ENVIRONMENTAL RADIOACTIVITY

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Abstract

Radon is a naturally occurring element that is formed from the nuclear decay of its parent radium. Due to its gaseous nature and the fact that it is an alpha-decay isotope, radon is responsible for the majority of the public exposure to ionizing radiation. Due to the higher abundance of ^{238}U relative to ^{232}Th and ^{235}U, ^{222}Rn is the most abundant of the three main radon isotopes. ^{222}Rn is an alpha decay isotope with a half-life of 3.8 days and it can be measured through its two β-decay daughters, ^{214}Pb and ^{214}Bi. In this experiment, ^{222}Rn contained in different environmental samples such as rocks, air and water will be measured using a Ranger equipped with a Geiger-Mueller tube to detect low level radiation. The aim of the experiment is to put in practice the concept of ingrowth, decay and secular equilibrium between ^{222}Rn and its progeny, and to identify the radionuclides based on their half-life. The experimental setup also includes some background measurements and an efficiency calibration of the Ranger detector.

1. Introduction to environmental radioactivity

"Naturally Occurring Radionuclides" refer to those radionuclides that are present in significant quantities on Earth. These radionuclides embrace ^{40}K and isotopes from the principal primordial series: ^{238}U, ^{235}U and ^{232}Th (Figure 1) series, known as the uranium, the actinium and the thorium series, respectively. Due to the shorter half-life of ^{235}U compared to ^{238}U and ^{232}Th, present ^{235}U abundance is only 0.72% of the natural uranium. All three of these series have numerous radionuclides in their decay chains before reaching a stable end nuclide, lead. The distribution of primordial radionuclides in the geosphere depends on the location of the geological media for which they are derived and the processes which concentrate them at a specific location in specific media (IAEA, 2003). At background concentrations, the naturally occurring radionuclides of these series contribute about one-half of the natural background external radiation to which all humans are continuously exposed to (UNSCEAR, 2000).

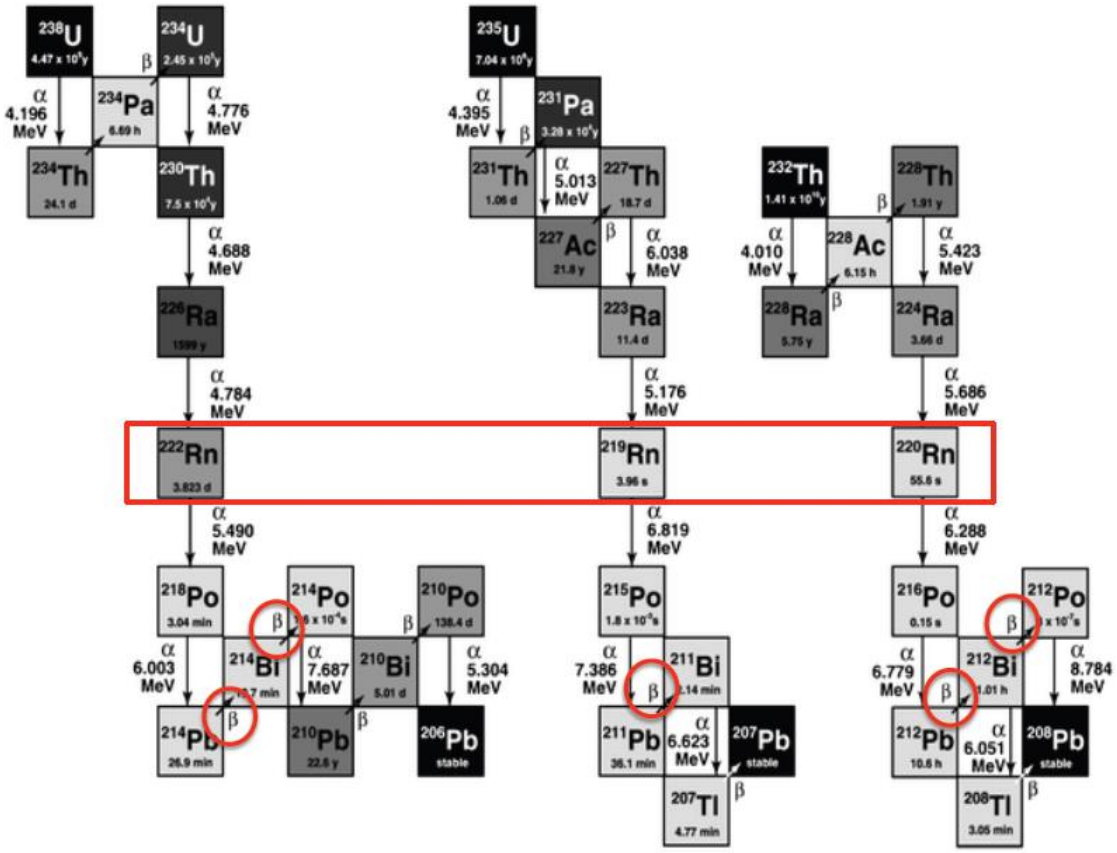

Figure 1: Decay chains of the Naturally Occurring Radionuclides of the three primordial series of ^{238}U (99.27% abundance), ^{235}U (0.72% abundance) and ^{232}Th. Rn isotopes of the three naturally decay chains are highlighted in the red box. Measured beta-counts using the LLM 500 are emphasized in red circles.

The main goal of this practice is to measure radon isotopes (or their daughter nuclides), which are formed during the decay of the primordial radionuclides. Unlike all the other intermediate isotopes in the primordial decay chains, radon is a noble gas. As such, radon exhales and escapes from the structures where it is formed (e.g. rocks, walls, water, etc.). Due to the gaseous nature of radon, it can be easily inhaled and that is the reason why nowadays it is responsible for the majority of the public exposure to ionizing radiation. Despite its short half-life (from seconds to 3.8 days), radon can accumulate in closed or poor ventilated environments such as buildings, basements and caves, and its concentrations can increase several orders of magnitude. While in open places ^{222}Rn concentrations range from 0.1 to 100 $Bq \cdot m^{-3}$, in mines and caves this number can increase to several $kBq \cdot m^{-3}$ (Cigna, 2005).

^{222}Rn is an alpha-decay isotope with a half-life of 3.8 days. This is the time period required for the activity of ^{222}Rn to be reduced to half its value. The detector used in this experiment will detect mainly the β-particles of the two daughters, ^{214}Pb and ^{214}Bi. For the planned measurements, the α-particles will be stopped by the sealing of the sample (e.g. for activated carbon). In other cases, our sample will only contain the daughters of radon (e.g. fiber filter of air and water), and thus again, we will mainly detect the β-particles.

In numerical terms, the time evolution of nuclide concentrations undergoing serial or lineal decay chain is governed by a set of first order differential equation, called Bateman equations. The Bateman equations are as follows:

$$A \rightarrow B \rightarrow C$$

$$\frac{dN_A}{dt} = -\lambda_A N_A$$

where λ $[s^{-1}]$ is the decay constant specific for each radionuclide $\lambda = \frac{\ln(2)}{T_{1/2}}$ and N the number of atoms. The solution to the equation is:

$$N(t) = N_0 e^{-\lambda t}$$

where N(t) is the number of atoms at time t and N_0 is the number of atoms at t_0.

In a case of A an B nuclide series in linear chain describing nuclide concentrations, assuming 0 concentrations of B at time 0 but a known concentration of the parent (A), then:

$$N_B = \frac{\lambda_A}{\lambda_B - \lambda_A} N_{A0}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

So then, by knowing the initial concentration of ^{222}Rn, one can calculate the time evolution of its daughters (Figure 2). While ^{222}Rn decays, ^{214}Pb grows up until reaching secular equilibrium with its progeny, in about 3-4 hours. The time that it takes for a daughter to reach secular equilibrium with its parent depends on the half-life of the progeny. In this case, it would depend on the ^{214}Pb decay rate.

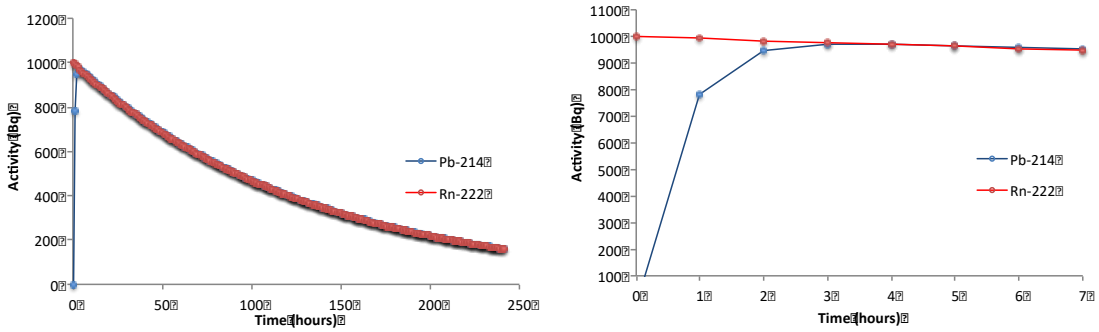

Figure 2: Decay curves of ^{222}Rn and its daughter ^{214}Pb. Secular equilibrium between ^{222}Rn ($T_{1/2}$=3.8 days) and ^{214}Pb ($T_{1/2}$=26.9 minutes) is reached in about 3-4 hours.

This type of relation between parent and daughter activity occurs when the half-life of the parent nuclide (A) is larger than that of the daughter nuclide (B). In such a situation, the decay rate of A, and hence the production rate of B, is approximately constant. The quantity of radionuclide B builds up until the number of B atoms decaying per unit time becomes equal to the number being produced per unit time; the quantity of radionuclide B then reaches a constant, equilibrium value. This

property can be used in this experiment to calculate the ^{222}Rn half-life by measuring the decay of their progeny ^{214}Pb ad ^{214}Bi, in secular equilibrium with their parent.

Since radon and its daughters are radioactive, a radiation dose is associated with their exposure. Inhalation of radon and its progeny can lead to their deposition in the lungs and result into bombardment of sensitive lung tissues with alpha radiation. Over a period of time, this may lead to malignant transformation and the formation of lung cancer. Indeed, indoor radon has been determined to be the second leading cause of lung cancer after tobacco smoking (Chen, 2005). A radiation dose assessment from radon inhalation (and its progeny) can be obtained as follow:

$$Dose\ [nSv] = C_0(\varepsilon_r + \varepsilon_d F) \cdot O$$

where C_0 is the mean annual radon activity concentration in $Bq \cdot m^{-3}$, ε_r (0,17 $nSv \cdot h^{-1}$) per $Bq \cdot m^{-3}$) and ε_d (9$nSv \cdot h^{-1}$ per $Bq \cdot m^{-3}$) are the dose conversion factors for radon and its short-lived progeny respectively, F is the equilibrium factor between radon and its short-lived progeny (F=0,4 UNSCEAR and ICRP recommended for indoors), and 0 is the occupational factor (time spent indoors).

The general aim of this experiment is to measure ^{222}Rn from different environmental compartments, such as rocks, air and water and understand the concept of decay, ingrowth and secular equilibrium between radon isotopes and its daughters (Pb and Bi).

2. Materials and Methods

The Ranger Detector

The Ranger Radiation Alert (Ranger) is an instrument that is optimized to detect low levels of radiation (Figure 3). It can measure alpha, beta, gamma, and x-ray radiation. The Ranger is more sensitive for beta particles and much less for gamma radiation. And, as mentioned above, alpha particles will not be measured because of the nature of the sample or the sealing applied to the sample. The Ranger uses a Geiger-Mueller tube to detect radiation. The tube generates a pulse of electrical current each time radiation passes through the halogen quenched tube. Each pulse is electronically detected and registered as a count. The unit displays the counts in the mode you choose. Our recommendation is to use counts per second (cps).

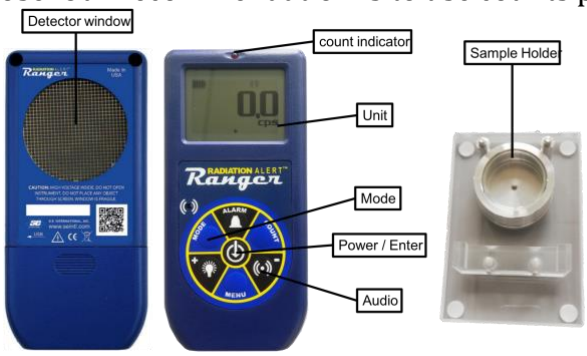

Figure 3: The Ranger Radiation Alert detector and sample holder.

How to operate the detector

1. Remove the plastic protection from the detector window. Take care the detector window is not touched either by the sample or the operator.
2. To switch on/off the detector press and hold the power/enter button until you hear a beep. Wait for 30 seconds until you hear another long beep which indicates that the detector is ready to count.
3. To silence the audio, press the audio button.
4. Place the unit on the sample holder.
5. Connect the Ranger to the computer using the cable and USB ports. In few seconds the Radiation Alert ® Observer USB Software will pop up. Make sure all settings are as displayed in Figure 4. The software displays a *Chart* with the raw count rate in the *y* axis and time in the *x* axis. The display also shows a *Grid* to summarize the data.
7. To save the data, go to: File, Save Chart, choose a location and name the file. We recommend to save the file regularly, specially at the end of the measurements. There is no need to save the Grid.

*For more detailed information about the Ranger please read the Manual from S.E International.

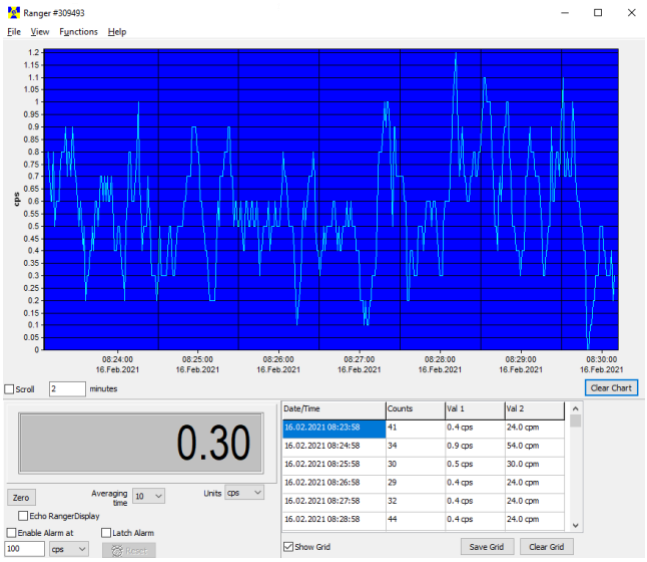

Figure 4: Display of the Radiation Alert ® Observer USB Software.

Data type and initial data treatment

Data are saved in a *.txt* file that can be easily imported into excel, python, etc. The first two columns report the data of interest, the date and time and the number of detected counts in one second. Please ignore the data from column 3 and higher. Each raw is equivalent to one second of measurement. The number of counts detected by the unit varies from moment to moment due to the random nature of radioactivity. A reading is expressed more accurately as an average over time. Thus, the raw data have to be binned to decrease the scatter i. e. sum of counts in a time interval divided by the number of integrated rows. We will employ different time intervals for binning the data depending on the rate of change of the raw count rate. There is a known issue for long measurements: the time interval (time elapsed from

one row to the next) seems to increase steadily over time. While this feature is negligible for the first 6 hours of measurement, it can be and issue for longer times, especially for measurements conduced over few days.

Types of samples

The experiment includes rocks, air and water. These are the required measurements. Additionally, you are very welcome to try any other material in which you are interested in (e.g. dried banana, nuts, soil, etc.). The most active samples are the rocks containing ^{238}U and a gas mantle containing ^{232}Th. Details on sample preparation are provided below. The most important part is that the sample is placed in the sample holder facing, but not touching, the detector window of the Ranger.

3. Procedures: sample preparation and measurements

Since the laboratories are closed on Thursday make sure to perform the experiments in the following order so that the whole of the practice can be completed within a week.

1. *Background measurements*

This part of the experiment measures the natural radiation that is always present in the environment and it can either come from space (i.e., cosmic rays) and from naturally occurring radioactive materials contained in the Earth and living organisms. Also, the detector noise can be a source of detector background. The background is expressed as a raw count rate usually in units of counts per second (cps) or counts per minute (cpm). The obtained count rate has to be subtracted from all other measurements, including the efficiency.

To perform these measurements the Ranger is placed on an empty sample holder. The aim is to see the differences between long and short measurements. Therefore, we suggest to perform several measurements lasting between 10 minutes and 1 hour. Then, bin the data in time intervals ranging from 1 minute to 1 hour.

2. *Efficiency calibration*

The detector efficiency relates to the number of particles counted by the detector compared to the number of beta particles emitted by the source. This depends on both the intrinsic detection efficiency and the geometrical efficiency. The intrinsic detection efficiency of any device operated in pulse mode is defined as the probability that a quantum of radiation incident on the detector will produce a recorded pulse. The geometrical efficiency is the ratio of the number of particles emitted towards the detector by the number of particles emitted by the source.

The good functioning (or efficiency ε) of the Ranger can be checked by using a known activity of a beta emitter. The following formula applies:

$$\varepsilon = \frac{1}{k}$$

where k is the calibration factor described as:

$$k = \frac{A}{cps}$$

where A activity expressed in Becquerels (Bq) (decays per second) and *cps* the net count rate (raw count rate minus the background) expressed in counts per second or "cps".

Why do we need to know the efficiency and the k factor? The detector provides a raw count rate, but we are interested in the activity of the sample. Both the raw count rate and the activity are a measure of radioactive decays over time. And,often, both are expressed in similar units: cps for the former and Bq for the latter. Bq is equal to cps only after we have subtracted the background and multiplied the raw count rate by the k factor.

In this experiment, the efficiency calibration will be performed using 1.9 g KCl salt, which in turn contains a known amount of the radioisotope ^{40}K (Philipsborn and Hoffmann, 1996). Note that the KCl needs to be ground, weighted, placed homogeneously with the diameter of the detector window, and properly sealed with adhesive film from both sides in order to not contaminate the Ranger detector. The salt will be counted for 60 minutes. A binning of 5 minutes is recommended.

3. *Measurement of Rn in environmental samples: air and water*

The aim of this exercise is to determine the ^{222}Rn activity in environmental samples (e.g. water and air) and make a dose assessment due to indoor Rn inhalation. To optimize time, it is suggested to perform the determination of Rn in water first and then in the air sample so you can leave it running over night (Monday to Tuesday)

Water contains radon isotopes and its progeny. Radon dissolves into groundwater and can be transported away from the source. However, once the water is exposed to air the radon is released. Therefore, groundwater from a well or a fountain in the mountain area is better for this experiment.

Filter approximately 2 to 3 litres of water through a fiber glass membrane placed on the porcelain funnel. After the whole volume of samples has been filtered, dry the filter on the hotplate for 5 minutes.Afterwards, place the filter under the Ranger. This measurement can usually be performed in less than 5 hours. Because the radon concentration is usually low and a rapid decay is expected, a binning time of 1 to 5 minutes is recommended.

For the air experiment, about 25 m^3 of air will be filtered using a dust-collecting pump, and by means of a glass fiber filter placed in a cardboard holder. Note that high radon concentrations can be found in closed environments, especially at underground level (e.g. go to -1 level of any of the buildings in Hönggerberg). Note

the initial and final pumped volume in the dust-collecting pump. Then place the filter in the sample holder. The measurement should last for about 10 hours. Please try binning times between 1 and 10 minutes.

To back-calculate the radon activity, used the k factor obtained for the efficiency calibration.

4. *Ingrowth and decay of ^{222}Rn and its progeny*

The half-life of ^{214}Pb ($T_{1/2}$=26.9 minutes) is shorter than that of ^{222}Rn ($T_{1/2}$= 3.8 days), allowing them to reach secular equilibrium after a certain time. This allows quantifying the ^{222}Rn half-life through the decay of ^{214}Pb and ^{214}Bi ($T_{1/2}$=19.7 minutes) monitored by Ranger. However, before reaching equilibrium, one can observe the ingrowth of ^{214}Pb and ^{214}Bi from their precursor ^{222}Rn. This is not the case for ^{220}Rn progeny, which ^{212}Pb ($T_{1/2}$=10.8 hours) is longer than its precursor ($T_{1/2}$=55 seconds).

The aim of this exercise is to trap the radon isotopes emitted from different sources (i.e. rock containing ^{238}U and/or gas mantle containing ^{232}Th) and measure the ingrowth and decay of their progeny.

For the ingrowth curve, following steps need to be executed:
1. Fill the bottom of the empty glass beaker with activated carbon.
2. Place the adhesive film on the top of the beaker.
3. Turn the beaker so that the activated coal sticks to the adhesive film.
4. Open the sample container and rapidly place the adhesive film with the activated carbon facing thew sample
5. Leave it like this for **5 minutes** to let the emanated ^{222}Rn be adsorbed on the granular activated carbon.
6. Cover the other side of the activated carbon with another adhesive film and make sure no activated carbon is being left outside. Now the sample is ready to place in the sample holder.
7. Measure the sample for 5 hours and bin the data every 5 or 10 minutes.
8. Apply fittings to the ingrowth curve.

In order to get the exponential decay of ^{222}Rn, a similar procedure than for the ingrowth curve needs to be done, with the difference that emanated ^{222}Rn needs to be absorbed by activated carbon for a longer time (2 hours). Measure the sample over three days and bin the data every 1 hour. *Due to the abovementioned issue of increasing time intervals in long measurements, we will probably not be able to obtain a correct quantitative number for the half-life of ^{222}Rn. Yet, you will see qualitatively the decay.

If the obtained half-life differs too much from the expected value of 3.8 days, the TA will provide with an alternative dataset that will allow a better quantification. The dataset corresponds to activated carbon that was exposed for 2 hours to an ^{238}U-rich rock. The data displays the activity (Becquerels; already background subtracted and multiplied by the k factor) every 1 hour (each row equals to 1 hour).

4. Results and discussion

In this section some guidelines are provided to ensure that the most relevant aspects of the experiment are discussed. You are welcome to focus in other aspects additionally.

Background measurements

- Which is the background count rate (cps or cpm) and its mean variation?
- Does the background's mean and variation change for short and long measurements?
- Discuss the importance of determining the radiation background compared to the type of samples that are measured in this experiment.
- How could the detection background be improved?

Calibration efficiency

- Which is the theoretical activity of ^{40}K of 1,9 g of KCl? Note that K contains 0.0117% of ^{40}K.
- Which is the calibration factor *k* (and its uncertainty) and efficiency?
- Which is the k factor for ^{222}Rn?

Ingrowth and decay of ^{222}Rn and its progeny

- What do the ingrowth and decay curves show?
- Use the Bateman equations to demonstrate that secular equilibrium between ^{222}Rn and ^{214}Pb is reached after seven ^{214}Pb half-lives.
- Which are the ^{222}Rn (and/or ^{220}Rn) half-lives obtained from the experimental numbers? Discuss the results comparing them to the bibliography.
- Calculate the activity of ^{226}Ra in the uranium rock considering secular equilibrium between ^{226}Ra and its daughters and assuming 100% ^{222}Rn adsorption efficiency on the activated coal.

Measurement of Rn in environmental samples: air and water

- Discuss the decay curve of the dust sample. How many isotopes can you see there? To check this, best it to display the activity in log scale. See if there are different slopes. Then apply a fit to each slope, orapply a double fitting.
- Which are the Rn concentrations in air and water at t_0? Compare the results obtained in water and air samples.
- How do the $T_{1/2}$ compare to the theoretical ones?
- Estimate the dose that one might receive if working in the place where the dust has been collected. Consider only the ^{222}Rn concentration and secular equilibrium between ^{222}Rn and ^{214}Pb.

- Compare the Rn values obtained in these environmental samples to other sources of dose to humans.

5. References

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6. APPENDIX

Safety procedures

There are few safety steps to be considered in this experiment:

1. In order to avoid contamination and/or damage of the detector, it is imperative that the detector window is not put in direct contact with the sample or touched with the fingers.
2. Please wear gloves when manipulating the salt and the activated carbon. These products might cause irritation to your skin and eyes, specially the activated carbon.
3. Before manipulation, please make yourself familiar with the safety sheet about the chemical products provided in the sample prep area. Make sure the container of activated carbon is properly closed after use because it is inflammable.
4. The samples provided in the laboratory produce low level ionizing radiation. Despite their activity is below the threshold of a dangerous radioactive source, it is good practice to keep the sample sealed and off direct contact. Specially, the samples produce radon, and thus, please make sure the sample container are well closed.
5. It is possible to leave the Ranger ON for several days and to perform overnight measurements. Please, when performing measurements out of normal hours (8 am to 6 pm), fill in the overnight measurement document and place it on the table next to your measuring device.
7. Please clean any surface and material before the experiment is finished.
8. Of course, please follow general laboratory rules or recommendations, including not eating food or drinking in the experiment room.
9. Pease keep track of the experiment using a notebook or any electronic device.

After you finish the measurements, what?

Each student needs to send preliminary plots and the .txt files to the VP assistant. He/she will check and ensure the student has enough and meaningful data. The, he/she will provide the signature so that the student can enroll to the next experiment.

Report writing, few recommendations

Here few recommendations are given regarding the structure and key aspects that the students usually forget:

Abstract: Use short sentences to introduce the topic, the setup and the performed measurements. Summarize the main findings and provide most important results (numbers) to make the abstract also quantitative. Briefly state the lessons learnt from the experiment.

Introduction: please introduce concepts of radioactivity, the types of decay, the natural decay chains and focus specially on radon and its daughter nuclides. It is important that in a final paragraph the aim of the experiment is stated.

Materials and Methods: describe the setup, the types of samples and the procedures used for sample preparation and measurement. It helps to subdivide this section is small paragraphs for each measurement (background, efficiency, rock ingrowth, rock decay, other samples (e.g. air, water). Explain the general approach used to treat data. Here you should briefly mention the binning, the background and k factor and any consideration regarding error calculation.

Results and discussion: I recommend putting results and discussion all together. Divide this section in background, efficiency, rock ingrowth, rock decay, other samples (e.g. air, water). For each section, use one paragraph to introduce the Figure and described what it is seen. In a second paragraph, discuss any data fitting done to extract the half-life(s) and provide an in-depth interpretation. Each section should have one or two main figures. You may add a table(s) if you think it is convenient.

Conclusions: begin again stating what was your aim. Then summarize the main findings from a critical point of view. Usually helps to dedicate one sentence to each measurement. Finally, suggest recommendations for future research and make recommendations based on your results.

Acknowledgements: please acknowledge any person and organization/institution involved in helping you to conduct the experiment

References: It is important to cite in the main body of the report the external literature (if possible peer-reviewed literature). Especially when discussing agreements/disagreements between your experimental results and the theoretical values. A list of the references has to be included after the acknowledgements.

Appendix: please mention the main safety measures followed to protect yourself and the Ranger from exposure to samples, etc.