MONITORING RAINWATER AND SEAWEED REVEALS
THE PRESENCE OF $^{131}$I IN SOUTHWEST AND
CENTRAL BRITISH COLUMBIA, CANADA
FOLLOWING THE FUKUSHIMA NUCLEAR ACCIDENT
IN JAPAN

by

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B.Sc., Michigan State University, 2009

A Thesis submitted in partial fulfillment
of the requirements for the degree of

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in the
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Abstract

Detailed analysis of $^{131}$I levels in rainwater and in three species of seaweed (Fucus distichus Linnaeus, Macrocystis pyrifera, and Pyropia fallax) collected in southwest British Columbia and Bella Bella, B.C., Canada was performed using gamma-ray spectroscopy following the Fukushima nuclear power plant accident on March 11, 2011. The maximum $^{131}$I activity was found to be 5.8(7) Bq/L in rainwater collected at the campus of Simon Fraser University in Burnaby, B.C. nine days after the accident. Concomitantly, the maximum observed activity in the brown seaweed Fucus distichus Linnaeus was observed to be 130(7) Bq/kg dry weight in samples collected in North Vancouver 11 days following the accident and 67(6) Bq/kg dry weight in samples collected from the Bamfield Marine Sciences Centre on Vancouver Island 17 days following the accident. The $^{131}$I activity in seaweed samples collected in southwest B.C. following the Fukushima accident was an order of magnitude less than what was observed in similar measurements performed in British Columbia following the Chernobyl accident. Iodine-131 activity in Fucus distichus Linnaeus remained detectable for 60 days following the accident and was detectable in each seaweed species collected. The Germanium Detector for Elemental Analysis and Radioactivity Studies (GEARS) was modeled using the Geant4 software package and developed as an analytical tool by the Nuclear Science group in the Simon Fraser University Department of Chemistry for the purpose of these measurements.

Keywords: Fukushima; Radiation monitoring; Seaweed; Detector development; $^{131}$I
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C.1 Reproduction of Fig. 3.4 from Sec. 3.1.2.

D.1 The pair production process in free space. Even under the assumption that $h\nu \geq 1.022$ MeV, this process is forbidden in free space by the conservation of energy and momentum.

H.1 An error ellipse for two correlated $\chi^2$ parameters defined by Eq. H.17. The ellipse is centered at $(a_0^*, a_1^*)$ which are the parameter values that correspond to $\chi^2_{\text{min}}$. 
## List of Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>abs.</td>
<td>Absolute</td>
</tr>
<tr>
<td>B.C.</td>
<td>British Columbia</td>
</tr>
<tr>
<td>BMSC</td>
<td>Bamfield Marine Sciences Centre</td>
</tr>
<tr>
<td>CDF</td>
<td>Cumulative distribution function</td>
</tr>
<tr>
<td>CERN</td>
<td>European Organization for Nuclear Research</td>
</tr>
<tr>
<td>eV</td>
<td>Electron volt</td>
</tr>
<tr>
<td>keV</td>
<td>Kiloelectron volt (10^3) eV</td>
</tr>
<tr>
<td>MeV</td>
<td>Megaelectron volt (10^6) eV</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full width at half maximum</td>
</tr>
<tr>
<td>Geant4</td>
<td>Geometry and Tracking, version 4</td>
</tr>
<tr>
<td>GEARS</td>
<td>The Germanium Detector for Elemental Analysis and Radioactivity Studies</td>
</tr>
<tr>
<td>HPGe</td>
<td>High purity germanium</td>
</tr>
<tr>
<td>LFR</td>
<td>Low frequency rejection</td>
</tr>
<tr>
<td>MCA</td>
<td>Multichannel analyzer</td>
</tr>
<tr>
<td>NORM</td>
<td>Naturally occurring radioactive materials</td>
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<tr>
<td>pp</td>
<td>Photopake</td>
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<tr>
<td>rel.</td>
<td>Relative</td>
</tr>
<tr>
<td>SFU</td>
<td>Simon Fraser University</td>
</tr>
<tr>
<td>SIMON</td>
<td>Subcritical Intense Multiplier of Neutrons</td>
</tr>
<tr>
<td>USB</td>
<td>Universal Serial Bus</td>
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</tbody>
</table>

The mathematical notation in this document expresses vector quantities as bold symbols \(\mathbf{p}_x, \lambda, \text{etc.}\) and their corresponding magnitudes in italics \(p_x, \lambda, \text{etc.}\) unless otherwise noted.

Orbital angular momentum is defined as \(L\) and total angular momentum as \(I\) throughout. All quantum mechanical operators are defined using a hat (orbital angular momentum operator \(\hat{L}\), for example).
Chapter 1

Introduction

Following the Tohoku earthquake and subsequent tsunami on March 11, 2011, the damaged Fukushima Dai-ichi nuclear power plant released radioisotopes from fission fragments and nuclear fuel into the environment. Some fission fragments were released as aerosols or gases and dispersed over a large geographic region through the atmosphere. The volatile fission fragment $^{131}$I represents $\sim$2.89% of the fission yield of enriched uranium [1] and is of particular concern to human health. The thyroid gland concentrates iodine in order to produce two iodine-rich metabolic hormones; radioactive iodine in the environment can accumulate in the thyroid and cause an elevated risk of thyroid cancer.

Iodine-131 is not naturally present in the environment, but with an 8.0252 day half-life [2], survives long enough to be transported in the atmosphere across the Pacific Ocean. The likely transport mechanism across the Pacific is the jet stream, which lies in the upper troposphere, which is consistent with the transport mechanism described in Ref. [3]. Therefore, during the current studies any $^{131}$I detected in rainwater or seaweed was attributed to releases from Fukushima.

Fallout of $^{131}$I following the Fukushima accident was widespread. Airborne $^{131}$I was observed in Europe [4–7], the Canary Islands off the northwest coast of Africa [8], South Korea [9] and in the northwest United States [10]. Iodine-131 concentrations in rainwater of $\leq$3.5 Bq/L were reported in Milano, Italy [4], Bordeaux, France [6], and Jeju, South Korea [9]. Measurements of $^{131}$I in Washington state, U.S.A., indicated that releases from Fukushima first arrived between March 16 and March 18. Subsequently, $^{131}$I was first detected on the European continent between March 21 and March 28 and in South Korea on March 28.
As part of the global effort to monitor fission fragment releases following the Fukushima accident, $^{131}$I was measured in rainwater collected at the Simon Fraser University (SFU) campus on Burnaby Mountain and in seaweed samples collected in North Vancouver, B.C., the Bamfield Marine Sciences Centre (BMSC) on Vancouver Island, B.C., located ~250 km west of Vancouver, and Bella Bella, B.C., located ~650 km north of Vancouver along the Pacific coast. The perennial brown seaweed species *Fucus distichus* Linnaeus (henceforth *Fucus*) was chosen as the primary biological monitor for $^{131}$I releases from Fukushima. *Fucus* was chosen because it is known to concentrate iodine present in the environment, is widespread in intertidal zones in the Pacific Northwest where it can accumulate iodine from rainfall, and was used as a biological monitor in B.C. following the Chernobyl disaster [3]. Although the exact mechanism for iodine uptake in *Fucus* remains an open question, a comparison of two *Fucus*-based measurements does not require that the iodine uptake mechanism be understood. In addition, *Macrocystis pyrifera* and *Pyropia fallax* samples collected in Bella Bella served as secondary biological monitoring systems. The monitoring campaign provided an opportunity to develop the equipment and techniques necessary to perform highly accurate analytical radiochemistry at SFU in order to assess $^{131}$I contamination in B.C. following Fukushima and to compare with measurements made following the Chernobyl accident.

Iodine-131 content in rainwater and seaweed was monitored following the analysis of gamma-ray decay spectra acquired using a shielded high purity germanium (HPGe) detector. HPGe detectors possess excellent energy resolution which facilitates the detection of weak sources of radiation that would otherwise be lost in the background if detectors with poorer energy resolution, such as inorganic scintillators were used [11]. HPGe detectors are therefore the preferred tool to measure complex spectra such as those found in environmental samples which contain many gamma-ray lines that must be resolved for proper analysis.
Chapter 2

Principles of gamma-ray decay

Gamma-ray emission is the nuclear analog to X-ray and optical transitions observed in atoms [12]. During gamma-ray decay, a nucleus in an excited state decays to its ground state by photon (gamma-ray) emission. Gamma-ray decay typically follows alpha and beta decay which populate excited states in the daughter nuclei; gamma decay can also follow electron capture by the atomic nucleus. Generally, gamma-ray decay is a fast process on the order of $\sim 10^{-9}$ s, though longer decay half-lives have been observed [12]. Selected properties of gamma-ray decay of importance to this work are discussed herein.

2.1 Quantization of angular momentum

Angular momentum in quantum mechanics is defined by the operator $\hat{L}$ given by

$$\hat{L} = \hat{r} \times \hat{p}$$

(2.1)

where $\hat{r}$ and $\hat{p}$ are the position and momentum operators. In three-dimensional space, the angular momentum operator is a three component vector

$$\hat{L} = (\hat{L}_x, \hat{L}_y, \hat{L}_z)$$

(2.2)

where $\hat{L}_x, \hat{L}_y$, and $\hat{L}_z$ are the one-dimensional angular momentum operators. Computing the expectation value of $\hat{L}^2$ yields information about the magnitude of the angular momentum and gives rise to the angular momentum quantum number $\ell$ shown in Eq. 2.3.

$$\langle \hat{L}^2 \rangle = \langle \hat{L}_x^2 + \hat{L}_y^2 + \hat{L}_z^2 \rangle = \hbar^2 \ell (\ell + 1)$$

(2.3)
The quantum number $\ell$ is either 0 or a non-negative integer. Due to angular momentum commutation relations, defined by Eq. 2.4, only one component of the angular momentum vector and its magnitude can be specified simultaneously.

$$[\hat{L}_i, \hat{L}_j] = i\hbar\hat{L}_k$$ \hspace{1cm} (2.4)

The $z$-axis is chosen as the quantization axis by convention. Computing the expectation value $\langle \hat{L}_z \rangle$ gives

$$\langle \hat{L}_z \rangle = \hbar m_\ell$$ \hspace{1cm} (2.5)

where $m_\ell$ are the magnetic substates for a given $\ell$ which define the projection of $\ell$ on the $z$-axis. The allowed values are $m_\ell = 0, \ldots, \pm \ell$.

### 2.2 Selection rules

Selection rules for transitions between excited angular momentum states arise from quantization and the conservation of angular momentum. If a nucleus exists in an excited initial angular momentum state $I_i$ and decays by gamma-ray emission to another state $I_f$, conservation of angular momentum expressed in vector form dictates that

$$I_i = \lambda + I_f$$ \hspace{1cm} (2.6)

where $\lambda$ is the multipole order of the emitted gamma ray which arises from the multipole expansion of the time-dependent Maxwell’s equations in free space. The photon emitted following a multipole transition of order $\lambda$ carries with it an angular momentum of $\lambda\hbar$. These three vectors $I_i$, $I_f$ and $\lambda$ must form a closed triangle, which restricts the possible values of $\lambda$ to the positive integers given in Eq. 2.7.

$$|I_i - I_f| \leq \lambda \leq I_i + I_f$$ \hspace{1cm} (2.7)

An exception occurs when $I_i = I_f = 0$ because no $\lambda = 0$ transitions occur in which a single gamma ray is emitted. Therefore, the lowest allowed multipole transition is $\lambda = 1$ [12]. The Weisskopf estimates of transition rates for decays of a given multipole order $\lambda$ predict that the transition rate is proportional to the product of $E_\gamma^{2\lambda+1}$ and the square of the transition matrix element of the multipole operator which depends on the structure of the initial and final states. This prediction agrees with experimental evidence, and indicates
that lower-order multipole transitions (corresponding to smaller values of $\lambda$) are favored [12].

The restriction on $\lambda$ constrains the change in the magnetic quantum number. In general, allowed values for $\Delta m_l = m_i - m_f$ are $0, \pm 1, \pm 2, \ldots, \pm (\lambda - 1), \pm \lambda$.

### 2.3 Gamma-ray emission following beta decay

Gamma-rays are emitted by nuclei as they transition from excited states to lower lying levels [11]. One important process which populates excited states in nuclei is beta decay, a three body decay in which a parent nucleus decays to a daughter nucleus (often in an excited state), a beta particle (an electron or positron) and an electron neutrino or its antiparticle. There are two types of beta decay distinguished by their decay products, $\beta^-$ decay given by Eq. 2.8 and $\beta^+$ decay given by Eq. 2.9

\[
\frac{A}{Z}X \rightarrow \frac{A}{Z+1}Y + \beta^- + \nu_e \quad (2.8)
\]

\[
\frac{A}{Z}X \rightarrow \frac{A}{Z-1}Y + \beta^+ + \nu_e \quad (2.9)
\]

where $A$ is the mass number, $Z$ is the atomic (proton) number, $X$ is the parent nucleus, $Y$ is the daughter nucleus, $\beta^-$ is a fast electron, $\beta^+$ is a positron, $\nu_e$ is the electron antineutrino, and $\nu_e$ is the electron neutrino.

The recoil energy of the daughter nucleus is very small, so the energy of the decay is shared by the beta particle and the neutrino, which is undetectable in a typical laboratory setup. Beta decay transitions possess a fixed decay energy or Q-value; this energy is shared between the beta particle and the neutrino because the kinetic energy of the nucleus is approximately zero [11]. Therefore, the beta particles have an energy spectrum which ranges from 0 to to the “endpoint energy” which is equal to the Q-value of the transition. If the daughter nucleus is populated in an excited state, the endpoint energy is shifted by the energy difference between the states of interest [11]. In practice, transitions to a number of different excited states can occur; the probability of a particular de-excitation pathway occurring is referred to as a “branching ratio.”

A specific example of interest for this work is the beta decay of $^{131}$I which populates excited states in $^{131}$Xe, which is shown in Eq. 2.10. In its ground state, $^{131}$Xe is stable and does not emit any subsequent radiation [2].

\[
^{131}_{53}I \rightarrow ^{131}_{54}Xe^* + \beta^- + \nu_e \quad (2.10)
\]
A partial level scheme for this transition is shown in Fig. 2.1. The beta decay half-life of $^{131}$I is 8.0252 days, while the half lives of the gamma-ray emitting spin $5/2^+$ 364.5 keV and spin $1/2^+$ 80.2 keV states in $^{131}$Xe shown in Fig. 2.1 are 67.5 ps and 454 ps, respectively [2]. Since the $^{131}$Xe isotope is not naturally occurring and the excited states which decay by gamma-ray emission have a half-life much shorter than the beta decay half-life of $^{131}$I, the observation of the 364.5, 284.3 and 80.2 keV gamma-rays in $^{131}$Xe are characteristic of the presence of $^{131}$I.

![Diagram](image)

**Figure 2.1:** The partial decay scheme of $^{131}$I; widths of gamma-ray lines represent relative intensities of the decay. $^{131}$I undergoes beta decay ($Q(\beta) = 606$ keV, branching ratio 89.6%) to the 364.5 keV state in $^{131}$Xe which then decays primarily by the emission of a 364.5 keV gamma-ray to the ground state of $^{131}$Xe. Beta decay branches to other excited states have small branching ratios and can be ignored in the context of the presented work.
2.4 Angular correlation

Consider a nucleus in an excited angular momentum state $I_o$ decaying to its ground state $I_f$ through an intermediate state $I_i$ by a gamma-ray cascade as shown in Eq. 2.11.

\[ I_o \gamma_1 \rightarrow I_i \gamma_2 \rightarrow I_f \]  

(2.11)

where $\gamma_1$ and $\gamma_2$ are the gamma rays associated with the transition. If the magnetic substates in state $I_i$ are populated anisotropically due to decays from state $I_o$, the direction of propagation of $\gamma_2$ is correlated to that of $\gamma_1$; the theory and formalism associated with this process is well understood [13]. This process is illustrated in the following example. Assume a nucleus transitions from an initial state with $I_o = 0$ to a final state with $I_f = 0$ through an intermediate state with $I_i = 1$ as shown in Fig. 2.2. Let the emission of the first gamma ray $\gamma_1$ define the z-axis. The angular distribution of $\gamma_1$ is proportional to $\sin^2 \theta_1$ for decays corresponding to transitions between the $m_o = 0$ and $m_i = 0$ states and proportional to \( \frac{1}{2}(1+\cos^2 \theta_1) \) for transitions between the $m_o = 0$ and $m_i = \pm 1$ states [12]. Due to the choice of the z-axis, $\theta_1 = 0$ which implies that the $m_o = 0 \rightarrow m_i = 0$ transition is not allowed.

![Diagram](image)

Figure 2.2: Anisotropic decay can arise from an unequal population of substates. For the two gamma-ray decay between states with angular momentum 0 → 1 → 0, the transition between the $m_o = 0$ and $m_i = 0$ states and $m_i = 0$ to $m_f = 0$ states are prohibited [red] when the quantization axis is defined by $\gamma_1$. The only allowed transitions in this case correspond to decays where $\Delta m = \pm 1$ [blue].
Thus, the population $p(m_i) = p(0) = 0$, and the angular distribution of $\gamma_2$ relative to $\gamma_1$ is governed by Eq. 2.12

$$W(\theta_2) \propto p(-1) \left[ \frac{1}{2} (1 + \cos^2 \theta_2) \right] + p(0)(\sin^2 \theta_2) + p(+1) \left[ \frac{1}{2} (1 + \cos^2 \theta_2) \right]$$

$$\propto 1 + \cos^2 \theta_2$$

(2.12)

where $p(-1) = p(+1) = \frac{1}{2}$, $p(0) = 0$, and $\theta_2$ is the angle between $\gamma_2$ and the z-axis defined by $\gamma_1$.

An example of a process which exhibits correlated gamma-ray decay due to anisotropic intermediate magnetic substate population is the decay of $^{60}$Co to the $^{60}$Ni ground state shown in Fig. 2.3. The angular distribution function can be written according to Eq. 2.13:

$$W(\theta) = \sum_{\text{even } k}^{k_{\text{max}}} A_k Q_k(\gamma_1, \gamma_2) \cos^k \theta$$

(2.13)

where $A_k$ is the angular distribution coefficient which can be calculated provided some properties of the transition are known [13], $Q_k(\gamma_1, \gamma_2)$ is the solid angle correction factor for the gamma rays impinging on a detector volume, and $\theta$ is the angle between the two emitted gamma rays. The $Q_k$ factors can be calculated using Eq. 2.14 and Eq. 2.15, where $\beta$ is the incident angle on the detector and $\epsilon_i(\beta)$ is the detection efficiency of gamma-ray $i$ at angle $\beta$. [14].

$$Q_k(i) = J_k(i)/J_0(i)$$

(2.14)

$$J_k(i) = \int_0^{\beta_{\text{max}}} \epsilon(\beta) \cos^k(\beta) \sin(\beta) d\beta$$

(2.15)

The angular correlation coefficients for the two gamma cascade of $^{60}$Co are well known: $A_0 = 1$, $A_2 = 1/8$ and $A_4 = 1/24$ [13], see Appendix A for more information.
Figure 2.3: The decay scheme of $^{60}\text{Co}$. $^{60}\text{Co}$ undergoes beta decay ($Q(\beta) = 318$ keV) to the 2505.7 keV state in $^{60}\text{Ni}$ which then decays by a two gamma cascade to the $^{60}\text{Ni}$ ground state. Both the 1173.237 keV and 1332.492 keV gamma rays have multipole order $\lambda = 2$. The $Q(\beta) = 1491$ keV branch is a weak channel which comprises only 0.12% of decays and can be ignored for analysis of gamma-ray angular correlation. Transitions from the $I = 4^+$ state to the second $I = 2^+$ state at 2158.6 keV are hindered by a factor of 441 relative to the $I = 2^+$ state at 1332.5 keV due to the $E_\gamma^5$ dependence on the transition rates predicted by the Weisskopf estimate.
Chapter 3

Principles of gamma-ray spectroscopy

Gamma-ray spectroscopy is a powerful tool used in a wide variety of fields, including national defense, research, medicine, and environmental monitoring. Gamma-rays provide a unique probe of small structures like the atomic nucleus due to their short wavelength (on the order of $10^{-12}$ m) and are therefore commonly used to study nuclear interactions. Cosmic gamma-rays provide an insight into a variety of cosmic events such as supernovae, black holes, and solar processes and are an important aspect of modern astronomical observation [12].

An analogy to this method can be made from the study of emission spectra of the chemical elements. When an atomic electron in a high energy state with energy $E_H$ transitions to a lower energy state with energy $E_L$, a photon is emitted with energy equal to the energy difference of the states involved in the transition as shown in Eq. 3.1.

$$E_{\text{photon}} = E_H - E_L \quad (3.1)$$

The set of emission energies associated with a particular element is unique and can be used as an unambiguous method of identification as shown in Fig. 3.1. Additionally, such spectra provide information about the electron energy levels inside the atom. Similarly, gamma-ray decay energies can be used as a method of isotopic identification as shown in Fig. 3.2 as well as to provide information regarding nuclear energy levels.
3.1 Interactions of gamma-rays with matter

Photons are neutral particles and therefore cannot be directly detected. In order to detect gamma ray photons and measure their energy, interactions which result in the complete or partial transfer of photon energy to an atomic electron are utilized. In these reactions, a gamma ray is either completely adsorbed in the detection material or scattered through an angle [11], producing high energy electrons. The important processes in radiation detectors which give rise to this behavior are photoelectric absorption, Compton scattering, and pair production; these interactions are described in the following sections. Gamma-ray detectors have two important features: 1) the detector material is chosen such that incident gamma-rays have a reasonable probability of interaction and 2) they function as a detection system for the secondary electrons produced following interactions with the detector medium [11]. Because the incident energy of a gamma ray (and the resultant secondary electrons) is \( \sim 1 \) MeV, the detector material must be chosen to have sufficient electronic stopping power; as a consequence, gas-filled detections systems are generally not used [11].

3.1.1 Photoelectric absorption

Photoelectric absorption is a process in which a photon transfers all of its energy to an atomic electron (called the photoelectron) [11]. The electron is ejected from the atom with
energy given by Eq. 3.2

\[ E_{e^-} = h\nu - E_b \]  

where \( h\nu \) is the energy of the incident photon and \( E_b \) is the binding energy of the electron in the atom. The atom is ionized in this process and the vacancy left by the photoelectron is filled by a free electron or by an electron in another atomic shell [11]; a schematic diagram of this process is shown in Fig. 3.3. During this process, an X-ray or Auger electron can be emitted as the absorber atom returns to the ground state. Because the photoelectric effect requires the presence of a nucleus, the innermost electrons occupying the 1s orbital are preferentially ejected from the atom. If the atom returns to the ground state by X-ray emission, a characteristic X-ray with energy equal to the binding energy of an electron in the 1s orbital of the interaction medium is emitted. Photoelectric absorption is the primary interaction mechanism for low energy gamma rays; the photoelectric absorption cross section
\( \sigma \) is a function of both the gamma-ray energy and the atomic number \( Z \) of the interaction medium approximated by Eq. 3.3

\[
\sigma \sim C \times \frac{Z^n}{E_{\gamma}^{3.5}}
\]  

(3.3)

where \( C \) is a constant, \( E_{\gamma} \) is the incident gamma-ray energy and \( n \) is between 4 and 5 depending on the gamma-ray region of interest [11].

Figure 3.3: The photoelectric absorption of an incident gamma ray and resultant photoelectron. The photoelectron energy is given by Eq. 3.2. When the vacancy left by the photoelectron is filled, energy is released either as an X-ray or Auger electron.

The photoelectric absorption process can occur on an atomic electron but not a free electron, as the presence of the massive nucleus is necessary in order to satisfy the conservation laws for energy and momentum. A detailed description of the violation of the conservation laws for photoelectric absorption on a free electron is provided in Appendix B.

### 3.1.2 Compton scattering

The Compton scattering process involves an incident photon scattering off an electron. During this interaction, some of the incident photon energy is transferred to the recoiling electron [11]; this process is illustrated in Fig. 3.4. The amount of energy transferred is dependent on the scattering angle. For some scattering angle \( \theta \in [0, \pi] \), the energy of the scattered photon is given by Eq. 3.4 and the energy of the recoiling electron by Eq. 3.5:

\[
h_{\nu}' = \frac{h\nu}{1 + \frac{\alpha (1 - \cos \theta)}{1 + \alpha (1 - \cos \theta)}}
\]  

(3.4)

\[
E_{e^-} = h\nu - h\nu' = h\nu \left( \frac{\alpha (1 - \cos \theta)}{1 + \alpha (1 - \cos \theta)} \right)
\]  

(3.5)

where \( \alpha = \frac{h\nu}{m_0c^2} \) and \( m_0 \) is the electron mass; a derivation of the Compton scattering equations is presented in Appendix C. From Eqs. 3.4 and 3.5, two special cases can be
CHAPTER 3. PRINCIPLES OF GAMMA-RAY SPECTROSCOPY

Figure 3.4: The Compton scattering interaction showing the recoil electron and gamma ray deflected by a scattering angle $\theta$. The energies of the scattered gamma ray and recoil electron are given by Eq. 3.4 and 3.5, respectively.

identified. The first case is when the scattering angle $\theta \sim 0$. In this case, $h\nu' \sim h\nu$ and $E_{e^-} \sim 0$ since $\cos \theta \sim 1$. The second special case occurs when the scattering angle $\theta = \pi$. In this case, the incident gamma is back-scattered and the electron recoils in the direction of the incident gamma ray. The back-scattered gamma ray has energy given by Eq 3.6

$$h\nu'|_{\theta=\pi} = \frac{h\nu}{1 + 2\alpha}$$

and the electron energy is given by Eq. 3.7; this energy represents the maximum recoil energy of the electron in the Compton scattering process.

$$E_{e^-}|_{\theta=\pi} = h\nu \left( \frac{2\alpha}{1 + 2\alpha} \right)$$

The differential Compton scattering cross section $d\sigma/d\Omega$ is given by the Klein-Nishina formula shown in Eq. 3.8.

$$\frac{d\sigma}{d\Omega} = Zr_0^2 \left( \frac{1}{1 + \alpha(1 - \cos \theta)} \right)^2 \left( \frac{1 + \cos^2 \theta}{2} \right) \left( 1 + \frac{\alpha^2(1 - \cos \theta)^2}{(1 + \cos^2 \theta)[1 + \alpha(1 - \cos \theta)]} \right)$$

where $r_0$ is the electron radius. Equation 3.8 shows a linear dependence on the atomic number $Z$ of the absorber material. Compton scattering is the primary interaction mode for intermediate energy gamma-rays as the probability of scattering per absorber atom decreases as the gamma-ray energy increases [11].

3.1.3 Pair production

The pair production process occurs when a gamma ray with energy twice the rest mass of the electron ($2m_0c^2 = 1.022$ MeV) interacts with the electric field of the nucleus [11].
From the conservation of energy and momentum, it can be shown that pair production in free space is prohibited, see Appendix D for details. In this process, the photon creates an electron/positron pair; if \( E_\gamma > 1.022 \text{ MeV} \), the excess energy is shared between the two particles. The probability of pair production increases with the incident gamma-ray energy, and is therefore the primary interaction process for high energy gamma rays. The electron and positron travel only a short distance (< a few millimeters) in the absorber medium before losing their kinetic energy. Once the positron has a kinetic energy similar to the kinetic energy of the absorber electrons, it will interact with an electron in the medium and annihilate, creating two photons of energy \( m_0c^2 = 511 \text{ keV} \) emitted back-to-back as shown in Fig. 3.5 [11]. The annihilation process occurs quickly, and in practice, these elimination photons are emitted in coincidence with the initial gamma ray.

![Figure 3.5: The pair production process involves a gamma-ray interaction with the electric field of the nucleus and the subsequent creation of an electron/positron pair. When the positron has kinetic energy roughly equal to the thermal energy of the atomic electrons, an annihilation reaction creating two photons with energy \( E_\gamma = m_0c^2 = 511 \text{ keV} \) occurs.](image)

3.2 Semiconductor diode radiation detectors

Radiation detectors based on organic or inorganic scintillators are resolution limited due to the statistical fluctuations in the number of photoelectrons produced during a detection event. The construction of semiconductor diode radiation detectors from material such as high purity germanium (HPGe) reduces the statistical limit on resolution by increasing the number of information carriers per event [11]. In a semiconductor diode detector,
charged particles create electron-hole pairs as they move through the detector material. The average energy required to create an electron-hole (the “ionization energy”) is generally both constant and independent of the ionization mechanism. At 2.96 eV, the ionization energy of HPGe is low compared to the energy regime of interest for gamma-ray spectroscopy (∼10 keV–10 MeV), leading to a large number of electron-hole pairs created during each interaction. Since the number of electron-hole pairs is directly related to the energy of the charged particle which creates them, the large number of electron-hole pairs created in an HPGe detector during a detection event results in superior energy resolution due to reduced statistical fluctuations. Other advantageous properties of semiconductor diode detectors are their relatively small size, good timing characteristics, and the flexibility during fabrication which allows for the construction of detectors of varying sizes and shapes dependent on intended use [11].

Impurities in semiconductor diode detectors can result in semiconductors of “n-type” or “p-type.” An n-type semiconductor contains a trace impurity with an additional valence electron compared to the bulk material. The additional valence electron is loosely bound to the impurity site in the crystal lattice and can be easily promoted to the conduction band without the formation of the hole which occurs during the normal excitation process [11]. Similarly, p-type semiconductors contain small concentrations of impurities with one less valence electron than the bulk material. The unformed covalent bond in the crystal lattice behaves like the hole which remains when a valence electron is promoted to the conduction band [11]. An electron captured by this site is slightly less bound to the impurity atom than an electron which occupies a normal bonding site within the valence band, for more information see Figs. 11.3 and 11.4 in Ref. [11]. For HPGe detectors, the bulk material is typically of p-type due to the fabrication process; typical doping materials which change the properties of the bulk are lithium for the making the bulk of the n-type and boron for making the bulk of the p-type; concentrations generally range from $10^{11}$–$10^{15}$ atoms/cm$^3$ [11]. Lithium and boron are also the metals used to fabricate the electrical contacts on the surface of the bulk HPGe crystal.

Semiconductor diode detectors take advantage of the properties which occur at the interface of p- and n-type semiconductors. At the p-n junction, there is a sharp discontinuity in electron density. Consequently, electrons in the conduction band can migrate from the n-type material to the p-type, causing a net positive charge of the n-type material and a net negative charge of the p-type [11]. The resultant electric field gradient from the charge
buildup when this process is at equilibrium is called the “contact potential.” The field of the contact potential is sufficient to prevent the continued migration of charge, and a steady-state distribution is established. The region over which this effect manifests is known as the “depletion region” and has some beneficial properties for radiation detection [11].

When ionizing radiation interacts with the sensitive volume of the detector, a series of electron-hole pairs are created along the interaction pathway. As a consequence of the electric field present in the depletion region, any electrons created by ionization of the detection medium are transported towards the n-type material while the corresponding holes are transported towards the p-type. As electrons leave occupied valence sites to fill existing holes, they leave behind a new hole in their place [11]. The hole behaves like a positive point charge because it represents the absence of a negatively charged electron. As a result, there is a migration of charge due to both the movement of the electrons opposite to the direction of the electric field and the holes along the direction of the electric field. The motion of these charges within the electric field forms the electrical signal of the detector [11].

The depletion region of an unbiased p-n junction is very small, and has a contact voltage on the order of 1 V. Thus, the motion of the electrons and holes are slow [11]. Applying a reverse bias (negative on the p-type side, positive on the n-type) enhances the natural potential difference of the junction and the flow of charge in one direction while simultaneously preventing the flow of charge in the opposite direction. This increased potential extends the depletion depth according to Eq. 3.9

$$d = \left( \frac{2\varepsilon V}{eN} \right)$$

where $\varepsilon$ is the dielectric constant of the detector material, $V$ is the applied voltage, $e$ is the electric charge and $N$ is the impurity concentration. For a given applied voltage, the only way to increase the size of the depletion depth is to decrease the concentration of impurities in the detector material. HPGe fabrication techniques have been refined to impurity concentrations on the order of $10^9$ atoms/cm$^3$, or approximately 1 impurity in $10^{12}$ atoms of germanium [11]. HPGe detectors typically operate at bias voltages of 3500–5000 V.
CHAPTER 3. PRINCIPLES OF GAMMA-RAY SPECTROSCOPY

with depletion regions of several cm, making them ideal for detailed spectroscopic studies.

In principle, the active volume of an HPGe detector is the entire depleted region between the n and p contacts. The contacts are typically constructed by implantation or evaporation of dopant onto the surface of the germanium crystal or the diffusion of dopant into the germanium. These processes can lead to contacts with thicknesses ranging from tens of microns to a few millimeters depending on the fabrication method. This creates a dead layer on the crystal which has a non-negligible effect on the detection of low energy gamma-rays [11].

3.3 Germanium detector response to a monoenergetic gamma-ray source

The response of HPGe detectors to a monoenergetic gamma-ray source is complicated due to the interactions detailed in Sec. 3.1. The overall detector response is a combination of the energy dependent contributions of the photoelectric absorption, Compton scattering and pair production processes, as well as a response from the signal processing electronics. For simplicity, the detector response to a monoenergetic gamma ray with $E_\gamma > 1.022$ MeV is assumed in order to account for pair production effects. If the gamma-ray energy is less than 1.022 MeV, only the responses which arise from photoelectric absorption and Compton scattering must be considered. A schematic diagram of the detector response to a gamma ray with $E_\gamma > 1.022$ MeV is shown in Fig. 3.6; each feature of this spectrum can be described with respect to the interaction processes which cause them.

The photopeak of energy $E_\gamma$ in Fig. 3.6 occurs when the sum of the kinetic energy of all electrons created following the photoelectric absorption of the incident gamma ray are deposited in the detector. The detector response is a single delta function of the electron kinetic energy which corresponds to the energy of the incident gamma-ray [11]. The X-ray escape peak occurs when the X-ray created following the full photoelectric absorption of the incident gamma ray escapes the detector, and has energy $E_\gamma - E_X$; for germanium, the characteristic X-ray has energy $E_X = 11$ keV.

During pair production, an energetic electron is created along with two coincident 511 keV gamma-rays. If all subsequent interactions involving both the pair production electron and the secondary electrons created by the two 511 keV gamma rays take place within the detector volume, the full photopeak energy is detected. However, one or both 511 keV
gamma-rays can escape the detector, resulting in the detection of the single escape peak with energy $E_\gamma - 511$ keV or double escape peak with energy $E_\gamma - 1.022$ MeV as shown in Fig. 3.6. The annihilation peak at 511 keV is due to the detection of gamma rays from pair production events which occur outside the detector. The so-called Compton continuum which arises from the electron energy distribution as a function of Compton scattering angle $\theta$ in Eq. 3.5 is shown in Fig. 3.7 for the case of a monoenergetic gamma-ray of energy $E_\gamma = h\nu$. The Compton edge at energy $h\nu - E_C$ is equal to the maximum electron energy which occurs at scattering angle $\theta = \pi$ given in Eq. 3.7.

The effects of the gamma-ray interactions are further complicated due to resolution losses due to the statistical fluctuations of the number of electron-hole pairs created during the interaction processes, incomplete charge collection and electronic noise [11]. The full width half max (FWHM) of a photopeak in a monoenergetic gamma-ray spectrum is given by Eq. 3.10

$$\text{FWHM}^2 = W_D^2 + W_X^2 + W_E^2$$

(3.10)

where $W_D^2$ is due to contributions from the statistical fluctuations, $W_X^2$ is the contribution from the incomplete charge collection, and $W_E^2$ is noise due to the electronics of the signal processing. The contributions due to statistical fluctuations can be expanded as shown in Eq. 3.11

$$W_D^2 = \left(2\sqrt{2\ln 2}\right)^2 F\rho E_\gamma \approx 5.55 \times F\rho E_\gamma$$

(3.11)
Figure 3.7: The resultant gamma-ray energy spectrum for an incident gamma-ray with energy \( E_\gamma = h\nu \) which arises from the energy distribution of energetic electrons from the Compton scattering process. The energy distribution of the Compton continuum is given by Eq. 3.5. The Compton edge at energy \( h\nu - E_C \) is given by the maximum electron energy which occurs at scattering angle \( \theta = \pi \) as shown in Eq. 3.7.

where \( F \) is the Fano factor (which relates the observed variance in the number of charge carriers created to the expected variance), \( \rho \) is the ionization energy of the detector material (2.96 eV for germanium) and \( E_\gamma \) is the incident gamma-ray energy. At the common calibration energy of 1332.492 keV from \(^{60}\text{Co}\), Eq. 3.11 predicts the FWHM contribution from statistical noise to be \( \sim 1.32 \) keV if a Fano factor of 0.08 is used [11]. In practice, HPGe detectors demonstrate energy resolutions of \( \sim 2 \) keV at 1332.492 keV, indicating that either the contributions to the FWHM from incomplete charge collection and electronic noise are significant or that the estimated Fano factor is incorrect; careful measurements can establish reasonable estimates for \( W_X \) and \( W_E \) for most detector systems [11]. The energy broadening introduced by Eq. 3.10 degrades the sharp delta function features which arise from the gamma-ray interactions with the detector and give rise to experimental spectra which show the Gaussian-like features of Fig. 3.6.
3.4 Germanium detector response to a polyenergetic gamma-ray source

The response of an HPGe detector to polyenergetic gamma-rays can be separated into two cases: 1) gamma-rays not emitted in a cascade and 2) gamma-rays emitted in a cascade. In the first case, the gamma rays are emitted independently and the detector response is a linear combination of the monoenergetic response functions due to each gamma ray where the overall contribution of the features which arise from a single decay is defined by the intensity of the decay channel. In the second case, the response of the detector is complicated due to summing effects. A gamma-ray cascade occurs when two or more gamma rays are emitted in quick succession where the lifetime of the intermediate states are short in comparison to the time resolution of the detector. Because the intermediate state lifetimes are shorter than the time resolution, there is a probability that two or more gamma rays will register as a single event. If both gamma-rays are detected at the full photopeak energy, a “sum peak” with energy equal to the sum of the photopeak energies of the participating gamma rays will be present in an experimental system. Features which arise from coincident partial energy deposits are also present and contribute additional counts to the background. Summing effects can be quite significant in experimental spectra, and special care must be taken when analyzing data in those cases.
Chapter 4

Gamma-ray spectroscopy at Simon Fraser University

The SFU Department of Chemistry recently underwent a $\sim$ $50$ million facilities renovation which in part supplied the Nuclear Science group lead by Profs. Krzysztof Starosta and Corina Andreoiu with new laboratory space designed to be used with radioisotopes. The new facilities include a radiation vault that will house a neutron generator [17], a “hot” laboratory with stainless steel counter tops and a fume hood that is graded for radioisotope use, and a laboratory for detector development. These facilities and the existing resources of the Nuclear Science group are part of an active and ongoing commitment by SFU Chemistry to cutting-edge nuclear science research.

SFU currently has the facilities, equipment, and expertise necessary to take advantage of opportunities to use gamma-ray spectroscopy in fundamental research and applied science. At least two major projects at SFU are envisioned for the existing gamma-ray spectroscopic equipment at SFU: the ongoing efforts of SFU Nuclear Science laboratory providing environmental monitoring services to the scientific community as well as the general public, and the future Subcritical Intense Multiplier of Neutrons (SIMON) project. Applications in nuclear science and materials science within the Department of Chemistry are being considered as well as projects of interest to Archaeology, Criminology and Earth Science.
4.1 The Germanium Detector for Elemental Analysis and Radioactivity Studies (GEARS)

The Germanium Detector for Elemental Analysis and Radioactivity Studies (GEARS) is an Ortec GEM Profile Series HPGe coaxial gamma-ray detector which was operational at the time of the Fukushima accident. GEARS is coupled to an Ortec DSPEC jr. 2.0 multichannel analyzer (MCA) with bias voltage provided by a built-in power supply. Gamma-ray decay spectra are collected by a computer running the Ortec Maestro32 software.

A low-activity lead shield with dimensions 50 cm $\times$ 50 cm $\times$ 64 cm ($l \times w \times h$) constructed from 10 cm thick lead bricks was used to house the GEARS detector. The shield reduces low-level background seen by the detector due to naturally occurring radioactive materials (NORM) [18]. Most background radiation is cosmogenic or primordial such as the strong $^{40}$K line and gamma rays characteristic of the uranium and thorium decay chains [18]. Some background radiation, however, has been introduced into the environment by human activity. For example, $^{137}$Cs background radiation is a legacy of atmospheric nuclear weapons testing. The lead shield is coupled with a Cu/Cd graded-$Z$ shield to decrease the flux of low energy gamma rays and X-rays coming from the background and the lead shield [11]; this setup is shown in Fig. 4.1. This shielding arrangement reduces the background radiation measured by the GEARS detector by a factor of $\sim$ 100 and strongly suppresses or eliminates many background lines, see Fig. 4.2. The suppression or elimination of gamma-ray lines depends on both the energy of the gamma ray and the gamma-ray source. Higher energy gamma rays penetrate the shield with higher probability. Gamma rays and X-rays which originate from radioactive material in the lead shield and the air are shielded less effectively than gamma rays of terrestrial origin and from radioisotopes present in the building material.

4.2 The data acquisition system and analysis software

4.2.1 The DAQ

Digital signal processing for GEARS is provided by the DSPEC jr. 2.0 zero dead time multichannel analyzer (MCA) manufactured by Ortec. The DSPEC electronics include a low frequency rejection (LFR) filter for noise reduction during pulse processing. The parameters of the LFR filter as well as other settings related to spectral acquisition and
detector operation such as the energy threshold and detector high voltage are adjustable in the included software. The DSPEC is connected to a data acquisition computer through a USB 2.0 interface and spectra are acquired using the Maestro 32 general purpose MCA software. Documentation of these devices can be found in Refs. [19, 20].

4.2.2 RadWare

RadWare is an interactive graphical software package for the analysis and presentation of gamma-ray data. RadWare includes a least squares fitting program named gf3 for the analysis of one-dimensional gamma-ray spectra [21]. The gf3 program can simultaneously fit up to fifteen peaks above a quadratic background. Peaks are fit with a Gaussian or a skewed Gaussian function and may be positioned above a step function. The $\chi^2$ goodness of fit statistic, peak shape parameters, peak height, width, area, and energy can be extracted from the results of the fit. While developed specifically to aid gamma-ray spectroscopy, gf3 is a general purpose histogram analysis program. Information regarding RadWare gf3 commands and details of the fitting function can be found in Ref. [21].
Figure 4.2: Examples of background spectra obtained with GEARS in an unshielded [red] and shielded [blue] configuration; the shielded configuration shows an $\sim 100$ fold decrease in the overall background while many background peaks associated with NORM are either eliminated or strongly suppressed. Spectra are normalized to the same live time.

4.2.3 ROOT

ROOT is an object oriented, C++ based software package for data analysis initially developed at the European Organization for Nuclear Research (CERN) in the mid-1990’s. ROOT scripts in C++ can be written to aid in the visualization of multidimensional histograms and to perform iterative curve fits to functions defined by the user. The ROOT framework also contains a set of useful data structures to facilitate the analysis of complex data. Optional cross compiling with the Monte Carlo code Geant4 (described in Sec. 4.3) allows the user to run these simulations with a ROOT graphical interface; all Geant4 simulations described herein were compiled in this fashion. Documentation for ROOT is available online and can be accessed via Ref. [22].
4.3 Geant4 Monte Carlo simulations for GEARS

Geant4 is a software package developed by CERN to simulate the interaction of radiation with matter [23,24]. Geant4 allows the user to create an experimental hall of custom dimension and incorporate objects of various geometries and materials into the hall. The GEARS detector, housing, graded-Z shield, lead shield, and other components were accurately modeled using this framework as shown in Fig. 4.3. Additionally, many types of radioactive decay, including gamma-ray decay, can be simulated using Geant4.

Geant4 simulations are used to aid the analysis of GEARS data, in particular, they can be used to perform spectral decomposition, described in detail in Sec. 5.6. Geant4 simulates the energy deposited in a detector, but does not simulate the response of the electronics involved in processing the signal. However, the effect of the electronics can be simulated using a response function, which in the case of GEARS is the energy-dependent peak shape response function.

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Figure 4.3: The GEARS geometry implemented in the Geant4 software package. The box superstructure is lead shielding, the large wire-frame cylinder is the Cu/Cd graded-Z shield, the small wire-frame cylinder is the aluminum can which houses the detector, and the solid cylinder is the HPGe crystal of the GEARS detector.
The peak shape for gamma-rays interacting with GEARS in a simulated spectrum is modeled using the normal distribution given by Eq. 4.1,

\[ f(E) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(E - E_0)^2}{2\sigma^2}\right) \] (4.1)

where \( E_0 \) is the energy of the gamma-ray and \( \sigma \) is related to the width of the distribution. Gamma-ray line shape is approximately Gaussian as a consequence of the Central Limit Theorem, though in practice, deviations from pure Gaussian shapes are observed. For more information on gamma-ray interactions with Germanium detectors and the processes responsible for peak shape, see Sec. 3.3. The energy dependent detector response is modeled using the full width at half maximum (FWHM), which is related to the Gaussian \( \sigma \) by Eq. 4.2. Further information on FWHM and peak shape in Germanium detectors is contained in Sec. 3.3.

\[ \text{FWHM} = 2\sqrt{2\ln 2} \sigma \approx 2.35\sigma \] (4.2)

The detector dead layer was included in the simulations, and all energy deposits in the defined dead layer were not recorded. The dead layer was set to 700 \( \mu \)m, as listed in the detector manual. Further information on detector construction and dead layers is found in Sec. 3.2.

### 4.4 Applications of gamma-ray spectroscopy at SFU

#### 4.4.1 Activation analysis

The Subcritical Intense Multiplier of Neutrons (SIMON) project at SFU is proposed to bring a pulsed deuterium/tritium neutron generator with a flux of \( 3 \times 10^8 \) neutrons/second to the existing radiation vault at the SFU Department of Chemistry. This project will pursue a research program that addresses topics of fundamental interest to the nuclear science community [17]. Such applications include mass and beta-decay lifetime measurements, spontaneous fission studies, and neutron activation analysis. A gamma-ray detector will be used in addition to the proposed detector systems envisioned for the SIMON project in order to monitor the neutron flux generated by the system during the course of operation.

Thin foils of materials with well known neutron absorption cross sections can be placed in the moderator for neutron activation. Their gamma-ray decay spectra can be observed with
a single gamma-ray detector with known absolute efficiency over a large range of energies. Neutron activation analysis of these samples can be used to monitor the neutron flux.

Neutron activation analysis can also be used as an analytical technique. Activation analysis does not require samples to be soluble, involves little preparation time, and is non-destructive. Coupled with nuclear identification methods such as gamma-ray spectroscopy, neutron activation can provide important information regarding chemical composition of materials for use in both basic and applied research [17].

4.4.2 Environmental monitoring

Gamma-ray spectroscopy with germanium detectors is a powerful technique which can be used to identify and measure the concentration of radioisotopes. This technique has been used extensively in environmental monitoring [3–10, 25, 26]. Of particular interest to health professionals, governments, and the general public is the monitoring of radioisotope emissions from nuclear accidents. Prevailing atmospheric conditions and the accident scenario can lead to widespread release of gaseous or aerosolized radioisotopes which can be distributed over large areas. Many of these radioisotopes have half-lives long enough to survive long journeys and can pose serious public health and environmental hazards if present in large quantities. Therefore, dedicated detection systems that can be brought into operation on short notice following a nuclear accident are of great use for both public education and informing government response on a local level.
Chapter 5

Calibration of the GEARS detector

Following the release of radioisotopes from the damaged Fukushima Dai-ichi nuclear power station, there was a limited window of opportunity to gather and analyze meaningful data. The time necessary to fully characterize the detector before analyzing backlogged samples was not available. Energy calibration can be completed quickly and was accomplished using a set of standard point sources. Due to time constraints, the efficiency of GEARS was initially estimated with a large error and samples were analyzed.

The future operation planned for the GEARS detector described in Sec. 4.4 requires that both the energy and efficiency response of GEARS is understood to a high degree of accuracy and precision. Therefore, it is critical to establish a standard set of repeatable procedures in order to perform energy and efficiency calibrations. Since both energy and efficiency calibrations were necessary in order to analyze the data taken during the Fukushima monitoring campaign, the standard calibration procedures needed for the future operation of GEARS were developed prior to analysis and a thorough calibration of GEARS was performed.

5.1 Energy calibration

Every radioisotope that decays via gamma-ray emission produces a characteristic energy spectrum, examples of such spectra are shown in Fig. 3.2 for $^{60}$Co and $^{137}$Cs. For a sample containing a mixture of radioisotopes, the spectrum measured by GEARS will be a linear combination of the spectra corresponding to each individual radioisotope contained in the sample plus natural background, see Sec. 3.4. In order to describe the isotopic composition
of such a spectrum, peak energies of each observed gamma-ray decay must be compared to a library of known decay energies. An energy calibration was performed to establish the detector response as a function of gamma-ray energy.

The Maestro32 software used by the GEARS detector writes decay spectra in a histogram with the arbitrary x-axis unit of channels. It is therefore necessary to understand the relationship between energy and channel number. A series of $^{137}\text{Cs}$, $^{152}\text{Eu}$, $^{133}\text{Ba}$, $^{56}\text{Co}$, $^{60}\text{Co}$ and $^{241}\text{Am}$ source measurements were performed. Peaks of known energy were fit using modified Gaussian functions using RadWare gf3 as described in Sec. 4.2.2 to establish a relationship between decay energy and channel number using the formalism described in Sec. 5.2

### 5.2 Least squares formalism

The least squares method is used to find the best set of parameters for a function $f$ which represents a set of measurements $(x_i, y_i, \sigma_i)$. It is implemented by defining the function given by Eq. 5.1

$$\chi^2 = \sum_i \sigma_i^{-2}(y_i - f(x_i))^2$$

followed by the minimization of $\chi^2$ with respect to the parameters of $f$. In some cases, minimization can be performed analytically rather than numerically, this is referred to as the “linearized $\chi^2$ method.” The linearized $\chi^2$ is specific to the problem, and an analytical solution must be derived for each application.

#### 5.2.1 First-order Fit

A linear relationship between gamma-ray energy and channel number can be described by $E = a_0 + a_1c$ where $E$ is energy, $c$ is channel number, and $a_0$, $a_1$ are coefficients which are determined by a least-squares fitting method for which

$$\chi^2 = \sum_{i=1}^{N} (E_i - a_0 - a_1c_i)^2$$

is minimized with respect to $a_0$ and $a_1$ for $N$ measurements where each measurement is a $(c_i, E_i)$ pair which corresponds to a known decay. Details of this particular method can be found in Appendix E.
5.2.2 Weighted First-order Fit

The fitting technique in Sec. 5.2.1 gives equal weight to each \((c_i, E_i)\) pair. However, it is not possible to fit every peak in a spectrum with equal precision due to the quality of the peaks and the background pattern in a particular spectral region. To correct this problem, the initial first-order fit equation shown in Eq. 5.2 was modified to include a weighting factor \(\sigma^{-2}\), where \(\sigma\) was the uncertainty in the channel number of a fitted peak given by an analysis program. The new, weighted first-order fit function is given by Eq. 5.3

\[
\chi^2 = \sum_{i=1}^{N} \sigma_i^{-2} (E_i - a_0 - a_1 c_i)^2
\]  

(5.3)

which can be solved using a similar method as Eq. 5.2, see Appendix F.

5.2.3 Second-order Fit

The weighted first order fit was not adequate to calibrate the detector to a satisfactory level, and it was decided that adding a non-linear term to the \(\chi^2\) calculation may bring the calibration better in line with the known source energies. The fit was again done by the \(\chi^2\) minimization of Eq. 5.4

\[
\chi^2 = \sum_{i=1}^{N} \sigma_i^{-2} (E_i - a_0 - a_1 c_i - a_2 c_i^2)^2
\]  

(5.4)

which can be solved using the method outlined in Appendix G.

5.3 Photopeak efficiency calibration of GEARS

Only a small number of gamma-rays emitted by a source will interact with the GEARS detector. Most will not hit the detector due to limited solid angle coverage, and most that reach the detector will pass through without interaction. Therefore, to accurately measure the activity of a radioactive sample, the absolute efficiency of gamma-ray detection must be known with a high degree of accuracy and precision. Only photopeak events are used to determine efficiency because the number of counts in the photopeak is not sensitive to scattering effects and noise [11]. The absolute photopeak efficiency of a detector system is related to the number of gamma rays observed in a photopeak of a given energy. The
CHAPTER 5. CALIBRATION OF THE GEARS DETECTOR

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definition of the relationship is shown in Eq. 5.5

\[ N(E) = \epsilon(E) \times \text{br}(E) \times I \]  

(5.5)

where \( N(E) \) is the number of counts in the photopeak at energy \( E \), \( \epsilon(E) \) is the efficiency for detecting a gamma ray at energy \( E \), \( \text{br}(E) \) is the branching ratio of the gamma ray at energy \( E \) and \( I \) is the absolute intensity of the source.

Efficiency calibrations can be relative or absolute. Relative efficiency measurements determine the photopeak efficiency at a given energy with respect to the photopeak efficiency of detecting a reference decay, while absolute efficiency measurements yield a direct photopeak detection probability as a function of energy.

The efficiency of a detector is typically measured using reference sources of known activity. However, the absolute activity of a reference source is reported by the vendor and adds systematic uncertainty due to source calibration. A source-independent absolute efficiency calibration method was developed and compared to an absolute efficiency calibration using an activity-calibrated source. The methods of these independent calibrations are presented in Secs. 5.4 and 5.5; a comparison of measured efficiencies of GEARS using a calibrated source and a source-independent method is shown in Sec. 5.7.

5.4 Absolute efficiency calibration of GEARS using an activity-calibrated \(^{60}\text{Co}\) point source

5.4.1 Relative efficiency calibration of GEARS using an uncalibrated \(^{152}\text{Eu}\) point source

Relative efficiency calibrations establish the efficiency of detecting a gamma-ray \( \gamma_i \) with respect to a reference gamma-ray, \( \gamma_{\text{ref}} \). A high energy gamma ray on the order of \( \sim 1 \text{ MeV} \) should be used as the reference to eliminate effects of the electronics (such as threshold settings) in the signal processing; for more information, see Sec. 4.2.1 and references therein. Proper calibration requires a source with multiple gamma ray photopeaks across a range of energies such as \(^{152}\text{Eu}\) or \(^{57}\text{Co}\). Since the branching ratios of gamma-ray sources used for calibration are well known, relative efficiency can be measured with a high degree of accuracy.

Ratios of efficiencies can be extracted from the ratio of the number of counts at a given energy \( E_i \) to the number of counts at energy \( E_{\text{ref}} \), where the number of counts is given
by Eq. 5.5; the formula for relative efficiency is shown in Eq. 5.6 and is independent of source intensity $I$ from Eq. 5.5. In Eq. 5.6, $N(E)$ is the number of counts contained in a photopeak of energy $E$, $\epsilon(E)$ is the absolute efficiency of detection at energy $E$, and $br(E)$ is the branching ratio for the gamma ray of energy $E$. It should be stressed that the ratio $br(E)/br(E_{\text{ref.}})$ is well known for the common calibration sources used for relative efficiency measurements.

$$\frac{N_i(E_i)}{N_{\text{ref.}}(E_{\text{ref.}})} = \frac{\epsilon_i(E_i)}{\epsilon_{\text{ref.}}(E_{\text{ref.}})} \times \frac{br(E_i)}{br(E_{\text{ref.}})} \quad (5.6)$$

The relative efficiency of GEARS was measured at several $^{152}\text{Eu}$ decay energies with the source secured 10.5 cm above the detector can. Relative efficiencies of GEARS were measured with respect to $E_{\text{ref.}} = 1408.011$ keV. The results of the relative efficiency measurement are shown in Table 5.1 and Fig. 5.1.

Table 5.1: Relative efficiency of GEARS measured with a $^{152}\text{Eu}$ source positioned 10.5 cm above the detector can. Errors at one standard deviation are shown in parentheses. Relative efficiency $\epsilon_{\text{rel.}} = \text{photopeak counts/scaling factor}$, where the scaling factor is a normalized, decay-independent peak intensity ratio which accounts for branching ratios from multiple decay channels and gamma-ray cascades. The scaling factors are normalized with respect to a scaling factor of 10000 for $E_{\text{ref.}} = 1408.011$ keV. Data from this table are shown in Fig. 5.1.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Photopeak counts</th>
<th>Scaling factor</th>
<th>$\epsilon_{\text{rel.}}$ [arb.]</th>
</tr>
</thead>
<tbody>
<tr>
<td>121.783</td>
<td>10807303(4675)</td>
<td>13620(160)</td>
<td>793(9)</td>
</tr>
<tr>
<td>244.692</td>
<td>2253868(2263)</td>
<td>3590(60)</td>
<td>628(11)</td>
</tr>
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<td>295.939</td>
<td>113066(839)</td>
<td>211(5)</td>
<td>536(13)</td>
</tr>
<tr>
<td>344.276</td>
<td>6234362(3481)</td>
<td>12750(90)</td>
<td>489(3)</td>
</tr>
<tr>
<td>367.789</td>
<td>195688(853)</td>
<td>405(8)</td>
<td>483(10)</td>
</tr>
<tr>
<td>411.115</td>
<td>454056(1087)</td>
<td>1070(10)</td>
<td>424(4)</td>
</tr>
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<td>598236(1204)</td>
<td>1480(20)</td>
<td>404(6)</td>
</tr>
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<td>778.903</td>
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<td>6190(80)</td>
<td>266(3)</td>
</tr>
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<td>867.388</td>
<td>491150(1107)</td>
<td>1990(40)</td>
<td>247(5)</td>
</tr>
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<td>6490(90)</td>
<td>208(3)</td>
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<td>670(8)</td>
<td>195(2)</td>
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<td>143813(554)</td>
<td>780(10)</td>
<td>184(2)</td>
</tr>
<tr>
<td>1408.011</td>
<td>1747387(1772)</td>
<td>10000(30)</td>
<td>174.7(6)</td>
</tr>
</tbody>
</table>
Figure 5.1: Relative efficiency of GEARS measured with a $^{152}\text{Eu}$ source positioned 10.5 cm above the detector can normalized with respect to $E_{\text{ref.}} = 1408.011$ keV. Data shown on this figure are in Table 5.1. Error bars are smaller than the data points.

5.4.2 Absolute efficiency measurement of GEARS at $^{60}\text{Co}$ decay energies

Following re-arrangement of Eq. 5.5, absolute efficiency $\epsilon_{\text{abs.}}$ is given by Eq. 5.7.

$$\epsilon_{\text{abs.}}(E) = \frac{N(E)}{I_{\text{abs.}} \times \text{br}(E)}$$  \hspace{1cm} (5.7)

where $N(E)$ is the number of gamma rays detected in a photopeak of energy $E$ detected during a measurement, $\text{br}(E)$ is the branching ratio associated with the gamma ray with energy $E$, and $I_{\text{abs.}}$ is the absolute intensity of the source. The absolute intensity of the source is the number of gamma rays emitted by the source during the live time of the measurement and is given by Eq. 5.8

$$I_{\text{abs.}} = A(t_0)e^{-(t-t_0)/\tau} \times t_{\text{live}}$$  \hspace{1cm} (5.8)

where $A(t_0)$ is the source activity reported at $t = t_0$, $t - t_0$ is the time elapsed since $t_0$, $\tau$ is the lifetime of the source and $t_{\text{live}}$ is the live time of the measurement. It should be noted that Eq. 5.8 is only valid when $t_{\text{live}} \ll \tau$. Substituting Eq. 5.8 into Eq. 5.7 yields Eq. 5.9,
the equation used to calculate absolute efficiency from an activity calibrated source.

$$\epsilon_{\text{abs}}(E) = \frac{N(E)}{A(t_0)e^{-(t-t_0)/\tau} \times t_{\text{live}} \times br(E)} \tag{5.9}$$

In order to effectively characterize the efficiency response of a detector, the error associated with the measured absolute efficiency must be known. The error $\delta\epsilon_{\text{abs}}$ can be calculated from Eq. 5.9 using standard error propagation techniques. However, when experimentally measuring absolute efficiency, careful source selection and a long measurement time ensure that $\delta A(t_0)$ is the dominating factor in the error associated with the absolute efficiency and the approximation given by Eq. 5.10 can be made.

$$\frac{\delta\epsilon_{\text{abs}}(E)}{\epsilon_{\text{abs}}(E)} = \frac{\delta A(t_0)}{A(t_0)} \tag{5.10}$$

The absolute efficiency of GEARS was measured using a $^{60}$Co source activity calibrated to 1% ($\delta A(t_0)/A(t_0) = 0.01$). As shown in Fig. 2.3, $^{60}$Co is a two gamma-ray cascade where the 2.505 MeV state in $^{60}$Ni is populated 99.88% of the time following the $\beta$-decay of $^{60}$Co. This makes $^{60}$Co an ideal calibration source since it can be safely assumed that

$$\text{br}(1173.237 \text{ keV}) = \text{br}(1332.492 \text{ keV}) = 1 \tag{5.11}$$

without loss of precision on the absolute efficiency measured at either energy. The calibrated $^{60}$Co point source was placed 10.5 cm above the center of the detector, an identical experimental setup as described in Sec. 5.4.1 and a high-statistics spectrum was analyzed to measure the GEARS efficiency. The background subtracted spectrum used for measuring the absolute efficiency is shown in Fig. 5.2 and the results of the measurement are shown in Table 5.2.

Table 5.2: Measured absolute efficiency using a 1% calibrated $^{60}$Co point source 10.5 cm above the GEARS detector using the spectrum shown in Fig. 5.2. Absolute efficiency, $\epsilon_{\text{abs}}$, is calculated using Eq. 5.9 and the error $\delta\epsilon_{\text{abs}}/\epsilon_{\text{abs}} = 1\%$ is calculated using the approximation shown in Eq. 5.10.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Photopeak counts</th>
<th>$A(t_0)$ [Bq]</th>
<th>$t - t_0$ [d]</th>
<th>$\tau$ [d]</th>
<th>$t_{\text{live}}$ [s]</th>
<th>$\epsilon_{\text{abs}}$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173.237</td>
<td>10391816(4400)</td>
<td>38480(385)</td>
<td>1911</td>
<td>2777.59</td>
<td>239890</td>
<td>0.224(2)</td>
</tr>
<tr>
<td>1332.492</td>
<td>9469637(4107)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.204(2)</td>
</tr>
</tbody>
</table>
Figure 5.2: The $^{60}$Co spectrum from a 1% calibrated source placed 10.5 cm above the center of the detector can which was used for measuring the absolute efficiency of GEARS. The 1173.237 keV photopeak, 1332.492 keV photopeak and 2505.729 keV sum peak are visible, as well as the features described in Sec. 3.3 and Sec. 3.4. Data obtained from this spectrum and the result of the absolute efficiency calibration are given in Table 5.2.

5.4.3 Scaled absolute efficiency curve for GEARS

A weighted first order $\chi^2$ fit to Eq. 5.12 was performed on the relative efficiency data shown in Fig. 5.1 for $E_\gamma \geq 244.692$ keV; a description of the fit formalism is given in Sec. 5.2.2 and Appendix F.

$$\log \epsilon_{\text{rel.}} = k \log E_\gamma + b$$  \hspace{1cm} (5.12)

In Eq. 5.12, $\epsilon_{\text{rel.}}$ is the relative efficiency, $E_\gamma$ is the gamma-ray energy, and $k$ and $b$ parametrize the line. Values of $k$ and $b$ from the fit are shown in Table 5.3. The 1σ (68.27%) confidence interval was calculated for the best fit line and defines the error on the relative efficiency curve, for details, see Appendix H.

The relative efficiency of the 1173.237 keV and 1332.492 keV gamma rays emitted by $^{60}$Co was calculated using Eq. 5.12 with the parameter values given in Table 5.3, the results are shown in Table 5.4. An absolute efficiency curve for GEARS from 244.692 keV to
Table 5.3: Fit parameter values from the weighted first order $\chi^2$ fit given to Eq. 5.12 to the relative efficiency data shown in Fig. 5.1. Errors on the parameter values are given for the $1\sigma$ (68.27%) confidence level.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td>-0.729(6)</td>
</tr>
<tr>
<td>$b$</td>
<td>4.538(19)</td>
</tr>
</tbody>
</table>

Table 5.4: Relative and absolute efficiency of $^{60}$Co. Relative efficiency is calculated using Eq. 5.12 with the parameter values given in Table 5.3. The error of the calculated relative efficiency is for the $1\sigma$ (68.27%) confidence level. Absolute efficiency for the $^{60}$Co peaks at 1173.237 keV and 1332.492 keV is given in Table 5.2 and calculated using Eq. 5.9.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>$\epsilon_{\text{rel.}}$ [arb.]</th>
<th>$\epsilon_{\text{abs.}}$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173.237</td>
<td>199.5(8)</td>
<td>0.224(2)</td>
</tr>
<tr>
<td>1332.492</td>
<td>181.9(8)</td>
<td>0.204(2)</td>
</tr>
</tbody>
</table>

1408.011 keV was constructed using the above data. A multiplicative factor $S$ was derived using a $\chi^2$ fit which scaled the relative efficiency values calculated for $^{60}$Co using Eq. 5.12 to absolute efficiency measured using the calibrated $^{60}$Co source, the data used to calculate the scaling factor are given in Table 5.4, and the $\chi^2$ fit formalism is given in Appendix I. The fit and $1\sigma$ confidence intervals were scaled using the scaling factor $S$ in Eq. 5.13; the absolute efficiency curve of GEARS, fit, and $1\sigma$ confidence interval are shown in Fig. 5.3, and the absolute efficiencies for $^{152}$Eu and $^{60}$Co are shown in Table 5.5.

$$S = \frac{\epsilon_{\text{rel. 1173 keV}}(\epsilon_{\text{abs. 1173 keV}}) + \epsilon_{\text{rel. 1332 keV}}(\epsilon_{\text{abs. 1332 keV}})}{\epsilon_{\text{rel. 1173 keV}}^2 + \epsilon_{\text{rel. 1332 keV}}^2} = 0.00001123(10)$$

### 5.5 Absolute efficiency calibration of GEARS using the sum peak method with $^{60}$Co

The sum peak calibration method is a method in which gamma-gamma coincident events in a single detector can be used to determine the absolute efficiency of this detector independent of the source activity [14, 27]. This method utilizes the fact that the decay of $^{60}$Co to the $^{60}$Ni ground state shown in Fig. 2.3, a two step gamma-ray cascade, is a fast process on the order of $\sim 1$ ps. Due to the short intermediate state lifetime, the detector is unable to
Figure 5.3: Absolute efficiency of GEARS as a function of energy for point-like sources 10.5 cm above the detector can. The scaled \( \chi^2 \) best fit to Eq. 5.12 is shown as a solid line, the 1\( \sigma \) confidence interval is shown as a dashed line. Data shown are \(^{152}\text{Eu}\) [hollow squares], the absolute efficiency measured for \(^{60}\text{Co}\) [filled squares], and calculated absolute efficiency of 0.525(6)\% for the \(^{131}\text{I}\) peak of interest [filled circle]; error bars are smaller than the data points. Data from this figure are shown in Table 5.5.

resolve the two decays if both gamma rays are detected. If both gamma rays are detected, there is a probability that both decays will be detected at full photopeak energy; this results in a sum peak, which has energy \( E_{\text{sum}} = E_1 + E_2 \) where \( E_1 \) and \( E_2 \) are the energies of the first and second gamma decay in the cascade [27]. The probability of detecting the sum peak is a fundamental property of the detector related to the absolute efficiency, and is independent of the source intensity.

5.5.1 Event classification

For a gamma-ray cascade consisting of two decays as in a \(^{60}\text{Co}\) source of intensity \( I \), there are nine ways to classify events based on interactions with the detector. The probability of gamma-ray \( i \) being detected at any energy (that is, detected as a partial energy deposit or the full energy in the photopeak) is defined as \( t_i \) and the probability of gamma-ray \( i \) being
Table 5.5: Absolute efficiency of GEARS for selected $^{152}$Eu and $^{60}$Co decay energies. Errors at one standard deviation are shown in parentheses. The relative efficiency measurement is described in Sec. 5.4.1, the absolute efficiency measurement is described in Sec. 5.4.2 and the scaling factor $S$ for calculating absolute efficiency is given in Eq. 5.13. Data from this table are shown in Fig. 5.3.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy [keV]</th>
<th>$\epsilon_{\text{rel.}}$ [arb.]</th>
<th>$\epsilon_{\text{abs.}}$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{152}$Eu</td>
<td>244.69</td>
<td>628(11)</td>
<td>0.705(14)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>295.94</td>
<td>536(13)</td>
<td>0.602(16)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>344.28</td>
<td>489(3)</td>
<td>0.549(7)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>367.79</td>
<td>483(10)</td>
<td>0.542(12)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>411.12</td>
<td>424(4)</td>
<td>0.476(7)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>443.98</td>
<td>404(6)</td>
<td>0.454(8)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>778.9</td>
<td>266(3)</td>
<td>0.298(5)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>867.39</td>
<td>247(5)</td>
<td>0.277(6)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>964.13</td>
<td>231(3)</td>
<td>0.259(4)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1005.28</td>
<td>230(5)</td>
<td>0.259(7)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1112.12</td>
<td>208(3)</td>
<td>0.234(4)</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1173.237</td>
<td>199.5(8)</td>
<td>0.224(2)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1212.95</td>
<td>195(2)</td>
<td>0.219(4)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1299.12</td>
<td>184(2)</td>
<td>0.207(4)</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1332.492</td>
<td>181.9(8)</td>
<td>0.204(2)</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1408.01</td>
<td>174.7(6)</td>
<td>0.196(2)</td>
</tr>
</tbody>
</table>

detected in the photopeak as $e_i$. For a more detailed discussion of gamma-ray interactions with matter and HPGe detector operational characteristics, refer to Sec. 3.1 and Sec. 3.2. The definitions imply that $e_i \subseteq t_i$. For a detailed list of the event types, see Table 5.6.

### 5.5.2 Sum peak method formalism

Based on the event classification in Sec. 5.5.1, the total number of counts $N_T$ in a spectrum is given in Eq. 5.14 where $I$ is the source intensity and $Q_T$ is the associated $Q$-factor accounting for the effects of angular correlation defined in analogy to Eq. 2.14.

$$N_T = Q_T (N_{\text{Ib}} + N_{\text{IIa}} + N_{\text{IIB}})$$
$$= Q_T I \times [(1 - t_1)t_2 + t_1(1 - t_2) + t_1t_2]$$
$$= Q_T I \times (t_1 + t_2 - t_1t_2)$$

(5.14)

The number of counts in the photopeak for $\gamma_1$, $\gamma_2$, and the sum peak are shown in Eqs. 5.15,
Table 5.6: Event classification for a cascade of two gamma rays. N means the gamma ray is not detected, d means the gamma ray is detected and contains both full energy photopeak deposits and partial energy deposits, and p means the gamma ray is detected in the photopeak. By definition, $Ic$ and $IIc$ are subclasses of $Ib$ and $Ib$ respectively, and class $III$ events are subclasses of the corresponding class $II$ events. Events of class $Ia$ are not observable.

<table>
<thead>
<tr>
<th>$\gamma_1$</th>
<th>$\gamma_2$</th>
<th>Description</th>
<th>Class</th>
</tr>
</thead>
<tbody>
<tr>
<td>N</td>
<td>N</td>
<td>$(1-t_1)(1-t_2)$</td>
<td>$Ia$</td>
</tr>
<tr>
<td>N</td>
<td>d</td>
<td>$(1-t_1)t_2$</td>
<td>$Ib$</td>
</tr>
<tr>
<td>N</td>
<td>p</td>
<td>$(1-t_1)e_2$</td>
<td>$Ic$</td>
</tr>
<tr>
<td>d</td>
<td>N</td>
<td>$t_1(1-t_2)$</td>
<td>$IIa$</td>
</tr>
<tr>
<td>d</td>
<td>d</td>
<td>$t_1t_2$</td>
<td>$IIb$</td>
</tr>
<tr>
<td>d</td>
<td>p</td>
<td>$t_1e_2$</td>
<td>$IIc$</td>
</tr>
<tr>
<td>p</td>
<td>N</td>
<td>$e_1(1-t_2)$</td>
<td>$IIIa$</td>
</tr>
<tr>
<td>p</td>
<td>d</td>
<td>$e_1t_2$</td>
<td>$IIIb$</td>
</tr>
<tr>
<td>p</td>
<td>p</td>
<td>$e_1e_2$</td>
<td>$IIIc$</td>
</tr>
</tbody>
</table>

5.16, and 5.17, respectively:

$$N_1 = Q_{vV}I \times e_1(1 - t_2)$$  \hspace{1cm} (5.15)

$$N_2 = Q_{Vv}I \times e_2(1 - t_1)$$  \hspace{1cm} (5.16)

$$N_s = Q_{vv}I \times e_1e_2$$  \hspace{1cm} (5.17)

Appropriate $Q$-factors with $v$ corresponding to detection of the gamma-ray at full photopeak energy and $V$ corresponding to non-detection are included. It can be seen from the above equations that the $Q$-factors effectively modify the intensity of the source depending on the emission angles of the gamma rays in the cascade which are identified by the detection or non-detection of a gamma ray within the volume of the detector.

This system of four equations $N_T$, $N_1$, $N_2$, $N_s$ has 5 unknowns: $I$, $t_1$, $t_2$, $e_1$, and $e_2$ and is therefore unsolvable. The $Q$-factors are calculable using Geant4 simulations and are not treated as unknowns. Dividing Eq. 5.14, 5.15, and 5.16 by Eq. 5.17 reduces the number of variables to 4 by eliminating $I$, but also reduces the number of equations to 3, yielding
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another unsolvable system, shown below:

\[
N_1/N_s = \eta_1 = \frac{Q_{vV} 1 - t_2}{Q_{vv} e_2}
\]
(5.18)

\[
N_2/N_s = \eta_2 = \frac{Q_{vV} 1 - t_1}{Q_{vv} e_1}
\]
(5.19)

\[
N_T/N_s = \eta_T = \frac{Q_T t_1 + t_2 - t_1 t_2}{Q_{vv} e_1 e_2}
\]
(5.20)

It should be stressed that \(\eta_1\), \(\eta_2\) and \(\eta_T\) are all observables. To make Eqs. 5.18-5.20 a solvable system, we select another observable \(x = e_2/e_1\), the relative efficiency of measuring the full photopeak energy for \(\gamma_2\) with respect to the full photopeak energy for \(\gamma_1\). The value of \(x\) is given by Eq. 5.21

\[
x = \frac{e_2}{e_1} = \frac{181.9(8)}{199.5(8)} = 0.911(6)
\]
(5.21)

where the values of \(e_1\) and \(e_2\) are the calculated relative efficiencies for \(^{60}\)Co from Table 5.4. This substitution eliminates the unknown \(e_2\), the number of unknowns is reduced to 3, and the system given by Eqs. 5.18-5.20 becomes solvable.

After making this substitution, solving Eq. 5.18 and 5.19 for \(t_2\) and \(t_1\) respectively and substituting into Eq. 5.20 yields source activity independent equations for photopeak efficiency given by Eq. 5.22 and 5.23, where \(Q = (Q_T Q_{vv})/(Q_{vV} Q_{Vv})\).

\[
e_1 = \sqrt{\frac{1}{x} \frac{Q_T}{Q_{vv}} \frac{1}{\eta_T + Q_1 \eta_2}}
\]
(5.22)

\[
e_2 = x e_1
\]
(5.23)

5.5.3 Error analysis for the sum peak method

Equation 5.22 is a function of \(x\), \(\eta_T\), \(\eta_1\), \(\eta_2\), \(Q_T\), \(Q_{vv}\), \(Q_{vV}\), and \(Q_{Vv}\). Since \(e_2\) in Eq. 5.23 can be expressed as a function of \(x\) and \(e_1\), evaluating the error on \(x\) and \(e_1\) is sufficient to calculate the errors on \(e_2\). The error on \(e_1\) at one standard deviation, \(\sigma_{e_1}\), is given in Eq. 5.24.

\[
\left(\frac{\sigma_{e_1}}{e_1}\right)^2 = \left(\frac{\partial e_1}{\partial x}\sigma_x\right)^2 + \left(\frac{\partial e_1}{\partial \eta_T}\sigma_{\eta_T}\right)^2 + \left(\frac{\partial e_1}{\partial \eta_1}\sigma_{\eta_1}\right)^2 + \left(\frac{\partial e_1}{\partial \eta_2}\sigma_{\eta_2}\right)^2
\]

+ \left(\frac{\partial e_1}{\partial Q_T}\sigma_{Q_T}\right)^2 + \left(\frac{\partial e_1}{\partial Q_{vv}}\sigma_{Q_{vv}}\right)^2 + \left(\frac{\partial e_1}{\partial Q_{vV}}\sigma_{Q_{vV}}\right)^2 + \left(\frac{\partial e_1}{\partial Q_{Vv}}\sigma_{Q_{Vv}}\right)^2
\]
(5.24)

Each of the partial derivatives in Eq. 5.24 must be calculated, expressions for each are given in Eqs. 5.25-5.32.
\begin{align*}
\left(\frac{\partial e_1}{\partial x}\right)^2 &= \frac{Q_T}{4x^3Q_{vv}(\eta_T + Q\eta_1\eta_2)} \quad (5.25) \\
\left(\frac{\partial e_1}{\partial \eta_T}\right)^2 &= \frac{Q_T}{4xQ_{vv}(\eta_T + Q\eta_1\eta_2)^3} \quad (5.26) \\
\left(\frac{\partial e_1}{\partial \eta_1}\right)^2 &= \frac{\eta_2^2Q^2Q_T}{4xQ_{vv}(\eta_T + Q\eta_1\eta_2)^3} \quad (5.27) \\
\left(\frac{\partial e_1}{\partial \eta_2}\right)^2 &= \frac{\eta_1^2Q^2Q_T}{4xQ_{vv}(\eta_T + Q\eta_1\eta_2)^3} \quad (5.28) \\
\left(\frac{\partial e_1}{\partial Q_T}\right)^2 &= \frac{\eta_1^2Q^3Q^3_{vv}}{4xQ_T(\eta_TQ_{vv}Q_{Vv} + Q_TQ_{vv}\eta_1\eta_2)^2} \quad (5.29) \\
\left(\frac{\partial e_1}{\partial Q_{vv}}\right)^2 &= \frac{Q_TQ_{vv}^2Q_{Vv}\eta_T + Q_TQ_{vv}\eta_1\eta_2}{4xQ_{vv}(\eta_TQ_{vv}Q_{Vv} + Q_TQ_{vv}\eta_1\eta_2)^3} \quad (5.30) \\
\left(\frac{\partial e_1}{\partial Q_{Vv}}\right)^2 &= \frac{Q_TQ_{vv}^2Q_{Vv}}{4xQ_{vv}(\eta_TQ_{vv}Q_{Vv} + Q_TQ_{vv}\eta_1\eta_2)^3} \quad (5.31) \\
\left(\frac{\partial e_1}{\partial Q_vV}\right)^2 &= \frac{Q^2Q_T\eta_1\eta_2}{4xQ_{vv}Q_{Vv}(\eta_T + Q\eta_1\eta_2)^3} \quad (5.32)
\end{align*}

Since the error calculation for the sum peak method depends on the errors for $Q$-factors which must be simulated in Geant4, a validation of the method was performed, and angular distributions were implemented in Geant4.

## 5.6 Sum peak method validation using Geant4

### 5.6.1 Single energy spectra

Geant4 simulations allow for individual components of complex, multi-component gamma-ray spectra to be disentangled. The 1173.237 keV and 1332.492 keV lines of $^{60}$Co were separately simulated with isotropic angular distribution. Sixteen simulations were performed for each energy with $10^7$ events/simulation. The source of radiation was placed 1 mm above the GEARS can in the Geant4 experimental hall. The mean number of counts in the photopeak and standard error on the mean were used to calculate the efficiency.

\[ \epsilon_{\text{sim.}} = \frac{N_{\text{pp}}}{N_{\text{sim.}}} \quad (5.33) \]

Example spectra of the energy deposited in the detector are shown in Fig. 5.4 for the purpose of verifying the method of Sec. 5.5. The FWHM response is set to zero to facilitate
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analysis. The results of the single energy simulations are shown in Table 5.7. Efficiencies for the single decay spectra are calculated using Eq. 5.33. Unlike true $^{60}$Co gamma-ray cascades, the single energy simulated spectra allow the detector efficiency at each decay energy to be calculated without any contributions to the spectrum from summing effects or background radiation.

Table 5.7: Geant4 simulation results for single energy peaks corresponding to $^{60}$Co decay energies. The source was simulated at 1 mm above the detector. $N_{pp}$ is the mean photopeak count value from 16 independent simulations. Error values shown are calculated from the standard error on the mean.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>$N_{pp}$</th>
<th>$N_{\text{sim.}}$</th>
<th>$\epsilon_{\text{sim.}}$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173.237</td>
<td>423630(188)</td>
<td>$10^7$</td>
<td>4.2363(19)</td>
</tr>
<tr>
<td>1332.492</td>
<td>380936(166)</td>
<td>$10^7$</td>
<td>3.8094(17)</td>
</tr>
</tbody>
</table>

5.6.2 Gamma-ray cascades

Geant4 was used to simulate a full $^{60}$Co spectrum following the implementation of gamma-ray cascades. A cascade consists of multiple gamma rays which are emitted in a defined sequence each time an event is generated. In this case, each simulated $^{60}$Co event consists of one decay at 1173.237 keV followed by one decay at 1332.492 keV, as shown in Fig. 2.3. Sixteen independent $^{60}$Co source simulations were conducted with $10^7$ cascades/simulation.

The source of radiation was placed 1 mm above the GEARS can in the Geant4 experimental hall to maximize the number of counts in the sum peak. Placing the source close to the detector tests the limit of the method for calculating efficiencies shown in Eq. 5.33, which does not account for any summing effects. The FWHM response of the detector was again set to zero to facilitate analysis. Both gamma rays in the simulated cascade were emitted isotropically. An example energy spectrum of a $^{60}$Co source simulated with isotropic gamma-ray decay used to validate the sum peak method is shown in Fig. 5.5; the ratios shown in Eqs. 5.18-5.20 were calculated using the mean and standard error measured from the simulated spectra where $Q_T = Q_{vv} = Q_{vV} = Q_{Vv} = 1$ because no angular distribution effects were implemented. Efficiency was calculated using Eq. 5.22 and Eq. 5.23 with the relative efficiency ratio $x$ given by Eq. 5.21 modified for the simulations as shown in Eq. 5.34.

$$x_{\text{sim.}} = \frac{\epsilon_{2 \text{ sim.}}}{\epsilon_{1 \text{ sim.}}} = \frac{3.8094\%}{4.2363\%} = 0.8992(6)$$  
(5.34)
Figure 5.4: Geant4 simulations of $10^7$ events at 1173.237 keV [top] and 1332.492 keV [bottom] with the simulated radioactive source 1 mm above the detector can. The numbers contained in Table 5.7 were calculated from a series of simulations of this type. FWHM = 0 was assumed to facilitate analysis of simulated data. Single energy spectra are useful to disentangle the contributions of each individual component of a composite spectrum. Due to interactions such as Compton scattering and pair production, not all gamma-rays are detected at full photopeak energy. Contributions to gamma-ray energy spectra are described in Sec. 3.3.
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Figure 5.5: An example Geant4 simulation of $10^7$ $^{60}$Co decay events with the simulated radioactive source 1 mm above the detector can. FWHM = 0 was assumed to facilitate analysis of simulated data. The sum peak method was validated by comparing efficiency values calculated using the single energy spectra described in Sec. 5.6.1 with the efficiency values calculated by applying the sum peak method to a simulated gamma-ray cascade. Results of the simulated $^{60}$Co source are presented in Table 5.8.

When incorrectly applied to the $^{60}$Co cascade simulated spectrum, Eq. 5.33 gives efficiency values of 3.3699(17)% and 3.0029(9)% for the 1173.237 keV and 1332.492 keV peaks, respectively. These efficiency values deviate by $\sim 20\%$ from the efficiency values calculated from the single energy spectra using the same equation. Calculating efficiency for the cascade spectrum with the sum peak method using Eq. 5.22 and Eq. 5.23 results in efficiencies which deviate by less than 0.5% from the values calculated from the single energy spectra. The results of the efficiencies calculated from the simulated $^{60}$Co cascade are shown in Table 5.8. The strong agreement between the efficiencies calculated from the single energy spectra and the efficiencies calculated from the cascade using the sum peak method serve as a validation of the sum peak method in the extreme case when summing effects are large and is evidence of the validity of the method.
Table 5.8: Geant4 efficiency calculation results for a $^{60}\text{Co}$ cascade. The source was simulated 1 mm above the detector. $N_{pp}$ is the mean photopeak count value from 16 independent simulations with $N_{sim} = 10^7$ events/simulation, the total number of counts is the integrated area of the entire simulated spectrum. The value of $N_{pp}/N_{sim}.$ is the calculated efficiency defined by Eq. 5.33, the sum peak method efficiencies for 1173.237 keV and 1332.492 keV are calculated using Eq. 5.22 and Eq. 5.23 with the efficiency ratio $x$ from Eq. 5.34, and the single energy efficiencies were calculated using the method defined in Sec. 5.6.1. The large discrepancy between $N_{pp}/N_{sim}.$ and the single energy $\epsilon_{sim}.$ is corrected when summing effects are accounted for as demonstrated by the Sum peak $\epsilon_{sim}.$ Error values given are the standard error on the mean.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>$N_{pp}$</th>
<th>$N_{pp}/N_{sim}.$ [%]</th>
<th>Sum peak $\epsilon_{sim}.$ [%]</th>
<th>Single energy $\epsilon_{sim}.$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173.237</td>
<td>336989(166)</td>
<td>3.3699(17)</td>
<td>4.249(4)</td>
<td>4.2363(19)</td>
</tr>
<tr>
<td>1332.492</td>
<td>300289(92)</td>
<td>3.0029(9)</td>
<td>3.821(4)</td>
<td>3.8094(17)</td>
</tr>
<tr>
<td>2505.729</td>
<td>16195(27)</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>total</td>
<td>3726706(337)</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

5.6.3 $Q_k$ factor definitions for Geant4 simulations

As described in Sec. 5.5.2, the $Q$-factors shown in Eqs. 5.14-5.17 must be calculated using Geant4 simulations with the proper angular distribution. Each $Q$-factor can be written as a sum in analogy to Eq. 2.13, with angular distribution coefficients given by Eqs. A.2-A.4 in Appendix A; these sums are shown in Eqs. 5.35-5.38.

\[
Q_T = A_0 Q_{0T} + A_2 Q_{2T} + A_4 Q_{4T} = Q_{0T} + (1/8)Q_{2T} + (1/24)Q_{4T}
\]  
\[
Q_{vv} = A_0 Q_{0vv} + A_2 Q_{2vv} + A_4 Q_{4vv} = Q_{0vv} + (1/8)Q_{2vv} + (1/24)Q_{4vv}
\]  
\[
Q_{vV} = A_0 Q_{0vV} + A_2 Q_{2vV} + A_4 Q_{4vV} = Q_{0vV} + (1/8)Q_{2vV} + (1/24)Q_{4vV}
\]  
\[
Q_{Vv} = A_0 Q_{0Vv} + A_2 Q_{2Vv} + A_4 Q_{4Vv} = Q_{0Vv} + (1/8)Q_{2Vv} + (1/24)Q_{4Vv}
\]

For the $^{60}\text{Co}$ cascade, equation 5.35 is the $Q$-factor correction for detection of a gamma ray at equal or less than full energy, Eq. 5.36 the $Q$-factor correction for detection of both gamma-rays at full energy, Eq. 5.37 the $Q$-factor correction for detecting the first gamma ray at full energy while the second gamma ray is not detected, and Eq. 5.38 the $Q$-factor correction for the detection of the second gamma ray at full energy while the first gamma ray is not detected. Each component in the sums defined in Eqs. 5.35-5.38 can be expressed
as the ratios shown in Eqs. 5.39-5.42

\[ Q_{kT} = \frac{J_{kT}}{J_{0T}} \]  
\[ Q_{kvv} = \frac{J_{kvv}}{J_{0vv}} \]  
\[ Q_{kvV} = \frac{J_{kvV}}{J_{0vV}} \]  
\[ Q_{kVv} = \frac{J_{kVv}}{J_{0Vv}} \]

where \( k = 0, 2, 4 \), corresponding to gamma-ray decay distributed as \( \cos^k \theta \). The \( J_k \) factors above and also shown in Eq. 2.15 correspond to the detection efficiency for a given energy and angular distribution integrated over the volume of the detector. The angular correlation algorithm is presented in Sec. 5.6.4, and the procedure for extracting the \( J \)-factors from simulated spectra is outlined in Sec. 5.6.5.

### 5.6.4 Angular correlation algorithm

In order to implement the angular correlation corrections described by Eq. 2.13 in Sec. 2.4 in Geant4, a novel algorithm which distributed gamma-ray emission according to \( \cos^k \theta \) \((k = 0, 2, 4)\) in spherical coordinates was created which utilizes the functionality provided by the Geant4 framework. To properly distribute decays according to \( \cos^k \theta \), a random number generator in \( \theta \) must be constructed such that the emission direction is distributed according to \( \cos^k \theta \). The differential solid angle in spherical coordinates must be accounted for which gives gamma-ray angular distribution probability density functions of the form shown in Eq. 5.43

\[ P_k(\theta) = \frac{k+1}{2} \cos^k \theta \sin \theta \]  

for \( k = 0, 2, 4 \). The constant \((k + 1)/2\) is the normalization factor so that

\[ \int_0^\pi P_k(\theta) \, d\theta = \int_0^\pi \frac{k+1}{2} \cos^k \theta \sin \theta \, d\theta = 1 \]

for each \( P_k \).

The random number generator of \( P_k(\theta) \) is constructed from a uniform random number generator \( r \in [0, 1) \) using the cumulative distribution functions (CDF) defined in Eq. 5.45 for \( k = 0, 2, 4 \).

\[ F_k(\theta) = \int_0^\theta P_k(\theta) \, d\theta = \frac{1}{2} \left( 1 - \cos^{k+1} \theta \right) = r \]  

Solving Eq. 5.45 for \( \theta \) gives

\[ \theta = \cos^{-1} \left( 1 - 2r \right)^{\frac{1}{k+1}} \]  

\[ \left( 1 - 2r \right)^{\frac{1}{k+1}} \]
which provides the emission angle $\theta$. This gamma-ray emission direction must be implemented in the Geant4 code.

Note that while $P_k(\theta)$ as defined in Eq. 5.43 is normalized, the corresponding $\cos^k \theta$ terms in the angular distribution function $W(\theta)$ shown in Eq. 2.13 are not. In order to account for the effects of normalization, $W(\theta)$ can be re-written in terms of normalized functions of $\cos^k \theta$ as shown in Eq. 5.47.

$$W(\theta) = \sum_{\text{even } k}^{\text{max } k} \frac{2}{k+1} A_k Q_k \left(\frac{k+1}{2} \cos^k \theta\right)$$

$$= 2A_0Q_0 \left(\frac{1}{2} \cos^0 \theta\right) + \frac{2}{3} A_2Q_2 \left(\frac{3}{2} \cos^2 \theta\right) + \frac{2}{5} A_4Q_4 \left(\frac{5}{2} \cos^4 \theta\right) \quad (5.47)$$

Additionally, Eqs. 5.35–5.38 must be re-written in terms of the coefficients modified to account for the normalization of the distribution functions; the corrected $Q$-factor definitions are given by Eqs. 5.48–5.51.

$$Q_T = 2Q_0T + (2/3)(1/8)Q_{2T} + (2/5)(1/24)Q_{4T} \quad (5.48)$$

$$Q_{vv} = 2Q_{0vv} + (2/3)(1/8)Q_{2vv} + (2/5)(1/24)Q_{4vv} \quad (5.49)$$

$$Q_{vV} = 2Q_{0vV} + (2/3)(1/8)Q_{2vV} + (2/5)(1/24)Q_{4vV} \quad (5.50)$$

$$Q_{Vv} = 2Q_{0Vv} + (2/3)(1/8)Q_{2Vv} + (2/5)(1/24)Q_{4Vv} \quad (5.51)$$

The angular distribution algorithm implemented in Geant4 has the following steps:

1. Initialize a vector $u = (0,0,1)$, the unit vector along the $z$-axis.

2. Initialize a vector $v = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ where $\theta$ is given by Eq. 5.46 for $k = 0, 2, 4$ depending on the distribution. The $\phi$ distribution is random, $\phi = 2\pi \times r'$, $r' \in (0,1)$. The random numbers $r$ and $r'$ are generated independently. The vector $v$ is distributed randomly according to $\cos^k \theta$ with respect to $u$.

3. Initialize a random vector $w$ in Geant4 which will become the direction of emission for the first gamma ray in the cascade.

4. Define a rotation angle and axis for the rotation of $u$ into $w$.

   - The angle $\Theta$ between $u$ and $w$ is calculable from the dot product shown in Eq. 5.52 where $\|u\|$ and $\|w\|$ are the magnitude of $u$ and $w$ respectively. By
construction, \( \mathbf{u} \) and \( \mathbf{w} \) are unit vectors.

\[
\cos \Theta = \frac{\mathbf{u} \cdot \mathbf{w}}{||\mathbf{u}|| ||\mathbf{w}||} = \mathbf{u} \cdot \mathbf{w}
\]  

- Define the rotation axis by the cross product of \( \mathbf{u} \) and \( \mathbf{w} \), which gives a vector \( \mathbf{t} = \mathbf{u} \times \mathbf{w} \) which is perpendicular to both \( \mathbf{u} \) and \( \mathbf{w} \).

5. Rotate \( \mathbf{u} \) into \( \mathbf{w} \).

6. Rotate \( \mathbf{v} \) around \( \mathbf{t} \) by \( \Theta \) into a vector \( \mathbf{v}' \).

7. For a \(^{60}\text{Co}\) cascade, emit the first gamma-ray with energy 1173.237 keV and momentum direction defined by \( \mathbf{w} \), and the second gamma ray with energy 1332.492 keV and momentum direction defined by \( \mathbf{v}' \).

This algorithm produces gamma rays with the distributions defined by Eq. 5.43. The results for the three correlation simulations is shown in Figs. 5.6-5.8. Although the gamma-ray emission directions are correlated in a single cascade, if each decay is observed independently, the directions should appear random. The emission angles in \( \theta \) and \( \phi \) fully define

Figure 5.6: The angle between two gamma-rays emitted in \( 10^4 \) \(^{60}\text{Co}\) decays simulated in Geant4. No angular correlation was implemented in this run. The fit is to \( f(\theta) = A\sin \theta \) where \( A \) is a free parameter.
Figure 5.7: The angle between two gamma-rays emitted in $10^4 \ ^{60}\text{Co}$ decays simulated in Geant4. Angular correlation according to $\cos^2 \theta$ was implemented. The fit is to $f(\theta) = A \cos^2 \theta \sin \theta$ where $A$ is a free parameter.

Figure 5.8: The angle between two gamma-rays emitted in $10^4 \ ^{60}\text{Co}$ decays simulated in Geant4. Angular correlation according to $\cos^4 \theta$ was implemented. The fit is to $f(\theta) = A \cos^4 \theta \sin \theta$ where $A$ is a free parameter.
the direction of the emitted gamma ray. The $\theta$ and $\phi$ distributions for the first and second gamma ray of the simulated $^{60}$Co cascade, as well as a 2D projection of $\theta$ and $\phi$ are shown for the $\cos^2 \theta$ and $\cos^4 \theta$ cases in Fig. 5.9 and Fig. 5.10, which confirm that each gamma-ray emission is random in space when viewed independently. Coupled with the correlated emission direction in a single cascade shown in Figs. 5.6-5.8, the independently random distribution is strong evidence that the angular correlation routine is properly implemented in Geant4.

Figure 5.9: Gamma-ray emission direction information for [left] the isotropically distributed first and [right] according to $\cos^2 \theta$ distributed second gamma rays in a simulated $^{60}$Co cascade of $10^6$ events. Top: a 2D projection of the emission angles $\theta$ and $\phi$; middle: distribution of $\cos \theta$; bottom: distribution of $\phi$. When the emissions are observed independently, the gamma-ray emission angles are randomly distributed.

5.6.5 Extracting $J$-factors from simulated spectra

In order to calculate the $Q$-factors defined in Eqs. 5.48–5.51, the associated $J$-factors must be extracted from simulated spectra. The following procedure describes the steps necessary
Figure 5.10: Gamma-ray emission direction information for [left] the isotropically distributed first and [right] according to $\cos^4 \theta$ distributed second gamma rays in a simulated $^{60}$Co cascade of $10^6$ events. Top: a 2D projection of the emission angles $\theta$ and $\phi$; middle: distribution of $\cos \theta$; bottom: distribution of $\phi$. When the emissions are observed independently, the gamma-ray emission angles are randomly distributed.

to calculate $Q_{vv}$, the $Q$-factor associated with the sum peak in the $^{60}$Co cascade. The procedure for calculating $Q_T$, $Q_{vV}$, and $Q_{VV}$ are identical for the proper choice of $Q$-factor component equations shown in Eqs. 5.39–5.42.

1. Initialize the source properties in Geant4. For a simulated $^{60}$Co cascade, the first gamma ray has $E_\gamma = 1173.237$ keV, and the second gamma ray has $E_\gamma = 1332.492$ keV.

2. Set the source location in the Geant4 experimental hall.

3. Repeat steps (a) and (b) $N$ times:

   (a) For each of the $k = 0, 2, 4$ cases, simulate one $^{60}$Co cascade of $10^7$ events where the 1173.237 keV gamma ray is distributed isotropically and the 1332.492 keV
gamma ray is distributed according to $\cos^k \theta$.

(b) Determine the number of counts in the peak of interest in each spectrum using gf3. For the calculation of $Q_{vv}$, the peak of interest is the sum peak.

4. Calculate the mean $\mu_k$ using Eq. 5.53 and standard error on the mean $\sigma_{\mu_k}$ using Eq. 5.54 from the data obtained in the previous step for $k = 0, 2, 4$

$$\mu_k = \frac{1}{N} \sum_{i=1}^{N} c_{ki}$$

$$\sigma_{\mu_k} = \sqrt{\frac{1}{N(N-1)} \sum_{i=1}^{N} (c_{ki} - \mu_k)^2}$$

where $c_{ki}$ is the number of sum peak counts in the $i^{th}$ simulation.

5. Define $J_{kvv} = \mu_k$ and $\sigma_{J_{kvv}} = \sigma_{\mu_k}$ for $k = 0, 2, 4$.

6. Using the $J$-factors from the previous step, calculate the component $Q$-factors $Q_{0vv}$, $Q_{2vv}$, and $Q_{4vv}$ using Eq. 5.40 and the associated standard deviations $\sigma_{Q_{0vv}}$, $\sigma_{Q_{2vv}}$, and $\sigma_{Q_{4vv}}$ using the standard error propagation formula; $Q_{0vv} = 1$ and $\sigma_{Q_{0vv}} = 0$ by construction.

7. Using the component $Q$-factors and associated standard deviations from the previous step, calculate $Q_{vv}$ using Eq. 5.49 and the associated standard deviation $\sigma_{Q_{vv}}$ using the standard error propagation formula.

The $Q$-factors used to perform the sum peak corrections in this work were calculated using $N = 16$ simulations in order to ensure that $\sigma_{Q_{vv}}/Q_{vv} < 1\%$. The results of the simulations, $J$-factors extracted by the above procedure, and the calculated $Q$-factors are summarized in Appendix J.

5.7 Absolute efficiency measurement results

Following the validation and simulation of the $Q$-factors in Geant4, the absolute efficiency of GEARS was calculated using the formalism derived for the sum peak method shown in Eq. 5.22 and Eq. 5.23. Error analysis for the efficiency of the 1173.237 keV peak was performed using Eq. 5.24. The efficiency of the 1332.492 keV peak was calculated using
Eq. 5.23 and the error was calculated using standard error propagation techniques. The sum peak method efficiency calculation was completed using the experimental spectrum used for the 1% source calibration shown in Fig. 5.2.

Table 5.9: Comparison between absolute efficiency for the 1173.237 keV and 1332.492 keV gamma rays associated with $^{60}$Co decay measured with a source activity calibrated to 1% and calculated using the sum peak method with the source secured 10.5 cm above the detector can. Error values at one standard deviation are shown in parentheses. Efficiencies for both methods were obtained from the experimental spectrum shown in Fig. 5.2.

<table>
<thead>
<tr>
<th>Method</th>
<th>$\epsilon_{1173}$ keV</th>
<th>$\epsilon_{1332}$ keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Co source calibrated to 1%</td>
<td>0.224(2)%</td>
<td>0.204(2)%</td>
</tr>
<tr>
<td>Sum peak method</td>
<td>0.233(2)%</td>
<td>0.213(4)%</td>
</tr>
</tbody>
</table>

A comparison between the two absolute efficiency calibration methods described in Sec. 5.4 and Sec. 5.5 is shown in Table 5.9. There is an $\sim$ 5% discrepancy between the two methods which is an $\sim$ 3σ effect. This discrepancy does not have a large impact on the presented results since it is insignificant when compared to statistical errors on the measurements. It should be noted that effects of radioactive decay signal processing by the electronics of GEARS are not included in the Geant4 simulations used to calculate the $Q$-factors. These effects are present in the experimental spectrum shown in Fig. 5.2 which was used to calculate the sum peak method absolute efficiency. Further work should add the electronic effects into the existing Geant4 simulation framework. The 1% source measurement method was chosen as the calibration standard in these studies; errors in the included tables are statistical errors at one standard deviation derived from this source, though a systematic error of 5% estimated on the basis of the sum peak method result cannot be excluded.
Chapter 6

Extended source efficiency correction

The source-based efficiency calibration described in Sec. 5.4 and accepted based on the results presented in Sec. 5.7 was completed using standard point sources, whereas rainwater and seaweed samples were measured in 24 mL, 60 mL, and 120 mL glass vials. Altering the source geometry and composition has a large effect on the efficiency of gamma ray detection; this effect must be quantified for proper data analysis. The corrected efficiency for extended sources can be calculated using Eq. 6.1, where the correction factor $C_s$ which accounts for the change in detection efficiency is source dependent.

\[ \epsilon_{\text{extended}} = \epsilon_{\text{point}} \times C_s \]  

Extended radioactive sources have been implemented in Geant4 to simulate the available sample configurations. Available configurations include 24 mL, 60 mL, and 120 mL vials. Gamma ray self absorption in extended source samples can be simulated in Geant4 provided that the elemental composition and density of the samples are known. Simulations implementing gamma ray self absorption were used to correct for extended source efficiency.

The validation of Geant4 simulation results has been attempted in a series of dilution experiments using radioactive salts. First a sample of purified UO$_2$Cl$_2$ crystals, a water soluble uranium salt containing depleted and physically separated $^{238}$U obtained from the SFU Department of Chemistry was used to construct a point-like source. The point-like source was measured in two positions: 1) placed on the detector can and 2) at 10.5 cm above the detector. Extended sources with identical geometry to those used in the measurements
described in Chapter 7 were constructed by dissolving the UO$_2$Cl$_2$ point source sample to prepare a series of dilutions which served as standard calibration sources with constant activity distributed in different volumes. The UO$_2$Cl$_2$ from the point source was transferred to a 24 mL glass vial, dissolved completely in distilled water, and measured on GEARS. Subsequent serial dilution measurements were performed using 60 mL and 120 mL vials; during each transfer, the smaller vial was thoroughly rinsed into the larger volume with distilled water.

The $^{238}$U decay chain up to $^{234}$U is in secular equilibrium (defined as the parent and daughter nuclei possessing equal activities) due to the use of depleted UO$_2$Cl$_2$ starting material; thus, the number of excited state $^{234}$U atoms populated following the $\beta$-decay of $^{234}$Pa which decay to the ground state by gamma-ray emission is constant with respect to the measurement duration. To analyze the GEARS efficiency for each extended source, the 258.227, 945.94, 1001.03, 1193.73, and 1737.75 keV [2] gamma-ray decays of excited states in $^{234}$U were measured after each dilution. Following each measurement, the activity ratio with respect to the point source was calculated over this range of energies. Two independent measurements were completed.

The results of the two measurements and Geant4 simulations for the $^{234}$U decay energies for the 24 mL, 60 mL and 120 mL vials are shown in Figs. 6.1, 6.2, and 6.3. The two measurements in the 24 mL vial were consistent. Geant4 simulations of this extended source were completed, and found to be consistent with the experimental values. The measurements in the 120 mL vial were also consistent with one another. However, the experimental results were $\sim 50\%$ below the ratio calculated using Geant4 simulations. This disagreement could arise from an inhomogeneous distribution of UO$_2$Cl$_2$ in the vial due to the formation of so-called radiocolloids or adsorption on the glass walls, phenomena which have been previously observed in dilute solutions of radioactive salts [28]; the effect of adsorption on the GEARS efficiency would have been difficult to spot without the aid of Geant4 simulations for extended sources.

The 24 mL and 60 mL vials had the same height but different radii, while the height of the 120 mL glass vial was twice that of the 60 mL and 24 mL vials. The impact of the height on the solid angle (and therefore detection efficiency) would be magnified by the distribution of the uranium adsorbed on the glass. The adsorption of uranium on the glass walls would explain why a large deviation from the Geant4 simulations was observed for this volume only. A control experiment to test the adsorption of UO$_2$Cl$_2$ on the walls of the glass vials
was completed and non-reversible adsorption on glass was observed following the rinsing procedure used to construct the extended sources for calibration. Methods of constructing solid sources of uniformly distributed UO$_2$Cl$_2$ should be investigated and pursued in the future.

![Figure 6.1: Extended source/point source efficiency ratio at prominent gamma-ray energies of $^{234}$U. The results from two independent experiments (a) and (b) are shown with Geant4 simulations for the serial dilution measurements. Results shown are for the 24 mL vial, the initial step of the serial dilution process.](image-url)
Figure 6.2: Extended source/point source efficiency ratio at prominent gamma-ray energies of $^{234}$U. The results from two independent experiments (a) and (b) are shown with Geant4 simulations for the serial dilution measurements. Results shown are for the 60 mL vial, the second step of the serial dilution process.
Figure 6.3: Extended source/point source efficiency ratio at prominent gamma-ray energies of $^{234}$U. The results from two independent experiments (a) and (b) are shown with Geant4 simulations for the serial dilution measurements. Results shown are for the 120 mL vial, the final step of the serial dilution process. Experimental results are consistent but show a $\sim 50\%$ discrepancy from Geant4 simulations attributed to adsorption of $\text{UO}_2\text{Cl}_2$ on the walls of the vial.
Chapter 7

Iodine-131 monitoring results

7.1 Sampling procedure

7.1.1 Rainwater

Rainwater samples were collected at the SFU campus approximately once per day between March 16 and April 14, 2011. Following collection, the rainwater was filtered using 110 mm diameter Whatman filter paper to remove particulates and transferred to 24 mL and 120 mL glass vials. The filtering process removed particulates which could represent radioactive “hot spots” which can settle on the bottom of the glass vials and skew the results due to a change in detection efficiency. The filter paper was discarded, though in future operation, measuring the filter paper using GEARS is highly recommended. The vials were placed on the GEARS detector and measured. The 364.489 keV line indicative of $^{131}$I decay was selected for quantitative analysis. Rainwater collection began on March 16, 2011 and continued through April 14, 2011.

7.1.2 Seaweed

*Fucus* samples were gathered primarily from the Bamfield Marine Sciences Centre (BMSC) on Vancouver Island and from the Burrard Inlet in North Vancouver. Seaweed samples were collected in North Vancouver between March 15 and May 10, 2011; samples from BMSC were collected between March 22 and May 10, 2011. The sample collection site from North Vancouver was located near a storm drain, potentially exposing the seaweed to $^{131}$I contained in sewage effluent and runoff. Once obtained, samples were rinsed with tap...
water to remove sand, rocks, aquatic animals and epiphytes from the surface of the seaweed, and air dried until most of the rinsing water had evaporated (∼4 hours). The air-dried seaweed was placed in a commercially available food dehydrator and dried overnight at a temperature of ∼70 °C. Samples were ground into a powder using a mortar and pestle and transferred to 24 mL, 60 mL, or 120 mL glass vials and weighed. Iodine is concentrated in the blade of the seaweed, so portions of the stipe which were difficult to grind could be safely discarded [29]. Seaweed collection began in North Vancouver on March 15, 2011 and at BMSC on March 22.

7.2 Confirmation of $^{131}$I contamination in environmental samples

The presence of $^{131}$I was confirmed following the observation of the characteristic 364.489 keV and 284.3 keV gamma rays as shown in Fig. 2.1 in both rainwater and seaweed samples, as well as a lifetime measurement of the 364.489 keV gamma-ray line in a single seaweed sample as shown in Fig. 7.1. To perform the lifetime fit, activity ratios in a single sample were measured as a function of time. The sample used to generate these data was collected on March 28, 2011 in North Vancouver and initially measured on March 31, 2011. The same sample was re-measured on April 9, 2011 and May 4, 2011. The data was fit with the exponential function given in Eq. 7.1.

$$\frac{A(t)}{A(t_0)} = e^{-(t-t_0)/\tau}$$

where $A(t)$ is the activity at time $t$, $A(t_0)$ is the activity measured on $t_0 = March 31, 2011$, $t$ is the time since $t_0$ in days, and $\tau$ is the best fit lifetime in days. A re-measurement of the sample was performed on November 9, 2012 and no $^{131}$I was observed. The decrease to zero $^{131}$I activity justifies the exclusion of a constant in the exponential fit. The best fit lifetime was $\tau = 11.2(1.1)$ days, in agreement with the literature value of 11.5779(7) days [2].

Activity concentrations of $^{131}$I were obtained from integration of the 364.489 keV gamma-ray line. The activity concentration in rainwater and seaweed samples at the collection time was corrected for measurement delay, $^{131}$I half-life, the efficiency of the detector, and the known branching ratio (0.815) of the 364.489 keV gamma ray [30].
CHAPTER 7. IODINE-131 MONITORING RESULTS

Figure 7.1: Activity in North Vancouver seaweed of the 364.489 keV gamma-ray line relative to the original measurement as a function of time. The data were fit using Eq. 7.1. The best fit lifetime is $\tau = 11.2(1.1)$ days, in agreement with the literature value of 11.5779(7) days [2].

7.3 Determination of $^{131}$I detection efficiency

The $^{131}$I detection efficiency was determined for a point source of $^{131}$I at 10.5 cm for the $E_\gamma = 364.489$ keV peak of interest. This was achieved based on the relative efficiency of $^{131}$I detection determined using Eq. 5.12 with the coefficients from Table 5.3 and scaled to the accepted absolute efficiency measured using the 1% $^{60}$Co source shown in Table 5.9 using Eq. 5.13. The measured absolute efficiency for the detection of a 364.489 keV gamma-ray point source at 10.5 cm was found to be 0.525(6)%.

The absolute efficiency of $^{131}$I detection was calculated assuming a point source of $^{131}$I at 10.5 cm. During the Fukushima monitoring campaign, extended sources of various geometries, fill levels, and elemental compositions (corresponding to water and seaweed) were used. This necessitated expanding the extended source efficiency corrections described in Chapter 6. Only the 24 mL and 120 mL glass vials were used during the rainwater monitoring campaign; each of these was filled to capacity and a single correction factor for each volume was calculated in Geant4 where the volume of each vial was constructed of water.
Seaweed samples were measured in all three vials filled to various degrees and Geant4 simulations were conducted to assess how detection efficiency varied with vial fill level in order to properly correct the seaweed data. The density of processed seaweed was measured from four samples and found to be 0.86(10) g/cm$^3$; the exact elemental composition of the seaweed samples was not known and assumed to be 100% carbon for the purpose of the Geant4 self absorption simulations. The simple elemental approximation of seaweed as 100% carbon does not have a significant impact on the gamma-ray self absorption, as the major components of seaweed (carbon, nitrogen, and oxygen) have nearly identical attenuation coefficients for photons, as shown in Tab. 7.1 [2].

Table 7.1: X-ray mass attenuation coefficients for carbon, nitrogen, and oxygen at 400 keV from Ref. [31]. The small differences in attenuation due to the presence of nitrogen and oxygen in seaweed are negligible when calculating self absorption of $^{131}$I gamma rays in seaweed samples approximated as 100% carbon.

<table>
<thead>
<tr>
<th></th>
<th>Carbon</th>
<th>Nitrogen</th>
<th>Oxygen</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.09546 cm$^2$/g</td>
<td>0.09557 cm$^2$/g</td>
<td>0.09566 cm$^2$/g</td>
</tr>
</tbody>
</table>

Simulations with $E_\gamma = 364.489$ keV were performed with vials 1/4 full, 1/2 full, 3/4 full and filled to capacity and the ratio of the extended source efficiency to the measured point source efficiency of 0.525(6)% was calculated at each fill level. A least squares fit with a second order polynomial was performed on these data. Efficiency corrections for partially filled vials were made using the correction factor obtained from the fit and Eq. 6.1. These data and the accompanying fits are shown in Fig. 7.2. The calculated efficiency correction factors were used to determine the activity of $^{131}$I in measured samples.

7.4 **Iodine-131 monitoring results**

7.4.1 **Rainwater**

Iodine-131 was first observed in rainwater on March 19, 2011, eight days after the tsunami damaged the reactors at the Fukushima Dai-ichi plant on March 11. Maximum activity was observed on March 20, nine days after the tsunami. The activity of $^{131}$I in rainwater was below the detection limit of GEARs by early April, 2011 and was no longer monitored once the 364.489 keV gamma ray was consistently not observed. Several gamma-ray spectra illustrating the change in activity measured during the rainwater monitoring campaign are
CHAPTER 7. IODINE-131 MONITORING RESULTS

Figure 7.2: Extended source efficiency correction curves for the 24 mL [black], 60 mL [red] and 120 mL [blue] vials; extended source efficiency is defined in Sec. 6 and calculated using Eq. 6.1. Data points are Geant4 simulated extended/point source ratios corresponding to the vial being 1/4 full, 1/2 full, 3/4 full and filled to capacity; error bars are smaller than the data points. The curves shown are second order polynomial fits to the ratio of the extended source efficiency to point source efficiency at 10.5 cm with a vial fill level of x cm.

shown in Fig. 7.3, the extended source efficiency corrected data collected during the rainwater monitoring campaign are contained in Table 7.2, and a time profile of $^{131}$I concentration in Burnaby Mountain rainwater is shown in Fig. 7.4.

7.4.2 Seaweed

The maximum measured $^{131}$I activity concentration occurred on March 22 for local seaweed and on March 28 for seaweed from BMSC, 11 and 17 days after the earthquake, respectively. Iodine-131 concentration in seaweed demonstrates a significant positive correlation with precipitation, and $^{131}$I levels in Fucus have been shown to reflect observed levels in rain [3]. Seawater samples in Korea collected at the surface and at depths up to 2000 m did not contain measurable amounts of $^{131}$I [9], which indicates that rainwater is the primary source of $^{131}$I uptake in Fucus. By mid-May, one month after $^{131}$I was last observed in rainwater,
Figure 7.3: Three gamma-ray spectra measured by GEARS during the rainwater monitoring campaign. The 364.489 keV peak is characteristic of $^{131}$I and indicates its presence in the samples collected at SFU. The 295.224 keV and 351.9 keV peaks are natural background radiation from the $^{238}$U decay chain. Damage to the Fukushima Dai-ichi plant occurred on March 11. No $^{131}$I was observed in the sample collected on March 18, while the spectrum from the sample collected on March 20 contained the highest measured $^{131}$I activity. Rainwater collection was halted shortly after March 30, when $^{131}$I content dropped below the detection limit.

The $^{131}$I content in the seaweed samples had decreased to below 5 Bq/kg dry weight, therefore the seaweed monitoring campaign was halted.

Gamma-ray spectra showing the change in $^{131}$I content measured in North Vancouver seaweed are shown in Fig. 7.5. The extended source efficiency corrected data collected from North Vancouver seaweed over the course of the monitoring campaign are shown in Tabs. 7.3 and 7.4; the corresponding data from the BMSC are shown in Tabs. 7.5 and 7.6. All activity concentrations and mass measurements for seaweed samples are reported for dry weight. Time profiles of $^{131}$I activity concentration in North Vancouver and BMSC seaweed samples are shown in Fig. 7.6. The variation observed in the maximum activity and time profiles of the BMSC and North Vancouver seaweed samples is largely due to differences in the process of $^{131}$I uptake which cannot be controlled. Such processes are dependent on...
Figure 7.4: Time profile of $^{131}$I activity concentration in rainwater collected at the SFU campus on Burnaby Mountain following the Fukushima accident. The maximum activity concentration of $^{131}$I was observed on March 20, nine days after the tsunami. By April, no more $^{131}$I was present in collected rainwater, indicating no continuous releases from the Fukushima reactors. Gaps in the collection correspond to days when there was no rainfall.

the weather patterns at each sample location, the age of each seaweed sample collected, the specific location of the collection sites at the sampling location, and the genetic diversity of each sampling population.

Two samples of *Macrocystis pyrifera* (giant kelp), two samples of *Fucus*, and one sample of *Pyropia fallax*, formerly known as *Porphyra fallax* [32] collected in Bella Bella, BC were received and processed following the procedure outlined above, the data are shown in Table 7.7.
Figure 7.5: Three gamma-ray spectra from North Vancouver *Fucus* measured by GEARS during the seaweed monitoring campaign. The 365 keV and 284.305 keV peaks are characteristic of $^{131}$I and indicate its presence in the seaweed collected in North Vancouver. The 295.224 keV and 351.9 keV peaks are natural background radiation from the $^{238}$U decay chain. No $^{131}$I was observed in the sample collected on March 15, while the spectrum from the sample collected on March 28 exhibits two gamma-ray peaks characteristic of $^{131}$I loading. There is a marked decrease in $^{131}$I activity by mid April, and *Fucus* samples from North Vancouver were not collected past May 10 due to negligible $^{131}$I content.
Table 7.2: Activity concentration of $^{131}$I in rainwater collected at the SFU campus on Burnaby Mountain following the Fukushima accident on March 11, 2011. Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were determined by Geant4 simulation. The measurement live time was $\geq 99\%$ of the measurement duration. These data are shown in Fig. 7.4.

<table>
<thead>
<tr>
<th>Collection date</th>
<th>Measurement live time [s]</th>
<th>Sample volume [L]</th>
<th>Counts in 364.489 keV $^{131}$I photopeak</th>
<th>Efficiency correction factor</th>
<th>Activity concentration [Bq/L]</th>
</tr>
</thead>
<tbody>
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<td>16 Mar</td>
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<td>0.023(1)</td>
<td>0(34)</td>
<td>5.89(2)</td>
<td>0.0(8)</td>
</tr>
<tr>
<td>18 Mar</td>
<td>100945</td>
<td>0.023(1)</td>
<td>0(35)</td>
<td>5.89(2)</td>
<td>0.0(6)</td>
</tr>
<tr>
<td>19 Mar</td>
<td>151045</td>
<td>0.023(1)</td>
<td>349(34)</td>
<td>5.89(2)</td>
<td>4.1(4)</td>
</tr>
<tr>
<td>20 Mar</td>
<td>85465</td>
<td>0.023(1)</td>
<td>262(28)</td>
<td>5.89(2)</td>
<td>5.8(7)</td>
</tr>
<tr>
<td>25 Mar</td>
<td>92145</td>
<td>0.023(1)</td>
<td>242(29)</td>
<td>5.89(2)</td>
<td>4.9(6)</td>
</tr>
<tr>
<td>27 Mar</td>
<td>72109</td>
<td>0.023(1)</td>
<td>102(21)</td>
<td>5.89(2)</td>
<td>2.4(5)</td>
</tr>
<tr>
<td>28 Mar</td>
<td>70045</td>
<td>0.023(1)</td>
<td>77(19)</td>
<td>5.89(2)</td>
<td>2.0(5)</td>
</tr>
<tr>
<td>29 Mar</td>
<td>88231</td>
<td>0.023(1)</td>
<td>72(20)</td>
<td>5.89(2)</td>
<td>1.5(4)</td>
</tr>
<tr>
<td>30 Mar</td>
<td>80068</td>
<td>0.023(1)</td>
<td>31(19)</td>
<td>5.89(2)</td>
<td>0.7(4)</td>
</tr>
<tr>
<td>31 Mar</td>
<td>65534</td>
<td>0.119(1)</td>
<td>28(15)</td>
<td>3.242(12)</td>
<td>0.26(14)</td>
</tr>
<tr>
<td>01 Apr</td>
<td>162184</td>
<td>0.119(1)</td>
<td>162(29)</td>
<td>3.242(12)</td>
<td>0.65(12)</td>
</tr>
<tr>
<td>04 Apr</td>
<td>69455</td>
<td>0.119(1)</td>
<td>0(31)</td>
<td>3.242(12)</td>
<td>0.0(3)</td>
</tr>
<tr>
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<td>36567</td>
<td>0.119(1)</td>
<td>0(23)</td>
<td>3.242(12)</td>
<td>0.0(4)</td>
</tr>
<tr>
<td>07 Apr</td>
<td>78797</td>
<td>0.119(1)</td>
<td>74(31)</td>
<td>3.242(12)</td>
<td>0.6(3)</td>
</tr>
<tr>
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<td>65568</td>
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<td>0(31)</td>
<td>3.242(12)</td>
<td>0.0(3)</td>
</tr>
<tr>
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<td>0.119(1)</td>
<td>0(37)</td>
<td>3.242(12)</td>
<td>0.0(4)</td>
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Table 7.3: Activity concentration of $^{131}$I in Bq/kg dry weight from Fucus collected in North Vancouver following the Fukushima accident on March 11, 2011, part (a). Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were calculated from the appropriate best fit curves shown in Figs. 7.2. The $^\dagger$ denotes a single large sample which was divided into three samples measured at different times. The measurement live time was ≥99% of the measurement duration.

<table>
<thead>
<tr>
<th>Collection date</th>
<th>Collection location</th>
<th>Sample mass [g]</th>
<th>Counts in $^{131}$I photopeak [s]</th>
<th>Efficiency correction factor</th>
<th>Activity concentration in Bq/kg</th>
<th>Activity concentration error (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15 Mar</td>
<td>49.3124 N 123.0872 W</td>
<td>3.5693</td>
<td>89989</td>
<td>0.44</td>
<td>11.39(3)</td>
<td>130(7)</td>
</tr>
<tr>
<td>22 Mar</td>
<td>49.3124 N 123.0872 W</td>
<td>3.6629</td>
<td>702.0872 W 66619</td>
<td>0.44</td>
<td>7.02(3)</td>
<td>130(7)</td>
</tr>
<tr>
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<td>8.87(3)</td>
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<td>13.6534</td>
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<td>10.50(3)</td>
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<td>05 Apr†</td>
<td>49.3124 N 123.0872 W</td>
<td>10.9545</td>
<td>1092.0872 W 8844</td>
<td>0.44</td>
<td>6.032(17)</td>
<td>56(3)</td>
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</tbody>
</table>
Table 7.4: Activity concentration of $^{131}$I in Bq/kg dry weight from *Fucus* collected in North Vancouver following the Fukushima accident on March 11, 2011, part (b). Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were calculated from the appropriate best fit curves shown in Figs. 7.2. These data are shown in Fig. 7.6. The measurement live time was $\geq 99\%$ of the measurement duration.

<table>
<thead>
<tr>
<th>Collection date</th>
<th>Collection location</th>
<th>Measurement live time [s]</th>
<th>Sample mass [g]</th>
<th>Counts in $^{131}$I photopeak</th>
<th>Efficiency correction factor</th>
<th>Activity concentration [Bq/kg]</th>
</tr>
</thead>
<tbody>
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<td>49.3124 N 123.0872 W</td>
<td>20926</td>
<td>45.9654</td>
<td>487(35)</td>
<td>5.203(19)</td>
<td>51(4)</td>
</tr>
<tr>
<td>11 Apr</td>
<td>49.3122 N 123.0872 W</td>
<td>20803</td>
<td>71.8451</td>
<td>607(38)</td>
<td>4.803(14)</td>
<td>35(2)</td>
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<tr>
<td>11 Apr</td>
<td>49.3115 N 123.0864 W</td>
<td>69327</td>
<td>31.1776</td>
<td>1238(57)</td>
<td>8.143(19)</td>
<td>27.4(1.1)</td>
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<tr>
<td>11 Apr</td>
<td>49.3111 N 123.0856 W</td>
<td>82553</td>
<td>47.9947</td>
<td>1643(65)</td>
<td>5.203(19)</td>
<td>28.6(1.1)</td>
</tr>
<tr>
<td>17 Apr</td>
<td>49.3124 N 123.0872 W</td>
<td>101097</td>
<td>22.7575</td>
<td>1542(65)</td>
<td>8.97(2)</td>
<td>26.9(1.1)</td>
</tr>
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<td>17 Apr</td>
<td>49.3124 N 123.0872 W</td>
<td>102893</td>
<td>22.6537</td>
<td>1170(59)</td>
<td>9.52(2)</td>
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<tr>
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<td>49.3124 N 123.0872 W</td>
<td>63226</td>
<td>38.8693</td>
<td>550(42)</td>
<td>7.167(17)</td>
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<td>76177</td>
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<td>168885</td>
<td>74.2620</td>
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<td>8.01(2)</td>
<td>2.14(16)</td>
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</table>
Table 7.5: Activity concentration of $^{131}$I in Bq/kg dry weight from *Fucus* collected at BMSC on Vancouver Island, part (a). Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were calculated from the appropriate best fit curves shown in Fig. 7.2. The measurement live time was $\geq 99\%$ of the measurement duration. These data are shown in Fig. 7.6.

<table>
<thead>
<tr>
<th>Collection date</th>
<th>Collection location</th>
<th>Measurement live time [s]</th>
<th>Sample mass [g]</th>
<th>Counts in $^{131}$I photopeak</th>
<th>Efficiency correction factor</th>
<th>Activity concentration [Bq/kg]</th>
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</thead>
<tbody>
<tr>
<td>22 Mar</td>
<td>48.83 N 124.15 W</td>
<td>91250</td>
<td>9.3063</td>
<td>639(47)</td>
<td>8.90(2)</td>
<td>29(2)</td>
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<tr>
<td>25 Mar</td>
<td>48.8370 N 125.1433 W</td>
<td>73278</td>
<td>19.4958</td>
<td>1595(61)</td>
<td>10.71(3)</td>
<td>44.9(1.7)</td>
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<tr>
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<td>48.8370 N 125.1433 W</td>
<td>16055</td>
<td>11.2668</td>
<td>354(30)</td>
<td>9.45(2)</td>
<td>67(6)</td>
</tr>
<tr>
<td>30 Mar</td>
<td>48.8356 N 125.1366 W</td>
<td>11495</td>
<td>10.1661</td>
<td>115(19)</td>
<td>9.85(2)</td>
<td>50(8)</td>
</tr>
<tr>
<td>01 Apr</td>
<td>48.8356 N 125.1366 W</td>
<td>2942</td>
<td>40.2153</td>
<td>96(15)</td>
<td>6.429(19)</td>
<td>55(9)</td>
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<td>-</td>
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<td>218(33)</td>
<td>11.88(4)</td>
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<tr>
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<td>87450</td>
<td>60.3971</td>
<td>1463(63)</td>
<td>5.203(19)</td>
<td>36.0(1.5)</td>
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<tr>
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<td>14942</td>
<td>51.9189</td>
<td>219(25)</td>
<td>5.203(19)</td>
<td>36(4)</td>
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Table 7.6: Activity concentration of $^{131}$I in Bq/kg dry weight from *Fucus* collected at BMSC on Vancouver Island, part (b). Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were calculated from the appropriate best fit curves shown in Fig. 7.2. The measurement live time was $\geq 99\%$ of the measurement duration. These data are shown in Fig. 7.6.

<table>
<thead>
<tr>
<th>Collection date</th>
<th>Collection location</th>
<th>Measurement live time [s]</th>
<th>Sample mass [g]</th>
<th>Counts in $^{131}$I photopeak</th>
<th>Efficiency correction factor</th>
<th>Activity concentration [Bq/kg]</th>
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</thead>
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<td>48.8356 N 125.1366 W</td>
<td>73537</td>
<td>48.9233</td>
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<td>5.203(19)</td>
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<td>10.9988</td>
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<td>9.08(2)</td>
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<td>10.81(4)</td>
<td>16.7(1.4)</td>
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<td>87681</td>
<td>37.3040</td>
<td>152(25)</td>
<td>6.651(19)</td>
<td>2.2(4)</td>
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Figure 7.6: Time profile of $^{131}$I activity concentration in *Fucus* collected in North Vancouver [black] and *Fucus* collected at BMSC [red]. Maximum measured activity was observed on March 22 for seaweed collected in North Vancouver and March 28 for seaweed collected at BMSC, 11 and 17 days after the tsunami, respectively. By mid-May, there was very little observed $^{131}$I activity in seaweed, and the monitoring campaign was halted.
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Table 7.7: Activity concentration in Bq/kg dry weight of a variety of seaweed species collected from Bella Bella, BC on 1 April 2011 following the Fukushima accident. Error values at one standard deviation are shown in parenthesis; for further details regarding error analysis, see Sec. 5.7. Efficiency correction factors were calculated from the appropriate best fit curves shown in Fig. 7.2. The measurement live time was ≥99% of the measurement duration. The variation between collected samples suggest different accumulation rates of 131I as a function of seaweed size, age, or habitat.

<table>
<thead>
<tr>
<th>Species</th>
<th>Measurement live time [s]</th>
<th>Sample mass [g]</th>
<th>Counts in 131I photopeak</th>
<th>Efficiency correction factor</th>
<th>Activity concentration [Bq/kg]</th>
</tr>
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<td>145(22)</td>
<td>9.26(2)</td>
<td>90(9)</td>
</tr>
<tr>
<td>pyrifera</td>
<td>48048</td>
<td>2.9702</td>
<td>340(34)</td>
<td>9.85(2)</td>
<td>90(9)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>14564</td>
<td>223(25)</td>
<td>10.70(3)</td>
<td>40(3)</td>
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<td>Fucus</td>
<td>14564</td>
<td>7.0597</td>
<td>564(44)</td>
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<td>(a)</td>
<td></td>
<td>21025</td>
<td>26(13)</td>
<td>8.07(2)</td>
<td>15(8)</td>
</tr>
<tr>
<td>(b)</td>
<td></td>
<td>71949</td>
<td>8.90(2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pyropia</td>
<td>21025</td>
<td>2.4901</td>
<td>26(13)</td>
<td>8.07(2)</td>
<td>15(8)</td>
</tr>
<tr>
<td>fallax</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Chapter 8

Discussion

The main results of the monitoring campaign are the time profiles shown in Figs. 7.4 and 7.6, and Tabs. 7.2–7.6 which include the maximum measured activities in seaweed and rainwater. When constructing these time profiles, special care was taken to properly account for the efficiency of GEARS and in particular for the discrepancy between Geant4 simulations and the extended source calibration discussed in Chapter 6. For the rainwater samples, only the 24 mL and 120 mL vials were used during the monitoring campaign. The Geant4 simulations and extended source measurement using the 24 mL vial were consistent, therefore Geant4 simulations were accepted as accurate for the 24 mL vial efficiency corrections. The 120 mL vials were only used for rainwater measurements following March 31, 2011 when the $^{131}$I content was approaching negligible levels. Seven measurements were taken using the 120 mL vials; four of these were below the detection limit of GEARS and the other three were corrected to below 1 Bq/L using the Geant4 simulations. Because the activity concentration measured in these cases was very low, a large uncertainty in the extended source correction due to adsorption on the glass does not significantly impact the interpretation of the results. For the Fucus samples, which were not in solution, adsorption effects on the glass walls of the vials were not considered.

Iodine-131 was first observed in rainwater at SFU on March 19 and maximum activity concentration was measured on March 20. This result is consistent with results reported from Washington state where airborne $^{131}$I activity concentration was first observed on March 18, with a maximum occurring on March 20 [10]. Airborne $^{131}$I activity concentration in Washington state was higher than what was observed in Europe, the Canary Islands, and South Korea. Concurrent rainwater measurements at SFU demonstrate larger $^{131}$I activity...
concentration compared to values reported in Europe and South Korea [4, 6, 9]; these data are shown in Table 8.1.

Table 8.1: Comparison of maximum airborne $^{131}$I activity and maximum $^{131}$I activity in rainwater across multiple sampling locations. The results of the current work (*) are consistent with $^{131}$I arrival on the west coast of North America between March 16 and March 18 measured in Seattle, WA.

<table>
<thead>
<tr>
<th>Sample location</th>
<th>Airborne [mBq/m$^3$]</th>
<th>Rainwater [Bq/L]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milano, Italy [4]</td>
<td>0.46(3)</td>
<td>0.89(12)</td>
</tr>
<tr>
<td>Jeju, S. Korea [9]</td>
<td>0.89</td>
<td>2.81</td>
</tr>
<tr>
<td>Bordeaux, France [6]</td>
<td>2.36(6)</td>
<td>3.5(3)</td>
</tr>
<tr>
<td>Pacific NW, NA</td>
<td>4.4(1.3) [10]</td>
<td>5.8(7)*</td>
</tr>
</tbody>
</table>

The results of the current work were also compared to measurements performed following the Chernobyl nuclear accident [3]. Similar experimental conditions (using a shielded HPGe detector, measuring *Fucus*) allow for a direct comparison of $^{131}$I loading in *Fucus* samples collected from the Vancouver area and BMSC; the results summarized in Table 8.2 demonstrate that the $^{131}$I activity concentration observed in seaweed following the Fukushima accident was an order of magnitude less than what was observed following the Chernobyl accident.

Table 8.2: Maximum measured $^{131}$I activity concentration in Bq/kg dry weigh for *Fucus* collected following the Chernobyl accident [3] compared to the current work following the Fukushima accident. Errors values at one standard deviation are shown in parentheses.

<table>
<thead>
<tr>
<th>Sample location</th>
<th>Chernobyl [Bq/kg]</th>
<th>Current work [Bq/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vancouver area</td>
<td>4750(380)</td>
<td>130(7)</td>
</tr>
<tr>
<td>BMSC</td>
<td>4930(680)</td>
<td>67(6)</td>
</tr>
</tbody>
</table>

Dose estimates for Canadian residents following the Chernobyl accident were estimated to be $\sim 1 \mu$Sv [33]. The observed $^{131}$I activity in the current study implies an upper limit for the radiation dose attributable to releases from Fukushima of $\sim 0.1 \mu$Sv, an order of magnitude less than what was reported following Chernobyl. For comparison, the annual effective dose from naturally occurring radiation in Vancouver is 1.3 mSv, and the dose limit above background for the general population is 1 mSv [34]; dose estimates are summarized in Table 8.3.
Table 8.3: Dose estimates for Canadian residents following the Chernobyl and Fukushima accidents compared to the dose from natural background radiation in Vancouver [33,34].

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Dose</th>
<th>Dose limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chernobyl</td>
<td>$\sim 1 \mu$Sv</td>
<td>1 mSv/yr</td>
</tr>
<tr>
<td>Fukushima</td>
<td>$\sim 0.1 \mu$Sv</td>
<td>1 mSv/yr</td>
</tr>
<tr>
<td>Vancouver background</td>
<td>1.3 mSv/yr</td>
<td>N/A</td>
</tr>
</tbody>
</table>
Chapter 9

Conclusions

The measured $^{131}$I activity in seaweed in the current work is an order of magnitude less than what was observed in BC following the Chernobyl accident. The maximum observed $^{131}$I concentration in rainwater of $5.8(7)$ Bq/L is a factor of 16 lower than the government action levels of 100 Bq/L set on drinking water by Health Canada following a nuclear emergency [35]. The estimated dose in Vancouver attributed to the Fukushima accident is four orders of magnitude less than the annual dose from background radiation. Therefore, the short- and long-term impact of Fukushima on human health and the environment in Canada is expected to be insignificant.

Iodine-131 was observed in rainwater samples for $\sim$ 1 month following the Fukushima accident, while $^{131}$I activity in Fucus samples collected in southwest B.C. was observed for $\sim$ 2 months. The observations made in the current work confirm that Fucus can be used as an efficient system to measure the distribution of small amounts of $^{131}$I in the environment, as previously proposed [3]. This result is particularly relevant when governments and coastal communities need to choose an indicator source and monitoring timeframe when $^{131}$I contamination in the environment is a concern.

Additionally, the current work establishes GEARS as a high-precision tool for radiochemical analysis. A well shielded, absolutely calibrated HPGe detector is a powerful instrument for detecting small amounts of radiation as demonstrated by the low levels of activity measured with high precision in rainwater and seaweed. There is significant interest in monitoring and quantifying radioisotopes present in the environment, and the Nuclear Science group at SFU is actively pursuing this research opportunity.
Appendix A

Angular distribution coefficients

The angular correlation function for successive decays is

\[ W(\theta) = A_0 + A_2 \cos^2 \theta + A_4 \cos^4 \theta \]  \hspace{1cm} (A.1)

where \( A_0, A_2, \) and \( A_4 \) can be calculated if the transition probabilities between states are known, see Ref. [13]. The values for a nucleus undergoing two consecutive \( \Delta I = 2 \) quadrupole decays are given by Eqs. A.2–A.4.

\[ A_0 = 1 \]  \hspace{1cm} (A.2)

\[ A_2 = \frac{1}{8} \]  \hspace{1cm} (A.3)

\[ A_4 = \frac{1}{24} \]  \hspace{1cm} (A.4)

For some analytical applications, it is useful to write Eq. A.1 in terms of the orthonormal Legendre polynomials in \( \cos \theta, P_i(\cos \theta) \) as shown in Eq. A.5.

\[
W(\theta) = a_0 P_0(\cos \theta) + a_2 P_2(\cos \theta) + a_4 P_4(\cos \theta)
\]

\[ = a_0 + a_2 \left[ \frac{1}{2} (3 \cos^2 \theta - 1) \right] + a_4 \left[ \frac{1}{8} (35 \cos^4 \theta - 30 \cos^2 \theta + 3) \right] \]  \hspace{1cm} (A.5)

\[ = \left[ a_0 - \frac{a_2}{2} + \frac{3a_4}{8} \right] + \left[ \frac{3a_2}{2} - \frac{30a_4}{8} \right] \cos^2 \theta + \left[ \frac{35a_4}{8} \right] \cos^4 \theta \]

To find the coefficients \( a_0, a_2, \) and \( a_4, \) set Eq. A.5 equal to Eq. A.1 with the proper values of \( A_0, A_2, \) and \( A_4 \) and solve.

\[ \frac{35a_4}{8} = \frac{1}{24} \]

\[ \Rightarrow a_4 = \frac{1}{105} \]  \hspace{1cm} (A.6)
\[
\frac{3a_2}{2} - \frac{30a_4}{8} = \frac{1}{8} \\
\frac{3a_2}{2} - \frac{1}{28} = \frac{1}{8} \\
\Rightarrow a_2 = \frac{3}{28}
\]
\(\text{(A.7)}\)

\[
a_0 - \frac{a_2}{2} + \frac{3a_4}{8} = 1 \\
a_0 - \frac{3}{56} + \frac{1}{280} = 1 \\
\Rightarrow a_0 = \frac{21}{20}
\]
\(\text{(A.8)}\)

For convenience, the coefficients \(a_0, a_2,\) and \(a_4\) can be renormalized by dividing through by \(21/20\) so that \(\tilde{a}_0 = 1\).

\[
\tilde{a}_0 = 1 \\
\tilde{a}_2 = \frac{5}{49} \\
\tilde{a}_4 = \frac{4}{441}
\]
\(\text{(A.9)}\) \(\text{(A.10)}\) \(\text{(A.11)}\)
Appendix B

Conservation laws for photoelectric absorption

Figure B.1: The photoelectric absorption of an incident gamma ray on a free electron in the center of mass frame. The initial electron kinetic energy and momentum of the electron are equal to 0 due to the choice of reference frames. This process violates the conservation laws and indicates that photoelectric absorption can only occur on electrons bound to an atomic nucleus.

Consider the simple case shown in Fig. B.1, the photoelectric absorption of a gamma-ray photon on a free electron in the center of mass reference frame. The initial momentum of the system given by Eq B.1 is equal to the momentum of the gamma ray since the electron is at rest in the center of mass frame, and thus, $p_e = 0$.

\[
p_i = p_\gamma + p_e = p_\gamma = \frac{h\nu}{c} \quad \text{(B.1)}
\]

The initial energy is the sum of the gamma-ray energy and the rest energy $m_0c^2$ of the
electron shown in Eq. B.2.

\[ E_i = E_\gamma + E_e \]
\[ = h\nu + m_0c^2 \]

(B.2)

Since the gamma ray disappears in the absorption process, the final momentum and energy of the system are equal to the final momentum and energy of the electron defined by Eq B.3 and Eq. B.4.

\[ p_f = p_e' \]

(B.3)

\[ E_f = \sqrt{(p_e'c)^2 + (m_0c^2)^2} \]
\[ = \sqrt{(p_fc)^2 + (m_0c^2)^2} \]

(B.4)

From the conservation of momentum, Eq. B.4 can be re-written as follows using the initial momentum given in Eq. B.1:

\[ p_f = p_i = h\nu/c \]

(B.5)

Substituting the expression for momentum in Eq. B.5 into Eq. B.4 and removing the square root gives the result shown in Eq. B.6

\[ E_f^2 = (h\nu)^2 + (m_0c^2)^2 \]

(B.6)

The conservation of energy implies that

\[ E_f = E_i \Rightarrow E_f^2 = E_i^2 \]

(B.7)

and it follows from Eq. B.6 and Eq. B.2 that

\[ (h\nu)^2 + (m_0c^2)^2 = (h\nu + m_0c^2)^2 \]

(B.8)

After cancelling terms, Eq. B.8 implies that

\[ 2h\nu(m_0c^2) = 0 \]

(B.9)

where the gamma-ray frequency \( \nu = 0 \) since Planck’s constant \( h \) and the electron rest energy \( m_0c^2 \) are non-zero. Since a gamma-ray photon cannot have frequency \( \nu = 0 \), Eq. B.9 implies that photoelectric absorption cannot occur on a free electron.
Appendix C

Compton Scattering

The process of Compton scattering is described in Sec. 3.1.2; Fig. 3.4 from that section is reproduced here as Fig. C.1 for reference. Equations 3.5–3.7 in Sec. 3.1.2 are derived from Eq. 3.4 which gives the energy of a gamma ray following Compton scattering. The energy of the scattered gamma ray can be derived from the conservation of energy and momentum; the derivation below assumes the center of mass reference frame for convenience. For consistency, the bold symbol $\mathbf{p}$ denotes the momentum vector while $p$ denotes the vector magnitude.

The conservation of momentum is given by Eq. C.1 and energy by Eq. C.2.

\[
\mathbf{p}_\gamma = \mathbf{p}_\gamma' + \mathbf{p}_e' \quad \text{(C.1)}
\]
\[
E_\gamma + E_e = E_\gamma' + E_e' \quad \text{(C.2)}
\]

with the terms of Eq. C.2 given by Eqs. C.3–C.6. The initial electron momentum $p_e = 0$. 

Figure C.1: The Compton scattering interaction showing the recoil electron and gamma ray deflected by a scattering angle $\theta$. The energies of the scattered gamma ray and recoil electron are given by Eq. 3.4 and 3.5 in Sec. 3.1.2.
due to the choice of reference frame.

\[ E_\gamma = h\nu \]  
\[ E_e = m_0c^2 \]  
\[ E_\gamma' = h\nu' \]  
\[ E_{e'} = \sqrt{(p_{e'}c)^2 + (m_0c^2)^2} \]  

Substituting Eqs. C.3–C.6 into Eq. C.2 and solving for \( p_{e'}^2c^2 \) gives the expression shown in Eq. C.7.

\[ p_{e'}^2c^2 = (h\nu - h\nu' + m_0c^2)^2 - (m_0c^2)^2 \]  

Using Eq. C.1,

\[ p_{e'} = p_\gamma - p_\gamma' \]  

and from the scalar product,

\[ p_{e'}^2 = p_{e'} \cdot p_{e'} \]
\[ = (p_\gamma - p_\gamma') \cdot (p_\gamma - p_\gamma') \]  
\[ = p_\gamma^2 + p_\gamma'^2 - 2p_\gamma p_\gamma' \cos \theta \]  

where \( \theta \) is the angle between the incident and scattered gamma rays as shown in Fig. C.1. Multiplying through by \( c^2 \) gives an alternative expression for \( p_{e'}^2c^2 \) shown in Eq. C.10.

\[ p_{e'}^2c^2 = p_\gamma^2c^2 + p_\gamma'^2c^2 - 2p_\gamma p_\gamma'c^2 \cos \theta \]  

Setting Eq. C.7 equal to Eq. C.10, substituting in \( p_\gamma c = h\nu, p_\gamma'c = h\nu' \), and cancelling like terms gives the expression shown in Eq. C.11.

\[ (h\nu)(h\nu') + h\nu'm_0c^2 - (h\nu)(h\nu') \cos \theta = h\nu m_0c^2 \]  

Solving for \( h\nu' \) yields

\[ h\nu' = \frac{h\nu m_0c^2}{m_0c^2 + h\nu(1 - \cos \theta)} \]
\[ = \frac{h\nu}{1 + \alpha(1 - \cos \theta)} \]  

where \( \alpha = h\nu/m_0c^2 \) as presented in Eq. 3.4.
Appendix D

Pair production

The process of pair production is described in Sec. 3.1.3. This process cannot occur in free space and requires the presence of another participant such as the nucleus in order to satisfy the conservation of energy and momentum. Consider the interaction of a gamma ray in free space which leads to the production of an electron and positron as shown in Fig. D.1. The conservation of momentum is given by Eq. D.1 and Eq. D.2

\[
\begin{align*}
p_{\gamma x} &= p_{-x} + p_{+x} = p_- \cos \theta_- + p_+ \cos \theta_+ \\
p_{\gamma y} &= p_{-y} + p_{+y} = p_- \sin \theta_- + p_+ \sin \theta_+
\end{align*}
\]  

(D.1)  

(D.2)

Figure D.1: The pair production process in free space. Even under the assumption that \( h\nu \geq 1.022 \text{ MeV} \), this process is forbidden in free space by the conservation of energy and momentum.
and the conservation of energy by Eq. D.3

\[ E_\gamma = E_- + E_+ = \sqrt{(p_-c)^2 + (m_0c^2)^2} + \sqrt{(p_+c)^2 + (m_0c^2)^2} \]  

(D.3)

where \( p_- \) and \( E_- \) are the electron momentum and energy and \( p_+ \) and \( E_+ \) are the positron momentum and energy. The x-axis is defined by the direction of propagation of the gamma-ray photon, so \( p_{\gamma z} = h\nu/c \) and \( p_{\gamma y} = 0 \); the gamma-ray energy is \( E_\gamma = h\nu \). Thus, Eq. D.1 and Eq. D.3 yield two equivalent expressions shown in Eq. D.4 and Eq. D.5.

\[ h\nu = p_- c \cos \theta_- + p_+ c \cos \theta_+ \]  

(D.4)

\[ h\nu = \sqrt{(p_-c)^2 + (m_0c^2)^2} + \sqrt{(p_+c)^2 + (m_0c^2)^2} \]  

(D.5)

Equation Eq. D.4 implies

\[ h\nu_{\text{max}} = p_- c + p_+ c \]  

(D.6)

However, according to Eq. D.5

\[ h\nu > p_- c + p_+ c \]  

(D.7)

Equations D.6 and D.7 are contradictory, and therefore indicate that the conservation of momentum and energy cannot be simultaneously satisfied for pair production in free space and the interaction shown in Fig. D.1 is not possible.
Appendix E

First-order $\chi^2$ fit formalism

To minimize $\chi^2 = \sum_{i=1}^{N} (y_i - a_0 - a_1 x_i)^2$ with respect to $a_0$ and $a_1$, take the partial derivative of the function with respect to the coefficients:

\[
\frac{\partial \chi^2}{\partial a_0} = 0 = \sum_{i=1}^{N} y_i - a_0 \sum_{i=1}^{N} 1 - a_1 \sum_{i=1}^{N} x_i \tag{E.1}
\]

\[
\frac{\partial \chi^2}{\partial a_1} = 0 = \sum_{i=1}^{N} y_i x_i - a_0 \sum_{i=1}^{N} x_i - a_1 \sum_{i=1}^{N} x_i^2 \tag{E.2}
\]

Which can be re-written as the following matrix:

\[
\begin{bmatrix}
\sum 1 & \sum x_i \\
\sum x_i & \sum x_i^2
\end{bmatrix}
\begin{bmatrix}
a_0 \\
a_1
\end{bmatrix}
= 
\begin{bmatrix}
\sum y_i \\
\sum y_i x_i
\end{bmatrix}
\tag{E.3}
\]

and solved using Cramer’s Rules, yielding $a_0$ and $a_1$:

\[
a_0 = \frac{(\sum y_i)(\sum x_i^2) - (\sum x_i)(\sum y_i x_i)}{(\sum 1)(\sum x_i^2) - (\sum x_i)(\sum x_i)} \tag{E.4}
\]

\[
a_1 = \frac{(\sum 1)(\sum y_i x_i) - (\sum y_i)(\sum x_i)}{(\sum 1)(\sum x_i^2) - (\sum x_i)(\sum x_i)} \tag{E.5}
\]
Appendix F

Weighted first-order $\chi^2$ fit formalism

To minimize $\chi^2 = \sum_{i=1}^{N} \sigma_i^{-2}(y_i - a_0 - a_1 x_i)^2$ with respect to $a_0$ and $a_1$, take the partial derivative of the function with respect to the coefficients:

$$\frac{\partial \chi^2}{\partial a_0} = 0 = \sum_{i=1}^{N} \sigma_i^{-2} y_i - a_0 \sum_{i=1}^{N} \sigma_i^{-2} x_i - a_1 \sum_{i=1}^{N} \sigma_i^{-2} x_i^2$$  \hspace{1cm} (F.1)

$$\frac{\partial \chi^2}{\partial a_1} = 0 = \sum_{i=1}^{N} \sigma_i^{-2} y_i x_i - a_0 \sum_{i=1}^{N} \sigma_i^{-2} x_i - a_1 \sum_{i=1}^{N} \sigma_i^{-2} x_i^2$$  \hspace{1cm} (F.2)

Which can be re-written as the following matrix:

$$\begin{bmatrix}
\sum \sigma_i^{-2} & \sum \sigma_i^{-2} x_i \\
\sum \sigma_i^{-2} x_i & \sum \sigma_i^{-2} x_i^2
\end{bmatrix}
\begin{bmatrix}
a_0 \\
a_1
\end{bmatrix}
= 
\begin{bmatrix}
\sum \sigma_i^{-2} y_i \\
\sum \sigma_i^{-2} y_i x_i
\end{bmatrix}$$  \hspace{1cm} (F.3)

and solved using Cramer’s Rules, yielding $a_0$ and $a_1$:

$$a_0 = \frac{(\sum \sigma_i^{-2} y_i)(\sum \sigma_i^{-2} x_i^2) - (\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} y_i x_i)}{(\sum \sigma_i^{-2})(\sum \sigma_i^{-2} x_i^2) - (\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i)}$$  \hspace{1cm} (F.4)

$$a_1 = \frac{(\sum \sigma_i^{-2})(\sum \sigma_i^{-2} y_i x_i) - (\sum \sigma_i^{-2} y_i)(\sum \sigma_i^{-2} x_i)}{(\sum \sigma_i^{-2})(\sum \sigma_i^{-2} x_i^2) - (\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i)}$$  \hspace{1cm} (F.5)
Appendix G

Second-order $\chi^2$ fit formalism

To minimize $\chi^2 = \sum_{i=1}^{N} (y_i - a_0 - a_1 x_i - a_2 x_i^2)^2$ with respect to $a_0$, $a_1$, and $a_2$ take the partial derivative of the function with respect to the coefficients:

$$\frac{\partial \chi^2}{\partial a_0} = 0 = \sum_{i=1}^{N} \sigma_i^{-2} y_i - a_0 \sum_{i=1}^{N} \sigma_i^{-2} x_i - a_1 \sum_{i=1}^{N} \sigma_i^{-2} x_i^2 - a_2 \sum_{i=1}^{N} \sigma_i^{-2} x_i^4$$ (G.1)

$$\frac{\partial \chi^2}{\partial a_1} = 0 = \sum_{i=1}^{N} \sigma_i^{-2} y_i x_i - a_0 \sum_{i=1}^{N} \sigma_i^{-2} x_i^2 - a_1 \sum_{i=1}^{N} \sigma_i^{-2} x_i^3 - a_2 \sum_{i=1}^{N} \sigma_i^{-2} x_i^4$$ (G.2)

$$\frac{\partial \chi^2}{\partial a_2} = 0 = \sum_{i=1}^{N} \sigma_i^{-2} y_i x_i^2 - a_0 \sum_{i=1}^{N} \sigma_i^{-2} x_i^3 - a_1 \sum_{i=1}^{N} \sigma_i^{-2} x_i^4 - a_2 \sum_{i=1}^{N} \sigma_i^{-2} x_i$$ (G.3)

Which can be re-written as the following matrix:

$$\begin{bmatrix}
\sum \sigma_i^{-2} \\
\sum \sigma_i^{-2} x_i \\
\sum \sigma_i^{-2} x_i^2
\end{bmatrix}
\begin{bmatrix}
\sum \sigma_i^{-2} y_i \\
\sum \sigma_i^{-2} y_i x_i \\
\sum \sigma_i^{-2} y_i x_i^2
\end{bmatrix}
= 
\begin{bmatrix}
a_0 \\
a_1 \\
a_2
\end{bmatrix}$$ (G.4)

and solved using Cramer's Rules, yielding $a_0$, $a_1$, and $a_2$:

$$\det[A] = (\sum \sigma_i^{-2})(\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i^4) + (\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i^3)$$

$$+ (\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i^3) - (\sum \sigma_i^{-2})(\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i^3)$$ (G.5)

$$- (\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i) - (\sum \sigma_i^{-2} x_i^2)(\sum \sigma_i^{-2} x_i)(\sum \sigma_i^{-2} x_i^2)$$

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\[
\text{det}[A_0] = (\sum \sigma_i^{-2} y_i) (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} x_i^4) + (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} x_i^3) (\sum \sigma_i^{-2} y_i x_i) \\
+ (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^3) - (\sum \sigma_i^{-2} y_i) (\sum \sigma_i^{-2} x_i^3) (\sum \sigma_i^{-2} x_i^3) \\
- (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^4) - (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} x_i^3) (\sum \sigma_i^{-2} y_i x_i^2)
\]

\text{(G.6)}

\[
\text{det}[A_1] = (\sum \sigma_i^{-2}) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^4) + (\sum \sigma_i^{-2} E_i) (\sum \sigma_i^{-2} x_i^3) (\sum \sigma_i^{-2} x_i^2) \\
+ (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^3) - (\sum \sigma_i^{-2}) (\sum \sigma_i^{-2} x_i^3) (\sum \sigma_i^{-2} y_i x_i^2) \\
- (\sum \sigma_i^{-2} y_i) (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} x_i^4) - (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^2)
\]

\text{(G.7)}

\[
\text{det}[A_2] = (\sum \sigma_i^{-2}) (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} y_i x_i^2) + (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^2) \\
+ (\sum \sigma_i^{-2} y_i) (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} x_i^3) - (\sum \sigma_i^{-2}) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^3) \\
- (\sum \sigma_i^{-2} x_i) (\sum \sigma_i^{-2} y_i x_i) (\sum \sigma_i^{-2} x_i^4) - (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} x_i^2) (\sum \sigma_i^{-2} y_i x_i^2)
\]

\text{(G.8)}

\[
a_0 = \frac{\text{det}[A_0]}{\text{det}[A]} \\
a_1 = \frac{\text{det}[A_1]}{\text{det}[A]} \\
a_2 = \frac{\text{det}[A_2]}{\text{det}[A]}
\]

\text{(G.9-G.11)}
Appendix H

Confidence intervals for first order \( \chi^2 \) fits

Let the sums outlined in Appendix F for minimizing the weighted first order \( \chi^2 \) equation \( \chi^2 = \sum \sigma_i^{-2} (y_i - a_0 - a_1 x_i)^2 \) be re-written as follows:

\[
\begin{align*}
 s_0 &= \sum \sigma_i^{-2} \quad \text{(H.1)} \\
 s_1 &= \sum \sigma_i^{-2} x_i \quad \text{(H.2)} \\
 s_2 &= \sum \sigma_i^{-2} x_i^2 \quad \text{(H.3)} \\
 v_0 &= \sum \sigma_i^{-2} y_i \quad \text{(H.4)} \\
 v_1 &= \sum \sigma_i^{-2} y_i x_i \quad \text{(H.5)} \\
 v_2 &= \sum \sigma_i^{-2} y_i^2 \quad \text{(H.6)}
\end{align*}
\]

where the limits from \( i = 1 \) to \( i = N \) are dropped from the sums for convenience. The minimization condition defined in Appendix F, Eq. F.3 can be written as a \( 2 \times 2 \) system of equations shown in Eq. H.7.

\[
\begin{align*}
 a_0^* s_0 + a_1^* s_1 &= v_0 \\
 a_0^* s_1 + a_1^* s_2 &= v_1
\end{align*} \quad \text{(H.7)}
\]
Equation H.7 can be solved for \( a_0 \) and \( a_1 \) in terms of the sums defined in Eqs. H.1-H.6; the solutions are shown in Eq. H.8 and Eq. H.9.

\[
a_0^* = \frac{v_0s_2 - v_1s_1}{s_2s_0 - s_1^2} \tag{H.8}
\]

\[
a_1^* = \frac{v_1s_0 - v_0s_1}{s_2s_0 - s_1^2} \tag{H.9}
\]

Similarly, \( \chi^2 \) can be re-written for any \( a_0 \) and \( a_1 \) using Eqs. H.1-H.6 as

\[
\chi^2 = \sum \sigma_i^{-2} (y_i - a_0 - a_1x_i)^2
\]

\[
= \sum \sigma_i^{-2} y_i^2 + a_0^2 \sum \sigma_i^{-2} + a_1^2 \sum \sigma_i^{-2} x_i^2
\]

\[
- 2a_0 \sum \sigma_i^{-2} y_i - 2a_1 \sum \sigma_i^{-2} y_i x_i + 2a_0 a_1 \sum \sigma_i^{-2} x_i
\]

\[
= v_2 + a_0^2 s_0 + a_1^2 s_2 - 2a_0 v_0 - 2a_1 v_1 + 2a_0 a_1 s_1
\]

\[
= v_2 - a_0 v_0 - a_1 v_1 + a_0(a_0 s_0 + a_1 s_1 - v_0) + a_1(a_1 s_2 + a_0 s_1 - v_1)
\]

The system of equations defined by Eq. H.7 imply

\[
\chi^2_{\text{min}} = v_2 - a_0^* v_0 - a_1^* v_1 \tag{H.11}
\]

which follows from Eq. H.10 since \( a_0^* s_0 + a_1^* s_1 - v_0 = 0 \) and \( a_0^* s_1 + a_1^* s_2 - v_1 = 0 \) in the minimum.

The uncertainties of the fit parameters correspond to a change in \( \chi^2_{\text{min}} \) by \( c_{\nu,\alpha} \), which depends on the number of fit parameters \( \nu \) and desired confidence level \( \alpha \). Useful values of \( c_{\nu,\alpha} \) which correspond to 1\( \sigma \), 2\( \sigma \), and 3\( \sigma \) confidence intervals for \( \nu = 2 \) are shown in Table H.1; additional \( c_{\nu,\alpha} \) values can be found in Ref. [36].

<table>
<thead>
<tr>
<th>( \alpha )</th>
<th>( c_{2,\alpha} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1( \sigma ) (68.27%)</td>
<td>2.30</td>
</tr>
<tr>
<td>2( \sigma ) (95.45%)</td>
<td>6.71</td>
</tr>
<tr>
<td>3( \sigma ) (99.73%)</td>
<td>11.8</td>
</tr>
</tbody>
</table>

In order to calculate the uncertainties associated with \( a_0^* \) and \( a_1^* \), first expand \( \chi^2 \) using a Taylor expansion around the point \((a_0^*, a_1^*)\) as shown in Eq. H.12 where \( da_i \equiv (a_i - a_i^*) \)
and each derivative is evaluated at \((a_0^*, a_1^*)\).

\[
\chi^2 = \chi^2(a_0^*, a_1^*) + \frac{\partial \chi^2}{\partial a_0} da_0 + \frac{\partial \chi^2}{\partial a_1} da_1 + \frac{1}{2} \frac{\partial^2 \chi^2}{\partial a_0^2} da_0^2 + \frac{1}{2} \frac{\partial^2 \chi^2}{\partial a_1^2} da_1^2 + \frac{\partial^2 \chi^2}{\partial a_0 \partial a_1} da_0 da_1
\]

When evaluated at \((a_0^*, a_1^*)\),

\[
= \chi^2(a_0^*, a_1^*) + \frac{1}{2} \frac{\partial^2 \chi^2}{\partial a_0^2} da_0^2 + \frac{1}{2} \frac{\partial^2 \chi^2}{\partial a_1^2} da_1^2 + \frac{\partial^2 \chi^2}{\partial a_0 \partial a_1} da_0 da_1
\]

The first derivative terms are dropped since the minimization condition implies that

\[
\frac{\partial \chi^2}{\partial a_0} = \frac{\partial \chi^2}{\partial a_1} = 0
\]

Evaluating the second derivatives yields

\[
\frac{\partial^2 \chi^2}{\partial a_0^2} = 2s_0
\]

\[
\frac{\partial^2 \chi^2}{\partial a_1^2} = 2s_2
\]

\[
\frac{\partial^2 \chi^2}{\partial a_0 \partial a_1} = 2s_1
\]

which can be used to re-write Eq. H.12 as

\[
s_0 da_0^2 + 2s_1 da_0 da_1 + s_2 da_1^2 = c_{\nu, \alpha}
\]

with the change in \(\chi^2\) defined by Eq. H.18.

\[
\Delta \chi^2 = \chi^2 - \chi^2(a_0^*, a_1^*) = c_{\nu, \alpha}
\]

Equation H.17 defines an ellipse centered at \((a_0^*, a_1^*)\), shown Fig. H.1. The uncertainty \(\Delta a_1\) can be found since the line parallel to \(a_1\) at \(a_1^* + \Delta a_0\) is tangent to the ellipse defined by Eq. H.17 and there has to be one and only one solution to

\[
s_0 \Delta a_0^2 + 2s_1 \Delta a_0 da_1 + s_2 da_1^2 = c_{\nu, \alpha}
\]

which can be re-written as a quadratic equation in \(da_1\):

\[
s_2 da_1^2 + 2s_1 \Delta a_0 da_1 + s_0 \Delta a_0^2 - c_{\nu, \alpha} = 0
\]

Equation H.20 has only one solution if

\[
4s_2^2 \Delta a_0^2 - 4s_2(s_0 \Delta a_0^2 - c_{\nu, \alpha}) = 0
\]

\[
\Rightarrow \Delta a_0 = \pm \sqrt{\frac{s_2c_{\nu, \alpha}}{s_0 s_2 - s_1^2}}
\]
APPENDIX H. CONFIDENCE INTERVALS FOR FIRST ORDER $\chi^2$ FITS

Figure H.1: An error ellipse for two correlated $\chi^2$ parameters defined by Eq. H.17. The ellipse is centered at $(a_0^*, a_1^*)$ which are the parameter values that correspond to $\chi^2_{\text{min}}$.

Similarly, it can be shown that

$$\Delta a_1 = \pm \sqrt{\frac{s_0 c_{\nu, \alpha}}{s_0 s_2 - s_1^2}}$$  \hspace{1cm} (H.22)

The values of $\Delta a_0$ and $\Delta a_1$ define the range of possible values of $a_0$ and $a_1$ in the parameter space for a change in $\chi^2_{\text{min}}$ of $c_{\nu, \alpha}$. Note that every point on the ellipse defined by Eq. H.17 corresponds to an $(\overline{a}_0, \overline{a}_1)$ pair where $\overline{a}_0 \in [a_0^* - \Delta a_0, a_0^* + \Delta a_0]$ and $\overline{a}_1 \in [a_1^* - \Delta a_1, a_1^* + \Delta a_1]$. For a fixed value of $\overline{a}_1$ lying on the ellipse, the corresponding value of $\overline{a}_0$ can be calculated by observing that

$$s_0(\overline{a}_0 - a_0^*)^2 + 2s_1(\overline{a}_0 - a_0^*)(\overline{a}_1 - a_1^*) + s_2(\overline{a}_1 - a_1^*)^2 - c_{\nu, \alpha} = 0$$  \hspace{1cm} (H.23)

where the values of $p_0$, $p_1$, and $p_2$ are given by Eqs. H.24-H.26.

$$p_0 = s_0$$  \hspace{1cm} (H.24)

$$p_1 = 2s_1(\overline{a}_1 - a_1^*) - 2s_0 a_0^*$$  \hspace{1cm} (H.25)

$$p_2 = s_0(a_0^*)^2 - 2s_1 a_0^*(\overline{a}_1 - a_1^*) + s_2(\overline{a}_1 - a_1^*)^2$$  \hspace{1cm} (H.26)
Equation H.23 has two solutions given by Eq. H.27 and Eq. H.28

\[
\begin{align*}
(\bar{\alpha}_0)^+ &= \frac{-p_1 + \sqrt{p_1^2 - 4p_0p_2}}{2p_0} \\
(\bar{\alpha}_0)^- &= \frac{-p_1 - \sqrt{p_1^2 - 4p_0p_2}}{2p_0}
\end{align*}
\]  

(H.27)  

(H.28)

except when \( \bar{\alpha}_1 = a_1^* \pm \Delta a_1 \) where in each case there is a single solution \( \bar{\alpha}_0 \) which corresponds to

\[
\bar{\alpha}_0 = \frac{-p_1}{2p_0}
\]

(H.29)

with \( p_1 \) evaluated at \( \bar{\alpha}_1 = a_1^* \pm \Delta a_1 \).

The confidence interval for a linear function \( f(x_i) = a_0^* + a_1^*x_i \) is computed for each \( x_i \) based on the set of \( (\bar{\alpha}_0, \bar{\alpha}_1) \) values corresponding to the ellipse defined by Eq. H.17. The pointwise confidence interval \( C_\alpha \) is given by Eq. H.30:

\[
C_\alpha(x_i) = [f_{\min}(x_i), f_{\max}(x_i)]
\]

(H.30)

where \( \alpha \) is the confidence level corresponding to a change in \( \chi^2_{\min} \) by \( c_{\nu,\alpha} \), and \( f_{\min}(x_i) \) and \( f_{\max}(x_i) \) are the minimum and maximum values of \( f(x_i) \) calculated over the error ellipse by evaluating \( f(x_i) = \bar{\alpha}_0 + \bar{\alpha}_1x_i \) at each \( (\bar{\alpha}_0, \bar{\alpha}_1) \) pair.
Appendix I

$\chi^2$ formalism for scaling data

To minimize $\chi^2 = \sum_{i=1}^{N} \sigma^{-2}_i (y_i - a_0 x_i)^2$ with respect to $a_0$ take the partial derivative

$$\frac{\partial \chi^2}{\partial a_0} = 0 = -2 \sum_{i=1}^{N} \sigma^{-2}_i x_i (y_i - a_0 x_i)$$

and solve for $a_0$ which yields

$$a_0 = \frac{\sum_{i=1}^{N} \sigma^{-2}_i x_i y_i}{\sum_{i=1}^{N} \sigma^{-2}_i x_i^2}$$
Appendix J

Geant4 simulations of $Q$-factors for the sum peak method

The $Q$-factors for Geant4 simulations described in Sec. 5.6.3 were calculated following the implementation and verification of the angular distribution algorithm described in Sec. 5.6.4 and the extraction of the $J$-factors from simulated spectra described in Sec. 5.6.5. Sixteen independent $^{60}\text{Co}$ spectra were simulated in Geant4 with $10^7$ simulated decays/spectrum. The simulated $^{60}\text{Co}$ source was positioned 10.5 cm above the GEARS can to replicate experimental conditions. In each simulated decay event, the first gamma ray in the $^{60}\text{Co}$ cascade ($E_\gamma = 1173.237$ keV, see Fig. 2.3) was simulated with an isotropic angular distribution, while the second gamma ray in the cascade ($E_\gamma = 1332.492$ keV, see Fig. 2.3) exhibits the effects of the angular distribution algorithm.

The data obtained following the analysis of the simulated spectra described in Sec. 5.6.5 are shown in Table J.1. The number of counts is the average $\mu_k$ from the 16 simulations and the quoted error is the standard error on the mean $\sigma_{\mu_k}$ for $k = 0, 2, 4$. These values define the $J$-factors according to the procedure in Sec. 5.6.5.

The $J$-factors determined from Table J.1 were used to calculate the component $Q$-factors $Q_{kT}$, $Q_{kvv}$, $Q_{kvV}$, and $Q_{kVv}$ for $k = 0, 2, 4$ according to the procedure of Sec. 5.6.5 using Eqs. 5.39-5.42 in Sec. 5.6.3. The results of the $Q$-factor calculations are shown in Table J.2 where the quoted error is calculated using standard error propagation techniques. The $Q$-factors shown in Table J.2 were used to calculate the absolute efficiency of GEARS using the sum peak method as described in Sec. 5.5 and Eq. 5.22 and Eq. 5.23. A comparison
between the sum peak method and the calibrated source efficiency calibration described in Sec. 5.4 is shown in Sec. 5.7, Table 5.9.

Table J.1: Results from the analysis of simulated \( ^{60}\text{Co} \) spectra where the second gamma ray in the \( ^{60}\text{Co} \) cascade (\( E_\gamma = 1332.492 \) keV) has an angular distribution governed by \( P_k(\theta) \) where \( P_k(\theta) \propto \cos^k \theta \sin \theta \) for \( k = 0, 2, 4 \). These results define the \( J \)-factors following the procedure described in Sec. 5.6.5 and were used to calculate the Geant4-simulated \( Q \)-factors defined in Sec. 5.6.4 and Eqs. 5.48-5.51.

\[
\begin{array}{cccc}
\text{Energy [keV]} & \# \text{ counts} & \# \text{ counts} & \# \text{ counts} \\
1173.237 & 229346(72) & 225061(130) & 221056(107) \\
1332.492 & 208055(118) & 203711(112) & 199854(99) \\
\text{Sum peak} & 489(7) & 1390(8) & 2250(8) \\
\text{Total} & 2444274(333) & 2423855(363) & 2405588(365) \\
\end{array}
\]

Table J.2: \( Q \)-factor components [top 3 rows] and final \( Q \)-factor values [bottom row] from Geant4-simulated \( ^{60}\text{Co} \) spectra. In the calculation of efficiencies defined by the sum peak method, only the ratios of the \( Q \)-factors are important. However, it is convenient to examine the \( Q \)-factors where the factor of 2 in Eqs. 5.48-5.51 is removed. The effect of the angular distribution can be examined by comparison to the isotropic distribution where each \( Q \)-factor is equal to 1.

\[
\begin{array}{cccccc}
\text{1173.237 keV} & \text{1332.492 keV} & \text{Sum peak} & \text{Total} \\
Q_{0vV} & 1 & 1 & 1 & 1 \\
Q_{2vV} & 0.9813(6) & 0.9791(8) & 2.84(5) & 0.9916(2) \\
Q_{4vV} & 0.9639(6) & 0.9606(7) & 4.60(7) & 0.9842(2) \\
\frac{1}{2}Q_{0V} & 2.09784(5) & 2.09760(7) & 2.314(4) & 2.099040(17) \\
\frac{1}{2}Q_{2V} & 1.04892(3) & 1.04880(3) & 1.157(2) & 1.049520(9) \\
\end{array}
\]
Bibliography


[29] A. Salomon. Personal communication.


