Student Project

Radioactive Salt and Gamma Spectroscopy

Thomas Gwise

There are ways to identify radioactive elements by looking at the radiation they emit. Through investigating some table salt substitute, I will guide you through one of these processes. Salt substitute, a food seasoning that uses potassium chloride in place of sodium chloride, was chosen as a subject because it is fairly common and 0.0117% of all potassium is the naturally occurring radioactive isotope of potassium, potassium-40 (K-40). Figure 1 shows the energy peak from radioactive K-40 in a sample of table-salt substitute. The graph is output of a gamma ray spectroscopy program.

Gamma spectroscopy is a computationally intense technique for identifying elements by the radiation they emit. It is employed in a diverse group of fields, from oil exploration deep under the oceans to astronomical investigations of the heavens. As a student of statistics, I find gamma spectroscopy particularly interesting because it is so dependent on statistical methods. Every time a gamma spectrum is analyzed a series of statistical applications, from differencing processes to control charting and goodness of fit tests, are performed. By the end of this article, one will see that commercial gamma spectroscopy programs are in essence specialized statistical packages locked into one data format.

Gamma Spectroscopy

The best way to start this tour is with a clear description of gamma spectroscopy. Gamma spectroscopy, as the name implies is the investigation of gamma spectra. What are gamma spectra? We will review some basic physics to make the rest of the explanation understandable to those unfamiliar with the terminology. Gamma rays are a subset of the electromagnetic spectrum. Recall that the electromagnetic spectrum consists

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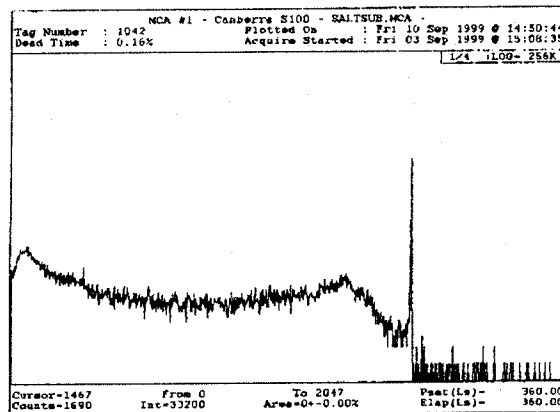
Figure 1.

of all forms of electromagnetic radiation. Visible light and radio-waves are examples of electromagnetic radiation. The whole electromagnetic spectrum is made up of the following types of radiation in order of ascending energy: radio waves, microwaves, infrared radiation, visible light, ultraviolet light, x-rays and gamma rays. The different kinds of electromagnetic radiation are really all the same basic stuff. Electromagnetic radiation can be thought of as little packets of energy called photons. The energy levels of photons determine their common names. For example, a photon with an energy level of 200 kilo electron volts (kilo eV or keV) will be called a gamma ray. Another way in which we refer to subsets of the spectrum is to name the different energy levels according to their wavelengths. The equation $E = hc / \lambda$, where h = Plank's constant and c = speed of light, relates photon energy to wavelength. Convenience normally determines how one identifies waves in the spectrum. Referring to 100 kilo hertz (3000 meter) radio-waves as 4.1E-10 eV waves would be cumbersome.

Back to gamma spectroscopy. A gamma spectrum can be thought of as a type of organized photograph. In a photograph the light photons interacting with the film over a controlled time

Table 1		
Common Name	Wavelength (m) (Approximately)	Energy (eV) (Approximately)
Gamma Rays	$< 1 \times 10^{-10}$	$> 1.2 \times 10^4$
X-Rays	1×10^{-15} to 1×10^{-8}	124 to 1.2×10^9
Ultraviolet	1×10^{-8} to 4×10^{-7}	3.1 to 124
Visible Light	4×10^{-7} to 7×10^{-7}	1.77 to 3.1
Infrared	1×10^{-6} to 1×10^{-4}	1.24×10^{-2} to 1.24
Microwaves	1×10^{-3} to 3×10^{-2}	1.24×10^{-5} to 1.24×10^{-3}
Radio Waves	1×10^{-2} to 3×10^3	4.1×10^{-10} to 1.24×10^{-4}
Electric Waves	$> 3 \times 10^3$	$< 4.1 \times 10^{-10}$

period causes an image to be recorded. A gamma spectrum is the result of recording gamma photons interacting with a crystal over a controlled time period. The big conceptual difference is that in gamma spectroscopy we organize the photons according to energy level rather than form a picture. To collect a spectrum we use a detector, a series of amplifiers and a computer. Photons interact with the detector creating electronic pulses proportional to their respective energy levels. These pulses go through a series of amplifiers and then to a computer that tallies them in bins according to their pulse size. At the end of a prescribed collection time, each bin will contain a number corresponding to the number of photons counted for a given energy level. The results are usually represented graphically. An example of such a graph is seen on the opening page. The graph resembles a histogram with the energy level as the x-axis and the number of photons detected in each energy gradation as the y-axis. The spectroscopy software used here does not label the axes, but the display does show that for the selected channel (number 1,467) 1,690 events have been recorded. Generally, the elements of the graph are some gamma peaks and what is called a

Gamma into the detector

γ

Each gamma's energy causes an electric pulse in the detector

Pulses

Detector

Analog to Digital Converter and Amplifiers

Refines and amplifies pulses

Computer

Pulses are assigned to a channel by size

Graph

The peak indicates that a large number of pulses (gammas) of the same size were detected.

X-Axis =Pulse size (energy level)
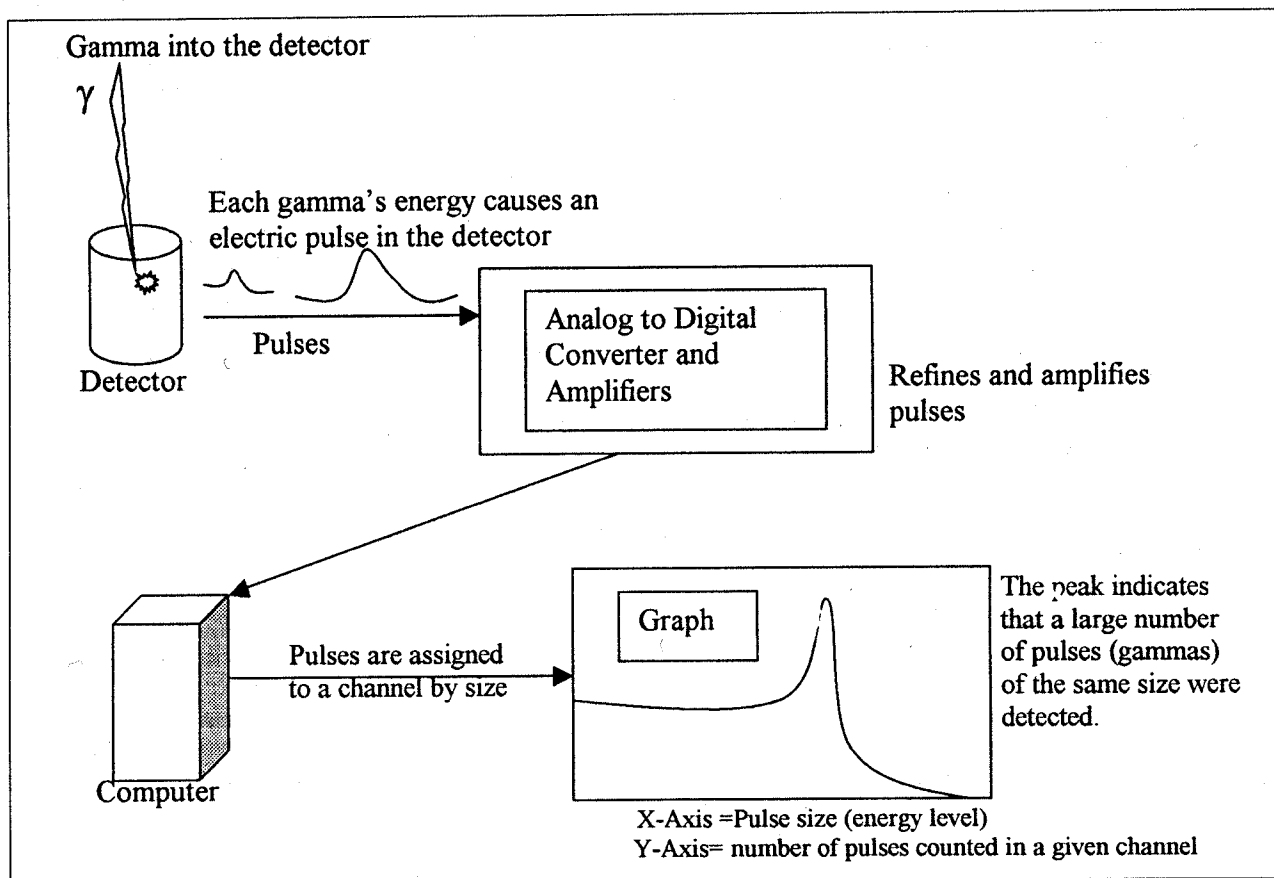Y-Axis= number of pulses counted in a given channel

Figure 2.

continuum. The continuum, without being too technical, is the rest of the graph made up of background radiation and residual energy from photons not completely absorbed in the detector. Existence of a peak in the graph indicates the detection of a large number of gamma rays of the same energy level. The flow chart, Figure 2, should help to visualize the process.

Gamma spectroscopy is useful because it can detect and identify gamma rays. Many radioactive isotopes of elements, or radionuclides, have unique gamma ray signatures. From a gamma spectrum it is often possible to tell the type and amount of radioactive element present in a sample. This fact makes gamma spectroscopy a useful tool in many fields. A geologist may want to know the quantity of thorium in a certain area. A medical technologist may need to know if he has been exposed to the radioactive iodine recently administered to a patient. Astronomers use a similar technique to investigate far off stars. In fact, different variations of this technique turn up in many surprising places.

Statistics Employed

Now that we know the nature and some uses of gamma spectroscopy, we will discuss some of the statistics involved. Since the object of the process is to match energy levels with peaks, we need to calibrate the computer's pulse counting bins, or channels, with respect to energy. Recall that pulses are sorted into bins according to their size and pulse size is proportional to energy. In our machine, the ratio of channels to keV is approximately one channel per keV. A gamma calibration source emitting gamma rays with energies of 59.5, 88, 122, 166, 279, 662, 898, 1173, 1332 and 1836 keV was placed on a germanium detector and a count, the term used for acquiring data, was taken for a period of 1000 seconds. Five more counts were taken. Each spectrum was inspected to determine the channels at which each peak had a maximum.

The channel numbers and the energy level vectors were entered into the analysis program Minitab and linear regression was then used to find a function relating energy to channel number. One must remember that the calibration problem is slightly more complicated than a standard linear regression problem.

I will restate the situation. In sample analysis, we have data stored in bins. We would like to assign an energy level to each channel. Using a known source of gamma rays and linear regression, we can obtain a representation of channel as a function of energy level. That is not the end. We need to identify the sample gamma rays based on channel number. To do this we invert the
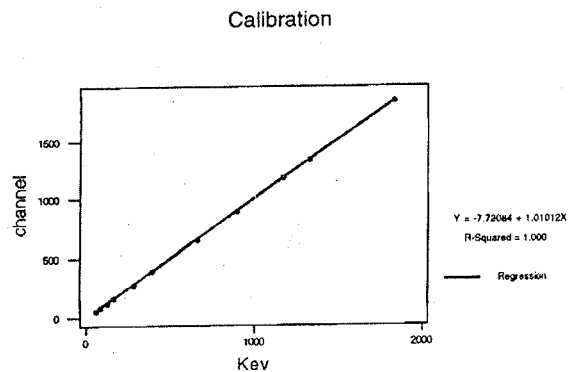
Calibration

Figure 3.

regression equation. It is important to note that this is not the same as using channel as the predictor and energy as the response in the initial regression. Our regression equation is:

$$\text{Channel} = -7.72084 + 1.01012 * \text{Energy.}$$

On the graph above, Y values represent channel numbers and X values represent energy levels in Kev. Solving this equation for energy, we get

$$\text{Energy} = .98998 * \text{Channel} + 7.643488.$$

When analyzing specimens of unknown radioactive material, we use the second equation above to assign energy levels to peaks found in the spectrum. Once the value for energy is found, a confidence interval for that value can be calculated. To see the details of these computations and for further discussion of regression as it is used in calibration see *Regression Analysis, Concepts and Applications* by Graybill and Iyer (pp. 427–431).

When viewing the graph of our original regression, it may seem that the line looks "too perfect." One should note that the amplifiers used for gamma spectroscopy are designed to have this linear property.

The plots in Figure 4, labeled "Calibration Residuals" and "Calibration Residuals vs. Fits" are for the regression above where "Channel" is the response variable. Inspection of these residual plots indicates that our standard regression assumptions of normal data and constant variance appear to hold.

Other parameters requiring calibrations are peak shape and efficiency. The fact that energy absorption properties in the detector will cause the peaks to be wider at the bottom for higher energy gamma rays necessitates a calibration of peak shape with respect to energy. Efficiency calibration is the standard process of relating a measurement to a known value. Because many radiation sources are isotropic, that is, they are emitted in equal number in all directions, only a fraction of their output can be incident on the detector. These two calibrations

Calibration Residuals vs Fits
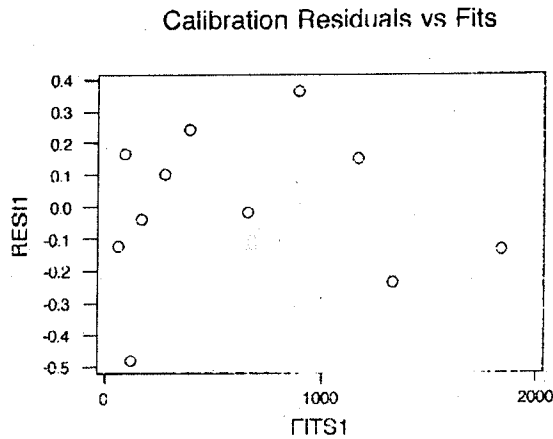
Calibration Residuals

Figure 4.

are more complicated than energy calibration and sometimes employ nonlinear regression procedures.

Now that we have our orientation with respect to energy, we can look at the rest of the analysis process. Many of the procedures used in gamma spectroscopy are complicated and a thorough discussion here would be very lengthy. In the interest of brevity, I will list, with a short description, some of these computations, many of which are statistical in nature. Most spectroscopy software packages run through a set of procedures similar to the following.

Peak Search

The second difference of the spectrum is obtained through a process similar to the differencing done in time series analysis. This process is analogous to finding the second derivative. Using the second difference, we are able to locate relative maxima, the peaks, in the spectrum.

Fitting

Least squares procedures identical to those used in linear regression are performed to fit continuous functions to the discrete data.

Integration

The area under each peak is proportional to the number of gamma rays counted for that energy. Numerical integration can be used to determine the true number of photons present.

Calibration

The number of counts in a peak can be related though the efficiency calibration to the units we desire. In other words, depending on our sample, we can get a number of photons emitted per unit area or volume or mass, etc.

Goodness of Fit

Chi-square fit tests are automatically
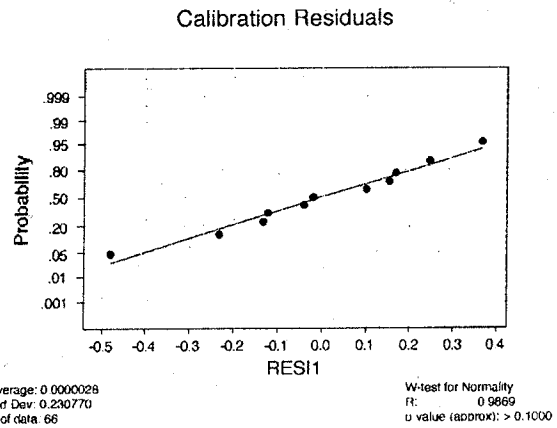
performed on the functions to alert the analyst to poorly fitting functions.

Propagation of Errors

The uncertainty in the count is determined and combined with the uncertainty of the calibrations to report a confidence interval or "error" with the measurement results.

Quality Control

Many commercial spectroscopy programs automatically keep quality control charts of key parameters.

Minimum Detectable Calculations

Minimum detectable concentrations or minimum detectable activities (MDA's) are determined for each sample count through a procedure based on the t-test showing what could loosely be thought of as the "power" of the analysis.

Data Collection

Aside from the statistical procedures, another aspect of gamma spectroscopy that may interest statisticians is the probabilistic nature of the data collection. The process of amassing data into a spectrum is a Poisson process. Recall that a Poisson process is a counting process with independent increments, and that the number of events counted in any interval of a given length has a Poisson distribution. Gammas arriving at a detector can be shown to meet these conditions. Further explanation of the Poisson process can be found in *Introduction to Probability Models*.

Another very interesting fact is that for a given gamma energy, the energy absorbed in the crystal will not be exactly the same, but will vary slightly. This is caused by certain characteristics of the detector crystal. The slight differences in detected energy level will cause peaks to have the shape of the normal distribution. That means for a given
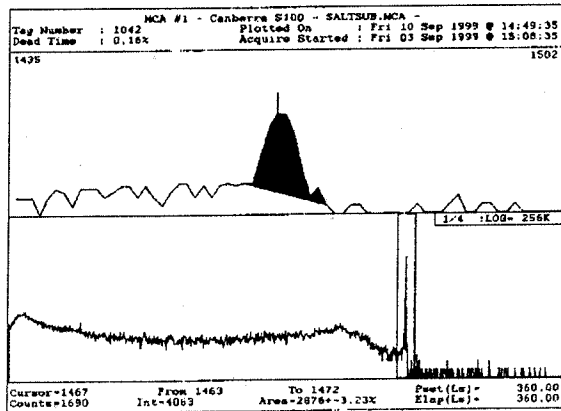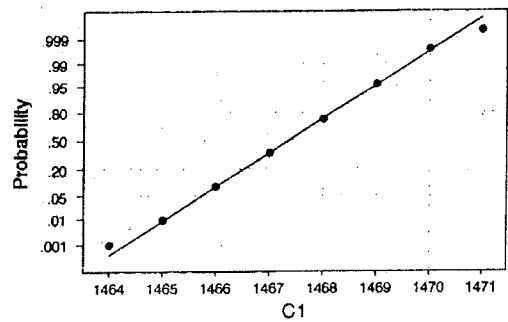
Figure 5.

Saltsub data

Figure 6.

gamma energy, the probability of collecting a certain number of counts over time is distributed as Poisson and the exact location on the x axis of the counts representing that gamma ray energy are distributed normally with the "true" energy level as the mean.

The plot of the salt substitute spectrum in Figure 5, with a close-up of the region between channels 1435 to 1502 in the upper frame, shows the Gaussian shape of the K-40 peak. To more closely investigate the shape of the K-40 peak, the data in the peak region were analyzed using Minitab. Keep in mind that we are investigating the energy spectrum, which is continuous, but our data are discrete. This fact will be important when choosing a test of normality. First, we look at a histogram of the data.

The shape of the histogram appears to be close to normal. Notice that the average of the peak data, when entered into the regression equation gives a result of 1460.3 keV. The energy level for K-40 given by The Table of Isotopes (Firestone et al., 1999) is 1460.8 keV. Our calibration is fairly accurate considering that the peak bounds, on which the mean will depend, were determined by eye rather than using an analytical method. Figure 6 is a normal plot of the data. The plot indicates that there is only a slight deviation from normality in the tails of the peak. This deviation is explained

in the theory of charge collection in detectors, and the transition from the peak to the continuum portion of the spectrum. Because the spectrum is continuous, but the data are discrete and forced into a small finite set of bins, the Anderson-Darling test of normality, based on the empirical distribution function and the default normality test in Minitab, indicates that the data are far from normal with a p-value of 0.000. The Ryan Joiner test of normality, which is based on a regression fit of the normal plot, gives an R-value of .9998 and a p-value reported as greater than .1. This seems to be in better agreement with the plot and supports our belief that the data is distributed normally.

To read more about the normality tests mentioned here, see *Goodness-of-Fit Techniques*, edited by D'Agostino and Stephens. Also, another reference is a paper entitled *Normal Probability Plots and Tests for Normality* by Thomas A. Ryan and Brian L. Joiner (1976) which is available on the Minitab website (*http://www.minitab.com/resources/whitepapers/index.htm*).

Measurement Results

To round out the tour, we will compare our measurement results with some standard reference information. The amount of K-40 we detected in our analysis agrees with the data found in the

Table 2. Character Histogram; Histogram of C1, N = 4053 Each * represents 35 obs.

Midpoint	Count	
1464	6	*
1465	61	**
1466	540	****************
1467	1690	***
1468	1423	***
1469	313	*********
1470	18	*
1471	2	*

periodic table and the chart of the nuclides. I have outlined the information and calculated an estimate for the amount of radioactivity that is expected in our sample of the salt substitute.

- .0117% of natural potassium is radioactive potassium 40 (periodic table).
- Potassium is 50% of the salt substitute (from product label).
- Our sample of salt substitute weighs 578 grams.
- The specific activity of K-40 is 1E-5 Curies/gram (Ci/gram). (*Handbook of Health Physics*).
- 1 Curie = 3.7E10 disintegrations per second. (*Handbook of Health Physics*).
- Efficiency of our detector for this sample = 0.01066 counts per gamma (prior calibration)
- The counting time for this sample was 360 seconds.
- The number of counts in the 1460.8 keV peak was 4053.
- The number of 1460.8 keV gammas per K-40 disintegration =0.1067 (*Handbook of Health Physics*).

.000117 * 578 grams*(50%) = .033813 grams of K-40 [percentage of K-40 in natural K]
.033813 gram*1E-5 Ci/gram [grams K-40* specific activity]
=3.38E-7 Ci [K-40 radioactivity in the sample]
= 338,130 pico Ci. (pCi) [unit conversion]
338,130 pCi / 578grams = 585 pCi/gram [K-40 radioactivity per gram of specimen]

Based on standard reference material, the amount of K-40 radioactivity we expect to see in a gram of salt substitute is 585 pCi.

Our measurement of the sample showed:
4053 counts / 360 seconds = 11.2583 counts/second
(11.2583 counts/second) / (0.01066 counts/gamma) = 1056.129 gammas/second
(1056.129 gammas/second) / (0.1067 gammas/disintegration) = 9898.11 disintegrations/second
(9898.11 disintegrations/second) / {3.7E10 (disintegrations/ second)/Ci}
=2.68E-7 Ci
(2.68E-7 Ci) / 578 grams = 4.63E-10Ci/gram
=463 pCi/gram

The result of our measurement is 463 pCi/gram. That translates to about 16 potassium 40 atoms self-destructing every second per gram of our sample. A commercially produced software package calculated a value of 437 pCi/gram ± 5.6%. The confidence interval is calculated by considering the variances for all measurement and calibrations. The results of our measurement are within reasonable agreement with the references when one considers the allowed variability on food labels. So yes, salt substitute contains small amounts of radioactivity, but so do all foods high in potassium, such as bananas.

Conclusion

With the aid of some salt substitute, we have taken a brief look at the intersection of statistics and gamma spectroscopy. We discussed the properties of gamma counting which allow us to model it as a Poisson process. The shapes of the gamma peaks being investigated are Gaussian. In fact, almost all of the tools used to analyze the data are statistical methods. Even the analysis report should please the statistician. Accompanying all measurements are well-founded confidence intervals and in the MDA value, an indication of the "power" of the analysis. This brief introduction to gamma spectroscopy shows why I say that this little corner of the physics world is almost all probability and statistics.

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