BAYESIAN ALGORITHMS FOR AUTOMATED ISOTOPE IDENTIFICATION

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THESIS

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ABSTRACT

Handheld radio-isotope identifiers (RIIDs) are widely used in the United States for nuclear security, but these detectors generally have poor performance in isotope identification. While much research is being conducted on alternative detector materials, there is much evidence that the primary problem with these automated identifiers is with the algorithms used for making identifications. We propose a new algorithm using Bayesian statistics that uses peak positions and areas to identify the source while allowing for calibration drift and shielding.
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CHAPTER 1

INTRODUCTION

1.1 Introduction

Many hand-held automated radio-isotope identifiers (RIIDs) have been deployed by the International Atomic Energy Agency (IAEA) and others to detect nuclear threats, track radioactive material shipments, and more. Most of these detectors use sodium iodide (NaI) spectrometers due to their efficiency, decent stability over a wide range of operating conditions, and their low cost [4]. However, evaluations of these commercial detectors have demonstrated that these identifiers perform poorly in isotope identification; in one of these evaluations, the detectors correctly identified approximately one-third of the radio-isotopes correctly [5].

While NaI(Tl) detectors are the most common commercially available handheld detectors, their low resolution, temperature drift, and nonlinearity of the energy response leave much room for improvement. Other detectors, such as LaCl$_3$(Ce), are promising replacements for NaI due to their superior energy resolution and more linear energy response but have not yet become widespread in usage due to their significantly higher cost [27].

While these other detectors have better energy resolutions than NaI, the issue with the currently-used detectors is not just their low energy resolutions but rather the algorithms that automate isotope identification [5, 8]. Improving the resolution of the detector allows for more sophisticated identification algorithms [12], but a trained spectroscopist can generally identify isotopes from low resolution spectra, even when automated algorithms fail [24], suggesting that there is much room for improvement in the algorithms themselves.
1.2 Motivation

While it is strongly desired for these identifiers to be accurate across all radioactive isotopes, the US Department of Homeland Security requires all future RIIDs produced to be able to meet a minimum identification standard set forth in ANSI N42.34. This standard requires the correct identification of the special nuclear material (SNM), industrial sources, and medical gamma sources, listed below: [1]:

- **SNM**: Uranium ($^{233}$U and $^{235}$U), $^{237}$Np, Pu
- **Industrial**: $^{57}$Co, $^{60}$Co, $^{133}$Ba, $^{137}$Cs, $^{192}$Ir, $^{204}$Tl, $^{226}$Ra, $^{241}$Am
- **Medical**: $^{18}$F, $^{67}$Ga, $^{51}$Cr, $^{75}$Se, $^{89}$Sr, $^{99}$Mo, $^{99m}$Tc, $^{103}$Pd, $^{111}$In, Iodine ($^{123}$I, $^{125}$I, $^{131}$I), $^{153}$Sm, $^{201}$Tl, $^{133}$Xe

Most of the current commercial spectrometers combine NaI spectrometers with a proprietary automated identification algorithm. However, these identifiers have an unacceptable identification accuracy. One published evaluation of these handheld identifiers by Blackadar et al. found that the correct isotopes were identified correctly in less than a third of all trials. When applied only to SNM, industrial, and medical gamma sources, the commercial identifiers produced correct identifications in less than a fourth of all measurements and produced false negatives in a third of all trials [3].

1.3 Existing Methods for Automated Isotope Identification

The performance of the currently-used automated identification algorithms does not generally meet the requirements of the users nor the ANSI standard [5, 27, 23]. Unknown shielding, low-count spectra, calibration drift, and other effects complicate the identification process, often leading to an incorrect identification.

There are several broad classes of algorithms for automated identification: library comparisons, region of interest (ROI) methods, template matching, and expert interaction [8]. While the algorithms on commercial detectors are
generally proprietary knowledge, only a few of these are practical for use on a RIID

Library comparison methods typically compare the peak centroid positions to an isotope library. These algorithms do not use the areas of the observed peaks to make identifications due to the complexity of automating an algorithm to identify areas accurately. This approach is generally used for high-resolution detectors due to the ease of accurately locating many peak centroids. However, inaccurate peak positioning (due to calibration drift and nonlinearity in the detector response function), which is common in NaI-based handheld detectors, and missing peaks due to shielding can lead to incorrect identifications [8].

ROI methods compare regions of the data to library ROIs by monitoring count rates above background levels. However, because library ROIs should not overlap, it can be difficult to use this method for larger isotope libraries or for isotopes with peaks close together [8].

Template matching compares the data to a spectral template library. By measuring the goodness of fit, whether with Mahalanobis distance, Euclidean distance, or another metric, the user can determine which template the data most closely matches. This method can be used to not only determine radioisotopes but also shielding materials, but source mixtures can be difficult to account for [8]. While template matching can handle shielding and calibration drift with a suitable template library, a library capable of handling these issues is too large for use on a handheld identifier.

Expert interaction algorithms generally compare peak energies and areas to a spectral library. However, this method requires the attention of a trained spectroscopist, which is not necessarily within the skill set of first responder and is not practical for general use. It is also not practical to use this type of algorithm on an automated RIID (as it is contradictory to the nature of an automated algorithm).

1.4 Peak Finding

Many algorithms for isotope identification must first employ a peak finding method to quantify the centroid energy and possibly the area of all photopeaks in a spectrum [12]. Peak identification methods have been specially
studied in the past 20 years to quantify peak energy, peak shape, and peak efficiency calibrations [11, 14, 26, 25, 20]. In the case of high-resolution detectors, a peak-finding algorithm combined with a spectral library lookup method can determine the presence of a given isotope(s) with a single measurement [12, 7, 2].

Some methods use a smoothing algorithm as a pre-processing stage in peak identification. However, smoothing introduces a bias that reduces the maxima and minima of spectra [9]. Further research is being conducted on smoothing algorithms that minimize bias without being excessively computationally intensive [9, 13, 17].

Further, other algorithms use some form of spectral decomposition to “condition” an observed spectrum. By inverting the detector response function (DRF), a “true” spectrum is estimated with improved resolution and ideally less noise [12, 21].

1.5 Proposed Solution

To combat many of these issues, we propose a new algorithm that will take measured peak energies and areas and compare them to library values while allowing for calibration drift and shielding. The detection of the peaks and measurement of their areas in a data set will be left to other methods such as wavelet analysis [26, 25]; this algorithm will take a list of peak energies and areas and return a probability for each isotope in the library. This algorithm uses Bayes’ theorem and a three-part probability model to assign these probabilities.

Our isotope library uses all of the isotopes in the ANSI Standard N42.34-2006 for hand-held RIIDS. This library contains special nuclear materials, medical radionuclides, naturally occurring radioactive materials, and industrial radionuclides [1]. Further, we have included a few extra isotopes for interesting test cases (\(^{152}\)Eu and \(^{177}\)Lu). For each isotope, this library contains the peak energies and branching ratios [6].

This work presents our first attempt at applying Bayesian analysis to the problem of isotope identification for any type of spectrometer. The functions presented in the three-part model are not necessarily optimal, but are reasonable starting points. As demonstrated in Chapter 6, the algorithm
produces accurate identifications for single source problems. Notably, the current version algorithm takes approximately 0.3 seconds to run in Matlab on an older laptop with an Intel Core2 Duo processor, suggesting it may be computationally cheap enough to eventually be used for handheld detectors.

In Chapter 3, a brief overview of Bayesian theory is presented, and in Chapter 4 we develop the Bayesian Radioisotope Identifier (BRAIN) algorithm. Chapter 5 presents an experiment to justify the probability models used in BRAIN, and Chapter 6 shows the identification results of this algorithm for several spectral problems.
CHAPTER 2

BACKGROUND: GAMMA SPECTROSCOPY

2.1 Gamma Spectroscopy

The nuclear decay of a parent radionuclide generally leaves a daughter nuclide in a highly excited state; the nuclide reaches a lower energy state by emitting a gamma ray with 100 keV or more. For example, $^{60}$Co decays to an excited state of $^{60}$Ni by beta emission and subsequently relaxes by emitting gammas with energies 1173 and 1332 keV respectively.

Because nuclei can only exist in certain well-defined energy states, the energies of the gamma rays emitted have very specific energies and branching ratios (a measure of how frequently a given emission occurs out of all possible decay chains). By measuring the emitted gamma radiation, a radiation source can be identified by its spectrum. We use the tables of emission energies and branching ratios in Browne and Firestone for all work presented in this thesis [6].

When a gamma ray interacts within a NaI scintillator, a count event is observed in a bin corresponding to the energy deposited in the detector. Ideally, the gamma ray entering the detector has the same energy as it did when it was emitted by a radioisotope and all of its energy is deposited in the detector. In this ideal case, the energy observed produces a photopeak in the spectrum.

However, many of the counts in a spectrum are not part of a photopeak. Instead, the corresponding gammas have lost energy via another interaction prior to entering the detector, do not immediately deposit all of their energy within the detector, or are the result of a different nuclear process.
2.2 Continuum Effects

In addition to the photopeaks, a gamma ray spectrum also contains a continuum of counts. Practically speaking, the spectrum will always exhibit a Compton continuum, and beta-emitter or high energy gamma emitter will also have a bremsstrahlung continuum.

2.2.1 Compton Continuum

In Compton scattering, a gamma photon scatters inelastically off an electron. By applying conservation of momentum and energy, one can show that the energy given to the electron is bounded by:

\[
E'_e(\theta = 0) = 0
\]

\[
E'_e(\theta = \pi) = E_\gamma \frac{E_\gamma}{m_e c^2} \frac{m_e c^2}{1 + \frac{E_\gamma}{m_e c^2}}
\]

By combining this result with the angular probability distribution given by the Klein-Nishina equation, one can derive the electron recoil energy distribution, shown in Figure 2.1. Since \( \theta = \pi \) gives the maximum electron recoil energy, this distribution has a sharp edge (the Compton edge), above which the distribution is zero. However, in the case of multiple scattering events (more significant for larger detectors), higher order continua can be seen.

2.2.2 Bremsstrahlung Continuum

Any source that emits \( \beta \) particles (or creates them via pair production) will have a bremsstrahlung continuum as well, though this is only significant with \( \beta \)-energies greater than 1 MeV [16]. As the energetic beta slows via electromagnetic interactions, a continuous x-ray spectrum is produce. This continuum has counts at lower energies (up to a few hundred keV), with most counts at the lowest energies energies [16].
2.3 Peaks From Other Processes

Not all of the peaks in a spectrum are due to a simple gamma emission by a radioisotope. High energy gammas can undergo pair production, causing up to three other peaks in the observed spectrum. Other nuclear processes, such as neutron absorption, can cause the emission of gamma rays as well. Finally, Compton scattering will produce a low-energy backscattering peak.

2.3.1 Pair Production

When a gamma ray with energy greater than $2m_e c^2 = 1.022$ MeV interacts with the strong electric field around a nucleus, the spontaneous creation of an electron-positron pair can occur. By energy conservation, any “leftover” energy from the pair production is shared by the electron and positron in the form of kinetic energy. For the relevant (to isotope identification) gamma ray energies (under 3.5 MeV), the electron and positron travel at most a few millimeters before losing their kinetic energy. When the positron annihilates with an electron, two gammas with energy 0.511 MeV are produced.

For the observed spectra, pair production has a few effects. First, if either the electron or positron escape, the corresponding 0.511 MeV is never
deposited in the detector, so a single escape peak is observed with centroid energy 511 keV less than the original incident gamma. If both the positron and electron escape, a double escape peak with energy 1.022 MeV less than the original gamma is observed. Finally, the annihilation process produces a wide peak at 0.511 MeV (width is due to the Doppler shift). These can be seen in the spectra of $^{232}$Th in Figure 2.2.

For automated isotope identification, the effects of pair production can be negated by comparing the locations of the observed peaks. Because single/double escape peaks are always 511/1022 keV lower than the photopeak, these peaks can be removed from consideration and do not obfuscate a spectrum significantly.

Figure 2.2: $^{232}$Th spectrum showing escape peaks and annihilation peaks
2.3.2 Backscatter Peak

Besides creating a continuum, Compton scattering also produces a backscatter peak. In the limit that the incident gamma has much more than \( \frac{m_e c^2}{2} \approx 255 \) keV of energy, the scattered gamma energy is independent of its initial energy [18]:

\[
E'_\gamma(\theta \approx \pi) = \frac{m_e c^2}{2} = 0.25 \text{ MeV}
\]

Because the energy of this peak can be easily calculated from the energy of the photopeak, these peaks do not pose a problem to automated isotope identification; we can simply filter out backscatter peaks by calculating the backscatter energy for each of the photopeaks in an observed spectrum.

2.3.3 Other Nuclear Processes

Other forms of radiation can cause secondary emission of gamma particles. In particular, neutron absorption causes a 2.2 MeV gamma via \(^1\text{H}(n,\gamma)^2\text{H}\). This interaction is especially useful because very few isotopes emit a gamma ray near this energy, so a 2.2 MeV peak is a sign of a neutron-emitting source.

However, we are interested in identification of gamma-emitting sources. Since a given detector has a known FWHM (full width at half maximum) versus energy curve, we can filter these extra peaks out of our data by measuring their widths. These secondary gamma rays are Doppler-broadened, so they are significantly wider than the other peaks in the spectrum.

2.4 Shielding

One of the largest problems for nuclear isotope identification is shielding. Gammas (and other radiation) are attenuated by shielding. A monoenergetic beam of radiation with intensity \( I(x) \) and energy \( E \) is attenuated approximately as [28]:

\[
I(x) = I(0)e^{-\mu x}
\]

where \( \mu \) is the total linear attenuation coefficient and \( x \) is the distance traveled in a medium. However, the total linear attenuation coefficient is a
function of the shielding material and the energy of the gamma particle; $\mu$ is shown for a few materials in Figure 2.3.

Figure 2.3: Total linear attenuation coefficient as a function of gamma-ray energy. [16]

As shown above, the linear attenuation coefficient $\mu$ is much larger at lower energies; that is, a gamma with lower energy is much more likely to interact than a higher energy gamma. In observed spectra, low-energy or small peaks may not be resolvable if any shielding is present. In the $^{152}\text{Eu}$ spectra in Figure 2.4, the peaks at lower energies are no longer visible. Also, since gammas at all energies are attenuated, the total counts observed in the spectra differ between the shielded and unshielded cases; with more shielding, one could completely hide the presence of a gamma-emitting source.

Our algorithm evaluates the data by comparing the ratios of the areas of neighboring peaks. Without knowledge about the activity of the source and the geometry of the measurement, it isn’t possible to judge on the area of a single peak. By comparing the ratio of peaks, the activity and geome-
try information is unnecessary for isotope identification, though it would be needed for determining the source activity if that was a quantity of interest. However, because the attenuation via shielding is a function of energy, the area ratios are reduced by a shielding material-dependent factor:

\[
\frac{A_1(\text{observed})}{A_2(\text{observed})} = \frac{A_1(\text{exact}) e^{-\mu(E_1)x}}{A_2(\text{exact}) e^{-\mu(E_2)x}} = \frac{A_1(\text{exact})}{A_2(\text{exact})} e^{- \left( \mu(E_1) - \mu(E_2) \right) x} \leq \frac{A_1(\text{exact})}{A_2(\text{exact})}
\]

When \( E_1 \approx E_2 \), we have \( \mu(E_1) \approx \mu(E_2) \) and the observed ratio approaches the expected library ratio. Also, a larger difference between the energies of these peaks will result in a larger uncertainty in the area ratio because the difference between the attenuation coefficients is larger and shielding can affect the area ratios more.

2.5 Summary

Various interactions of gamma radiation with materials add features to observed spectra, and shielding material reduces the relative size of lower energy peaks to higher energy peaks. In the field of isotope identification, we seek to identify radioisotopes in a wide variety of circumstances; we generally do not
have information about shielding material or activity of the sources. It is of paramount importance for a spectroscopist to understand all of the features of a gamma spectrum and the effects that these features have on isotope identification algorithms.
3.1 Introduction to Bayesian Statistics

Bayesian statistical methods differ from frequentist methods in that they are based on the notion that knowledge, or lack thereof, can be expressed as a probability distribution. Whereas a particular parameter $M$, such as the isotope responsible for a given data set, would be considered unknown but fixed by a frequentist, in a Bayesian view something is known about it. Before taking any data, a prior distribution $P(M)$ contains any a priori knowledge and assumptions about the parameter. In the case of isotope identification, we would assume that a spectrum is produced by a radioactive isotope that must be in our spectral library. If we don’t know if one isotope is any more likely than another, we would say that all isotopes in the library are equally likely and therefore uniformly distributed.

Next, data $D$ are obtained to obtain information about parameter $M$, which is expressed in terms of the density of the data $P(D|M)$ conditioned on the parameter $M$. The information from the data in the likelihood function $P(D|M)$ is combined with the prior information $P(M)$ to find the posterior distribution $P(M|D)$.

\[
P(M|D) \propto P(D|M)P(M) \quad (3.1)
\]

The preceding equation is a simple form of Bayes’ Theorem. A more complete form, says that the posterior probability is precisely [15]:

\[
P(M|D) = \frac{P(D|M)P(M)}{P(D)} \quad (3.2)
\]

Here, $P(D)$ is a normalization factor and is obtained by requiring that the sum of all probabilities is one [10]:
Decisions on the parameter are then based on the posterior distribution, whether by a point estimate (mean, median, etc.), a posterior probability set (analogous to a confidence interval), or by a Bayesian decision rule (choosing $M$ to minimize a cost function) [22].

### 3.2 A Simple Example and Motivation for Improvement

Suppose we observe a spectra containing one peak at 662 keV. For this example, suppose our spectral library contains only $^{137}$Cs and $^{241}$Am (both of which have a peak at 662 keV, although $^{241}$Am has several other, larger peaks). Before the measurement, we assume no prior knowledge, so both isotopes are assumed to be initially equally likely.

$$P^{(137}\text{Cs}) = P^{(241}\text{Am}) = \frac{1}{2}$$

Next, we need to compute the likelihood of each isotope. For this example, we will assume an (overly) simple probability model, where

$$P(iso_i|M) = \frac{\text{number of peaks of } iso_i \text{ observed}}{\text{number of peaks of } iso_i \text{ expected}}$$

$^{137}$Cs has 10 gamma peaks and $^{241}$Am has 148 photopeaks [6]. This simple model ignores the fact that most of these peaks are much too small to be seen; in practice, a NaI scintillator would only see one peak of $^{137}$Cs and a few peaks of $^{241}$Am, depending on the detector’s efficiency.

We apply this probability model to obtain the posterior likelihoods of each
isotope and then the normalization $P(D)$:

\[
P(D|^{137}\text{Cs}) = \frac{1}{10}
\]
\[
P(D|^{241}\text{Am}) = \frac{1}{148}
\]
\[
P(D) = P(D|^{137}\text{Cs})P(^{137}\text{Cs}) + P(D|^{241}\text{Am})P(^{241}\text{Am})
\]
\[
= \frac{1}{10} \times \frac{1}{2} + \frac{1}{148} \times \frac{1}{2} = \frac{79}{1480}
\]

Using Bayes’ Rule, we finally compute the posterior probability for each isotope.

\[
P(^{137}\text{Cs}|D) = \frac{\frac{1}{10} \times \frac{1}{2}}{\frac{79}{1480}} = \frac{74}{79} = 93.7\%
\]
\[
P(^{241}\text{Am}|D) = \frac{\frac{1}{148} \times \frac{1}{2}}{\frac{79}{1480}} = \frac{5}{79} = 6.3\%
\]

The probability function used to compute the likelihood has tremendous room for improvement, as it does not account for many important factors: the relative size of the peaks, how closely they match the library peaks, how much of the library spectrum was actually observed, how much of the data was accounted for by that isotope, and more. However, the basic ideas of this calculation are the very same as the algorithm presented in Chapter 4.
CHAPTER 4

THEORY: BAYESIAN RADIOISOTOPE AUTOMATED IDENTIFIER (BRAIN)

4.1 Introduction

To improve automated identification results on low-resolution identifiers, we have developed a new algorithm that uses peak energies and areas to make identifications. The Bayesian Radioisotope Identifier (BRAIN) algorithm uses probability models to score the isotopes in the spectral library to obtain identifications and a confidence-of-prediction.

This algorithm does not begin with a raw spectrum. A peak energy and area measurement algorithm, such as the wavelet transform method [26, 25], must be coupled to this algorithm. The wavelet transform method is currently being developed and will be combined with BRAIN in the future. For results presented in this work, peaks are detected manually, i.e. peaks are identified by eye and fit using a gaussamp fit in Origin; this fitting is discussed more in the experiment sections.

BRAIN uses a spectral library containing all of the isotopes in the ANSI Standard N42.34-2006 for hand-held RIIDS. This library contains special nuclear materials, medical radionuclides, naturally occurring radioactive materials, and industrial radionuclides [1]. Further, we have included a few extra isotopes for interesting test cases ($^{152}$Eu and $^{177m}$Lu). For each isotope, this library contains the peak energies and branching ratios convolved with our detector’s efficiency [6].

Most importantly, BRAIN is computationally cheap (running in 0.3 seconds in Matlab on an older laptop with an Intel Centrino 2 processor) while providing accurate identifications, as demonstrated in Section 6.
4.2 Probability Models for BRAIN

4.2.1 Bayes’ Theorem

From a list of peak energies and areas produced either manually or by an automated method, this algorithm will give each isotope a posterior probability that is a measure of how likely an isotope is to be responsible for the data relative to the other isotopes in the library. To this end, BRAIN uses Bayes’ Theorem and our proposed probability models.

Applying Bayes’ Theorem to the problem of assigning a probability to isotope $iso_i$ (a list of peak centroid energies and branching ratios convolved with the detector efficiency), given data set $data$ (a list of peak centroid energies and areas, as output by a peak identification algorithm), gives:

$$P(iso_i|data) = \frac{P(data|iso_i)P(iso_i)}{P(data)} (4.1)$$

To determine the final probability that an isotope is responsible for the given data, $P(iso_i|data)$, each of the three terms on the right must be computed for each isotope.

The first term, $P(iso_i)$ is the prior probability, i.e. the probability that a particular isotope is present, regardless of any observed data. Currently, a (non-informative) uniform prior is assumed, meaning all isotopes are assumed to be equally likely to be present before considering any data. There are several possibilities for better priors, some of which are situational-dependent (e.g. use the frequency that a given isotope crosses through a shipping lane, etc). An exploration of other priors and the sensitivity of the final posterior distributions to the priors is planned for the future; the non-informative prior is used as a demonstration of the method.

The second term, $P(data|iso_i)$, is the likelihood, the probability that $iso_i$ will produce the observed data set. A four-part model is developed in the following sections to compute this quantity. $P(data|iso_i)$ will be the product of four terms: $f_{LPI}$, $f_{DPI}$, $f_{CP}$, and $f_{PA}$, which respectively account for the percentage of library peaks identified, the percentage of data peaks identified, the peak centroid positions, and the peak areas.

$$P(data|iso_i) = f_{LPI}f_{DPI}f_{CP}f_{AR} (4.2)$$
Naturally, these probabilities will be highly dependent on the peak identification algorithm used. As discussed in Chapter 7, a refinement of these models will be conducted once BRAIN has been merged with an automated peak identification algorithm. For now, this algorithm will use approximated probability functions for these terms, which are explored briefly in Chapter 5 as well.

Lastly, $P(data)$ is the probability of observing this data and is a normalization factor. An expansion from basic probability theory gives:

$$P(data) = \sum_{i=1}^{n} P(data|iso_i)P(iso_i) + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} P(data|iso_i \cap iso_j)P(iso_i \cap iso_j) - \ldots$$

However, our algorithm truncates this expansion at the first order term:

$$P(data) \approx \sum_{i=1}^{n} P(data|iso_i)P(iso_i) \quad (4.3)$$

The expansion is truncated for three reasons. First, our model does not currently allow for the possibility of multiple isotopes (see Section 7), though this is a future goal for this algorithm. Second, for every isotope, the quantity $P(iso_i|data)$ is weighted by the same factor of $P(data)$; numerical error in this term does not affect the final relative probabilities assigned to each isotope. Third, truncation of this sum is equivalent to assuming that only a single source is present in the spectrum; under this assumption, Equation 4.3 is exact, as shown in Chapter 3.

In fact, it is simpler to think of $P(data)$ as the normalization factor. By requiring that the sum of the probabilities of each isotope equals one (applying the assumption that there is one isotope responsible for the data), we immediately obtain:
\[ 1 = \sum_{i=1}^{n} P(\text{iso}_i|\text{data}) = \sum_{i=1}^{n} \frac{P(\text{data}|\text{iso}_i) P(\text{iso}_i)}{P(\text{data})} = \sum_{i=1}^{n} \frac{P(\text{data}|\text{iso}_i) P(\text{iso}_i)}{P(\text{data})} \]

Thus, to compute the probability that isotope \( \text{iso}_i \) is responsible for the observed data set, BRAIN begins with a set of prior probabilities \( P(\text{iso}_i) \), calculates the likelihood function \( P(\text{data}|\text{iso}_i) \) for each isotope, and normalizes the product of the likelihood and the prior to obtain the posterior probabilities for all isotopes in the library. BRAIN currently chooses the isotope with the maximum posterior probability. Note that in the case of a non-informative prior, the posterior probability is directly proportional to the likelihood function. Then, BRAIN’s decision rule is equivalent to a maximum likelihood estimator (which is equivalent to a Bayes’ decision rule with uniform costs) in the case of a non-informative prior.

### 4.2.2 Peak Matching

Before computing any probabilities, the algorithm must match the data peaks with the library peaks. A simple nearest neighbor test is used to find the corresponding peak; that is, for \( \text{iso}_i \) in the library, each peak in the data is paired with the closest peak of \( \text{iso}_i \). Each of the peaks in the data are not uniquely matched to single isotope, but rather to the closest peak for each isotope.

However, only library peaks within a certain neighborhood of the data peaks will be allowed. Since calibration drift is generally more significant at higher energies, the peak identification algorithm uses a variable sized neighborhood that grows wider at higher energies. A linearly-increasing neighborhood is used here, though this term could be optimized for specific detectors with knowledge of how their calibrations tend to drift.

Given a data peak at energy \( E_D \), the energy neighborhood in which to
check for a library peak will be \([E_D - w, E_D + w]\), where \(w\) varies linearly between 10 and 20 keV. Assuming the detector has a maximum detectable energy of 3000 keV, the neighborhood \(w\) is given by:

\[
w(E_D) = 10 + \frac{E_D}{3000} \times 10
\]  

(4.4)

Thus, the neighborhood width will vary linearly from ±10 keV to ±20 keV as the peak energy increases from zero to the maximum detectable energy.

For each isotope in the library, this stage of the algorithm will then return a list of expected (library lookup) peak positions and areas that have been matched to the data peaks. The energy centroids from the library lookup will be used for the peak positions term \(f_{PP}\), while the areas will be used in the total peaks identified term \(f_{PI}\) and the area ratios term \(f_{AR}\).

### 4.2.3 Peaks Identified: \(f_{LPi}\) and \(f_{DPI}\)

After the data peaks have been matched to library peaks, the algorithm can begin calculating the various parts of the probability model to obtain the likelihood of each isotope:

\[
P(data|iso_i) = f_{LPi}f_{DPI}f_{PP}f_{AR}
\]  

(4.5)

The first two terms, \(f_{LPi}\) and \(f_{DPI}\), are very similar and are computed in the same manner (in fact, they are both calculated with the same custom function in Matlab, but with the arguments reversed).

\(f_{LPi}\) is the percentage of the peaks in the library isotope that have been found in the data, weighted by the library branching ratio. This gives a rough estimate of how well the data matches an isotope in the library without accounting for how close each of the matches are (which is taken into consideration with \(f_{PP}\), see Section 4.2.4) nor the areas of the peaks (which are used by \(f_{AR}\), see Section 4.2.5). This will also immediately assign a probability of zero to isotopes that don’t have any matched peaks, which will often greatly reduce the number of computations needed.

Similarly, \(f_{DPI}\) is the percentage of data peaks matched with library peaks, weighted by the peak areas. This term is especially important in data sets with many peaks, some of which might be close to other isotope peaks.

For the sake of notation, let \(iso_i\) contain a list of \(n\) peak energies and areas:
\[ \text{iso}_i = \{E_j, A_j\}_{j=1}^{j=n} \]

Similarly, the data set D contains a list of m peak energies and areas:

\[ D = \{\bar{E}_k, \bar{A}_k\}_{k=1}^{k=m} \]

For a given isotope \text{iso}_i in the library, let \( J = \{j\} \) be the index set of matched library peaks; that is, if \( j \in J \) then the library peak with energy \( E_j \) and area \( A_j \) was matched to one of the peaks in the data. If the library for \( \text{iso}_i \) contains \( n \) peaks, then the percentage of library peaks identified \( f_{LPI} \) is:

\[
f_{LPI}(\text{iso}) = \frac{\sum_{j \in J} A_j}{n} = \frac{\sum \text{(library peak areas that were matched to data peaks)}}{\sum \text{(all library peak areas)}} \tag{4.6}
\]

Similarly, let \( K = \{k\} \) be the index set of matched data peaks; that is, if \( k \in K \) then the data peak with energy \( \bar{E}_k \) and area \( \bar{A}_k \) was matched to one of the peaks in the library. If the library for \( \text{iso}_i \) contains \( n \) peaks, then the percentage of data peaks identified \( f_{DPI} \) is:

\[
f_{DPI}(\text{iso}) = \frac{\sum_{k \in K} \bar{A}_k}{n} \tag{4.7}
\]

Let’s consider a simple example. Suppose a detector identifies peaks at 208 keV and 662 keV with equal areas (area 1). Before computing \( f_{LPI} \) and \( f_{DPI} \) for \( ^{137}\text{Cs} \), the algorithm will match the main photopeak of \( ^{137}\text{Cs} \) at 661.7 keV with the data peak, but will not match the 208 keV data peak. The 661.7 keV peak has an branching ratio of 85.2, while all of the library gamma peaks combined have a branching ratio of 93.2 [6]. Then \( f_{LPI}^{^{137}\text{Cs}} \) will be the area of the 661.7 keV peak over the total area of the \( ^{137}\text{Cs} \) peaks.
\[
f_{LPI}^{(137\text{Cs})} = \frac{\sum_{j \epsilon J} A_j}{\sum_{i=1}^{n} A_i} = \frac{85.2}{93.2} = 0.914
\]

On the other hand, when computing these factors for \(^{241}\text{Am}\), the algorithm will match the data peaks with the library peaks at 208.0 keV and 662.4 keV, which only accounts for a very small percentage of the \(^{241}\text{Am}\) peaks.

\[
f_{LPI}^{(241\text{Am})} = \frac{\sum_{j \epsilon J} A_j}{\sum_{i=1}^{n} A_i} = \frac{0.00079 + 0.00036}{77.94} = 1.5 \times 10^{-5}
\]

However, while \(^{137}\text{Cs}\) seems more likely by the measure \(f_{LPI}\), \(^{137}\text{Cs}\) only accounts for half of the data:

\[
f_{DPI}^{(137\text{Cs})} = \frac{\sum_{k \epsilon J} \bar{A}_j}{\sum_{i=1}^{n} \bar{A}_i} = \frac{1}{2}
\]

\[
f_{DPI}^{(241\text{Am})} = \frac{\sum_{k \epsilon J} \bar{A}_j}{\sum_{i=1}^{n} \bar{A}_i} = \frac{2}{2} = 1
\]

These terms are important for the final isotope predictions, but they can also be used to reduce computation time. If isotope \(iso\_i\) has \(f_{LPI} = 0\), then none of the peaks were matched, and the final probability the isotope \(P(iso_i|data) = 0\). Thus, none of the other calculations need to be done if either of these terms are zero, reducing the total number of computations drastically (in some cases, by 1-2 orders of magnitude). Further, once the models are expanded to allow for multiple sources, isotopes where the single isotope probability \(f_{LPI} = 0\) will not need to be included in the set of possible isotopic mixtures, reducing computations even more.
4.2.4 Peak Positions: $f_{PP}$

To compute $P(data|iso_i)$, the model must account for the peak positions in the data versus the peak positions in our isotope library. The peak positions will be accounted for in the $f_{PP}$ term. This term is constructed by considering three constraints.

First, the model should assign a low probability if a peak is observed in the data but is not matched with a library peak. This penalty should be harsher at higher energies, as high-energy peaks are attenuated less by shielding and are less likely to be missed.

The model should also assign a high probability if the data peak and the library peak are very closely matched.

Finally, to accommodate for drift calibration, if a data peak at a high energy is matched to a library peak, it should not be penalized as harshly as a lower-energy isotope would be for a greater distance between peaks, as drift calibration has a more pronounced effect at higher energies. That is, suppose the data peak has centroid energy $E_D$ and the library peak has energy $E_L$, then the distance between peaks is $d = |E_D - E_L|$. If $d$ is fixed, then the probability $f_{PP}$ should be larger at higher energies.

To account for these three rules, this algorithm uses an offset $\cos^2(x)$ function to account for peak positions. The $\cos^2(x)$ is chosen for its general shape; a Gaussian shape or other similar curve works as well. Preliminary tests haven’t strongly favored any curve, but later during algorithm optimization this term may be changed.

The $\cos^2(x)$ needs to be offset so that the minimum probability is non-zero. If zero was an allowable value, then if a single extra peak was extrapolated from the spectra the correct isotope would be given a probability of zero and thrown out from consideration. However, because higher energy peaks are less likely to be attenuated out, higher energy peaks are weighted more heavily than lower energy peaks. That is, this minimum will be set to a lower value so that missing a higher energy peak is penalized more harshly. For simplicity, this offset, $b$, is a linearly decreasing function of peak energy that is set to vary from 0.3 to 0.1 as the peak energy increases. Again assuming that the maximum detectable energy is 3000 keV, $b$ has been chosen to be:

$$b(E_D) = 0.3 - \frac{E_D}{3000} \times 0.2$$
If the distance between the data peak and the corresponding library peak \( d = |E_D - E_L| \) is greater than the neighborhood \( w \), then \( f_{PP} = b \). That is, if the data peak is not matched with the library peak, a small probability of \( b \) is assigned.

Within the neighborhood of the peak centroid \([E_D - w, E_D + w]\), the probability varies from \( b \) to 1 with a \( \cos(x)^2 \) dependence. The \( \cos(x)^2 \) is scaled appropriately so that it continuously increases from \( b \) to 1. As shown in Figure 4.1, the probability of an isotope giving a peak at energy \( E_D \) is:

\[
 f_{PP}(E_D) = \begin{cases} 
 b + (1 - b) \cos^2\left(\frac{\pi}{2} \frac{E_D - E_L}{w}\right) & d < w \\
 b & d \geq w 
\end{cases}
\]

![Figure 4.1: \( f_{PP} \) for different data peak energies](image)

When the data contains multiple peaks \( E_1, E_2, \cdots, E_n \), \( f_{PP} \) is computed for each of the \( n \) peaks. As the peak locations are independent events, the probability of obtaining all of these peaks is the product of the probability of obtaining each of them individually. Then
To obtain a more realistic model for the peak positions, an experiment is performed in Chapter 5. By taking the same spectra many times ($10^4$ measurements with each setup), the actual variation in the observed peak energies from the expected energies can be found precisely. However, this will be extremely sensitive to the peak identification algorithm used; analysis using the manual peak fitting with Origin is included in Chapter 5, but the model for $f_{PP}$ will not be changed until an automated peak identification method has been coupled to BRAIN.

4.2.5 Peak Area Ratios: $f_{AR}$

For many problems, simply using peak positions is not enough to yield accurate isotope identifications; if it did, library comparison methods would be the standard for handheld identifiers. The algorithm needs to also account for the areas of the identified peaks. To do so, BRAIN will compare the ratio of areas of neighboring peaks; comparing area ratios eliminates the need for activity and geometry information which is not generally available.

Suppose the data contains two peaks with energies $E_1$ and $E_2$ and areas $A_1$ and $A_2$ respectively. To determine the probability $f_{AR}$ of obtaining peaks with these areas, the algorithm will compare the area ratio $r = \frac{A_1}{A_2}$ to the area ratio $R$ of the matched library peaks.

Consider the qualitative illustration of hypothetical $^{60}$Co peaks in Figure 4.2. Ignoring the efficiency curve of the detector, these peaks have nearly identical branching ratios of 99.9 and 99.9824 respectively [6]. In the top spectrum, the ratio $r$ is less than the library ratio $R = 1$, which is possible in the presence of shielding. [6] In this case, the algorithm should assign a high probability (or equivalently a low penalty) to this ratio. In the bottom spectrum, $r$ is somehow larger than the $R$; no shielding setup will produce this effect.

We therefore apply the following logic:

1. $\frac{A_1}{A_2}$ close to the library value: no penalty
Figure 4.2: Qualitative illustration of the two primary peaks of $^{60}$Co. The ideal case corresponds to the library values, while the reduced peaks are from hypothetical data. Top: Effects of shielding on the peak areas. Bottom: The higher energy peak has been reduced more than the lower energy peak.

2. $\frac{A_1}{A_2}$ smaller than the library value: small penalty, as the ratio will decrease as more shielding is added

3. $\frac{A_1}{A_2}$ larger than the library value: harsh penalty with increasing ratio

To obey the first rule, if the ratio $r$ is within a neighborhood around the library value for the area ratio $R$, no penalty (a probability of one) will be applied. The right bound, $B_r$, is set five percent above the expected (library) value to allow for statistical fluctuation in the counts, the uncertainty in the area measurement, and a possibly inexact detector efficiency curve.

$$B_r = 1.05 \times R$$

The left bound, $B_l$, is set further from the library ratio $R$ to allow for an unknown shielding setup. Since the linear attenuation coefficient is higher at lower gamma energies, a fixed amount of shielding will cause lower energy peaks to be proportionally reduced in area more than higher energy peaks. If the lower energy peak has energy $E_1$, the left bound is:

$$B_l = R \times e^{-E_1/3000}$$
If the ratio $r$ is within the neighborhood $(B_l, B_r)$, a probability of one is assigned (i.e. no penalty applied). Ratios outside of the neighborhood $(B_l, B_r)$ are penalized depending on their value. For ratios $r$ below $B_l$, a mild penalty is introduced, as it is possible with shielding to observe such a low area ratio; for ratios above the right bound $B_r$, an exponentially-increasing penalty is applied, as it should not be possible to increase the area ratio.

Then, for an observed area ratio $r = \frac{A_1}{A_2}$, the probability $f_{AR}$ given to this ratio is:

$$f_{AR}(A_1, A_2) = \begin{cases} 
1 - e^{-r} + e^{-B_l} & r < B_l \\
1 & B_l < r < B_r \\
e^{B_r-r} & B_r < r
\end{cases} \quad (4.8)$$

If the data contains more than two peaks, this term is computed sequentially. Given $n$ peaks ($n \geq 2$):

$$f_{AR}(A_1, A_2, \cdots, A_n) = \prod_{i=2}^{n} f_{AR}(A_{i-1}, A_i) \quad (4.9)$$

However, if there are multiple isotopes present, using the area ratios becomes more complicated. If peaks at energies $E_1$ and $E_2$ are from two different isotopes, then their area ratio will depend on the relative activity of the sources and the branching ratios. As such, this model currently assumes
that only one isotope is present in the data. To cope with this problem, the multiple source code will either have to try different isotope ratios (e.g. 10\% isotope A, 20\%, ...) or to deconvolve the data into subsets.

4.2.6 Complete Algorithm

Suppose the data has a set of energies $\{\bar{E}_k\}_{k=1}^m$ and areas $\{\bar{A}_k\}_{k=1}^m$. For a given isotope $iso_i$ with $n$ peaks, the algorithm first matches the data energies with the nearest library energies. If the nearest library energy is suitably close to the data energy, the two are considered to be a match and the index $j$ of the library peak (i.e. the matched data peak has energy $E_j$). The percent of library and data peaks identified $f_{LPI}$ and $f_{DPI}$ are then computed. For all isotopes with $f_{LPI} \neq 0$, the algorithm then computes the peak positions term $f_{PP}$ and the area ratios term $f_{AR}$. For each isotope, these results are combined to compute $P(data|iso_i)$, the probability that a given isotope would produce this data set:

\[
P(data|iso_i) = f_{LPI} f_{DPI} f_{PP} f_{AR}
\]

\[
= \left( \sum_{j \in J} \frac{A_j}{n} \right) \left( \sum_{k \in K} \frac{\bar{A}_k}{m} \right) \left( \prod_{m=1}^{n} f_{PP}(E_m) \right) \left( \prod_{q=2}^{n} f_{AR}(A_{q-1}, A_q) \right)
\] (4.10)

After calculating $P(data|iso_i)$ for every isotope in the library, the results are combined to compute $P(data)$ and finally $P(iso_i|data)$, the probability that $iso_i$ is responsible for the observed data.

In Chapter 5, we experimentally observe the peak position probability $f_{PP}$ and area ratio probability $f_{AR}$ as found with Origin 9.0. In Chapter 6, the algorithm is tested on a series of single-source spectra. A discussion of future improvements and tests can be found in Chapter 7.
CHAPTER 5

EXPERIMENT: JUSTIFICATION OF PROBABILITY MODELS

5.1 Introduction

In a perfect world, all spectra would be collected for a long enough period that the statistical fluctuations and discontinuities would be negligible. However, we seek to implement these algorithms for handheld detectors that are used in scenarios where it is infeasible, if not impossible, to take long dwell time measurements. Instead, a spectrum might only be taken for a few seconds. Measurements cannot always be made close to the source, and unknown layers of shielding may further mask spectra.

The probability models currently used by BRAIN are reasonable first estimates; however, future work will explore their efficacy and will optimize them. To this end, an experiment is conducted to find the experimentally observed deviations of the peak centroids and areas from their expected values.

By taking spectra under the exact same testing conditions over and over, we can run our peak-fitting software and observe the statistical fluctuations in peak centroid energy and area. For the data presented in this chapter, each spectrum has been observed for 10,000 trials without adjusting the experimental geometry. All of the peaks are currently fitted using a \textit{gaussamp} fit in Origin 9.0. In the future, the analysis will be re-run with an automated peak identification method [25], as the peak-fitting method used will likely change the distribution of observed peaks.

First, we will look at general results from several laboratory isotopes; afterwards, we will focus on how these distributions change as the count time is changed. All of the spectra were taken with the same detector gain settings, so one channel width corresponds to 3.6 keV.
5.2 Justification Experiment: General Results

5.2.1 $^{60}\text{Co}$

$^{60}\text{Co}$ is one of the easiest gamma-emitting isotopes to identify. It has two large, easily-distinguished photopeaks at 1173 and 1332 keV respectively with nearly identical branching ratios [6], as seen in the sample spectrum in Figure 5.1.

From the 15 second spectra, we observe that the two photopeaks are easily resolvable and are in a region with little continuum. These results are reflected in Figure 5.2, where we see little deviation in the fit peak centroid energies. From $10^4$ trials, all peaks were resolved and were found to be within 5 keV of the expected value ($\pm 1.3$ channels). These easily resolvable, low signal-to-noise peaks were all within a few keV of the mean. As shown in the following subsection, this will not be typical of all spectra.

![Figure 5.1: Spectrum of $^{60}\text{Co}$.](image-url)
Figure 5.2: Deviation of observed peak centroid of $^{60}$Co in $10^4$ trials with 15 second observations. Top: 1173 keV peak. Bottom: 1332 keV peak.

5.2.2 $^{152}$Eu

$^{152}$Eu has several resolvable peaks, though all are in a region with a significant continuum (see the spectrum in Figure 5.3). The results of fitting the peaks at 128, 245, 344, and 779 keV are presented in Figure 5.4. At low energies, the deviation from the expected energy bin is extremely small; for the 128 keV peak, 9943 of the spectra deviated less than 0.25 keV from the expected value. For the 779 keV peak, only 1508 of the $10^4$ trials deviated less than 0.25 keV, and 9942 deviated less than 3.65 keV.

5.3 Justification Experiment: Changing Count Times

To gauge the effects of count time on the peak position deviation, the spectra of $^{152}$Eu was taken for 1 second, 2 seconds, 3 seconds, 5 seconds, 10 seconds, and 15 seconds. Figure 5.5 shows the peak fitting results for the 964 keV peak; this peak is used here because it has the smallest continuum under the photopeak, so deviation of the peak centroid should be due to statistical
energy fluctuation only.

Increasing the measurement time has a dramatic effect on the observed peak centroids. The centroid deviation is reduced by a factor of $\approx 3$ by changing the measurement time from 1 second to 5 seconds and by a factor of $\approx 2$ by increasing it from 5 seconds to 15 seconds.

5.4 Conclusions of the Justification Experiment

Regardless of peak energy or area, the peak centroids appear to be approximately normally distributed about the expected peak energy. The variance, however, is a function of centroid energy and area. These results suggest significant improvement for BRAIN, as the model for peak positions does not account for the area of the peak or the signal-to-noise ratio. These improvements will wait until an automated peak identification algorithm has been coupled to BRAIN, as the exact distributions will likely be a function of the algorithm used for finding and fitting peaks. Once such an algorithm has been coupled to BRAIN, the thousands of spectra collected for this ex-
Figure 5.4: Deviation of observed peak centroid of $^{152}\text{Eu}$ in $10^4$ trials with 15 second observations. Top left: 128 keV peak. Top right: 245 keV peak. Bottom left: 344 keV peak. Bottom right: 779 keV peak.

Experiment can be used again to characterize the peak distributions given the peak algorithm.
Figure 5.5: Deviation of observed peak centroid of the 964 keV $^{152}$Eu in $10^3$ trials with at different time scales. From top to bottom: 1 second, 2 second, 3 second, 5 second, 10 second, and 15 second count time.
CHAPTER 6

EXPERIMENT: TESTING THE ALGORITHM

To test BRAIN, various spectra are taken using laboratory sources and a 1024 channel NaI detector. We also present the results of some shielded spectra and demonstrate the effects of varying temperatures on the energy calibration of a NaI scintillator. For each spectrum presented, the primary peaks were fit using the \textit{gaussamp} fit in Origin, and the energy centroids and areas from these fits were given to BRAIN.

6.1 Ideal Spectra

In an ideal scenario, gamma spectra would be taken without shielding materials present and for a suitably long count time. While these handheld detectors are almost never used this way in practice, understanding how an identification algorithm behaves in ideal circumstances will help improve it for practical scenarios.

The spectra of $^{60}$Co, $^{137}$Cs, $^{133}$Ba, $^{152}$Eu, and $^{232}$Th were taken by a detector shielded from the background by lead plates, though a background subtraction is not performed for any of these spectra. The linear energy calibration used was based on the 1173 and 1332 peaks of $^{60}$Co, the 661.7 keV peak of $^{137}$Cs, and the 2614 keV peak of $^{232}$Th. As noted in each of the sections, this linear fit does not produce a perfect calibration, but it is used regardless to mimic the calibration used by automated handheld identifiers.

6.1.1 $^{137}$Cs

$^{137}$Cs is a standard calibration isotope for handheld detectors because it has a single prominent photopeak at 661.66 keV, plus a few small characteristic x-ray peaks that are usually not seen by a NaI detector. The Origin fitting
Figure 6.1: Spectrum of $^{137}$Cs.

Table 6.1: $^{137}$Cs Identification Results: Five Most Probable Sources

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am</td>
<td>0.00046%</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>99.90 %</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>0.072%</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>0.02%</td>
</tr>
<tr>
<td>$^{153}$Sm</td>
<td>0.00048%</td>
</tr>
</tbody>
</table>

of Figure 6.1 identified a photopeak at 658.8 keV with area 539000, though the area is not used by BRAIN because only one peak is present; to compare peak areas by the ratio test, at least two peaks must be resolved.

As in Table 6.1, BRAIN correctly identifies the source with a posterior probability of 99.9%.

6.1.2 $^{60}$Co

$^{60}$Co is one of the easiest gamma-emitters to identify, as discussed in Chapter 5. The fitting in Origin of the spectrum in Figure 6.2 recognized peaks at 1173.6 and 1335.8 keV with areas 23492 and 27935 respectively. BRAIN
Figure 6.2: Spectrum of $^{60}$Co.

Table 6.2: $^{60}$Co Identification Results

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Co</td>
<td>99.995%</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>0.0052%</td>
</tr>
<tr>
<td>Other</td>
<td>0</td>
</tr>
</tbody>
</table>

correctly identifies $^{60}$Co with a posterior probability of 99.995% (Table 6.2).

6.1.3 $^{133}$Ba

$^{133}$Ba is a simple isotope to identify by eye but can give algorithms with low resolutions detectors trouble due to the overlapping peaks at 276.4, 302.9, 356.0, and 383 keV (note that in Figure 6.3, one can deduce the presence of the peaks at 276 and 383 by the shoulders in the Gaussian peaks, but these are not easily resolved by peak fitting algorithms). Using Origin, peaks at 295 and 345 keV are detected, which differ slightly from the true gamma ray energies. The error in peak centroid energy is due to both the overlapping peaks and the nonlinearity of the energy calibration.

BRAIN predicts $^{133}Ba$ with probability 62% (Table 6.3); the lower cer-
Figure 6.3: 10 minute spectrum of $^{133}$Ba.

Table 6.3: $^{133}$Ba Identification Results

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}$Ba</td>
<td>62.0%</td>
</tr>
<tr>
<td>$^{67}$Ga</td>
<td>10.9%</td>
</tr>
<tr>
<td>$^{131}$I</td>
<td>13.1%</td>
</tr>
<tr>
<td>$^{177m}$Lu</td>
<td>7.13%</td>
</tr>
<tr>
<td>$^{75}$Se</td>
<td>5.45%</td>
</tr>
</tbody>
</table>

tainty of this prediction is primarily due to the undetected peaks at 276 and 383 keV.

6.1.4 $^{232}$Th

$^{232}$Th is an isotope that BRAIN completely fails to identify for a simple reason. $^{232}$Th has a rich spectra (at least 9 peaks can be seen in Figure 6.4), but none of these are directly emitted by $^{232}$Th. The thorium series decay chain includes many daughters with half-lives ranging from a few years (e.g. $^{288}$Ra has a half-life of 5.7 years) to less than a second ($^{212}$Po has a half-life of 0.3 $\mu$s). These other isotopes are responsible for most of the observed
Table 6.4: $^{232}$Th Identification Results

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{57}$Co</td>
<td>0.17%</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>0.016%</td>
</tr>
<tr>
<td>$^{75}$Se</td>
<td>0.028%</td>
</tr>
<tr>
<td>$^{89}$Sr</td>
<td>99.9%</td>
</tr>
</tbody>
</table>

spectrum, including the tell-tale peak at 2614 keV.

Because most (if not all) of the detectable peaks in the spectrum of $^{232}$Th is not in our spectral library, it is impossible for BRAIN to make a correct identification (Table 6.4). Expanding the library is discussed in Chapter 7.

6.1.5 $^{152}$Eu

While the $^{232}$Th demonstrated that our library is missing many peaks that are vital for identifications, $^{152}$Eu has the opposite problem; the extreme wealth of gamma peaks makes correct peak identification difficult. Our library contains 156 gamma peaks (138 of which are over 100 keV) for $^{152}$ Eu. Not all of these peaks are visible in Figure 6.5, but there are still significantly
more peaks visible than for most isotopes.

During the peak matching stage of the algorithm, peaks are often mismatched even with little energy calibration drift. While BRAIN is capable of handling this drift when less peaks are present, it fails in the many peak case. Improving the library and changing the peak matching routine is discussed in Chapter 7 to solve this problem.

Giving BRAIN the four most easily-resolved peaks with a good calibration yields the correct result (Table 6.5), though not with less confidence than the identifications of sources with fewer expected peaks.
Figure 6.6: Spectrum of $^{220}$Ra.

6.2 Shielded Spectra

As discussed in Chapter 2, shielding has three primary effects on gamma spectra: a suppression of lower energy peaks, a reduction of the area ratio between peaks, and a reduction of total counts across all energies. In the field, almost all observed spectra are going to be affected by some (usually unknown) amount of shielding.

6.2.1 Shielded $^{152}$Eu

In the case of $^{152}$Eu, 2.1 cm of lead is enough to remove all but the four peaks at higher energies. The peaks with observed centroids at 778, 964, 1086, and 1408 keV are still easily resolved despite the shielding.

BRAIN correctly predicts $^{152}$Eu with nearly the same confidence as in the unshielded case (63.1% shielded, 55.2% unshielded). The improved confidence is due to a reduction in the likelihood of some of the other isotopes.
Figure 6.7: Spectrum of $^{152}$Eu shielded with 2.1 cm Pb.

Table 6.6: Shielded $^{152}$Eu Identification Results

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{152}$Eu</td>
<td>63.1%</td>
</tr>
<tr>
<td>$^{123}$I</td>
<td>0.20%</td>
</tr>
<tr>
<td>$^{99}$Mo</td>
<td>36.7%</td>
</tr>
</tbody>
</table>

6.3 Severe Temperature Drift

While the small temperature changes in the room over the course of a day are enough to influence the peak centroids (see Chapter 5), in the field these detectors may undergo much larger temperature shifts. For example, at a border crossing a detector might be calibrated in a warm car and then taken outside to make an identification, which could have a temperature shift of more than 10°C.

To simulate these effects, the detector was stabilized at room temperature and a spectrum of $^{152}$Eu was recorded. The detector was then placed in a freezer for one hour. Without restabilizing the detector, the spectrum was measured again; the two spectra are shown in Figure 6.8.
Figure 6.8: Spectrum of $^{152}$Eu was taken with a detector calibrated and stabilized at room temperature. The detector was placed in a freezer for one hour and the spectrum was measured again without restabilizing the detector.

Table 6.7: Chilled Detector: $^{152}$Eu Identification Results

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Posterior Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{152}$Eu</td>
<td>77.5%</td>
</tr>
<tr>
<td>$^{123}$I</td>
<td>0.11%</td>
</tr>
<tr>
<td>$^{99}$Mo</td>
<td>14.4%</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>8.0%</td>
</tr>
</tbody>
</table>

Interestingly, of all of the $^{152}$Eu spectra presented here, the chilled detector spectrum yields the greatest confidence in the correct identification (Table 6.7). This is because the drifted calibration puts the peak centroids further from the other competing isotopes (most importantly $^{99}$Mo).

6.4 Summary

For single isotope sources, BRAIN works well for isotopes that don’t have too many peaks, even in the presence of shielding or large calibration drift.
CHAPTER 7

CONCLUSION

7.1 Conclusions and Future Work

We have described an automated isotope identification algorithm that implements Bayesian statistics and should be computationally cheap enough to be usable on handheld RIIDs. The Bayesian Radioisotope Identifier (BRAIN) algorithm uses peak centroid positions and areas to make identifications, unlike many other methods. While in early stages, this algorithm seems to handle calibration drift and peak misidentification well and has tremendous room for further improvement.

As demonstrated in Chapter 6, this algorithm can account for missing peaks, calibration drift, “unknown” shielding, and more. Early results are promising, but much work is still needed to develop the algorithm.

First, this algorithm needs an improved library. An improved library will contain less peaks than the current library, which contains every known gamma peak and area for each isotope, many of which would not be detectable with a low-resolution detector. Undetectable peaks should be removed from the library, and unresolvable close peaks should be combined. This will greatly increase the accuracy of the peak matching step and allow BRAIN to deal with greater calibration drift. In the future, BRAIN will be combined with the adaptive library of [19].

Next, BRAIN needs to be coupled to a peak identification method. The current manual peak identification is useful for testing early versions of the algorithm, but manual detection is not appropriate for automated detectors. Further, some of the probability models used by BRAIN may be sensitive to the peak identification algorithm (e.g. a peak algorithm may find peak centroids with less accuracy/greater variation, etc.).

Then, while all of these probability models are reasonable starting points,
less subjective models are needed. While deriving these functions rigorously
may be possible for certain idealized cases, the analytical results will depend
on (among other things) the method used to obtain peak energies and ar-
reas, the detectors, and the particular geometry of the source and detector.
The justification experiment in Chapter 5 showed that while our estimated
probability models were reasonable, they did not perfectly reflect the ob-
served distributions. Once a peak identification method has been coupled to
BRAIN, this analysis will be re-run and then the probability models used by
BRAIN will be replaced.

Most importantly, this model needs to be extended to allow for multiple
sources. One possible (untested) solution is the following:

1. Run the single source version of the code and obtain posterior estimates
   for each of the single-source cases.

2. If an isotope has non-zero (or suitably large) probability, pass it to the
   multiple-source code.

3. For each combination of isotopes, form “super isotope” matrix that
   contains the peaks and areas of the isotopes in consideration. To al-
   low for different isotopic ratios, make separate “super isotopes” with
   different ratios. Use Kong’s library algorithm on each “super isotope”.

4. Run each “super isotope” through the single source code.

5. Re-normalize and compare posterior probabilities to make an identifi-
   cation.

This method for the multiple-source problem has several advantages over
other methods considered. First, it eliminates the problem of trying to split
the data into partitions that each correspond to a single isotope (which is a
complicated problem in the case of isotopes with close energy peaks). Second,
it uses the same probability models as the single source code, allowing the
multiple-source results to be compared alongside the single-source results.
Finally, it shouldn’t greatly lengthen runtimes. By only considering isotopes
with non-negligible single-isotope probabilities, the number of combinations
that need to be tested can be drastically reduced. Even if this method
increased the run-time by an order of magnitude, the expected run-time of
the code would still only be a few seconds.
We are also interested in applying this type of methodology to the case of low-count spectra. It is possible that the same algorithm (with appropriate probability models) would work, but this has not been tested. If it does work, a more general version of the algorithm that chooses its probability distributions based on the number of counts and the uncertainties will be developed. Another route to be explored is the analysis of real-time spectra with sequential Bayesian analysis [10] using similar methodologies.

Finally, instead of using a nearest-neighbor lookup to match data peaks to library peaks, the algorithm could instead run for all peaks within a larger neighborhood and keep the set that maximizes the final probability. While this could increase computation time by up to an order of magnitude, calibration drift would become significantly less important.
REFERENCES


