period 2

Collecting and counting the air sample

Ask the lab technologist to begin collecting an air sample. Switch to the operating voltage determined in the previous period. Count background for one hour. Retrieve the air sample and begin counting it immediately, until the end of the period.

Analysis

The ultimate result sought in this experiment is the atmospheric concentration of ^{222}Rn. Assuming secular equilibrium, i.e.$\lambda_{\mathrm{Rn}} n_{\mathrm{Rn}} = \lambda_A n_A = \lambda_B n_B = \lambda_C n_C$ (where $A_i(t) = \lambda_i n_i(t)$). We can compute this from the solution to the differential equation for the build-up of RaB during the filtering:

$$N_B = \frac{k(n_A + n_B)}{\lambda_B}(1 - \mathrm{e}^{-\lambda_B t}) + \frac{k n_A}{\lambda_B - \lambda_A}(\mathrm{e}^{-\lambda_B t} - \mathrm{e}^{-\lambda_A t}) \tag{1}$$

where k is the flow rate of the air sampler ($14L/s$), N is the number of atoms, n is the concentration of atoms (number per unit volume), λ the e-folding timescale related to the half-life $\lambda = \ln(2)/t_{\frac{1}{2}}$, and the subscripts A, B, and C correspond to RaA, RaB, and RaC. The only missing piece is N_B, which is what we will determine in our analysis of the filtered sample activity.

The activity measured in the air sample has contributions from numerous sources, largely falling within two categories: background radiation (e.g. ^{40}K, cosmic radiation) and radiation from the Thoron series. These background sources can be modeled with simpler functions and removed from the data to isolate the activity from RaB and RaC.

Remove the background radiation from the data by assuming the background radiation is constant and subtracting the average background count rate from the entire dataset. Next isolate the Thoron series by removing the first hour of data and fitting an exponential decay to the remaining data. This fit should have a decay rate that matches the decay rate of ^{212}Pb. You can then extrapolate this fit back to $t = 0$ and subtract it from the original data. The remaining data will have activity from the Radon decay chain including RaA,RaB, and RaC.

Compare the half-life of this exponential fit to the half-life for ^{212}Pb. Evaluate the goodness of fit. Are there other significant sources of radioactivity contributing here?

The activity from RaA is not included in our model. The main decay path for RaA (Po-218) is through an α decay that is not measured, but its decay products (including RaB) produce additional β particles that we do not model. The activity from RaA should be removed from the dataset by removing the first 15 minutes of the measurement (5 half-lives of RaA).

The remaining data should include only the RaB and RaC activity. Fit this data with equation 6 to determine the unknown parameters.

The times given here are rough guidelines, and may need to be adjusted based on the amount of observed activity. You should truncate the dataset where needed before fitting to models, and then extrapolate the model across all of the dataset to remove the unwanted signal contribution. Figure **??** shows the approximate timeline of the decay process. Each decay chain shows approximately where in timeline the particular decay contributes to the observed activity. In order to remove the unwanted signal, you need to find periods where the other decays do not contribute or can be removed by modeling the effect on the signal.

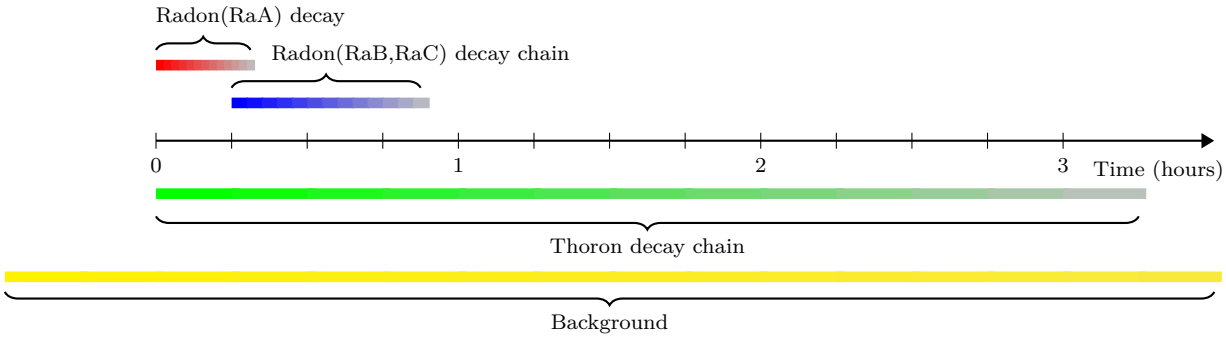

Figure 2: Schematic of the decay timeline. The Thoron decay chain is in secular equilibrium and can be removed by fitting an exponential decay to the signal once RaA,RaB, and RaC have decayed (about 1 hour into the measurement). The Radon(RaA) decay should be removed from the dataset (the first 15 minutes of the measurement) because it decays through α particles that are not measured but its decay products produce additional β particles. The Radon(RaB,RaC) decay can be analyzed with equation 6.

A model for β decay of the radon series

For a general treatment of radioactive decay chains, and for a detailed derivation of the following equations, refer to Whyte and Taylor [1962].

The variation of count rate with time will be a function of the decay constants of RaB and RaC and of their relative activities at the start of the counting period. Let $A_B(t)$ and $A_C(t)$ be, respectively, the actual activities of RaB and RaC.

The observed count rates in the detector can be expressed as the product of the actual activity and the detector efficiency ε:

$$A^{\text{obs}}(t) = \varepsilon A(t) \tag{2}$$

ε is a product of two factors primarily: a geometrical factor for the solid angle covered by the detector, and a factor for the fraction of β particles which have sufficiently low energy that they are stopped by the aluminum foil alpha absorber or the GM tube window before they reach the detector.

The total decay rate in the measurement includes contributions from RaB and RaC, with

different detection efficiencies so that the total count rate is

$$A^{\text{obs}}(t) = \varepsilon_b A_B(t) + \varepsilon_c A_C(t), \tag{3}$$

$$= A_B(t)\left(1 + \frac{\varepsilon_c}{\varepsilon_b} R(t)\right), \tag{4}$$

if we define a term R as the ratio of activities of RaC and RaB:

$$R = \frac{A_C(t)}{A_B(t)} = \frac{\lambda_C n_C(t)}{\lambda_B n_B(t)}. \tag{5}$$

The total observed activity is then given by (and derived in the appendix)

$$A^{\text{obs}}(t) = A_B^{\text{obs}}(0)\left[\left(1 + \frac{\varepsilon_C}{\varepsilon_B}\frac{\lambda_C}{\lambda_C - \lambda_B}\right)\mathrm{e}^{-\lambda_B t} + \frac{\varepsilon_C}{\varepsilon_B}\left(R - \frac{\lambda_C}{\lambda_C - \lambda_B}\right)\mathrm{e}^{-\lambda_C t}\right], \tag{6}$$

where ϵ_B and ϵ_C are the efficiencies, $A_B^{\text{obs}}(0)$ is the observed activity from RaB (including the efficiency factor). β energy spectra are continuous on the interval $[0, E_{\max}]$. The value of $E_{\max}$ varies between nuclides, and for RaB it's lower than for RaC. Thus, the efficiency for RaB is lower than for RaC. For an aluminum filter of $27\mu m$ thickness and a G-M tube window of $\rho x = 1.5 mg/cm^2$, $\varepsilon_B = 0.80$ for RaB and $\varepsilon_C = 0.95$ for RaC. These values don't include the geometric factor, which cancels out in the ratio.

Requirement: fit your background-subtracted data with Equation (6). Is the R value obtained from the fit consistent with the value predicted by Whyte and Taylor [1962] in Figure 3? For an approximate geometric efficiency of 50%, work out the initial activities of RaB and RaC. Compute the atmospheric concentration of radon using Equation (1). A typical concentration is $15\text{Bq}/m^3$. How does your result compare? Have we made any assumptions that were not stated explicitly?

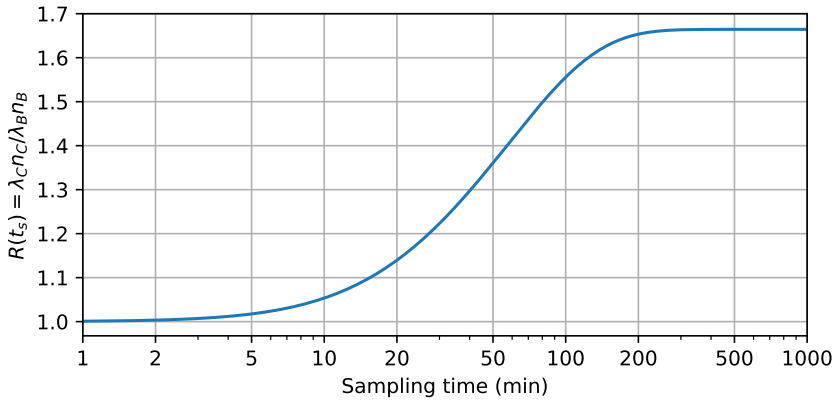

Figure 3: Predicted value of R as a function of air sampling duration (based on figure 1 of Whyte and Taylor [1962])

A Derivation of decay activity

Following Whyte and Taylor [1962], once the air flow over the sample has stopped the equations for the decay of RaB and RaC are (ignoring the small effect of RaA because of the relatively long decay timescale):

$$\frac{dN_B}{dt} = -\lambda_B N_B, \tag{7}$$

$$\frac{dN_C}{dt} = \lambda_B N_B - \lambda_C N_C. \tag{8}$$

Integrating the equation for RaB gives an exponential decay

$$N_B = N_B(0)e^{-\lambda_B t}, \tag{9}$$

or equivalently

$$A_B = A_B(0)e^{-\lambda_B t}. \tag{10}$$

The integral for RaC can be completed by first multiplying by $e^{\lambda_C t}$ then integrating

$$\frac{dN_C}{dt}e^{\lambda_C t} = \lambda_B N_B e^{\lambda_C t} - \lambda_C N_C e^{\lambda_C t}, \tag{11}$$

$$\frac{dN_C e^{\lambda_C t}}{dt} = \lambda_B N_B e^{\lambda_C t}, \tag{12}$$

$$\int_{t'=0}^{t'=t} dN_C e^{\lambda_C t} = \int_{t'=0}^{t'=t} \lambda_B N_B e^{\lambda_C t} dt, \tag{13}$$

$$\int_{t'=0}^{t'=t} dN_C e^{\lambda_C t'} = \int_{t'=0}^{t'=t} \lambda_B N_B(0) e^{\lambda_C t' - \lambda_B t'} dt', \tag{14}$$

$$N_C(t)e^{\lambda_C t} - N_C(0) = \frac{\lambda_B}{\lambda_C - \lambda_B} N_B(0)\left(e^{(\lambda_C - \lambda_B)t} - 1\right), \tag{15}$$

$$N_C(t) = N_C(0)e^{-\lambda_C t} + \frac{\lambda_B}{\lambda_C - \lambda_B} N_B(0)\left(e^{-\lambda_B t} - e^{-\lambda_C t}\right), \tag{16}$$

$$N_C(t) = N_B(0)\left(Re^{-\lambda_C t} + \frac{\lambda_B}{\lambda_C - \lambda_B}\left(e^{-\lambda_B t} - e^{-\lambda_C t}\right)\right). \tag{17}$$

$$\tag{18}$$

Expressed as an activity, this becomes

$$A_C(t) = A_B(0)\left(Re^{-\lambda_C t} + \frac{\lambda_B}{\lambda_C - \lambda_B}\left(e^{-\lambda_B t} - e^{-\lambda_C t}\right)\right). \tag{19}$$

Both A_C and A_B are measured in the experiment with different efficiencies

$$
\begin{aligned}
A_{\text{obs}}(t) &= \varepsilon_B A_B(t) + \varepsilon_C A_C(t), && (20)\\
&= \varepsilon_B A_B(0) e^{-\lambda_B t} + \varepsilon_C A_B(0) \left[R e^{-\lambda_C t} + \frac{\lambda_B}{\lambda_C - \lambda_B} \left(e^{-\lambda_B t} - e^{-\lambda_C t} \right) \right], && (21)\\
&= A_B(0) \left[\varepsilon_C e^{-\lambda_C t} \left(R - \frac{\lambda_B}{\lambda_C - \lambda_B} \right) + \varepsilon_B e^{-\lambda_B t} \left(1 + \frac{\varepsilon_C}{\varepsilon_B} \frac{\lambda_B}{\lambda_C - \lambda_B} \right) \right], && (22)\\
&= A_B^{\text{obs}}(0) \left[\frac{\varepsilon_C}{\varepsilon_B} e^{-\lambda_C t} \left(R - \frac{\lambda_B}{\lambda_C - \lambda_B} \right) + e^{-\lambda_B t} \left(1 + \frac{\varepsilon_C}{\varepsilon_B} \frac{\lambda_B}{\lambda_C - \lambda_B} \right) \right], && (23)\\
& && (24)
\end{aligned}
$$

References

A. Liblong. Decay chain of thorium-232, 2017a. URL https://commons.wikimedia.org/wiki/File:Decay_Chain_of_Thorium-232.svg.

A. Liblong. Decay chain of uranium-238, 2017b. URL https://commons.wikimedia.org/wiki/File:Decay_Chain_of_Uranium-238.svg.

G. N. Whyte and H. W. Taylor. A Radioactivity Experiment Using Activities Filtered from the Air. *American Journal of Physics*, 30(2):120–124, Feb. 1962. ISSN 0002-9505, 1943-2909. doi: 10.1119/1.1941929. URL https://pubs.aip.org/ajp/article/30/2/120/529510/A-Radioactivity-Experiment-Using-Activities.