Identification of radon/thoron daughters in basement air

D. M. Wood, Emeritus Associate Professor
Department of Physics, Colorado School of Mines

September 30, 2023

Contents

Reminder: green links below are clickable.

1 *Recap*

The measurement of radionuclides captured in a furnace filter in 2017 is described in the document *Measuring radiation accompanying radon*. From the fit half-life of about 47 ± 1.04 (standard error) minutes it was assumed that the excess count rate was from radon daughters and plausible candidates (and one for thoron, ^{220}Rn) were identified. The planned gamma ray spectroscopy discussed there is described in this document. Note that, unlike the Geiger-Müller detector used earlier, the gamma ray detector will not be sensitive to beta particles emitted during decay.

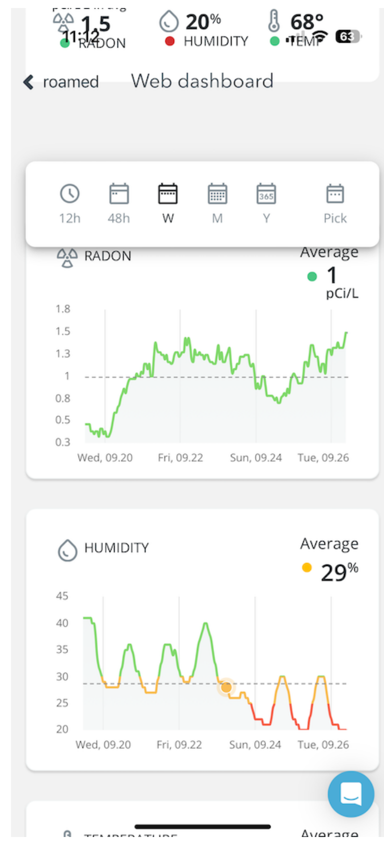

Figure 1: Typical data for radon levels and relative humidity in my garage, using a Wave Radon detector from Airthings.
By late September relative humidities have been as low as 20% and radon levels as high as 1.7 pCi/l, so I will attempt another sampling when the meteorology improves.

2 *Sample preparation*

My first attempt (low radon concentrations of about 0.5 pCi/l and 35% relative humidity as reported by a Wave Radon) using the balloon method was fairly successful. (Longer term data is shown in Fig. 1). I used a swatch of rabbit fur from Hobby Lobby and a plain gray 12 inch balloon (no printing) from a local supermarket and rubbed the balloon (minimizing handling) for about a minute. I was encouraged to hear static discharge sounds while rubbing; it is possible I left it suspended above the floor in the basement for *too long* (about 50 minutes) and some of the static charge dissipated. [Revision: probaby not. See the Appendix about the time dependence of the balloon charge.]

I gently deflated the balloon by sticking down a short length of Scotch tape (to inhibit tearing) near the neck and then carefully slicing through this with the corner of a box cutter blade, until air escaped fairly quickly (a few seconds). I then stuffed the deflated balloon into a small plastic pouch, sealed it, and cleaned the exterior with alcohol to avoid contaminating the sample chamber. I placed the pouch atop the scintillator can.

The gamma system is a 2" × 2" NaI:Tl scintillator and a photomultiplier tube encased in a thin sealed Al cylindrical can attached to a GammaSpectacular audio card spectrometer providing 700 V bias to the PMT. For this work the cylindrical can was placed inside a closed PVC cylindrical shield with a hollow cylindrical bore for the Al can, with 95 lbs of lead shot between the bore and inner PVC surface.

3 *Data acquisition*

The excellent PRA (Pulse Recorder and Analyser) free software from Marek Dolleiser (available via `gammaspectacular.com`) was used for pulse detection and data acquisition (including the count rate over time and the gamma ray pulse height histogram [the spectrum]). Gamma ray energy calibration was done with a ^{133}Ba and ^{137}Cs check sources. A cubic polynomial such as shown in Fig. 2 is adequate for the 700V bias voltage used.

Further analysis was done with InterSpec, another excellent free program from Sandia National Laboratory, for peak fitting and assignment of peaks to radionuclides.

I had already done an overnight gamma count and had found 18.28 counts/s for background. I counted the balloon packet for an approximately similar length of time.

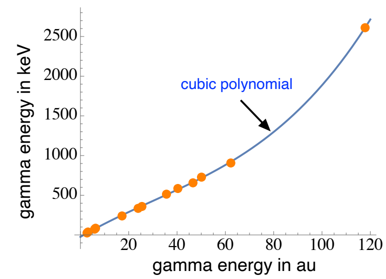

Figure 2: Typical cubic polynomial fit to calibration points from check sources. A ^{60}Co source will eventually be used to fill in data from 60-120 au.

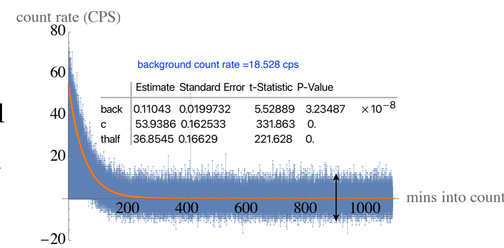

Figure 3: Parameters of fit to count rate. Vertical black arrow indicating spread due to Poisson statistical fluctuations of background rate. The Poisson standard deviation should be the square root of the mean background count rate.

4 *Analysis*

- I exported counts per 1 second interval, subtracted the background count rate, and fit to $a + b\, 2^{-t/t_{\frac{1}{2}}}$ to extract the best fit half-life and effective background (which should be near zero since I'd already subtracted the previously measured background rate). The value of 36.9 minutes [Fig. 3] was not very far from what I had measured years earlier with a Geiger-Müller counter.

- Using 5 photopeaks in the spectrum of ^{133}Ba and ^{137}Cs sources for calibration, I exported the gamma spectrum in keV and imported it to the fantastic InterSpec software (free from Sandia National Labs), where I fit the peaks and carefully assigned them to gamma peaks expected for ^{222}Rn and ^{220}Rn (thoron) daughter peaks (to be precise, to ^{214}Pb and ^{212}Bi peaks). The deviation in keV of the measured peak froom the nominal value is shown in red. Two prominent peaks which could not be assigned (no corresponding prominent vertical line) to radon or thoron daughters were excluded (large red Xs). Most other peaks are very well represented, although the expected intensities of the three peaks between about 200 and 400 keV increase with energy while the peak heights decrease. Note that the absolute efficiency of the NaI:Tl + PMT de-

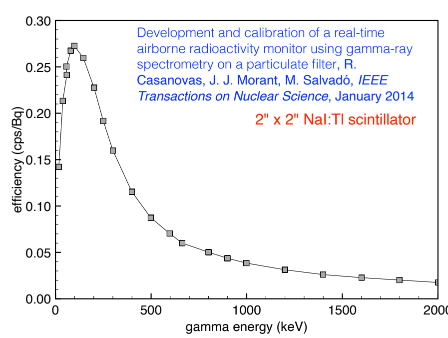

Figure 4: Efficiency of a 2" × 2" NaI:Tl detector much like the one I used.

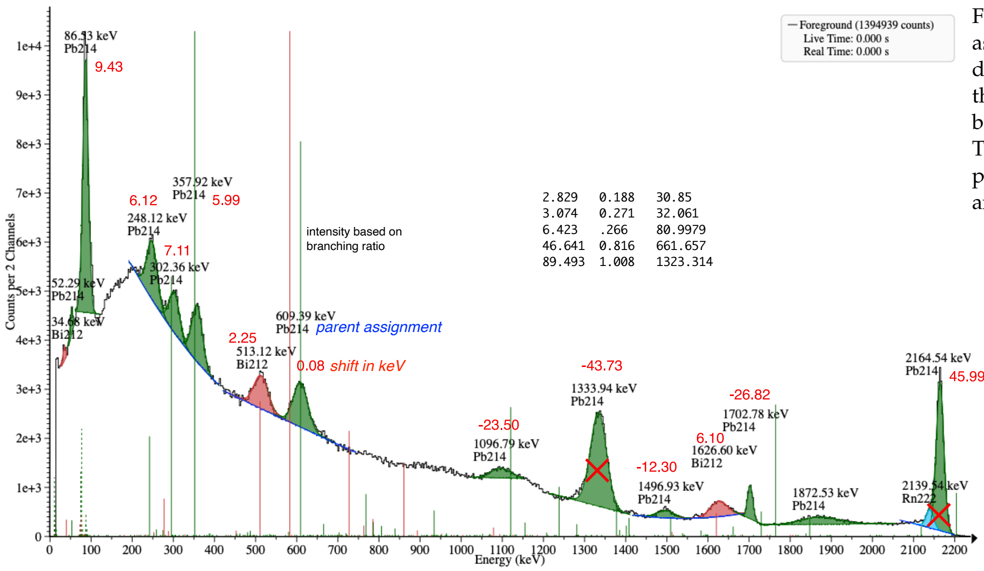

Figure 5: Initial balloon spectrum assuming presence of ^{214}Pb (a radon daughter, green peaks) and ^{212}Bi (a thoron daughter, red peaks). Vertical bars indicate relative brightness of lines. The 5 inset rows indicate calibration points between units of raw spectrum and keV.

tector was not applied but probably looks very much like what is shown in Fig. 4. The low-energy end (below about 200 keV) is in effect 'boosted' because of the efficiency peak for the gamma detector used.

It is *always* useful to subtract the background specrum to identify artifacts.

Not all features in a gamma spectrum are due simply to a well-defined gamma ray. Some are continuously distributed, such as the Compton scattering of photons from electrons, or (at high count rates) *sums of gamma energies* when two gammas arrive so close in time that the detector mistakes the count for one gamma with summed energies. There are also direct indications of special relativity: a gamma ray of energy above twice the electron rest energy can produce an electron/positron pair. (See below.)

- I knew from past experience that because of imperfect shielding, high-energy gammas would penetrate and could give high-energy peaks from *background* sources outside the sample chamber. Thus I subtracted the spectrum obtained from the background count from that for the balloon, even though the count times were somewhat different (and acknowledging this). Sure enough the two spectra (Fig. 6) it is clear that at higher gamma energies (roughly above 50 au or 700 keV) the count rate and features are dominated by background, including the ^{40}K peak at about 1460 keV.
- The initial assignment of peaks (before subtraction of the background spectrum) suggested the presence of multiple ^{214}Pb peaks and a couple of ^{212}Bi peaks (513 keV and around 1626 keV). However, expected strong ^{212}Pb peaks (notably, around 590 keV) were missing. The assigned ^{212}Bi peaks essentially disappeared in the subtracted spectrum shown in Fig. 7, thus leaving us with a mystery about the origins of the 513 keV peak. This is almost definitely due to the absorption in the scintillator crystal of one of the two gamma rays (generally of energy 511 keV) from the annhilation of an electron and a positron following the production of an elec-

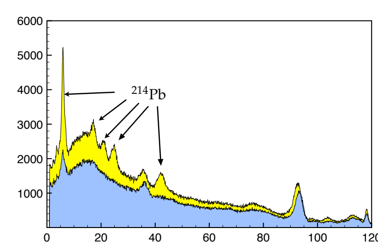

Figure 6: Gamma spectrum for balloon count and backgrouind count. The spectra are very similar (apart from detector drift-induced peak shifts) at high energies, indicating penetration through shielding of high-energy photons. At lower energies peaks due to radon daughters are evident.

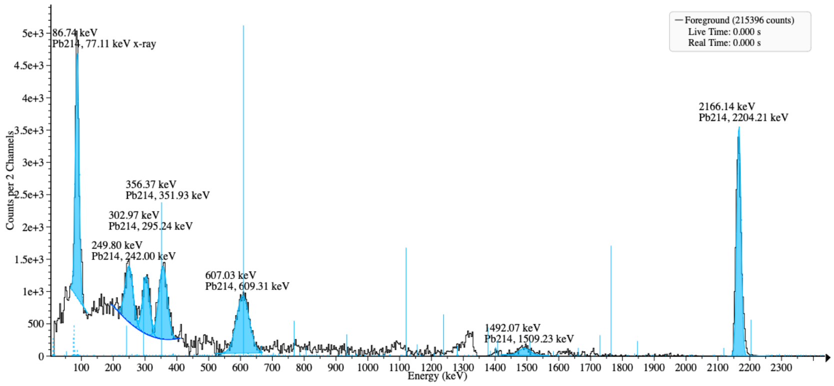

Figure 7: Gamma spectrum strictly due to radon daughters accumulated on the balloon.

tron/positron pair by a high energy gamma–there are plenty of ^{40}K 1460 keV gamma rays. The spectroscopist's curse–detector drift, the need for constant re-calibration (not done), and even energy-dependent peak shifts—is evident when we compare the background spectrum and baloon spectrum, shown in detector units ('arbitrary units' or au) in Fig. 7. In general when comparing measured gamma peak energies and their nominal values shifts around 8 keV or somewhat below are fairly typical for the NaI:Tl detectors of this size. The subtraction of background also partly restores the expected peak height trend seen in the vertical lines between 200 and 400 keV. Application of the detector efficiency curve would remove the enhancement of the first peak's height.

In order to proceed, pair production requires a 'momentum sink' to conserve momentum. There are plenty of high atomic weight Pb nuclei (to act as this sink and carry off little recoil kinetic energy) in the shielding, so in some sense the 511 keV gamma line 'comes from' the shielding, although the source of high-energy gamma rays probably is abundant ^{40}K in soil. This argument indicates why the 511 keV line is present in the *background* spectrum, not in the balloon spectrum.

5 *Conclusions*

We conclude that the clear ^{224}Pb peaks are thus at 77.11 keV (X-rays), 242, 295, 352, and 609, and 2204(?) keV. There is strong evidence for radon daughters (half-life 3.8 days), but not for thoron daughters.

Appendix: Maximizing charge on balloon

It occurred to me after my first gamma count of a balloon that it would be interesting to place a data-logging Geiger-Müller counter close to the balloon (but not so close as to induce a charge in the case which could pull the balloon and discharge it). I speculated that there may be a reasonable time at which a maximum (or maybe asymptotic) number of aerosol particles are bound to the balloon, as indicated by a maximum count rate on the counter, before the balloon begins to lose aerosols (because of relative humidity effects presumably). An interesting recent article [1] specifically examines how triboelectric charging depends on relative humidity and the size of what is being charged. It is a good introduction to this literature, uses familiar (to physicists) electrostatics, and experimentally tests the model they present. Surface charge densities (for very small metal spheres) and how they depend on relative humidity are also measured and discussed. For their situations, relative humidities above 50% are deadly to charge accumulation for small spheres.

In fact, something like this *did* occur, but without great clarity about the ideal time to expose the balloon to aerosols before beginning a gamma count. Results are shown in Fig. 8. If the balloon is too far from the Geiger-Müller detector only background is detected (black curve, top panel). I removed the plastic case and suspended the balloon about 2" above the (sensor face-up). On the second run (shown in red) in the figure, I was able to capture a maximum of the count rate before a gradual decline began. A third and final count showed that the maximum is relatively flat with no sudden onset of discharge (or decay of captured radionuclides).

The charge accumulated on the balloon appears to depend mostly on the relative humidity and the amount of rabbit fur rubbing (presumably accounting for the count rates shown in black and red in the top panel) and fairly weakly on how long the balloon is exposed to atmospheric aerosols. The shallow maxima of the count rate occur at 1272/12 = 106 minutes or 2282/12 = 190 minutes. These are already inconveniently long in comparison to the the half-lives of radon daughters: 3.1 minutes for ^{218}Po, 19.7 minutes for ^{214}Bi, and 26.8 minutes for For thoron, the half-life of its daughter ^{212}Bi is 60.5 minutes. The evolution of the most intense gamma lines with time is shown in the document *Measuring radiation accompanying radon* elsewhere on the website. But naturally the radionuclides on the balloon surface continue to be replenished until the balloon is deflated in put in the sealed pouch. It seems reasonable to expose the balloon to air for 30-60 minutes as a matter of convenience.

Finally, the bottom panel in Fig. 8 shows the strong dependence of the time scale for discharge of static electricity (under bulk room conditions with cool and warm temperatures) on relative humidity [2]. The (latex) balloons of interest are large enough that this data is probably relevant to balloon charges as well. This time scale evidently can change by an order of magnitude over a range of 20% in the relative humidity. For this reason it may well be worth waiting for dry conditions before attemping to concentrate aerosols on the balloon.

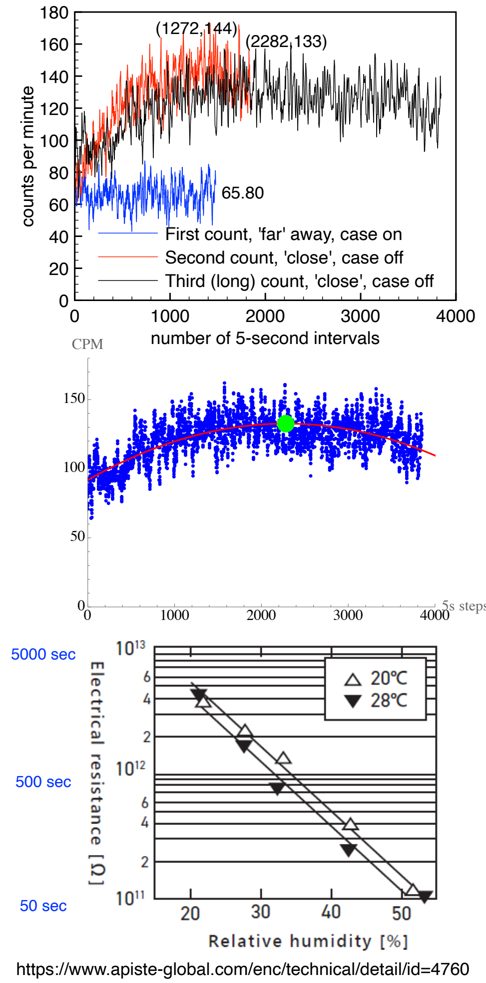

Figure 8: Count rate over time, top panel; maxima are labeled. Middle panel: location of maximum via quadratic fit. Bottom panel: extreme sensitivity (exponential dependence, indicated by straight lines on log scale) of discharge time on relative humidity.

References

[1] Reuben D. Cruise et al. “The effect of particle size and relative humidity on triboelectric charge saturation”. In: *Journal*

of Physics D: Applied Physics 55.18 (2022). ISSN: 13616463. DOI: 10.1088/1361-6463/ac5081.

[2] Apiste. *Relationship between static electricity and humidity*. URL: https://www.apiste-global.com/enc/technical/detail/id=4760.