

# Radioactivity in the Air

### Revisions

2017 R. M. Serbanescu

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## Introduction

Radon is a radioactive gas which is naturally present in the atmosphere as a decay product of the primordial radionuclides <sup>232</sup>Th, <sup>238</sup>U, and <sup>235</sup>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.

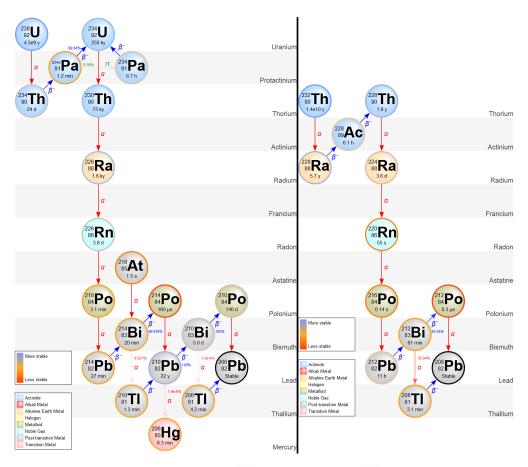


Figure 1: Decay chains of  $^{238}\mathrm{U}$  (left  $[?]) and <math display="inline">^{232}\mathrm{Th}$  (right [?])

In this experiment, you will count  $\beta$  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  $\beta$  activity from this chain to come solely from  $^{214}$ Bi and  $^{214}$ Pb, 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 <sup>220</sup>Rn, originating from <sup>232</sup>Th. <sup>220</sup>Rn is also called *thoron*; henceforth, mentions of "radon" will refer specifically to <sup>222</sup>Rn. From the thoron series, we can expect to observe activity from <sup>212</sup>Pb, <sup>212</sup>Bi, and <sup>208</sup>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, <sup>212</sup>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  $\alpha$  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 <sup>137</sup>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. N.B. you may have the opportunity to leave the apparatus counting past the end of the period, which will allow you to do a subtraction of the thoron series contribution; ask the technologist if this option is available.

#### 2.1Using 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  $mq/cm^2$  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  $\gamma$  rays, so this counter is most useful for  $\beta$  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  $^{137}\mathrm{Cs}$  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. Alternatively, if you've discussed this with the lab technologist, leave the apparatus counting at the end of the period; come in the next day to stop it and retrieve your data.

# Analysis

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

$$N_B = \frac{k(n_A + n_B)}{\lambda_B} (1 - e^{-\lambda_B t}) + \frac{kn_A}{\lambda_B - \lambda_A} (e^{-\lambda_B t} - e^{-\lambda_A t})$$
(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),  $\lambda$  is the half-life, 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 vs. time data for the air sample has contribution from numerous other sources, largely falling within two categories: background radiation (e.g. <sup>40</sup>K, cosmic radiation) and radiation from the thoron series. If these contributions can be modeled, the model curves can be subtracted from data to isolate activity from RaB and RaC.

#### **Background subtraction**

As a first step, subtract from the entire dataset the average background count rate determined on the day of the air sample counting. The remaining contribution should be solely from the thoron and radon series.

(If you've counted only during the lab period, skip this step, but read the paragraph regardless.) Next, isolate the radon series by looking at the data from t = 5hour onward, with t = 0 marking the end of air sampling (NOT the beginning of counting). N.B. the times given here are rough guidelines, and may need to be adjusted based on the amount of observed activity. You should also truncate the dataset where appropriate before doing fits. At this point, the RaB and RaC have decayed through many half-lives, and what remains is the  $^{232}$ Th subseries, which is now in secular equilibrium. You can fit this curve with a simple exponential function. Then, subtract the fitted function from the rest of the data. 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?

Finally, remove data from t=0 until t=15minute. By then, the initial RaA will have decayed through five half-lives, so it will be mostly depleted. RaA  $\alpha$  decays, so its decays are not observable with the G-M detector, but they do produce RaB, and the model we will use does not account for this.

# A model for $\beta$ 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 [?].

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  $\varepsilon$ :

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

 $\varepsilon$  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  $\beta$  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.

If we define a term for the ratio of initial activities of RaC and RaB:

$$R = \frac{A_C(0)}{A_B(0)} \tag{3}$$

we can write the total initial activity in terms of  $A_B(0)$ , R, and the detector efficiencies:

$$A^{\text{obs}}(0) = A_B^{\text{obs}}(0) + A_C^{\text{obs}}(0) = A_B^{\text{obs}}(0) \left(1 + \frac{\varepsilon_C}{\varepsilon_B} R\right)$$
(4)

Along with the general expression for radioactivity as a function of time:

$$A(t) = A(0) e^{-\lambda t} \tag{5}$$

we can derive an expression for the radioactivity of the RaB-RaC system as a function of time:

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

 $\beta$  energy spectra are continuous on the interval  $[0, E_{\rm max}]$ . The value of  $E_{\rm max}$  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 & Taylor in Figure 2? 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?

Revised in 2017 by Aaron Liblong. Previous version: 2013 by Ruxandra Serbanescu with help from Bogdan Scaunasu and Marko Korelek

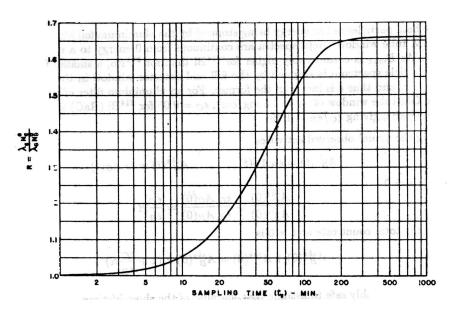


Figure 2: Predicted value of R as a function of air sampling duration (from Whyte & Taylor  $\cite{[?]}$ )