

Decay of Radon Daughters Sampled from Air

September 13, 2002

In this experiment you will study radioactive decays. You will experimentally verify that the time dependence is described by an exponential function. The investigation of a more complex decay scheme will allow you to apply coupled differential equations. Decay data taken with short-lived activities, sampled from the air, will allow you to determine the concentration of the radioactive noble gas ^{222}Rn in the air and therefore demonstrate the existence of *natural radioactivity*.

The formal discussion of the subject follows that made in *G.N. Whyte and H.W. Taylor, American Journal of Physics 30 (1962) 120-124*. In their experiment a Geiger counter, giving no energy information, was used to measure the combined effect of all Radon (^{222}Rn) decay products collected from the air by sucking a known volume of air through a filter.

We will use a NaI scintillation detector to measure the gamma radiation emitted in the decay of the Rn daughters. This will allow us to determine the activities of the Rn daughters *separately* through a statistical analysis of the gamma-ray spectra collected. We will use the data to examine the assumptions made in this publication.

1 Tasks

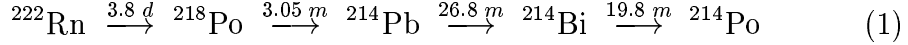
1. Read and understand the experiment description.
2. Solve the pre-lab problems given in the write-up.
3. Collect Radon decay daughters from the air on a filter.
4. Count the filter in order to quantify the activities of the unstable Radon daughters as a function of time.
5. Determine the half lives and activities of ^{214}Pb and ^{214}Bi by means of a statistical analysis. In your write-up you have to show graphs of the ^{214}Pb and ^{214}Bi activity versus time since end of filtering.
6. Determine the Rn activity per unit volume from this data.
7. Determine the ratio of the ^{214}Bi over the ^{214}Pb activity at the end of filtering (called $R(t)$ in *G.N. Whyte and H.W. Taylor*). Compare your measured value of $R(t)$ to the expectation as give in figure 1 of *G.N. Whyte and H.W. Taylor*. Is your data compatible with this paper? Explain why or why not.

2 Introduction

The radioactive noble gas ^{222}Rn ($T_{1/2}=3.8$ d) is a member of the ^{238}U decay series. It can be found in trace concentrations everywhere, as it is gaseous and relatively long lived. It is released by minerals into the air where it exists in some equilibrium concentration. There is another species of Rn present in the air: the ^{232}Th decay series, also naturally present in our environment, has ^{220}Rn ($T_{1/2}=55.6$ s) as a member. Due its short half live its equilibrium concentration is much lower than that of ^{222}Rn . We can hence neglect its effect in this experiment. Airborne Rn-gas is one of the principal components of our natural radiation exposure.

Rn decays into unstable elements having short half lives. Equation 1 shows the ^{222}Rn decay sequence. This sequence offers a convenient way to study radioactive decays in the laboratory without the need to produce costly isotopes at a particle accelerator or by neutron activation at a nuclear reactor.

Nature delivers them to us gratis! Here the decay sequence (half lives are given too):



${}^{222}\text{Rn}$ undergoes α -decay. The decay product, in this case ${}^{218}\text{Po}$, is left in a doubly charged state after the decay. The charged Po atoms tend to adhere to small dust particles (aerosols) suspended in the air. In order to collect the aerosols we will suck a calibrated volume of air through a filter. A suitable blower is available in the sub-basement. Our filter then catches the aerosols and together with them the Po. The ${}^{218}\text{Po}$ (from here on called activity A) α -decays quickly within a few minutes. You are then left with ${}^{214}\text{Pb}$ (activity B) which β -decays into ${}^{214}\text{Bi}$ (activity C), which is β -unstable as well and is already decaying while you are still collecting aerosols. However, ${}^{214}\text{Bi}$ is also being constantly replenished through the β -decay of ${}^{214}\text{Pb}$ yielding ${}^{214}\text{Bi}$. Let's examine the math describing both build-up and decay of the activities during the filtering. When the blower is *on*, activities A, B and C are building up on the filter. At the same time some of it already decays because these elements are unstable. The build up is described by the following differential equations:

$$\frac{dN_A}{dt} = k n_A - \frac{N_A}{\tau_A} \quad (2)$$

$$\frac{dN_B}{dt} = k n_B + \frac{N_A}{\tau_A} - \frac{N_B}{\tau_B} \quad (3)$$

$$\frac{dN_C}{dt} = k n_C + \frac{N_B}{\tau_B} - \frac{N_C}{\tau_C} \quad (4)$$

Here N_A , N_B and N_C are the numbers of atoms of activity A, B and C on the filter (unitless). n_A , n_B and n_C are the numbers of atoms of activity A, B and C per unit volume of air (unit [l^{-1}]). k is the volume of air passing through the filter per unit filtering time t (unit is [l/s]). In *G.N. Whyte and H.W. Taylor, American Journal of Physics 30 (1962) 120-124* the authors are assuming that the collection efficiency of the filter is 100%. You also note that the positive terms ($k n$) used in all three equations assumes equal efficiency for catching all three decay products of Rn on the filter. Your measurement of the parameter $R(t)$ effectively tests this assumption made by *G.N. Whyte and H.W. Taylor*.

How do we come up with these equations? Let's examine equation 3. $\frac{dN_B}{dt}$ describes the change of the number of nuclei of species B . It is hence a measure of the *decay rate* (unit is $[Bq]$ equivalent to $[s^{-1}]$). This is what we can directly measure with our radiation detector. The parent concentrations have then to be *inferred* from this data using an appropriate set of assumptions. We assume:

- The rate with which we capture activity B from the air is proportional to the blower speed, k , and the concentration, n_B , of species B in the air. The term $k n_B$ is positive (it is a gain) and measures this collection.
- While filtering is still in progress the sample of species B is already decaying. As in all radioactive decays the decay rate is proportional to the number of atoms on the filter. The shorter the live time τ the more “unstable” the sample is. Hence the decay rate has to be inversely proportional to τ .
- Species A is collected on the filter in parallel. It is decaying as well while filtering is still in progress. The decay of species A leaves species B behind. The decay rate of A , as given in equation 2 has hence to be booked as a gain (with “+” sign) for B . This couples the equations.

This set of assumptions allows us now to quantify the elemental concentrations n_A , n_B , and n_C from the measured decay rates. You also see that we do need differential equations. The fact that the change in number, dN , is proportional to the number of atoms, N , itself requires that this number is not much changed by the decay. In other words we need to require $dN \ll N$, otherwise the decay rate would be, to some extent, dependent on itself. The total change is then given as the sum, or better integral, of all small changes over all short time intervals considered.

Problem 1:

Show that the activity $A_A = -\frac{dN_A}{dt}$, or number of decays per unit time, of species A is given as $A_A = \frac{N_A}{\tau_A}$. Remember: $N_A(t) = N_A \cdot e^{-t/\tau_A}$, with $N_A(t)$ denoting the number of ^{218}Po nuclei at time t , N_A the number of ^{218}Po nuclei at time $t = 0$, and $\tau_A = \frac{T_{1/2}^A}{\ln 2}$ the decay time.

Above equations 2, 3, 4 are solved by the following functions:

$$N_A(t) = k n_A \tau_A (1 - e^{-t/\tau_A}) \quad (5)$$

$$N_B(t) = k\tau_B(n_A + n_B)(1 - e^{-t/\tau_B}) + \frac{kn_a\tau_A\tau_B}{\tau_A - \tau_B}(e^{-t/\tau_B} - e^{-t/\tau_A}) \quad (6)$$

$$\begin{aligned} N_C(t) = & k\tau_C(n_A + n_B + n_C)(1 - e^{-t/\tau_C}) + \\ & \frac{k(\frac{n_B}{\tau_B} - \frac{n_A}{\tau_A} - \frac{n_B}{\tau_A})\tau_A\tau_B^2\tau_C}{(\tau_A - \tau_B)(\tau_B - \tau_C)}(e^{-t/\tau_C} - e^{-t/\tau_B}) + \\ & \frac{kn_A\tau_A^2\tau_C}{(\tau_A - \tau_B)(\tau_A - \tau_C)}(e^{-t/\tau_C} - e^{-t/\tau_A}) \end{aligned} \quad (7)$$

The ratio of activities C and B after the filtering has gone on for the time t is defined as:

$$R(t) = \frac{N_C(t) \cdot \tau_B}{N_B(t) \cdot \tau_C} \equiv \frac{A_C^0}{A_B^0}. \quad (8)$$

This ratio depends on the filtering time. We define the activities A_B^0 and A_C^0 as those present on the filter *at the end of filtering*. We further assume that the activities of A, B, and C in the atmosphere are in *secular equilibrium* which means the decay rate is determined by the long lived mother (in our case ^{222}Rn) of the decay series and is *equal* for all members of the series:

$$\frac{n_A}{\tau_A} = \frac{n_B}{\tau_B} = \frac{n_C}{\tau_C} \quad (9)$$

One task of this experiment will be to *measure* $R(t)$ and compare to the expectation discussed below.

Problem 2:

Show that expressions 5 and 6 are the solutions of differential equations 2 and 3. Show this by substituting the solutions into the differential equations. Proof of the third solution can be performed the same way but is tedious.

As soon as the filtering process is stopped the activities are described by a different system of differential equations. The term kn , describing the gain

of activities, vanishes because the air flow is given by $k = 0$ now. We have:

$$\frac{dN_B}{dt'} = -\frac{N_B}{\tau_B} \quad (10)$$

$$\frac{dN_C}{dt'} = \frac{N_B}{\tau_B} - \frac{N_C}{\tau_C} \quad (11)$$

The solutions for equations 10 and 11 are the time dependent activities $A_B(t')$ and $A_C(t')$. You note that we have to use a new time notation. t measures the time during filtering, t' the time after the filtering has been concluded. This is true to good approximation. You note that the decay of species A has been omitted here. This was done to simplify the equations. A has a half life of only 3 min. It hence decays quickly. If you wait a few half lives after end of collection before you start counting, then A will effectively have died out. In this limit equations 10 and 11 describe the data well. From a practical point of view: $A_A(t')$ is unmeasurable in our setup because it (a) doesn't live long enough and (b) it undergoes an α -decay which doesn't result in emission of γ -radiation; and that's what we measure with or NaI detector. The observable activities are given by:

$$A_B(t') = A_B^0 e^{-t'/\tau_B} \quad (12)$$

$$A_C(t') = A_B^0 \left[R(t) e^{-t'/\tau_C} + \frac{\tau_B}{\tau_B - \tau_C} (e^{-t'/\tau_B} - e^{-t'/\tau_C}) \right] \quad (13)$$

A_B^0 is the activity B at the end of filtering. Evaluate your measured activities relative to this time zero $t' = 0$. Remember we defined $R(t)$ earlier in equation 8.

A proof that the functions 12 and 13 are indeed solutions of equations 10 and 11 can be found in the appendix, if you are interested.

You see from this discussion that the “exponential” decay is not really a law of nature. It is derived from the assumption that the decay probability only depends on the number of decaying nuclei. If you have a balance of production and decay the time development of the observed activity (or equivalently the of the emitted photon flux) may well be described by a more complicated function, which however always contains exponentials. This follows naturally from the same assumption as shown in this experiment.

It is essential that you keep track of time in this experiments. Note the time you start and stop the blower, and the time you start counting the filter.

Try to keep the delay between end of filtering and beginning of counting to a minimum.

We see from equation 13 that knowledge of the duration of the filtering is important. This equation describes both build up and decay of the isotopes under study in this experiment.

As a second task you will have to measure the half lives of ^{214}Pb (which is relatively straight forward) and ^{214}Bi (which is a bit more involved).

Here is how it works:

3 Procedure

First read the above paper I have distributed to you. Familiarize yourself with the methods and mathematical tools discussed in this reference. It is assumed that you are by now familiar with the basic data taking and analysis techniques used in the “Gamma Spectroscopy” experiment. If you still have trouble consult the write-up for that experiment.

3.1 Data Collection

Put a new red filter into the blower in the sub-basement. The lab supervisor will help you with this. Switch the fan *on* and read the air flow as measured with the flow meter on the back of the fan. If the flow meter doesn't work (sometimes the little ball gets stuck) use a flow of $0.85 \text{ m}^3/\text{min}$, which I determined earlier. Note the time you started the fan into your lab book. Collect aerosols for a period of 30-60 min. Switch the blower off and retrieve the filter. Note the time of end of filtering. Bring the loaded filter to the lab and put it in front of the NaI detector. Put it as close as possible. The round filter will conveniently fit onto the Al cap of the detector. Close the Pb shield of the counting set-up using the two Pb bricks on the floor underneath the table. You should start counting the filter as soon as possible after ending the filtering. Don't waste time!

During the time you are collecting aerosols with the blower you should quickly re-check the energy calibration of the NaI detector. Use the ^{57}Co , ^{137}Cs and

^{22}Na sources for that. Apply the techniques and procedures described in the write-up of the experiment on “Gamma Spectroscopy”.

After the filtering is over you will measure the γ -ray flux emitted by the filter as a function of time. This will allow you to determine the wanted half lives. Collect 10 successive spectra using a measuring time of 10 minutes for each run. Activate PCA by clicking on the appropriate icon on the desktop of the data taking PC. Now call an automated data taking routine called “filter.nbs” by issuing the command “Alt O”. Select the “Run a Batch File” option. Enter the name of the batch file: “filter.nbe”. If you are ambitious have a look into the code using the option “Edit a Batch File” but DON’T CHANGE IT! When activated the code will ask you for a file name. Enter a name of your liking e.g. “filter”. Don’t give a file extension the program will automatically assign “.asc”. When you are done after 100 min you will have 10 new files. In our example the names would be “filter1.asc” to “filter10.asc”. Note the time the first run started into your log book.

3.2 Data Analysis

Transfer all data files collected to the UNIX workstation using FTP. Add “!” in front of the comment lines, as done previously. You are now ready to analyze your data with PAW. Use the MACRO “plot_histogram” as before. You will have to analyze 10 data files each containing a number of peaks. Before embarking on this find on the web which peaks are to be expected in the ^{214}Pb and ^{214}Bi decays. You should get good results if you analyze the 351 keV peak for ^{214}Pb and the 609 keV peak for ^{214}Bi . You will, at this point, need a current energy calibration to identify the gamma peaks of interest. Note that the PAW fit returns peak amplitude, mean and width. You will need the appropriate combination of these (the MACRO “plot_histogram” tells you what to do in its header) to get the peak area. Don’t forget to propagate the errors when you do this calculation. For simplicity assume that the errors are un-correlated (this is not really the case but analysis would be too complicated otherwise).

For every run note the peak counting rates and their errors into your notebook (make a table). As you have kept track of time you know when you started data taking. To convert the measured counting rate $r(t')$ into an activity $a(t')$ you will need to make correction for the detector efficiency ε and the

finite photon emission probability (called branching ratio) b . For the 351 keV peak $\varepsilon = 0.14 \text{ counts}/\gamma$ and $b = 0.35 \text{ } \gamma/\text{decay}$. For the 609 keV line: $\varepsilon = 0.08 \text{ counts}/\gamma$ and $b = 0.46; \gamma/\text{decay}$. Calculate the time development of the activity contained in the filter. Let $c(E_\gamma, t')$ be the integral of the peak you are going to analyze. This integral was measured at a time t' for a peak energy E_γ . The measured activity of the decaying nuclide, $a(t')$ is given as:

$$a(t') = \frac{c(E_\gamma, t')}{\varepsilon b t''}, \quad (14)$$

with t'' denoting the duration of the data taking in [s]; yet another time you need to keep track of! Energy dependent values of ε are given in figure 2 below. Assume that the ε 's are free of error, for simplicity.

This is a different task compared to the experiment on “Gamma Ray Spectroscopy”. There we were primarily interested in evaluating the *peak position* in order to determine the *photon energy*. In this experiment you will only use your measurement of the peak energy to identify the appropriate peak and associated nuclear decay. The quantity of interest is this time the area under the peak. This is a measure of the strength of the gamma emitter. As pointed out before systematic corrections need to be made to get the absolute activity from the peak integrals. You need to (a) convert into rates, (b) account for the fact that the characteristic gamma is only emitted in a fraction of the decays of interest (described by the *branching ratio*), and (c) that only some of the gammas emitted by the filter will result in a “count” under the full absorption peak (an energy dependent detection efficiency, containing both the geometrical factor and the finite interaction probability is given in figure 2). All this is summarized in equation 14.

As you go along in your data analysis assign a time in minutes since end of filtering to each run. A run has a certain length which means it corresponds to a time interval. In your analysis you need to assign a certain “point” in time to each run. How will you chose the time?

Problem 3:

What is the average time, $\langle t \rangle$, for a decay? To find the answer remember that an average over any function $f(t)$ is defined as:

$$\langle t \rangle = \frac{\int_0^\infty t \cdot f(t) dt}{\int_0^\infty f(t) dt} = \frac{\int_0^\infty t \cdot e^{-t/\tau} dt}{\int_0^\infty e^{-t/\tau} dt}$$

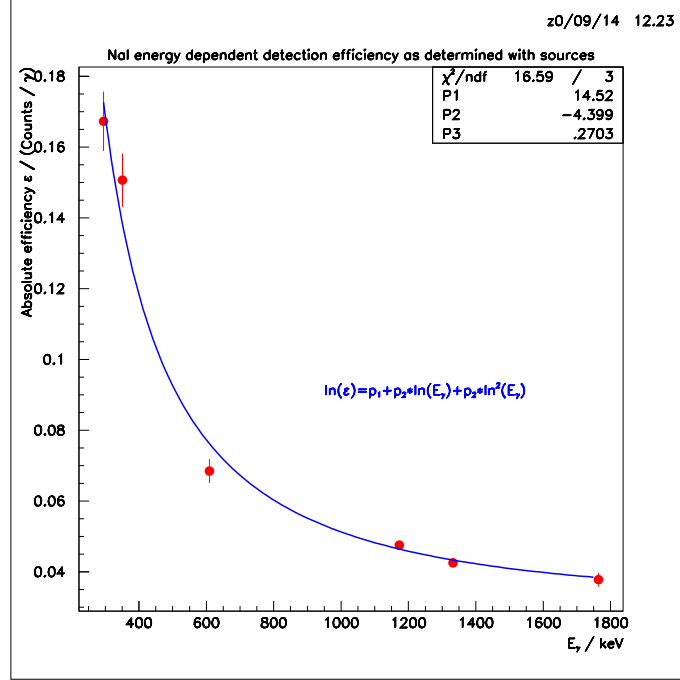


Figure 1: Energy dependent gamma ray detection efficiency of the NaI detector used in the lab. as determined from calibrated point sources placed onto the center of the Al cap.

In our case the mid point between start and stop is a good choice.

How do we get the Radon activity quantitatively? Use the assumption that the ^{222}Rn decay products are in secular equilibrium in the air, as defined in equation 9 ($n_B = \frac{n_A \tau_B}{\tau_A}$) and substitute into equation 6, which has been divided by τ_B to yield $A_B^0 = \frac{N_B}{\tau_B}$:

$$A_B^0 = k n_A \left[\underbrace{\left(1 + \frac{\tau_B}{\tau_A}\right) (1 - e^{-t/\tau_B}) + \frac{\tau_A}{\tau_A - \tau_B} (e^{-t/\tau_B} - e^{-t/\tau_A})}_{\equiv f} \right] \quad (15)$$

The specific activity is given as:

$$\frac{n_A}{\tau_A} = \frac{A_B^0}{f k \tau_A}. \quad (16)$$

You have measured A_B^0 by analyzing the time development of the 351 keV peak. You will need to calculate f for the specific filtering time t chosen. $\tau_A=4.4$ m.

Enter the run times, peak counting rates and their errors into one data file

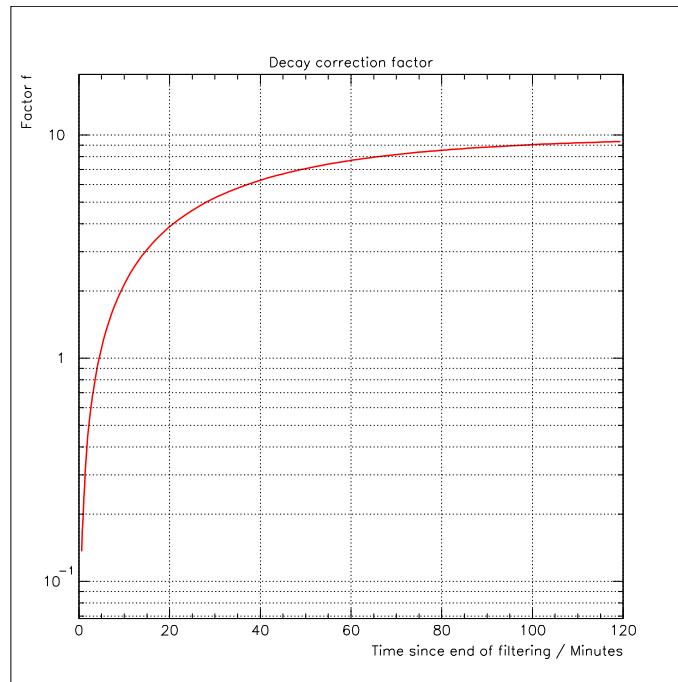


Figure 2: Correction factor f as a function of the filtering time t . You may read the correction factor from this graph.

per each gamma line. As in the previous experiment use the editor EMACS to create a data file with three columns: time [min], activity [Bq], error of the activity [Bq]. You have to convert the peak areas into activities first! Don't forget this or the result will be wrong. Use the PAW MACRO "plot_data" to plot and fit your result ("exec plot_data" at the PAW prompt) the way you have done this in the previous experiment. Use an exponential fit function

for the ^{214}Pb line. It is offered as one of the options of “plot_data”. This fit will yield the ^{214}Pb half live and activity at the end of filtering. The actual fit function for your measured rate $r_B(t')$ is: $r_B(t') = e^{p_1+p_2t}$. Obviously $p_2 = \frac{1}{\tau_B}$ and $a_B^0 = e^{p_1}$. Calculate the physical quantities from your fit parameters. Then go on and analyze the ^{214}Bi half life. You will need to perform a more complex fit as the Bi rate decreases due to radioactive decay as described by equation 13. The decay of ^{214}Pb into ^{214}Bi partially compensates this loss. The apparent half live of the 609 keV peak is therefore longer than the tabulated value. A custom fit function “bi.for” is on the disk. You will need to edit (use “emacs”) this file and enter the ^{214}Pb counting rate at the end of filtering at the marked position. Consult the PAW tutorial on the web on how to use a custom fit function. To fit your ^{214}Bi data use the following PAW commands:

```
vect/create par(2) r 6.8 5.e-2
vect/fit x y y_e 'bi.for' 2 par
```

The fit parameter p_1 is actually your measured value for $R(t)$ with measurement error. Compare it to the expectation value in figure 1 of *G.N. Whyte and H.W. Taylor*. Is your data consistent with that expectation? Explain. The fit parameter $p_1 = \tau_C$ is the mean live time of ^{214}Bi as determined from your data. Does it agree with the tabulated value?

There is an alternate route for the analysis available. You may use the PAW MACRO “analyz-spect”. This code will read all 10 gamma spectra collected, analyze the important peaks and then plot and fit their time dependence. WARNING: this is much more comfortable but the code is not yet fully debugged. But if you want give it a try...

Finally determine the specific ^{222}Rn activity in [Bq/l] from your data using equation 16. It is imperative to use consistent units or you will get a wrong result.

Problem 4:

The nuclei you are counting were not all collected at the same time; they have a range of ages. This means you don't really know “time zero”. How can you determine the half live of ^{214}Pb and ^{214}Bi in the presence of this “uncertainty”?

Appendix:

Why is function 12 the solution of equation 10. We can show this by substitution:

$$\begin{aligned}
 A_B(t') &= A_B^0 e^{-t'/\tau_B} \\
 \Rightarrow A_B(t') \cdot \tau_B &= A_B^0 \cdot \tau_B \cdot e^{-t'/\tau_B} \\
 \Rightarrow N_B(t') &= N_B^0 e^{-t'/\tau_B} \\
 \Rightarrow \frac{d}{dt'} N_B(t') &= -\frac{N_B^0}{\tau_B} \cdot e^{-t'/\tau_B} = \frac{N_B(t')}{\tau_B}.
 \end{aligned}$$

To show that 13 is the solution of equation 10 is a bit more complicated. Here is how it goes:

$$\begin{aligned}
 A_C(t') \cdot \tau_C &= A_C^0 \cdot \tau_C \cdot e^{-t'/\tau_C} + \frac{\tau_B \tau_C A_B^0}{\tau_B - \tau_C} (e^{-t'/\tau_B} - e^{-t'/\tau_C}) \\
 \Rightarrow N_C(t') &= N_C^0 e^{-t'/\tau_C} + \frac{\tau_C N_B^0}{\tau_B - \tau_C} (e^{-t'/\tau_B} - e^{-t'/\tau_C}) \\
 \Rightarrow \frac{d}{dt'} N_C(t') &= -\frac{N_C^0}{\tau_C} e^{-t'/\tau_C} - \frac{\tau_C N_B^0}{\tau_B - \tau_C} \left(-\frac{1}{\tau_B} e^{-t'/\tau_B} + \frac{1}{\tau_C} e^{-t'/\tau_C} \right) \\
 \frac{N_B}{\tau_B} - \frac{N_C}{\tau_C} &= \frac{N_B^0}{\tau_B} e^{-t'/\tau_B} - \frac{N_C^0}{\tau_C} e^{-t'/\tau_C} - \frac{N_B^0}{\tau_B - \tau_C} (e^{-t'/\tau_B} - e^{-t'/\tau_C}) \\
 &= -\frac{N_C^0}{\tau_C} e^{-t'/\tau_C} + N_B^0 \left(\frac{1}{\tau_B} e^{-t'/\tau_B} - \frac{1}{\tau_B - \tau_C} e^{-t'/\tau_B} + \frac{1}{\tau_B - \tau_C} e^{-t'/\tau_C} \right) \\
 &= -\frac{N_C^0}{\tau_C} e^{-t'/\tau_C} + N_B^0 \left(\frac{\tau_B - \tau_C}{\tau_B (\tau_B - \tau_C)} e^{-t'/\tau_B} - \frac{\tau_B}{\tau_B (\tau_B - \tau_C)} e^{-t'/\tau_B} + \right. \\
 &\quad \left. \frac{\tau_B}{\tau_B (\tau_B - \tau_C)} e^{-t'/\tau_C} \right) \\
 &= -\frac{N_C^0}{\tau_C} e^{-t'/\tau_C} + N_B^0 \left(-\frac{\tau_C}{\tau_B (\tau_B - \tau_C)} e^{-t'/\tau_B} + \frac{\tau_B}{\tau_B (\tau_B - \tau_C)} e^{-t'/\tau_C} \right) \\
 &= -\frac{N_C^0}{\tau_C} e^{-t'/\tau_C} + \frac{\tau_C \cdot N_B^0}{\tau_B - \tau_C} \left(-\frac{1}{\tau_B} e^{-t'/\tau_B} + \frac{1}{\tau_C} e^{-t'/\tau_C} \right) \\
 \Rightarrow \frac{d}{dt'} N_C(t') &= \frac{N_B}{\tau_B} - \frac{N_C}{\tau_C}
 \end{aligned}$$