RADIOACTIVITY IN THE AIR

REFERENCES

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INTRODUCTION

This experiment involves filtering outdoor air, and then measuring and identifying the radioactive substances deposited on the filter paper. The amount and type of radioactivity collected generally depend on the length of time for filtration, on the weather conditions on a given day and occasionally on emissions froth industrial sources.

The three natural radioactive families you are likely to observe originate with the isotopes ²³⁸U, ²³²Th and ²³⁵U. All three occur naturally with half-lives greater than the age of the earth. They each decay through a long chain of radioactive daughters, ending with stable ²⁰⁶Pb, ²⁰⁸Pb and ²⁰⁷Pb respectively. The members of the chain should be determined from the Chart of the Nuclides posted in the lab, or from any number of texts (see the references). In this experiment, the identification of the radioactive isotopes in your sample hinges on two criteria: the measured half-life and the type of radioactivity, alpha or beta. (Another possible means of identification could be by analysis of γ -ray energies. This method is an option suggested in the " γ -Ray Spectra" experiment)

²²²Rn (radon) is naturally present in the atmosphere because its progenitor, ²³⁸U, having a half-life of 4.47 x 10⁹ years (comparable to the age of the solar system) is naturally present in the earth. ²³⁸U decays by a succession of α and β decays (the "(4n + 2) series") to a chemically inert gas, ²²²Rn, which has a half-life of 3.82 days, long enough to diffuse out of the earth into the atmosphere. Once in the atmosphere, the ²²²Rn decays to chemically more active daughter products, starting with ²¹⁸Po.

The results of your experiment measuring the β -decay curves, will depend in part on how long the filtration is carried out. What is actually being measured here is the radioactivity of dust particles, and in a long filtration the short half-life daughter products already deposited on the filter will decay. (See Figure 1.)

The rate at which ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi (which are metallic atoms or ions) will accumulate on dust particles varies according to the availability of dust or smog. Thus in very clear conditions one might expect most of the ²¹⁸Po to α decay to ²¹⁴Pb without first becoming attached to dust since the half-life is only a few minutes. There will then be relatively more accretion of ²¹⁴Pb and ²¹⁴Bi onto atmospheric dust. On the other hand, under very smoggy conditions there will be much more ²¹⁸Po accretion on dust, and most of the observed radioactivity from filtered air may be decays from the filtered ²¹⁸Po. The *proportions* of ²¹⁴Pb, ²¹⁴Bi which are found initially on the filter may thus vary according to atmospheric conditions.

3.82^d
3.10^m
2¹⁸Po (RaA)
$$\longrightarrow$$
 Adheres to dust particles \longrightarrow 2¹⁸Po
 $\downarrow \alpha$
27.0^m
2¹⁴Pb (RaB) \longrightarrow Adheres to dust particles \longrightarrow 2¹⁴Pb
 $\downarrow \beta$
19.9^m
2¹⁴Bi (RaC) \longrightarrow Adheres to dust particles \longrightarrow 2¹⁴Bi
 $\downarrow \beta$
164^{µs}
2¹⁴Po
 $\cdots etc. \cdots$
2¹⁴Po

Fig. 1 – Model of successive radioactive decays from 222 Rn as tar as 214 Po. The series terminates at 206 Pb which is stable.

Another family of isotopes that you may detect comes from ²³²Th with a half-life of 1.40 x 10¹⁰ years. The "4n" succession of α and β decays produces a shorter-lived (55.6 seconds) isotope of radon, ²²⁰Rn (Th-Thoron), which can decay in the air through various isotopes, including ²¹²Pb (ThB), ²¹²Bi (ThC), ²⁰⁸Tl (ThC"), ultimately ending up as stable ²⁰⁸Pb. Some of these isotopes will be detectable in your experiment, either from their β activity or from their γ emissions.

The "4n + 3" series originates with 235 U (T_{1/2} = 7.04 x 10⁸ years) and produces very small amounts of 219 Rn (Actinon) with half-life of 3.9 seconds.

In this experiment you have an opportunity to investigate the various radioisotopes in the air at the time of sampling and, in particular, to evaluate the concentration of radon in the atmosphere. Your chief technique will involve following the beta particle emissions of the isotopes collected on filter paper. You will produce a mathematical model for the decay of these isotopes, with several parameters to be defined. You will then fit these parameters to your data.

THE EQUIPMENT

Counting Equipment

Geiger-Mueller Tubes: These are gas filled ionization detectors with an applied high voltage large enough that electron and ion multiplication takes place, so that every time ionizing radiation passes through the counter and produces ionization, there is a measurable negative charge accumulated on the centre wire anode. With the anode connected through a resistor to the power supply, a standard voltage negative pulse appears at the anode.

The Picker Scaler: This provides a high voltage power supply for the G-M tube, in addition to being a counter.

For the 1^{st} session of this experiment, you will be collecting data for a Cesium – 137 source, plotting your data in a Python program, taking the plateau curve of the data to determine the operating voltage, and then collecting data for background radiation.

For the 2nd session of this experiment, you will be using the air sampler in room 126 to retrieve air samples, then do an analysis of the model below (more detailed instructions follow)

For this experiment you will use the "Radioactivity In Air" (RIA) program to collect data from the Geiger-Mueller tubes. This program records the number of counts your Picker Scaler reads from the pulses shown in the oscilloscope.

For the oscilloscope, make sure these settings are upheld

- 1. Set the "Intensity" dial to maximum (for optimum brightness so you can observe the pulses accurately)
- 2. Set the "Position" (Vertical) dial to equilibrium (In other words, set it to be at the x-axis shown in the pulse terminal (the screen)
- 3. Set the "Position" (Horizontal) dial to equilibrium (You will see a straight line from one side of the pulse terminal to the other).
- 4. Set the "VOLTS/DIV" slider to so that the line in the pulse terminal appears clear and sharp
- 5. If there is a "FOCUS" dial on your oscilloscope, set it to equilibrium.
- 6. If there is a "SOURCE" switch on your oscilloscope, set it to "LINE" so that it doesn't flash every time there is a pulse.

Observe the construction of the G-M tube. Note the thin window at the end. *BE CAREFUL NOT TO DAMAGE IT*. This window has a thickness of between 1.4 and 2.0 mg/cm² which is thin enough to pass beta particles, but is thick enough to stop alpha particles of energy less than about 2 MeV. The gas is not very efficient for being ionized by γ rays so that this counter is most useful for beta particle detection.

To find out what kinds of pulses are produced by the GM tube, place a ¹³⁷Cs source in front of the GM counter, and connect it to the Picker Scaler "*detector input*" terminal with the high voltage set to a working value between 600 volts and 1,400 volts. **Note: When you reach the 1000 volts mark, switch off the oscilloscope.** Using the oscilloscope, observe the GM pulses appearing at the "*pulse input*" terminal. Note their voltage, shape and length. Note the effects of moving the source closer to and farther from the counter. Note the randomness of time of arrival of the pulses. Note the effects of varying the high voltage. Note the time the GM tube is "dead" after a pulse is produced. What is the "dead time" of this counter? Connect the oscilloscope to the "*output pulse*" terminal and observe the standardized pulse that the discriminator produces.

CAUTION: Some of the Picker Scaler connectors have high voltages on them, sufficient to damage an oscilloscope input. Be extra careful in connecting the oscilloscope to the 'pulse input' terminal. Moreover, connect the oscilloscope to the Picker Scaler 'pulse input' terminal only after the Picker Scaler has been set into operation. Also, do not switch the Picker Scaler high voltage setting while the oscilloscope is connected. When in doubt, consult a demonstrator.

Determine the operating voltage of the GM tube by taking a plateau curve. With ¹³⁷Cs as source near the front of the GM tube, measure and plot a curve of number of counts per standard time interval versus high voltage. The almost-horizontal flat portion of this curve is called the plateau. The operating voltage should be taken in this region where counter sensitivity is independent of high voltage. While taking the plateau curve, be sure that you monitor the pulses on the oscilloscope to make sure that they 'make sense'.

NOTE: In doing this experiment, make sure that you count long enough at each voltage setting, in order to make sure that counting statistics allow adequate determination of the count rate. In drawing your plateau curve, include error bars, using as a guide the statement that the standard deviation in the number, N, of random events observed is equal to \sqrt{N} .

To power the GM tube you will use the Picker Scaler which also serves the function of a discriminator, giving a more or less standard 3 Volt negative pulse at its *pulse output* terminal for any GM pulse which is large enough. You can then feed this pulse into the input (in) connector of the pulse shaper. The output (out) of the pulse shaper is a positive pulse which can be fed into the analyzer.

To open the "Radioactivity in the Air" program: go to Desktop and open it from there.

Before starting data collection, make sure the Picker Scaler and the Oscilloscope are switched on with the "Power" button and switch respectively. Make sure the voltage setting is at minimum before flipping the "High Voltage" switch into the "ON" position.

In the RIA program, set the "Total Sample Time (min)" box to the number of minutes you want to collect data and set the "Individual Sample Time (sec)" to 20 seconds (ideal).

To start data collection after setting the Picker Scaler to an appropriate voltage (again between 600V and 1000V), flip the "Count" switch on the Picker Scaler to the "ON" position and in the RIA program click the "Start" button.

If you want to stop data collection early or just want to pause data collection, click the "Stop" button in the RIA program.

Once you are finished with data collection, flip the "Count" switch on the Picker Scaler to the "OFF" position.

Note: Before starting a new data collecting session in the RIA program, make sure the "Count" switch is in the "OFF" position to reset the counter on the Picker Scaler to all zeros.

To export the data into a file type of your choice (preferably "txt" file), after waiting a couple of minutes after clicking "STOP" in the program, click "Export Data".

To clear the data from a previous data collection, select "Clear Data" in the program after stopping data collection.

Connect the GM tube to the Picker Scaler, the pulse shaper, and the DAQ card. Set an appropriate high voltage. Place your Cesium -137 source in the GM tube and follow the instructions above to record the data of the decay of the source. Do this for at least 1 hour.

Afterwards, take the Cesium – 137 source out of the GM tube and give it back to the lab technologist as you won't be using it again.

For the rest of the 1st session, collect background radiation (no source in the GM tube) from the room by the same method as above.

Follow pulses down the system, using an oscilloscope, to see what they look like and to check that they are satisfactory.

Sampling

You will want to have the air sampler run between one and two hours. Your counting equipment should be ready to receive the sample when you turn off the sampler. Minimize the time between the stop of the sampler and the beginning of counting. It will be important to record the actual times of sampling stop and data-taking beginning.

Approach to Data Analysis

There are various possible levels of sophistication in your approach to analyzing the data. You can do a constant background subtraction on your data and then, using a semi-logarithmic plot, find a rough half-life and total counting rate at the time sampling stopped.

At the most complex level (an extension to the experiment), you could do a full-blown analysis using the model described below, in addition to observing gamma-rays from the filter paper. You should read through the following pages and then decide for yourself the complexity of the analysis you wish to undertake.

Processing Your Data and/or Calculate Your Model

The data collected can be saved on a memory stick, or sent to your email as attachment. This option allows you to import the data into a Python script using the Poisson model.

THE MODEL

Your model for counts registered in the detector consists of three parts:

1. The first part is the constant background observed when no sample is placed under the detector. This is the result of cosmic rays and gamma-rays present in the room. It is easily determined by making a sufficiently long background count.

Since the amount of radioactivity in your sample is likely to be small, it is essential to determine the background (no sample) count rate accurately. Be sure to determine the background under identical conditions to those you will be using for your measurements, and count long enough to reduce the error in the background to ≤ 1 count per minute. As already indicated, if you observe *n* counts, the standard error is \sqrt{n} . (You can use the same setup for observing background as you use in the decay observations.)

2. The second part is the decay of long-lived isotopes that are accumulated on the filter paper. These counts will probably be dominated by the decay of the "4n" series, generated by ²³²Th, which results in ²²⁰Rn (55.6^s) \rightarrow ²¹⁶Po (0.145^s) \rightarrow ²¹²Pb (10.64^h) \rightarrow ²¹²Bi (1.00^h) \rightarrow ²¹²Po (0.3^{µs}) [alternative ²⁰⁸Tl (3.05^m)] \rightarrow ²⁰⁸Pb. The half-life of 10.64 hours of ²¹²Pb will probably dominate this activity. However you may find other long-lived activities.

These longer-lived components can be identified from a semi-logarithmic plot of the detector counts with background subtracted, for times after the half-hour activities from 222 Rn have decayed. Note: Only the first half hour data is relevant to the decay of the short lived products of 222 Rn.

Python Requirement:

Make a linear fit of the data after the half hour from your air sample. If a good linear fit is obtained, you can further subtract this component from the total data. You now will have corrected data which will lead to the third part of the model.

3. The third part deals with the decay products of 222 Rn and is detailed below.

Secular Equilibrium

When radon (²²²Rn) alpha-decays, the resulting ²¹⁸Po is highly ionized and some becomes attached to dust particles. (See figure I.) The subsequent beta decays produce ²¹⁴Pb and ²¹⁴Bi which are also highly ionized, have half-lives of the order of a half-hour and attach themselves to dust particles. The number of disintegrations per second of any isotope is proportional to the amount of that isotope. For example, the more ²¹⁸Po there is, the more will decay per second and soon the amount of ²¹⁸Po will build up to an equilibrium value which depends on the lifetime of ²¹⁸Po and the amount of the ²¹⁸Po being formed per second from its predecessor, radon. At equilibrium, the number of decays of ²¹⁸Po per second is equal to the number of ²¹⁸Po nuclei being created per second must be equal to the number of radon decays per second and in the same way the number of ²¹⁴Pb nuclei being created per second is equal to the number of ²¹⁸Po decays per second. This continues down the chain and means that the number of decays per second of radon, ²¹⁸Po, ²¹⁴Pb, and ²¹⁴Bi are equal. This is known as secular equilibrium and holds whenever the local parent (in this case radon) has a long life compared to the subsequent generations. Thus, the measurement of the disintegration rate of anyone of the series will give the activity of the radon present.

Thus we should be able to measure the beta activity from dust in the air and account for it by considering only the decay of ²¹⁴Pb and ²¹⁴Bi if we eliminate alpha particles by using an aluminum filter thick enough to absorb all the alpha particles. *If this model fits the data*, then by determining the amount of one component (in our case ²¹⁴Pb) we can determine the concentration of radon in the atmosphere.

The Mathematical Model for the Daughters of ²²²Rn

For a general treatment of radioactive decay chains, and for a detailed derivation of the following equations, see the *Whyte and Taylor* reference.

Although ²¹⁸Po, being an alpha emitter, is not counted directly, its continuous arrival and rapid decay during the sampling period increases the amount of ²¹⁴Pb present. However, once the filtering has stopped, the small number of ²¹⁸Po atoms present has little effect (except for a small augmentation of ²¹⁴Pb during the first 10 to 15 minutes) and the subsequent counting rate can be described solely in terms of ²¹⁴Pb and ²¹⁴Bi. The following treatment will neglect any build-up of ²¹⁴Pb from ²¹⁸Po on the filter paper.

The variation of counting rate with time will be a function of the decay constants of 214 Pb and 214 Bi and of their relative activities at the start of the counting period. If we call $A_B(t) =$ the actual activity of 214 Pb (RaB) at time t and $A_C(t) =$ the actual activity of 214 Bi (RaC) at time t, measured from when air sampling is stopped. We consider a ratio R which is at time t = 0. The relative amounts of isotopes "C" and "B" on the filter paper will be:

$$R = \frac{A_{\rm C}(0)}{A_{\rm B}(0)} \tag{1}$$

The actual observed counting rate in the detector is equal to the actual activity times a detector efficiency ϵ times a geometrical term. The detector efficiency is mainly the result of the beta particles of low energy being stopped by the aluminum foil alpha absorber and the GM tube window. Beta spectra are continuous from 0 to a maximum energy E_{max} . As E_{max} is considerably

higher for ²¹⁴Bi than for ²¹⁴Pb, a smaller fraction of the beta particle spectrum is stopped by the foil and detector window in the former than in the latter, and thus ε is larger in the former. For an aluminum filter of 27 µm thickness and a GM tube window of $\rho x = 1.5 \text{ mg/cm}^2$, $\varepsilon_C = 0.95 \text{ for }^{214}\text{Bi}$ (RaC) and $\varepsilon_B = 0.80$ for ²¹⁴Pb (RaB), giving $\varepsilon_C/\varepsilon_B = 1.19$.

Calling the actual observed activities:

 $A_B^{0bs}(t) = \epsilon_B A_B(t) \qquad \qquad A_C^{obs}(t) = \epsilon_C A_C(t)$

then at t = 0:
$$\frac{A_C^{obs}(0)}{A_B^{obs}(0)} = \frac{\varepsilon_C}{\varepsilon_B} \frac{A_C(0)}{A_B(0)} = \frac{\varepsilon_C}{\varepsilon_B} R$$

Thus, the total count rate at t = 0 is:

$$A_{B}^{obs}(0) + A_{C}^{obs}(0) = A_{B}^{obs}(0) \times \left(1 + \frac{\varepsilon_{C}}{\varepsilon_{B}}R\right)$$

It is reasonably safe to assume that, because of the short lifetime of ²¹⁸Po, very little gets attached to airborne particulates and that most of what does get onto the filter paper decays before the end of sampling. Thus we have decided to ignore any presence of 218Po on the filter paper.

You can derive the relations:

$$A_{B}^{obs}(t) = A_{B}^{obs}(0)e^{-\lambda_{B}t}$$

$$A_{C}^{obs}(t) = \frac{\varepsilon_{C}}{\varepsilon_{B}}A_{B}^{obs}(0)\left[\frac{\lambda_{C}}{\lambda_{C}-\lambda_{B}}e^{-\lambda_{B}t} + \left(R - \frac{\lambda_{C}}{\lambda_{C}-\lambda_{B}}\right)e^{-\lambda_{C}t}\right]$$

$$A_{B}^{obs}(t) + A_{C}^{obs}(t) = A_{B}^{obs}(0) \times \left[\left(1 + \frac{\varepsilon_{C}}{\varepsilon_{B}}\frac{\lambda_{C}}{\lambda_{C}-\lambda_{B}}\right)e^{-\lambda_{B}t} + \frac{\varepsilon_{C}}{\varepsilon_{B}}\left(R - \frac{\lambda_{C}}{\lambda_{C}-\lambda_{B}}\right)e^{-\lambda_{C}t}\right]$$

where λ is the radioactive decay constant $\lambda = \ln 2/(\text{half-life})$. The total observed counting rate as a function of time is: $A_B^{obs}(t) + A_C^{obs}(t)$.

Comparison of the Model to Your Data

Part 3 of the model yields a predicted decay curve with two parameters, $A_B^{obs}(0)$ and R. Your problem is to find if you can get a good fit and to establish the best values for the parameters. (This must be done on data which has first been corrected for parts 1 and 2.) A first approximation to $A_B^{obs}(0)$ can be estimated by extrapolating the count rate curve back to t = 0, noting that total

counts at (t = 0) is: $A_B^{obs}(0) + A_C^{obs}(0) = A_B^{obs}(0) \left(1 + \frac{\varepsilon_C}{\varepsilon_B}\right)$. A first approximation to R can be

estimated, noting that the *Whyte and Taylor* reference calculates (using certain assumptions) a predicted value of R as a function of sampling time. Their plot of this relation is shown in Fig. 2.



Fig 2. – Dependence of the ratio R on sampling time

Python Requirement:

One way of comparing your model to the data is by graphing data against model prediction, and using a least squares fit to a straight line through the origin. Generate this exact kind of fit of the data against the model described. If you can minimize χ^2 , and produce a reasonable value of χ^2 , you probably have a good fit.

Revised RMS in 2013 with help from Bogdan Scaunasu and Marko Korelek