Natural Radiation

There are a few radioisotopes that exist in our environment. Isotopes that were present when the earth was formed and isotopes that are continuously produced by cosmic rays can exist today if they have long enough half-lives. Here we will discuss 5 of these isotopes. Four were produced when the earth was formed: ^{40}K (half-life of 1.277×10^9 years), ^{238}U (half-life of 4.51×10^9 years, ^{232}Th (half-life of 1.4×10^{10} years), and ^{235}U (half-life of 7.1×10^8 years). ^{14}C (half-life 5280 years) is continuously produced in the upper atmosphere by cosmic rays entering the earth.

^{40}K

Most of the potassium found on earth is the stable ^{39}K. However, a small fraction, 0.0117%, of all potassium is ^{40}K. ^{40}K is radioactive with a half-life of 1.277×10^9 years. ^{40}K decays in the following way: ^{40}K has an 89.28% chance to undergo beta decay to the ground state of ^{40}Ca, and a 10.72% chance to undergo electron capture to ^{40}Ar. When electron capture occurs it is almost always to the excited state of ^{40}Ar, 99.53% of the time. From the excited state, ^{40}Ar decays to the ground state emitting a gamma particle with energy 1460.83 KeV. Thus when ^{40}K decays, there is a 89.28% chance a beta particle is emitted and a 10.72(0.9953) = 10.67% chance a gamma is emitted.

Although 0.0117% seems like a small amount, it means that one out of every 8500 potassium atoms is radioactive. This is a large amount! The average 70 Kg man has around 140 grams of potassium in his body. So the number of radioactive ^{40}K nuclei in his body is:

$$Number\ of\ ^{40}K\ nuclei = \frac{140g}{40g}(6.02 \times 10^{23})(0.000117) = 2.47 \times 10^{20} \tag{1}$$

which is a lot of nuclei. The activity due of ^{40}K decays in the average person can be calculated using the relation: activity $A = N\lambda$, or $A = N(ln2)/\tau$:

$$A = \frac{2.47 \times 10^{20} ln(2)}{1.277 \times 10^9 (365)(24)(3600) sec} = 4240 \frac{decays}{sec} \tag{2}$$

This amount of activity is equal to 4240/37000 = 0.11 μCi. Perhaps we should wear a radioactive sign around our necks! Fruits and vegetables can have as much as 0.4% potassium, and an average soil sample contains 2% potassium. Thus, one Kg of soil has an activity of 0.016 μCi due to the potassium content alone. By measuring the gamma spectrum of soil and food samples for long times, one or two hours, the potassium content can be measured to an accuracy of as good as 10%. ^{40}K is the largest contributor to our natural background radiation.

^{238}U

Another isotope found in the earth is uranium 238. Due to its long half-life some still remains since the formation of the earth. ^{238}U has a long decay series, undergoing alpha, beta and gamma decays until it finally becomes stable as lead 206, ^{206}Pb. We list the complete ^{238}U decay series:

Isotope	half-life	gamma energies
^{238}U	4.468×10^9 years	—-
^{234}Th	24.1 days	63.3 (3.67%) 92.38 (2.13%) 92.80 (2.10%) 112.8 (0.21%)
^{234m}Pa	1.17 minutes	765 (0.317%) 1001 (0.842%)
^{234}U	2.47×10^5 years	53.2 (0.123%)
^{230}Th	7.54×10^4 years	67.7 (0.377%)
^{226}Ra	1602 years	186.2 (3.59%)
^{222}Rn	3.823 days	510 (0.076%)
^{218}Po	3.05 minutes	——
^{214}Pb (99.98%)	26.8 minutes	53.2 (1.1%) 242.0 (7.25%) 295.2 (18.4%) 351.9 (35.6%) 785.9 (1.06%)
^{218}At (0.02%)	2 seconds	——
^{214}Bi	19.7 minutes	609.3 (45.5%) 768.4 (4.89%) 806.2 (1.26%) 934.1 (3.11%) 1120.3 (14.9%) 1238.1 (5.83%) 1377.7 (3.99%) 1408.0 (2.39%) 1509.2 (2.13%) 1729.6 (2.88%) 1764.5 (15.3%) 1838.4 (0.35%) 1847.4 (2.03%) 2204 (4.92%) 2447.7 (1.55%)
^{214}Po (99.98%)	164 microsec	799 (0.014%)
^{210}Tl (0.02%)	1.3 minutes	296 (80%) 795 (100%) 1310 (21%)
^{210}Pb	21 years	46.5 (4.05%)
^{210}Bi	5.01 days	——
^{210}Po	138.4 days	803 (0.0011%)
^{206}Pb	Stable	

In the first column we list the isotopes in the decay series. If the decay is Alpha emission, the atomic number is lowered by 2 and the atomic mass is lowered by 4. For a Beta decay process, the atomic number increases by 1 and the atomic mass remains unchanged. In the middle column we list the half-life of the radioisotope shown in the first column.

In the last column we list the energy of the gamma rays emitted by the isotope in the first column. The number in parentheses is the yield percentage of the gamma. The isotopes in the series are called the daughter isotopes of ^{238}U. From the table, one can see that as ^{238}U decays via its daughter isotopes many alpha, beta and gamma particles are emitted.

Consider the following question: During the long decay series, how many of the various daughter isotopes are present at any one time? The half-lives are listed in the center column. Are there a smaller number of isotopes with short half-lives than isotopes with long half-lives at any particular moment? Any particular isotope will have a certain rate at which it is produced, and a rate at which it decays. Consider the situation in which isotope A decays to isotope B, which decays to isotope C. Let N_A be the number of nuclei of isotope A, N_B be the number of nuclei of isotope B, and N_C be the number of nuclei of isotope C. Let λ_A, λ_B, and λ_C be the corresponding decay constants for the decays. The rate at which the number of nuclei of isotope B decreases is $\lambda_B N_B$, which is just the decays/sec or the activity. The rate of formation of isotope B is just the decay rate of isotope A, $\lambda_A N_A$. So the change in the number of nuclei of isotope B per unit time is given by:

$$Change\ of\ N_B\ per\ unit\ time = +\lambda_A N_A - \lambda_B N_B \tag{3}$$

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B \tag{4}$$

This rate-of-change equation applies to every radioactive nucleus in the decay series. The first nucleus, ^{238}U, will only have the decay term $-\lambda N$, and the final nucleus, ^{206}Pb, will only have the rate of formation term, $+\lambda N$. The solution to the series of differential equations is complicated. However, if we observe the decay series after a long time, long enough for the series to come into equilibrium, then the solution is simple. After a long time, the number of radioactive nuclei of a particular isotope remains constant in time, $dN_B/dt = 0$. The rate of formation, $+\lambda_A N_A$, is equal to the rate of decay, $-\lambda_B N_B$:

$$\lambda_A N_A = \lambda_B N_B \tag{5}$$

for every isotope in the decay series. This equilibrium condition is referred to as *secular equilibrium*. Since λN is equal to the activity of an isotope, **if the decay series is in secular equilibrium the activity of each isotope in the series is the same**. In terms of the half-lives of the isotopes, we have:

$$\frac{N_A}{\tau_A} = \frac{N_B}{\tau_B} = \frac{N_C}{\tau_C} \tag{6}$$

Isotopes with longer half-lives have more nuclei at any particular time in the decay series than isotopes with shorter half-lives.

In the experiment where we measure the spectrum of Brazil nuts, the decay series might not be in secular equilibrium. In fact, by measuring the counts under the appropriate photopeaks we can determine the age of the nuts. As before, suppose there are three isotopes A, B, and C, where $A \to B \to C$. If originally at $t = 0$ there were N_0 radioactive isotopes of A, then the number of A isotopes at time t is given by:

$$N_A = N_0 e^{-\lambda_A t} \tag{7}$$

Since $dN_B/dt = N_A\lambda_A - N_B\lambda_B$, we have

$$\frac{dN_B}{dt} = \lambda_A N_0 e^{-\lambda_A t} - \lambda_B N_B \tag{8}$$

The solution to this differential equation with the initial condition that $N_B(t = 0) = 0$ is (you should check the solution):

$$N_B = \frac{\lambda_A N_0}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t}) \tag{9}$$

Since the activity of B, $Act_B = \lambda_B N_B$ and the activity of A, $Act_A = \lambda_A N_A$ we have:

$$\frac{Act_B}{Act_A} = (\frac{\lambda_B}{\lambda_A})\frac{\lambda_A}{\lambda_B - \lambda_A}(1 - e^{-(\lambda_B - \lambda_A)t}) \tag{10}$$

In terms of the half lives of the isotopes, $\tau = ln2/\lambda$, the expression takes on the form:

$$\frac{Act_B}{Act_A} = \frac{\tau_A}{\tau_A - \tau_B}(1 - (\frac{1}{2})^{(\frac{\tau_A - \tau_B}{\tau_A \tau_B})t}) \tag{11}$$

How much time is needed for the series to reach secular equilibrium? The equation above describes the situation. If $\tau_A >> \tau_B$, then the ratio of activities reduces to $1 - (1/2)^{t/\tau_B}$. If $t >> \tau_B$ then the ratio of activities reduces to one and we have secular equilibruim. For the ^{238}U series, $\tau_A = 4.468 \times 10^9$ years. Since ^{234}U has the longest half-life of the series of $2.47x10^5$ years $\tau_A >> \tau_B$. Also, since ^{238}U was formed at a time much longer than this (at the earth's beginning), $t >> \tau_B$ and we can assume that today the ^{238}U series is at secular equilibrium. However, if radon, an inert gas, can escape the sample container, the activities of the isotopes before ^{222}Rn can be different than those after ^{222}Rn. Analyzing the combined 186 KeV gamma peak of ^{226}Ra and ^{235}U can assist in estimating how much radon has escaped the sample.

For the Brazil nuts sample, the situation is a bit more complicated. In the Appendix we derive the relevant equation to use. We find that

$$\frac{Act_B}{Act_A} \approx 1.5 - 1.30(\frac{1}{2})^{t/2.86} \tag{12}$$

where Act_B is the activity of the isotopes after ^{228}Th, and Act_A is the activity ob the isotopes before ^{228}Th. By measuring the ratio of activities we can determine the age of the Brazil nuts.

A good part of our natural background comes from the ^{238}U decay series. One of the isotopes ^{222}Rn is an inert gas. It can collect in rooms, and enter our lungs when we breath. When it decays, ^{218}Po is produced. ^{218}Po is not a gas, and can settle in the inner wall of the lungs. Having the remainder of the decay series take place inside the lungs can cause serious damage.

The best peaks in the gamma spectrum from the ^{238}U decay series to start your analysis with are the 609 KeV (^{214}Bi), 352 KeV (^{214}Pb), 295 KeV (^{214}Pb) and 1120 KeV (^{214}Bi). These peaks should be easy to find, and are also in the background spectra.

^{232}Th

Another isotope that makes up part of our natural background is ^{232}Th. Below we list the complete ^{232}Th decay series:

Isotope	half-life	gamma energies
^{232}Th	1.405×10^{10} years	63.8 (0.267%)
^{228}Ra	5.75 years	——
^{228}Ac	6.13 hours	57.7 (0.487%) 99.5 (1.26%) 129.0 (2.42%) 154.0 (0.722%) 209.3 (3.89%) 270.2 (3.46%) 328.0 (2.95%) 338.3 (11.3%) 409.5 (1.92%) 463.0 (4.40%) 772 (1.49%) 794.9 (4.25%) 835.7 (1.61%) 911.2 (25.8%) 964.8 (4.99%) 969.0 (15.8%) 1588.2 (3.22%)
^{228}Th	1.91 years	84.4 (1.19%)
^{224}Ra	3.64 days	241.0 (4.10%)
^{220}Rn	55 seconds	550 (0.07%)
^{216}Po	0.15 seconds	——
^{212}Pb	10.64 hours	238.6 (43.6%) 300.0 (3.30%)
^{212}Bi	60.6 minutes	39.9 (1.1%) 727.3 (6.67%) 785.4 (1.10%) 1620.5 (1.47%)
^{212}Po (64.06%) ^{208}Tl (35.94%)	304 nsec 3.1 minutes	—— 277.4 (6.6%) 510.77 (22.6%) 583.2 (84.5%) 763.1 (1.79%) 860.6 (12.5%) 2614.5 (99.8%)
^{208}Pb	stable	

Since the longest half-life other than the parent nucleus (^{232}Th) is 6.7 years for ^{228}Ra, the series is in secular equilibrium today. The best peaks in the gamma spectrum from the ^{232}Th decay series to start your analysis with are the 338 KeV (^{228}Ac), 911 KeV (^{228}Ac), 969 KeV (^{228}Ac), 238 KeV (^{212}Pb), and 583 KeV (^{208}Tl). These peaks should be easy to find, and are also in the background spectra.

^{235}U

A very small part of natural radiation is from the ^{235}U decay series. ^{235}U is found in ores with ^{238}U. On the average 0.7% of the uranium in ores is ^{235}U. We list the complete ^{235}U decay series below:

Isotope	half-life	gamma energy (KeV)
^{235}U	7.038×10^8 years	143.8 (10.96%) 163.33 (5.08%) 185.7 (57.0%) 205.3 (5.02%)
^{231}Th	25.5 hours	25.64 (14.1%) 84.2 (6.6%)
^{231}Pa	3.25×10^4 years	27.4 (10.5%) 300 (2.41%) 302.7 (2.87%)
^{227}Ac	21.6 years	70 (0.08%)
^{227}Th (98.6 %) ^{223}Fr (1.4%)	1.82 days 22 minutes	50.1 (8.4%) 236.0 (12.9%) 256.3 (7.0%) 300 (2.21%) 50.1 (34%) 79.7 (8.7%) 234.8 (3%)
^{223}Ra	11.43 days	144.2 (3.27%) 154.2 (5.7%) 269.5 (13.9%) 323.9 (3.99%) 338.3 (2.84%)
^{219}Rn	4 seconds	271.2 (10.8%) 401.8 (6.6%)
^{215}Po	1.78 millisec	——
^{211}Pb	36.1 minutes	404.9 (3.78%) 427.1 (1.76%) 832.0 (3.52%)
^{211}Bi	2.15 minutes	351.1 (13.02%)
^{211}Po (0.28%) ^{207}Tl (99.7%)	0.52 seconds 4.79 minutes	569.6 (0.0016%) 897.8 (0.26%) 897 (0.16%)
^{207}Pb	stable	

The series is at secular equilibrium today, since the longest half-life of the isotopes in the series is 3.25×10^4 years. ^{235}U is an interesting isotope because of its fission properties. A single neutron can initiate fission. When ^{235}U undergoes fission, it can release 3 neutrons. Each of these three neutrons can initiate another fission reaction, and a chain reaction can develop if the concentration of ^{235}U is large enough. Because of its fission properties, ^{235}U was the first radioisotope used for nuclear energy and weapons.

The peaks in the background gamma spectum due to ^{235}U are small and difficult to observe. However, with the high resolution of a Ge detector, the three gammas at 143 KeV, 163 KeV, and 205 KeV emitted by ^{235}U as well as the gamma at 236 KeV (^{227}Th) can sometimes be seen in a uranium sample. One can also use the double peak fit for the 185.7 KeV gamma from ^{235}U and the 186.2 KeV gamma from ^{238}U to obtain information regarding the natural abundance of ^{235}U.

1 Appendix

The dynamics of the radioisotope decay chain are described by the Bateman equations We need to apply the Bateman equations to the uptake and subsequent decay of Radium in Brazil nuts. There are two stages to consider: 1) the period when the Brazil nut is growing in the tree and 2) the period after the Brazil nut has fallen to the ground. In both cases there are essentially two half-lives that enter the calculation, the half-life of Ra^{228} and that of Th^{228}. This is because these two isotopes have significantly longer half-lives than the other isotopes in the decay series. The half-life of Ra^{228} is 5.75 years, the half-life of Th^{228} is 1.91 years, and the next longest half-life is only 3.64 days for Ra^{224}. Consequently there are essentially only two relevant activities. One activity is that of Ra^{228} and Ac^{228} which are equal. The other activity is that of Th^{228} and all its daughters, which also have the same activity as Th^{228} after a few weeks.

A. First Stage: Brazil Nut growing

Consider the first stage when the Brazil nut is growing in the tree. Let $N_A(t)$ be the number of Ra^{228} nuclei in the nut at time t while the nut is growing. Then N_A satisfies the differential equation:

$$\frac{dN_A}{dt} = \alpha - N_A \lambda_A \tag{13}$$

where α is the rate at which Ra^{228} is brought up into the nut, and λ_A is the decay constant for Ra^{228}. The solution to this equation is

$$N_A = \frac{\alpha}{\lambda_A}(1 - e^{-\lambda_A t}) \tag{14}$$

for the initial condition $N_A(0) = 0$. Since the activity of Ra^{228} is equal to $\lambda_A N_A$ we have

$$Act(Ra^{228}) = \alpha(1 - e^{-\lambda_A t}) \tag{15}$$

for the activity of Ra^{228} in the Brazil nut after a growing period of time t.

Using Eq. 5, we can determine the activity of Th^{228} in the Brazil nut during its growth. Let N_B be the number of Th^{228} nuclei in the nut at time t while the nut is growing. N_B satisfies the differential equation:

$$\frac{dN_B}{dt} = \lambda_A N_A - N_B \lambda_B \tag{16}$$

where λ_B is the decay constant for Th^{228}. The quantity $\lambda_A N_A$ is the activity of Ra^{228}, which is the rate at which Th^{232} is produced. We use the activity of Ra^{228} in this expression since both Ac^{228} and Ra^{228} have essentially the same activity due to the short half life of Ac^{228} compared to Ra^{228}.

Solving the above equation for N_B gives

$$N_B = \alpha(\frac{\lambda_A}{\lambda_B(\lambda_B - \lambda_A)} e^{-\lambda_B t} + \frac{e^{-\lambda_A t}}{\lambda_A - \lambda_B} + \frac{1}{\lambda_B}) \tag{17}$$

for the initial condition $N_B(0) = 0$. As before, the activity of Th^{228} is given by $\lambda_B N_B$, so we have

$$Act(Th^{228}) = \alpha(\frac{\lambda_A}{\lambda_B - \lambda_A} e^{-\lambda_B t} + \frac{\lambda_B e^{-\lambda_A t}}{\lambda_A - \lambda_B} + 1) \tag{18}$$

for the activity of Th^{228} in the Brazil nut after a growing period of time t.

The main factor in determining the age of the Brazil nut is the ratio of the activities of Th^{228} to that of Ra^{228}, $Act(Th^{228})/Act(Ra^{228})$. We will need to know what this ratio is when the nut falls from the tree. For a growing time of $t = 14\ months$ and $\lambda_A = ln(2)/5.75\ years^{-1}$ we have $Act(Ra^{228}) \approx 0.131\alpha$ from Eq. 5. Using these parameters and $\lambda_B = ln(2)/1.91\ years^{-1}$ gives $Act(Th^{228}) \approx 0.026\alpha$ from Eq. 8. The ratio of the Th^{228} to the Ra^{228} activity is therefore

$$\frac{Act(Th^{228})}{Act(Ra^{228})} \approx \frac{0.026\alpha}{0.131\alpha} \approx 0.20 \tag{19}$$

when the nut falls from the tree. Note that the rate of uptake, α, cancels out in this ratio.

B. Second Stage: After falling from the tree

Now consider stage two, after the nut has fallen from the tree and decays on its own. The number of Ra^{228} isotopes in the nut, N'_A satisfies the differential equation

$$\frac{dN'_A}{dt} = -N'_A\lambda_A \tag{20}$$

which is the same as before without the growth term α. The solution for times t after the nut falls from the tree is

$$N'_A(t) = N'_0 e^{-\lambda_A t} \tag{21}$$

where N'_0 represents the initial number of Ra^{228} nuclei just after the nut falls from the tree.

As in the initial stage, the number of Th^{228} nuclei, N'_B satisfies the differential equation

$$\frac{dN'_B}{dt} = \lambda_A N'_A - N'_B\lambda_B \tag{22}$$

After substituting $N'_0 e^{-\lambda_A t}$ for N'_A, and solving the differential equation, one obtains

$$N'_B(t) = \frac{\lambda_A N'_0}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t}) + N'_B(0)e^{-\lambda_B t} \tag{23}$$

Here $N'_B(0)$ is the initial number of Th^{228} nuclei. To compare with experiment, one needs to calculate the ratio of the activities of Th^{228}, $\lambda_B N'_B(t)$, to Ra^{228}, $\lambda_A N'_A(t) = N'_0 e^{-\lambda_A t}$. Using the equations above we obtain

$$\frac{Act(Th^{228})}{Act(Ra^{228})} = \frac{\lambda_B}{\lambda_B - \lambda_A}(1 - e^{(\lambda_A - \lambda_B)t}) + 0.20e^{(\lambda_A - \lambda_B)t} \tag{24}$$

since the initial ratio of $Act(Th^{228})/Act(Ra^{228})$ is approximately 0.20. Using the decay constants $\lambda_A = ln(2)/5.75\ years^{-1}$ and $\lambda_B = ln(2)/1.91\ years^{-1}$ we obtain the expression that can be compared with the data:

$$\frac{Act(Th^{228})}{Act(Ra^{228})} \approx 1.50 - 1.30(\frac{1}{2})^{t/2.86} \tag{25}$$

where t is in years and we have changed from base e to base 2. Note that the ratio of activities varies from 0.20 to 1.5 with an effective "half life" of 2.86 *years*.