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Radon-222 Radioactivity in Air

Introduction

Three radioactive families of isotopes U-238, Th-232, U-235 are present in the Earth with half-lives comparable to the solar system. The isotopes decay through radioactive series and produce intermediate inert gases isotopes of radon (Rn-222, Rn-220, and Rn-219, respectively), which seep into the air. The radon isotopes further decay into metallic ions along the decay chain and accumulate on dust particles. In this lab, we used an air sampler to collect dust particles on filter paper to identify the radioactivity of Rn-222 in the air. By removing the radioactivity from the background and Rn-220 through data analysis, we determined the radioactivity of Rn-222 in the air to be approximately 16.5 disintegrations per minute.

Theory

Many nuclei are unstable and can decay spontaneously to some other combination of nucleons that has a lower mass. Alpha decay (α) emits 2 neutrons and 2 protons, which is a helium atom. Beta decay (β) emits 1 electron. There is also gamma decay (γ) where a photon is emitted, but it isn't relevant to the decay processes studied. In this lab, an aluminum foil screen and a small detector window size stopped α particles and only allowed β particles through to the detector. [1]

Three radioactive families of isotopes U-238, Th-232, U-235 will be investigated in this lab [1]. The specific information of each isotope's decay is outlined in the below table.

Table 1. Radioactivity of U-238, Th-232, U-235 [1][2]

Isotopes	**U-238**	**Th-232**	**U-235**
Half-life	4.47×10^9 years	1.40×10^{10} years	7.04×10^8 years
Decay series	4n+2 series	4n series	4n+3 series
Radon (half-life)	Rn-222 (3.82 days)	Rn-220 (10.64 days)	Rn-219 (3.9s)
Decay products (downward in time, labelled by decay type)	| α Po-218 (3.1 min) | α Pb-214 (26.8 min) | β Bi-214 (19.9 min)	| α Pb-212 (10.64 hr) | β Bi-212 (61 min) | α Tl-208 (3.1 min) | β Pb-208 (stable)	Irrelevant since Rn-219 has a short half-life.

The main focus of this lab will be the radioactive 4n+2 series that originates from U-238. This series produces Rn-222 which decay into metallic ions and accumulate on the paper sample. However, the Geiger Muller tube detects decay events indiscriminately. There exists other decay series in the atmosphere and experimental environment, which mainly come from two sources:

1. The constant background radiation from cosmic rays and gamma-rays in the room.
2. Radioactivity from Th-232. This decay (4n series) produces Rn-220 and has longer half-lives. The 4n series will be dominated by Pb-212 with a half-life of 10.64 days.

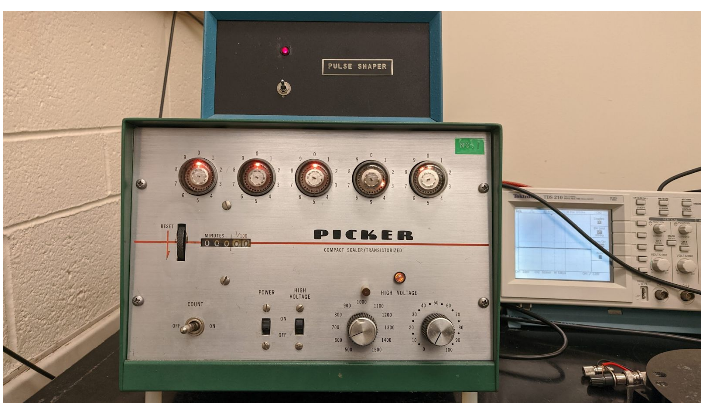
Note that although U-235 contributes to the 4n+3 series and produces Rn-219, we can neglect its effect because the half-life of Rn-219 is short compared to the half-life of both the Rn-222 (3.82 days) and Rn-220 (10.64 days) [1].

The radioactivity of both 1 and 2 mentioned above will be subtracted from the data collectively after modelling to a semi-logarithmic fit. Since the half-life of the metallic ions products of the Rn-222 are relatively short, we will only consider the first half an hour of data.

At secular equilibrium, the rate of production is equal to the rate of decay. Therefore, the number of decays per second of Rn-222, Po-218, Pb-214, and Bi-214 are equal. Assuming the secular equilibrium was reached at t=0 (after 45 minutes of air sampling), we can determine the rate of decay of Rn-222 by evaluating the rate of decay of Pb-214 at t=0 [1].

Procedure

Three recordings were measured: Cesium-137 source (figure 4), background radiation, and filter paper from the air sampler. The lab setup of the recording was the same across the three trials. The digital recording of the number counts can be read from the Picker Scalar, as well as from the "Radioactivity In Air" (AIR) program via a Data Acquisition (DAQ) card. Figure 1 shows the setup of the experiment.



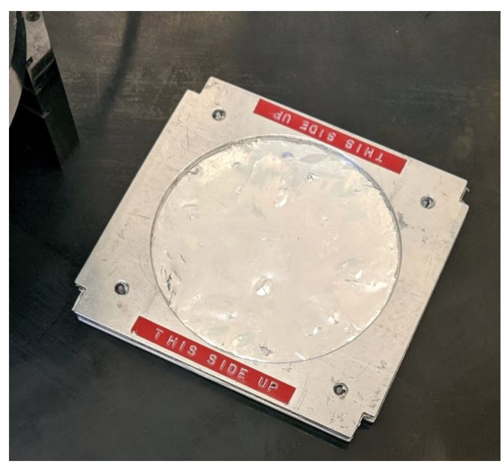


Fig 1: Setup of Experiment: Picker Scaler(left) and Geiger-Muller Tube Aluminum Filter (right). Note that the aluminum filter is thin enough for beta particles to pass, but not useful for alpha or gamma decay[1].

The Geiger-Mueller Tube is a gas-filled ionization detector with high voltage applied across the tube provided by the Picker Scaler. Figure 2 shows a block diagram of the setup. The Picker Scaler also acts as a counter for emitted particles from the radioactive decay. When an ionizing radiation bumps into an electron on the high voltage wire, there is a measurable negative charge accumulation. This is transferred into a negative voltage pulse, which is detected and logged by the LabView program.

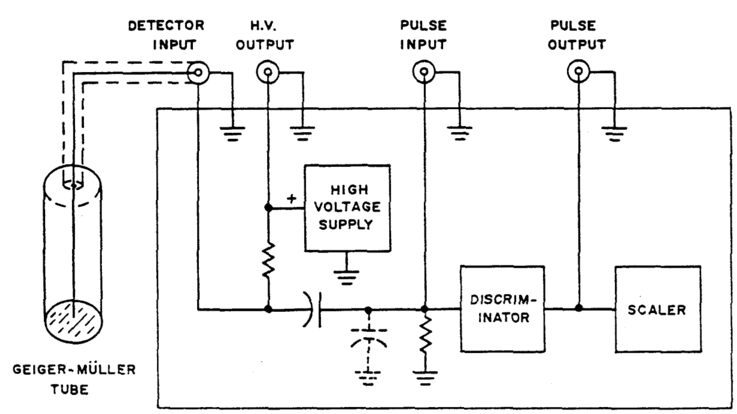


Fig 2: Block diagram of the setup [1]

In the first trial, we determined the optimal operating voltage. A sample of cs-137 was placed in front of the GM tube. The GM tube is then connected to the Picker Scaler, the pulse shaper, and the DAQ card. The detector input of the Picker Scaler was set to a voltage between 600V and 1000V at 50V increments, and at 1200V. For each voltage, the number of counts were taken at 15s interval for a total of 2 minutes. It was determined that a voltage of 800V the number of counts plateaued without showing unknown behaviours, hence the remainder of the lab was operated at a voltage of 800V. (see Fig 3)

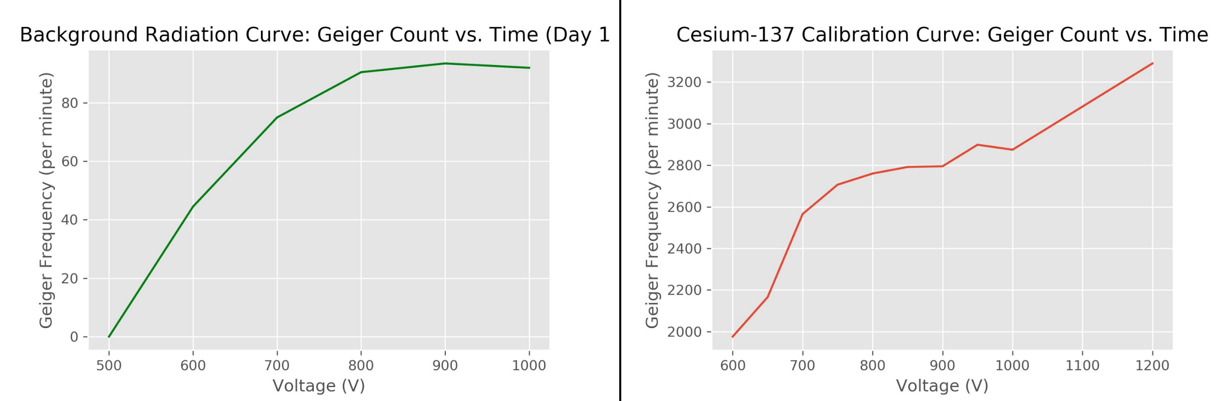


Fig 3: Radiation Curve for background (left) and Cesium-137 source (right). Note that we expected the Geiger frequency to plateau at a similar voltage. This voltage was determined to be 800V for both background and cesium. However, there was some unknown behaviour from the cesium source after 900V. Hence we chose 800V as the operating voltage.

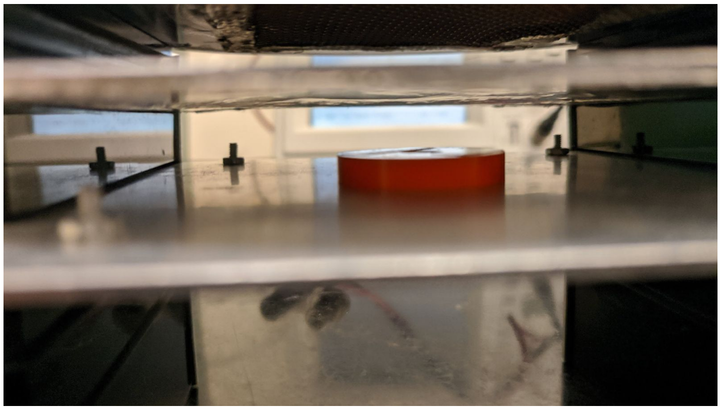
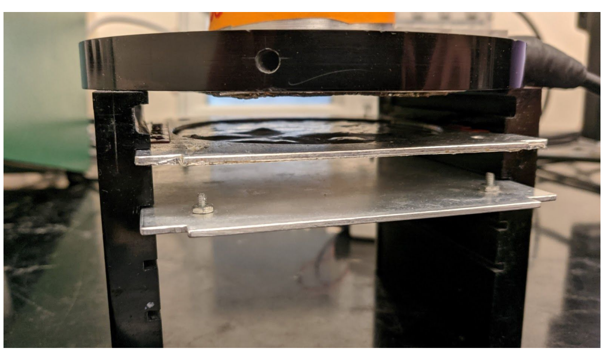


Fig 4: Setup for cesium measurements (left) and cesium sample (right)

In the second trial, we measured the background radiation in the room. No samples were placed in front of the GM tube. The background radiation from cosmic and gamma rays was measured via the same method in 1 with operating voltage of 800 V.

In the third trial, a paper filter was exposed to the air and collected particles in the air for 45 minutes (9:15 AM - 10:00 AM). The air sampler was taken at the west side of the McLennan Physical Laboratories (MP250) at the University of Toronto. On the day of sampling (11 March 2020), the atmosphere was partly cloudy and had an atmospheric pressure of 100 kPa). The paper was then placed in front of the GM tube in between two metal plates. The measurement of decay was conducted with the same method as 1 and 2. The number of counts were taken at 20s intervals for a total of 110 minutes.



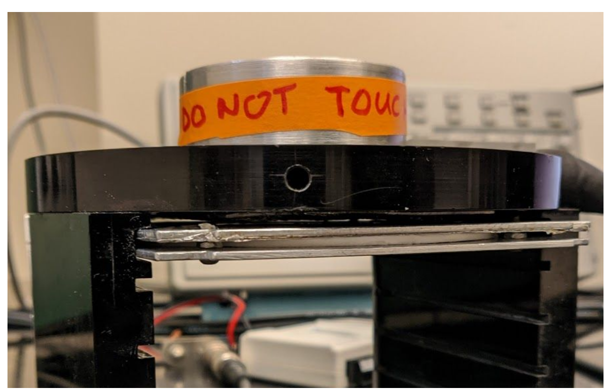


Fig 5. GM Tube Configuration for background (left) and filter paper (right) placed within the aluminum filter.

Data Analysis

Note that plots are not shown with error bars as the Picker Scaler records discrete counts of radioactive decay events. We trust the equipment to be accurate, so counts do not have errors.

The total Geiger-Muller tube measurements are shown below in figure 6. But only the short-lived Rn-222 decay series is of interest. It only appears in the first 30 minutes of detection. The GM tubes detect all decay events, so the data needs to be "cleaned up" to remove the background and long-lived Rn-220 decay before analyzing the Rn-222 series.

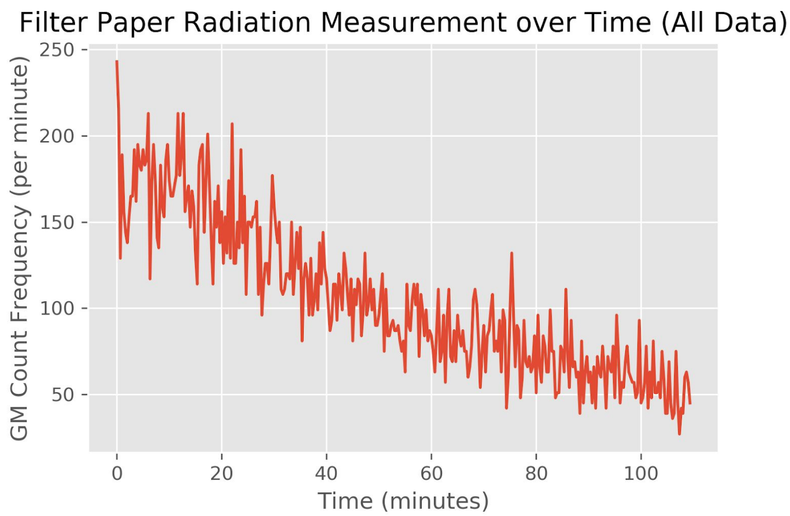


Fig 6. The total GM tube data shows Rn-222, Rn-220, and background radiation. Some "cleaning up" is required to extract only the Rn-222 data.

The first step is to create a fit of long-lived decay products from Rn-220 which is present throughout the data collection. This will be subtracted from the total data to leave only the short term Rn-222 decay, which is present in the first 30 minutes of data collection. Note that fitting for Rn-220 long-term decay also takes into account the background radiation. So we do not need to subtract away the background radiation again.

Radioactive decay means that the rate of disintegration is proportional to the amount of isotope that is currently present. This implies an exponential relationship. In curve fitting, a semilog plot should be used to fit a straight line. The known parameters are A(t), the measured data, and the k constant, related to half life.

$$A(t) = A_0 e^{-kt}$$
$$ln\, A(t) = ln(A_0) - kt$$
$$y(x) = b + mx$$

Using data after 30 minutes, both log ($\chi_v = 3.2$) and linear fit ($\chi_v = 3.18$) looked good. Both the residuals plots lacked any discernible trends (figure 8). From these 2 goodness of fit criteria, it seems that a log fit should be used since radioactive decay is inherently an exponential process. However when we used this fit, the mathematical model for Rn-222 daughters broke down and produced astronomical estimations. The log function explodes as we move back in time, so slight variations in the fitting data causes massive changes when we apply the mathematical model later on. The log model seemed to overfit the noise. To mitigate this, a linear fit was chosen instead. In addition, visually the data seems better approximated by a linear fit than logarithmic. See figure 7.

The linear fit used is:

$$GM\ count\ frequency\ (t) \ = \ -(0.93 \pm 0.05)t \ + \ (150 \pm 3) \quad (1)$$

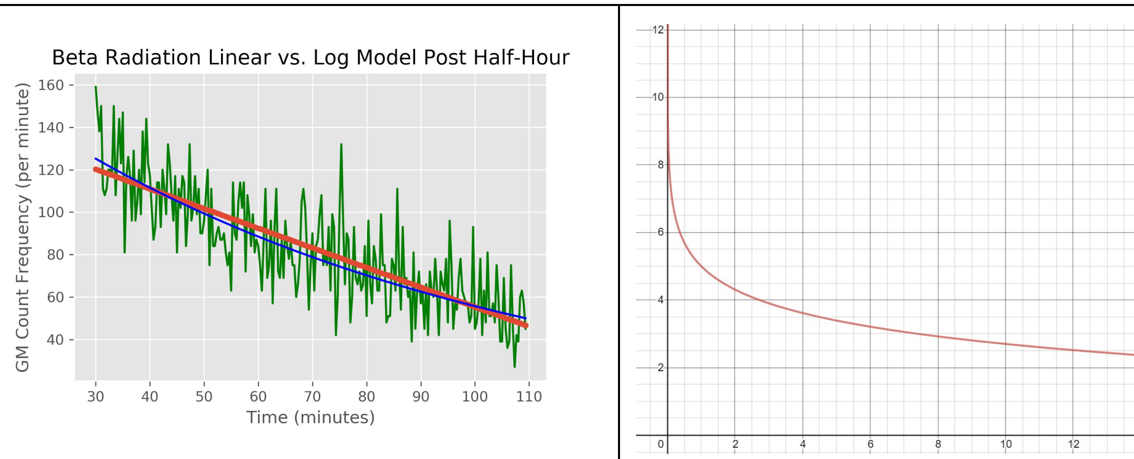
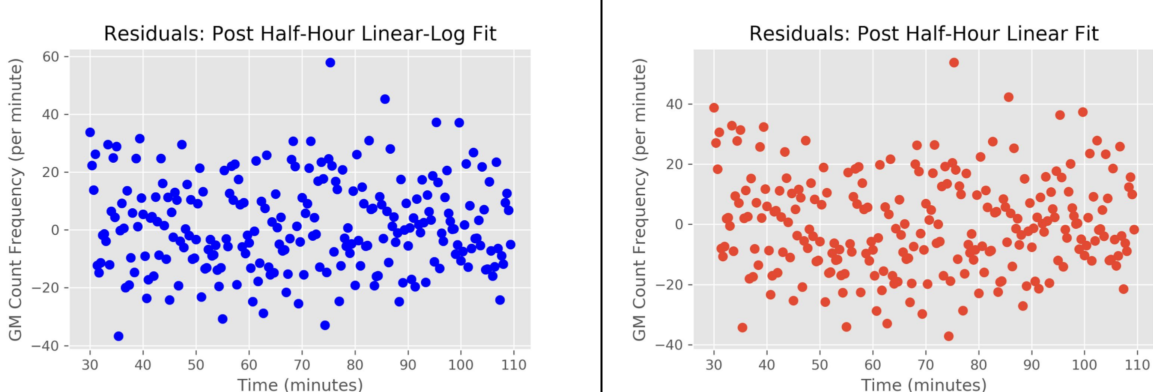


Fig 7. Both linear (left, red) and log (left, blue) fit suggested a good fit. The log fit overfit the noise and produced unphysical estimates when we extrapolated back in time 0-30 minutes. In addition, the "curvey-ness" characteristic of a logarithmic relationship (right) is not seen. So a linear fit was chosen over the true logarithmic nature of radioactive decay.



χ_v = 3.2	χ_v = 3.18

Fig 8. Residual plot for log (left) and linear fit (right). Both residuals appear random, indicating acceptable fit. Both reduced chi-squared are small and suggest appropriate fit.

The second step is to subtract away the Rn-220 long-lived decay series and background from the original data, to produce a "corrected" data. It only shows the Rn-222 short-lived decay series which is of interest, shown in figure 9 right.

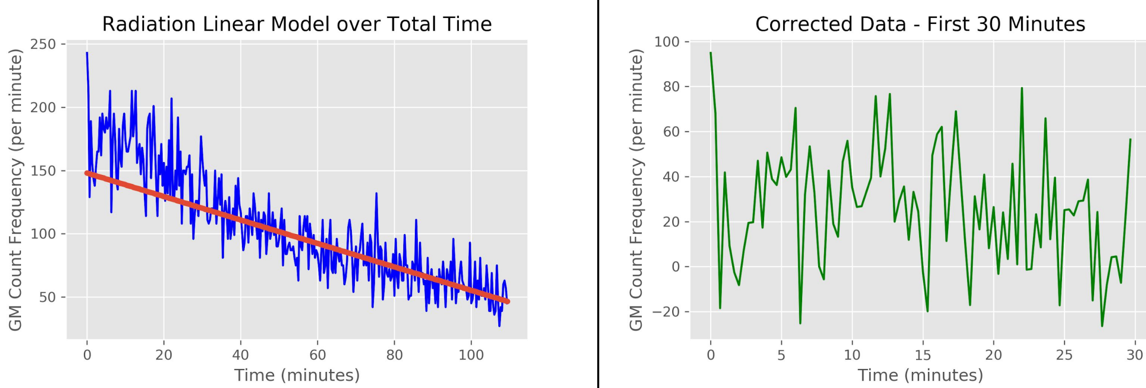


Fig 9. From 0-30min, the excess decay events above the trend line (left) is from Rn-222 short-lived decay series. Only the Rn-222 is of interest.

The last step is to apply the mathematical model of Rn-222 daughters on the "corrected" data. Details are found in the lab manual. At a high level, it models the detector activity as the sum of 2 radioactively decaying isotopes Pb-214 and Bi-214, where Pb-214 decays into Bi-214. The 2 parameters to be fit are:

$$R = \frac{Bi^{214}(0)}{Pb^{214}(0)}, \qquad \textit{where } Bi^{214}(t) \textit{ and } Pb(t) \textit{ are the radioactivity of the 2 isotopes}$$
$$A_{isotope}^{observed} = \varepsilon_{isotope} * A_{isotope}^{actual}, \qquad \textit{where } \varepsilon_{isotope} \textit{is the efficiency of the detector} \quad (2)$$

The model was created in Python. The fit was generated using SciPy's "curve_fit" function which takes in an initial guess for R and $A_{Pb-214}^{observed}$.

The output values are:

$$R = 0.734$$
$$A_{isotope}^{observed} = 13.245$$

The SciPy generated fit is shown in figure 10 left. This produced a reduced chi-squared χ_v=25, which is not particularly good. However the residuals plot (figure 10 right) appears random which indicates no systematic error in fitting.

Fig 10. Decay of Rn-222 series modelled (blue) on actual data (green). The reduced chi-squared χ_v is 25 and the residuals appear random.

Discussion

From the above data analysis, we obtained $A^{observed}_{isotope} = 13.245$. To get the actual rate of decay, we re-arranged (1) to divide the observed rate of decay by the efficiency of the detector for Pb-214, $\varepsilon_{Pb214}=0.8$. This leads to an actual decay rate of 16.5 GM detections per minute, for Pb-214. Since the system is in secular equilibrium, the decay rate of anyone of the series will give the activity of Rn-222 present. Therefore the activity of Rn-222 in the air is 16.5 disintegrations per minute.

Standard radon radioactivity measurements are given in Bq/m^3, which translates to number of radioactive decay events per second, per volume. The WHO states that radioactivity of radon in air is typically 5-15 Bq/m^3 [4]. Unfortunately, the amount of air that passed through the air sampler is unknown. The lab technicians turned on the air pump for 45 minutes. We did not examine the pump or ask the lab technicians about air volume. So we're left with a "decay per time" value without any means to convert it to a "decay per time, per volume" value. Thus, it's difficult to say if the experimental value agrees with literature.

Regardless, we can fairly certainly conclude that the decay rate is lower than usual. The weather on the day of testing was cloudy, 80% humidity, temperature hovering about 0°C [5]. It had rained 3.6mm the day before. It had been constantly humid and overcast for the previous 5 days. The rain and humidity likely caused dust particles to accumulate and fall to the ground, effectively purifying the air. With less dust particles to collect, the observed radioactivity was smaller than normal.

Sources of Errors

To reduce the time delay between retrieving the filter paper sample and start of detection, we ran from the air pump to the experimental setup. There was lots of jostling and shaking of the filter paper. Accumulated dust particles may have been knocked off the filter paper, leaving reduced radioactive decay.

The detector efficiency ε was assumed to be correct from the lab manual. However this value has a large sway effect, since it directly multiplies the actual activity level to get the observed activity level. The factors that affect this ε are the aluminum filter and GM tube window. We're placing a lot of trust in the lab manual and the permittivity of the filter has not changed or degraded.

The Geiger Muller tube has a "dead time" immediately following a decay detection, in which it cannot detect any other decay events. When viewed on an oscilloscope (n=12), this period was measured to be (1230±60) ns. There's a decay event roughly 2-3 times per second from the filter paper. It's highly unlikely that 2 decay events would happen within 1.2µs of each other. So the dead time is definitely a source of measurement error, but not significant.

The mathematical model for daughters of Rn-222 accounts for 214Pb β-decaying into Bi-214. But it does not account for any Po-218 ($t_{1/2}$= 3.1 minutes) decaying into Pb-214. There would've been a small amount of Po-218 present on the filter paper. But since it decays completely in 10-15 minutes, it was easier to neglect its effect and work with a simpler model. A more detailed analysis should account for this in the decay model.

Conclusion

This lab examined the radioactivity of Rn-222 (originating from U-238) in the air by filtering the dust particles that the decayed products accumulated on. The side effects from the background and series not of interest were eliminated through mathematical modelling. To prevent overfitting noises, we chose a linear fit over a logarithmic fit when modelling the radiation. The reduced chi-squared χ_v of the fitting was 25. The radioactivity of Rn-222 in air was found to be approximately 16.5 disintegrations per minute.

References

[1] Bogdon Scaunasu, Marko Korelek, Radioactivity In The Air Lab Manual, University of Toronto.
[2] C.M. Lederer; J.M. Hollander; I. Perlman (1968). Table of Isotopes (6th ed.). New York: John Wiley & Sons.
[3] S. T. Thornton, *Modern physics for scientists and engineers*. CENGAGE LEARNING, 2019.
[4]World Health Organization. *Radon and Health*. 30 June 2016. Available:
https://www.who.int/news-room/fact-sheets/detail/radon-and-health
[5] Toronto Historical Weather Data. Environment and Climate Change Canada. Available
https://toronto.weatherstats.ca/metrics

Appendix

Note: Since the number of counts were not direct measurements, but discrete counts from the DAQ card, uncertainties were not assigned.

For information about the mathematical modelling, see the accompanying Jupyter notebook, hosted on Aman's github: https://github.com/amanb2000/Radioactivity_Analysis

1. Raw calibration data from Cs-137

Table X: Raw Number of Counts for Cesium-137 taken at 15 s intervals.

	Voltage (V)									
Index	**600**	**650**	**700**	**750**	**800**	**850**	**900**	**950**	**1000**	**1200**
1	487	568	638	712	721	701	671	700	748	818
2	493	523	653	666	685	713	735	743	729	816
3	461	527	686	730	616	692	688	704	710	768
4	484	563	604	661	694	705	682	703	685	791
5	447	557	639	639	707	662	715	717	719	841
6	531	555	644	712	690	688	700	726	746	838
7	485	542	637	639	682	676	721	722	695	851
8	531	497	630		725	746	678	782	717	856
9	511									
10	494									
11	483									
12	521									

2. Raw background radiation count (day 1)

Table X: Raw Number of Counts for Background Radiation Taken at 15 s Intervals.

	Voltage (V)					
Index	500	600	700	800	900	1000
1	0	12	26	23	26	29
2	0	7	18	28	27	22
3	0	11	15	14	17	25
4	0	16	16	24	29	18
5	0	11	23	20	21	15
6	0	11	22	25	27	19
7	0	11	15	12	27	33
8	0	10	15	35	13	23

3. Raw background radiation count (day 2)

Table X: Background Radiation Table Taken at 20 s Intervals

	Voltage (V)				**Voltage (V)**	
Index	700	800		Index	700	8000
1	17	9		30	6	8
2	15	8		31	8	6
3	5	10		32	10	11
4	8	8		33	13	9
5	11	8		34	16	8
6	14	11		35	10	11
7	6	6		36	8	11
8	13	7		37	6	14
9	14	7		38	9	10
10	12	14		39	4	10
11	7	8		40	7	8
12	7	13		41	11	13
13	13	13		42	12	16
14	11	12		43	16	14
15	15	9		44	9	10
16	16	18		45	7	17
17	11	8		46	16	10
18	8	9		47	13	12
19	8	8		48	18	9
20	6	10		49	210	13
21	10	3		50	0	13
22	13	14		51		11
23	8	8		52		7
24	5	10		53		14
25	24	11		54		12
26	10	11		55		9
27	10	16		56		10
28	7	15		57		84
29	14	11				

4. Raw Number of Counts For Air Sampler Radiation, Taken At Intervals of 20 s

Index	Counts	Index	Counts	Index	Counts	Index	Counts
1	15	101	39	201	25	301	31
2	81	102	50	202	25	302	15
3	72	103	36	203	20	303	16
4	43	104	42	204	22	304	19
5	63	105	48	205	26	305	26
6	52	106	41	206	35	306	14
7	48	107	49	207	37	307	21
8	46	108	27	208	34	308	16
9	51	109	39	209	27	309	27
10	55	110	42	210	18	310	17
11	55	111	39	211	25	311	17
12	64	112	32	212	30	312	19
13	54	113	43	213	21	313	16
14	65	114	32	214	28	314	25
15	61	115	35	215	29	315	20
16	60	116	40	216	33	316	13
17	64	117	33	217	36	317	13
18	61	118	46	218	25	318	23
19	62	119	38	219	27	319	15
20	71	120	48	220	25	320	12
21	39	121	41	221	31	321	13
22	58	122	39	222	21	322	25
23	65	123	34	223	33	323	15
24	58	124	29	224	31	324	9
25	47	125	31	225	14	325	14
26	45	126	38	226	20	326	13
27	61	127	38	227	33	327	20
28	53	128	31	228	44	328	21
29	51	129	40	229	32	329	19
30	62	130	37	230	22	330	15
31	65	131	33	231	30		
32	58	132	44	232	29		
33	55	133	41	233	16		
34	55	134	36	234	20		

35	57	135	32	235	31		
36	59	136	39	236	23		
37	71	137	27	237	22		
38	59	138	37	238	24		
39	63	139	34	239	21		
40	71	140	39	240	22		
41	52	141	38	241	28		
42	55	142	28	242	17		
43	57	143	33	243	32		
44	49	144	44	244	22		
45	56	145	32	245	19		
46	53	146	34	246	28		
47	44	147	39	247	26		
48	38	148	33	248	21		
49	61	149	37	249	21		
50	64	150	30	250	33		
51	65	151	30	251	25		
52	48	152	32	252	25		
53	58	153	36	253	16		
54	67	154	40	254	17		
55	57	155	25	255	17		
56	47	156	37	256	26		
57	38	157	28	257	25		
58	54	158	28	258	21		
59	49	159	30	259	37		
60	57	160	31	260	24		
61	46	161	29	261	18		
62	52	162	29	262	31		
63	42	163	30	263	22		
64	51	164	27	264	23		
65	44	165	25	265	20		
66	58	166	27	266	21		
67	43	167	21	267	13		
68	69	168	38	268	27		
69	42	169	30	269	15		
70	42	170	29	270	21		
71	50	171	35	271	24		

72	45	172	38	272	19		
73	64	173	34	273	21		
74	46	174	38	274	15		
75	55	175	24	275	22		
76	36	176	36	276	14		
77	50	177	33	277	24		
78	50	178	28	278	21		
79	49	179	33	279	20		
80	51	180	27	280	26		
81	51	181	29	281	21		
82	54	182	28	282	14		
83	36	183	25	283	24		
84	49	184	21	284	22		
85	32	185	29	285	21		
86	38	186	37	286	26		
87	42	187	23	287	16		
88	42	188	25	288	32		
89	38	189	32	289	25		
90	48	190	19	290	15		
91	59	191	31	291	19		
92	53	192	37	292	18		
93	49	193	24	293	23		
94	46	194	23	294	26		
95	50	195	29	295	21		
96	37	196	23	296	20		
97	36	197	32	297	19		
98	37	198	28	298	19		
99	40	199	26	299	16		
100	40	200	29	300	17		