Visualization and Quantitation of Radioiodine Distribution in Silver Zeolite Cartridges

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VISUALIZATION AND QUANTITATION OF RADIOIODINE DISTRIBUTION IN SILVER ZEOLITE CARTRIDGES

A Thesis
Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Master of Science in
The Department of Physics and Astronomy

by
Daniel Joseph DiMarco
B.S., Louisiana State University, 2017 December 2019
Acknowledgements

I would like to thank my thesis committee for their help during this entire process. Dr. Kenneth Matthews has been an invaluable mentor during my entire graduate and undergraduate career at LSU. He is a valuable source of information and advice and I would not be where I am today without his guidance. I would also like to thank my committee members: Dr. Wei-Hsung Wang and Dr. Catherine Deibel. Dr. Wang helped me recognize that Health Physics is a passion for me and has been a great source of guidance. Dr. Deibel cultivated my interest in physics since the first year of my undergraduate career and I am very grateful to her.

I would also like to thank Dr. Joyoni Dey for her reconstruction algorithm used in the study. Her invaluable knowledge has made this experiment possible. I am also grateful to Amin Hamideh for his advice on cartridge prep, radiation safety, and iodine shipping. I would like to thank Cody Andre, Luke Best, Patrick Adams, and Doug Naden for their assistance with SPECT data acquisition. I would also like to thank Dr. Charlie Wilson for the use of his foodsaver device and his expertise on environmental regulations.

Additionally, I thank the Nuclear Regulatory Commission for their support through the NRC Graduate Fellowship. My graduate career was funded through their help and I hope to continue working closely with them.

Finally, I would like to thank my family and friends. My parents, Gerard and Kris, have been an enormous support to me throughout my entire academic career and I can never thank them enough. My brothers, Paul and Peter, have also been a big support even if they don’t always know what I am talking about. My fellow classmates in the medical and health physics program, in particular Garret Otis and Anthony Davila, have been great to work with and I wish
only the best for them in the future. I could not have accomplished this without the help of so many people and I want them to know I am forever grateful.
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Abstract

Iodine 131 is a major fission product released during a nuclear incident. This isotope provides a serious health hazard for humans and the environment, therefore nuclear power plants must monitor releases using air sampling. The air sampling is accomplished using air filter cartridges using silver zeolite as a filter media. During an emergency situation, silver zeolite is useful for its affinity to I-131 without adsorbing other radioactive gases. After leaving the plume, these cartridges are counted in a low background area. This measurement does not take into account the distribution of radioiodine in the cartridge. This study explores two methods of quantifying and visualizing the radioiodine within a silver zeolite cartridge, conjugate counting and single photon emission computed tomography (SPECT) imaging. Two silver zeolite cartridges were used as phantoms, one with I-131 and one with Ba-133, in the conjugate counting setup. The barium cartridge was used for efficiency and attenuation measurements, while the iodine cartridge was used for activity measurements. The iodine cartridge was also imaged using a SPECT system to determine the visibility of a pattern of I-131 placed in the cartridge. We found that the conjugate counting method provides an average activity measurement 8.8% different than the actual activity, compared to 28.4% different and 25.1% different for the individual faces. We were able to identify the pattern placed into the silver zeolite cartridge using the SPECT system and could determine that imaging a real-life cartridge with a lower activity could be done within 25 hours with a reasonable spatial resolution. We determined that the conjugate counting method is a reasonable addition to normal air monitoring for more accurate activity measurements. We also determined that SPECT imaging can be used to image a lower activity cartridge in a real-life scenario within a reasonable amount of time.
Chapter 1. Introduction

1.1. Background

1.1.1. Iodine-131

Nuclear power produces many different radionuclides as fission byproducts, including radioactive isotopes of iodine. These radioiodines are released in a plume with other radioisotopes during a nuclear release incident. [Wang, 2006] One isotope of iodine, iodine-131, is a significant health hazard for humans and the environment. Iodine-131 is a beta and gamma emitter; the dominant gamma emission (81% abundance) has an energy of 364 keV while the dominant beta emission (89% abundance) has a maximum energy of 606 keV. Iodine-131 has a physical half-life of 8.01 days, and a fission yield around 3%. [Nuclide Safety Data Sheet, 2019] The primary radiological hazard from I-131 is internal effects if inhaled or ingested. The biological half-life of I-131 is 120-138 days. [Nuclide Safety Data Sheet, 2019]

Iodine is an essential component for production of hormones in the thyroid gland; iodine is not produced within the body, so humans must get iodine from external sources. The thyroid gland is an important part of the endocrine system, producing the hormones triiodothyronine (T3) and tetraiodothyronine (T4), which affect the body’s metabolic rate. [Brent, 2012] Production of these hormones depends on iodine availability. The iodine in these hormones is released when the hormones are used in the body and returns to the thyroid to be recycled into new hormones. Because iodine accumulates in the thyroid, radioactive iodine can lead to localized radiation dose from I-131’s beta emission. These large doses can directly harm thyroid tissue, leading to reduced hormone production, or even thyroid cancer. Iodine-131 is used in radioisotope therapy because the thyroid specificity leads to a large amount of tumor ablation, and thyroid cancer control.
The general public can potentially be unintentionally exposed to radioiodine, especially I-131. A release of radioiodine from a nuclear power facility can contaminate the environment around the facility with I-131. [Cember, 2009] During a release incident, the iodine is emitted as part of a plume cloud. This cloud includes both vapor and particulate forms of I-131. The particulate form is deposited onto soil and vegetation and can contaminate waterways. [Cember, 2009] I-131 deposited onto soil can be taken up by plants, which are then eaten by grazing animals. I-131 then contaminates the meat and milk of these animals. I-131 deposited into waterways can contaminate fish and other aquatic animals. Direct consumption of contaminated crops or contaminated animal products (such as meat or milk) is a major route of radioiodine exposure for humans. The vapor form of I-131 is an inhalation hazard, especially for persons or animals inside the plume.

1.1.2. Environmental Monitoring

Both the Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC) require nuclear power plants to monitor emission of radioisotopes, where the monitoring is typically done through state governments (e.g. Louisiana Department of Environmental Quality).[10 CFR Part 50, 2018 and Louisiana DEQ Title 33 Part 15, 2019] These regulations involve both routine air monitoring and emergency air sampling. Routine air sampling is used to confirm integrity of operations. Emergency air sampling is used to determine the concentrations of radionuclides and noble gases inside the plume from a fission release event. In either case, sampling is performed by drawing air through a filter media to collect airborne radioisotopes. These filter media are often in the form of cartridges, small plastic canisters filled with the filter media.
Activated charcoal and silver zeolite are the most common types of media. [Wang, 2006] Activated charcoal is commonly used during routine sampling because it is relatively inexpensive, but it does not selectively absorb radioiodine and can easily be saturated by radioactive noble gases. Although more expensive, silver zeolite (AgZ) is preferred for emergency air sampling due to its affinity for gaseous iodine and relative insensitivity to radioactive noble gases. [Kravchick, 2008] High levels of radioactive noble gases in the plume could skew the estimates of radioiodine if charcoal was used.

After taking air samples inside the plume, the filter cartridges are taken to a low background area and then assayed for radioactivity. A sodium iodide (NaI) scintillation detector is a typical tool for measurement of activity in the cartridge by providing a measure of the gamma emissions from radioisotopes. [Maiello, 2011] This scintillation detector provides a rapid measure of activity in the cartridge, which lowers the amount of time needed for protective actions to be taken. Determining activity from counting of a cartridge requires that the detector system has been properly calibrated. Proper calibration requires knowledge of the radionuclide being measured as well as the geometry of the source. Many agencies involved in emergency response assessment use a barium-133 “check” source placed on top of a AgZ cartridge as the calibration standard for their equipment.[Montgomery, 1990] This approach does not accurately reflect the I-131 isotope energy or the distribution of isotope in the cartridge. To account for this inaccuracy, correction factors are used in activity calculations. However, radioiodine distribution is not well understood so an appropriate correction factor is not easily determined. [Maiello, 2011] Radioiodine distribution inside the cartridge is an important factor to consider when taking these measurements.
Currently, common assumptions are that the distribution in the cartridge is either homogenous or face-loaded. [Montgomery, 1990] The homogenous distribution assumes the radioiodine is uniformly distributed throughout the volume of filter media in a cartridge. Face-loaded, on the other hand, assumes that most of the radioiodine is distributed in the front layers of the filter media (typically 0.25” to 0.5” from the face where air flow enters the cartridge). Neither distribution is believed to be correct, resulting in uncertainty in iodine measurements with current methods of calibration and analysis. Furthermore, a check source placed on the surface of the cartridge (whether proximal or distal to the detector) has a decidedly different solid angle than a distributed source inside the cartridge, which leads to underestimates of radioiodine in the cartridge. [Hamideh 2019] Calibration with a check source is simple and convenient but determining a useful correction factor is essential to reliably gauge I-131 activity in the cartridge. One study determined that an erroneous assumption about the radioiodine distribution in a cartridge can cause up to a 9x underestimate of the iodine activity.[Hamideh, 2019] Other authors have recommended collecting “empirical data to determine the activity distribution in experimental cartridges exposed to radio-iodine” from which a correction factor can be determined that can then be applied to field cartridges.[Maiello, 2011]

1.2. Problem Statement

When determining activity from emergency air sampling cartridges, a cartridge is typically counted only on one face and with no knowledge of iodine distribution in the cartridge. The accuracy of correction factors used to convert counting measurement to I-131 activity, such as for non-uniform attenuation of gamma rays by the filter media, are limited by this lack of knowledge and simplifying assumptions.
The first part of this project evaluated conjugate counting as a method to gauge the I-131 activity in an air sampling cartridge that does not require explicit knowledge of the distribution. We hypothesized that this method is significantly more accurate at determining I-131 activity than the traditional approach of counting one face and making assumptions about the distribution.

The second part of this project assessed SPECT imaging of I-131 using pinhole collimation as a means to determine the distribution of I-131 in a filter cartridge. Visualization of the I-131 distribution, either for individual cartridge samples from the field or for a representative cartridge produced in controlled conditions, would facilitate accurate corrections for geometry of activity measurements. We hypothesized that pinhole SPECT imaging is capable of visualizing I-131 distribution at a level of resolution and sensitivity appropriate for use with AgZ filter media cartridges.

1.2.1. Conjugate Counting

Conjugate counting is a method of determining the activity of an object using measurements from two opposed views of the object.[Fleming, 1978] Note that while conjugate counting facilitates an accurate measurement of activity in the cartridge, the actual distribution of radioiodine in the cartridge as well as its effect on attenuation are not explicitly determined. Correction factors for object attenuation and detector efficiency are measured for the counting geometry and the object being counted.

The conjugate counting technique was historically used to determine radioisotope activity in organs, using either counting detectors or gamma cameras.[Thomas, 1976] However, the ability of gamma cameras to image radioisotope distributions in a patient, coupled with the fact that quantitation is a secondary concern in many situations, has overshadowed conjugate
counting in diagnostic nuclear medicine. By comparison, the ability of positron emission
tomography (PET) imaging to perform quantitation, in addition to visualization, is closely related
to the concept of conjugate counting.

The concept of conjugate imaging is illustrated in Figure 1.1. Two detectors were setup
and labeled as D1 for front facing detector and D2 for the back facing detector. Count rates were
collected from each detector where count rates from Detectors 1 were designated with R1 and
count rates from Detector 2 were designated with R2. The observed count rates R1 and R2
acquired by detectors viewing opposing faces of the source distribution are given by

\[
R_1 = AY \varepsilon_1 e^{-\mu_{eff} L_1} \\
R_2 = AY \varepsilon_2 e^{-\mu_{eff} L_2}
\]

where A is source activity, Y is decay yield and ε1 and ε2 are the detectors’ counting efficiencies
for the gamma emission that is being counted, μ_{eff} is the effective attenuation coefficient of the
source material, and L1 and L2 are the attenuation pathlengths through the source to the two
detectors. If the detectors and counting geometry are identical, ε1 = ε2 = ε. Computing the
conjugate mean R_{conj} of the count rates produces

\[
R_{conj} = \sqrt{R_1 R_2} = AY \varepsilon e^{-\mu_{eff} L}
\]

where \( L = \frac{L_1 + L_2}{2} \) is the total pathlength through the source.

Eqn. 1.2 allows the activity of an unknown source distribution to be calculated from the
conjugate mean count rate. The efficiency and attenuation factors can be measured or calculated
for given detector and source geometries. The correction factor for attenuation in the conjugate
counting method avoids the need to explicitly know the radioiodine distribution while still
effectively accounting for the impact of its spatial distribution.
Figure 1.1. Illustration of conjugate counting of a source distribution S using detectors D₁ and D₂ viewing opposite sides of the source. Distances d₁ and d₂ from source to detectors determine geometric efficiency. L₁ is the average attenuation pathlength to D₁, while L₂ is the average attenuation pathlength to D₂; total thickness L is the sum of L₁ and L₂.

Count rate measurements are made using an appropriate counting detector, such as a NaI scintillation detector or a high purity germanium detector (HPGe). Air filter cartridges are generally measured using NaI probes, as they are much easier for field use than a HPGe system and provide a more rapid measurement. One study on charcoal cartridges showed an increase in accuracy by flipping the cartridge halfway through a counting measurement.[Montgomery, 1990] This approach averaged over the effects of attenuation and distribution, rather than compensating for them as does conjugate counting; this averaging approach does not allow for correction of attenuation based on total pathlength through the source. The conjugate counting method described here assumes a homogeneous attenuator of uniform thickness; for a heterogeneous attenuator and/or non-uniform thickness, results will depend on the degree to which the source can be approximated satisfactorily by an effective attenuation coefficient and/or effective thickness. (In such situations, the literature reports conjugate imaging with gamma cameras as a solution. [Matthews, 1997])

1.2.2. SPECT Imaging

SPECT is an imaging technique to visualize the 3D distribution of a radioisotope in an object. [Bushberg, 2012] In SPECT imaging, a gamma camera acquires projection images of the
source distribution from multiple angles around the object; the projection data is reconstructed into the 3D source distribution with an appropriate algorithm, such as filtered backprojection or an iterative reconstruction algorithm.[Wernick, 2004] Acquisition parameters such as collimator type, number of and angular range spanned by the projection images, acquisition time per projection, and amount of source activity all heavily influence the resulting image quality.

Figure 1.2. An illustration of a SPECT system. Electronics [A], photomultiplier tubes [B], scintillation crystals [C], and collimators [D] are indicated.

SPECT imaging is a common method for imaging of iodine uptake in the thyroid.[Wernick, 2004] For diagnostic imaging, the isotopes I-123 and I-125 are commonly used because their low emission energies are well-suited to modern gamma cameras and collimators. Imaging of I-131 is often used to verify tracer uptake in the thyroid for ablation therapy using I-131, although image quality can be marginal; I-131 has a higher gamma energy than is optimal for gamma camera collimator design. [Wernick, 2004]
The choice of collimator is a key determinant of SPECT image quality. [Wernick, 2004] The collimator influences field of view, spatial resolution, and sensitivity (which affects noise and acquisition duration); collimator design is a trade-off between these factors. Collimators must also be designed appropriately for the gamma emission energy that is being imaged, to minimize degradation of image quality due to undesirable penetration of gamma rays through the collimator. Most diagnostic imaging uses parallel-hole collimators (for a large field of view) designed for low-energy gamma rays from Tc-99m. Fan-beam and pinhole collimators generally provide higher spatial resolution over a smaller field of view, due to magnification, compared to parallel-hole collimators. Collimators designed for medium-energy isotopes, such as In-111 typically have worse resolution and sensitivity than low-energy design, due to thicker septa required. Collimators for high-energy isotopes, such as I-131, are difficult to design with good resolution and sensitivity.[Wernick, 2004] Pinhole collimators are commonly used for thyroid imaging with I-123 and I-125; however, the 364 keV gamma of I-131 is higher than intended for typical pinhole collimator design, potentially degrading image quality for this isotope.

1.3. Objectives

This project had three phases. In the first phase, cartridges were fabricated using I-131; a known quantity of radioiodine spiked in a known pattern was required. The known activity allowed us to assess conjugate counting as a means to recover source activity from counting measurements. The known source distribution provided a means to assess the image quality achievable by pinhole SPECT imaging. For calibration purposes, a cartridge filled with a uniform distribution of Ba-133 was also fabricated during this phase.

Phase 2 was implementing the conjugate counting setup, followed by measurement and analysis of the Phase 1 cartridge. In this phase, we assembled a counting system based on a NaI
detector and associated electronics; the system was arranged so that each side of the cartridge could be counted with identical geometry. Calibration measurements were made to determine efficiency and attenuation corrections, so that source activity could be calculated from Eqn. 1.2. Conjugate counting results were compared to a typical field-measurement technique of counting one face of the cartridge using a detector calibrated with a Ba-133 check source.

Phase 3 was assessing the potential of pinhole SPECT imaging as a means to visualize the source distribution in an AgZ filter cartridge. The main objectives of this phase were (1) to determine if pinhole imaging coupled with an iterative reconstruction algorithm could resolve the known source distribution and (2) to gauge the acquisition parameters (especially acquisition time per projection) that might be required to visualize the very low-activity source distribution expected from emergency air sampling with an AgZ filter cartridge.
Chapter 2. Methods and Materials

2.1. Cartridge Production

2.1.1. Description of Cartridges

Two cartridges were fabricated using I-131; these were used for both conjugate counting measurements and for pinhole SPECT imaging. A cartridge filled with Ba-133 was made to use for calibration of the conjugate counting system. The methods to create these cartridges are described in the next section. The second and third sections of this chapter provide the methods for conjugate counting measurements and for pinhole SPECT imaging, respectively.

2.1.2. Iodine Cartridges

Shown in Figure 2.1, the AgZ air sampling cartridges used in this project were model “C” series cartridges (F&J Products, part #AGZC58). These cartridges nominally comprise 45g of loose granular AgZ contained within a plastic cylindrical housing.

Figure 2.1. Photo of a model "C" AgZ cartridge (F&J Products, part #AGZC58)

The faces of the housing have a grid of perforations to allow air flow; filter papers inside the faces contain the AgZ within the cartridge. The cartridge has a diameter of 2.26”, a height of 1.05”, and an adsorbent mesh size of 50 x 80.
To maintain a controlled distribution of I-131 within a cartridge, spots of I-131 were patterned onto filter papers, rather than directly adding the isotope to the loose AgZ granules. Whatman #3 filter papers (Sigma-Aldrich) were cut to fit the circular cross-section of a cartridge. Two patterns were chosen, which are illustrated in Figure 2.2. Pattern 1 comprised multiple spots; the spots were grouped in quadrants by size. An acrylic template (Figure 2.3), fabricated in-house, was used to guide the placement of drops for Pattern 1. Pattern 2 had four identical spots, one in the center of each quadrant.

Two variations of Pattern 2 were made, differing only in the size of the drops: larger in Pattern 2a and smaller in Pattern 2b. Of the two cartridges made for this project, one contained one each of Pattern 1, Pattern 2a and Pattern 2b; the other cartridge contained three filter papers of Pattern 1.

The I-131 was an aqueous solution (Cardinal Health) with a concentration of 1mCi/mL (used in cartridge 1) or 4 mCi/mL (used in cartridge 2). Food coloring was added to facilitate
visualizing the drops when placed on the filter paper. For Pattern 1, spots were pipetted (Eppendorf) in multiple drops onto the desired locations on the filter paper.

Figure 2.3. Photograph of acrylic guide used in producing Pattern 1

During pipetting, the filter paper was placed on top of plastic wrap (SE Grocers) inside a plastic petri dish (Scientific Products, D1906) to prevent accidental spillage or in case of leakage through the filter paper. For Pattern 1, drops of 3.5 \( \mu \text{L} \) were used for the largest spots, with 2.5 \( \mu \text{L} \), 1.5 \( \mu \text{L} \) and 0.85 \( \mu \text{L} \) drops were used to make the progressively smaller spots in each quadrant. Drops were applied to each spot on a filter paper, then allowed to dry; the process was repeated, adding drops to the same spots, until the desired activity had been placed. This iterative approach allowed a large activity to be placed onto each spot while controlling the spot sizes. Because a single activity concentration was used, larger spots represented more activity than smaller spots.
The same process was used for Pattern 2, except that drops were placed by using the tip of a paper clip that was dipped into the I-131 solution. A large paper clip was used for Pattern 2a and a small paper clip for Pattern 2b. The tips of the paper clips allowed for smaller overall spot sizes than could have been achieved with the available pipettes, but with less control of the volume between drops.

Before pipetting, an aliquot of I-131 solution was assayed in a dose calibrator (Capintec, Inc. Model 55tW) to determine the activity concentration. The planned total activity for each filter paper was approximately 12.3 MBq (0.33 mCi). After creating the filter papers, the total activity on each filter paper was calculated from the total volume pipetted onto the filter paper. The completed filter papers were also assayed in the dose calibrator to verify total activity. Until assembly into an AgZ cartridge, the filter papers were protected in plastic wrap.

![Diagram of AgZ cartridge with filter papers](image)

Figure 2.4. Illustration of the placement of spiked filter papers within the AgZ material of a cartridge

To assemble filter papers into an AgZ cartridge, one perforated face of the cartridge was first cut off, using the cutting disk of a Dremel tool, along the outer edge of the cylindrical cartridge. The loose AgZ was removed; the built-in filter papers behind the perforations in the cartridge faces were left in place. The AgZ and three radioiodine-laden filter papers were
interleaved into the cartridge, as shown in Figure 2.2; any excess AgZ was included in the
topmost layer. Black adhesive vinyl tape (3M Scotch) was used to affix the cartridge face back
onto the cartridge. One of the cartridges made for this project contained one each of Pattern 1,
Pattern 2a and Pattern 2b, with the Pattern 1 filter paper closest to the cartridge face. The other
cartridge contained three filter papers of Pattern 1; the three filter papers were rotated 90° from
each successive paper.

2.1.3. Barium Cartridge

To measure the counting efficiency of the scintillation detector used for conjugate
counting, a cartridge with similar geometry and known activity was required. To make this
efficiency phantom, the AgZ was removed from a cartridge and replaced with an aqueous Ba-
133 solution. Ba-133, often called “mock iodine”, is commonly used as a substitute for I-131 in
calibration measurements. Ba-133 and I-131 have similar γ emission energies (356 keV vs. 364
keV, respectively), while the 10.5 yr half-life of Ba-133 makes it convenient to use as a
calibration source. [Wang, 2006]

The zeolite was removed through a hole in one face, and the inside of the cartridge was
cleaned. Both faces were sealed with waterproof silicone sealant to minimize leakage. Using two
syringe needles to pierce the cured sealant, the cartridge was filled with water to 3/4ths of its
volume through one needle, with the other needle allowing the cartridge to vent. A syringe
containing Ba-133 was assayed in the dose calibrator, the Ba-133 was injected into the cartridge,
and the syringe was re-assayed; the difference in activity pre- and post-injection yielded the Ba-
133 activity in the cartridge. The remaining volume of the cartridge was filled with water. The
needle holes were then covered with silicone sealant. For additional protection against leakage,
the Ba-133 cartridge was sealed in a food-saver type polyethylene bag using a commercial vacuum sealing machine (Foodsaver).

### 2.2. Conjugate Counting

#### 2.2.1. Setup

The conjugate counting setup is shown in Figure 2.5. The scintillation detector was a 3” diameter by 3” tall NaI crystal with integral photomultiplier tube (Model #12|12/3+POB-14P, Alpha Spectra Inc.). The detector was coupled to a voltage divider with preamp (Model 276, Ortec). High voltage power (Model 3102D, Canberra), a spectroscopy amplifier (Model 572A, Ortec), and multichannel buffer (Model 926, Ortec) were located in a NIMbin (Model 4006, Ortec). Spectroscopy software (Maestro v.6.08, Ortec) running on a Windows XP laptop recorded the gamma spectra.

![Figure 2.5. Photograph of conjugate counting setup showing NaI detector, Minibin, multichannel analyzer, and iodine cartridge at 0.5 m](image_url)

For all counting measurements, the detector was placed on its side with the flat face looking towards the source; distances from the face of the detector were marked with a meter
stick. The cartridges were laid on their side, with one face looking toward the detector face. The spectroscopy system was calibrated with a Cs-137 source (662 keV). For measurements of both Ba-133 and I-131, a region of interest was defined at a 40 keV width centered on the respective photopeak energy, serving as an energy window around the photopeak. For I-131 this was a region of 364 ±20 keV and a region of 356±20 keV for Ba-133.

2.2.2. Efficiency and Attenuation Measurements

With the sources in identical cartridges, the geometric factors in efficiency were the same when the cartridges were located at identical distances from the detector face. Thus with an energy window bracketing the photopeaks, absolute peak efficiency for the counting setup was measured using the Ba-133 cartridge and then applied to the I-131 cartridge measurements. Because of the similar emission energies, no correction was made for differences in attenuation in NaI for the two isotopes.

Absolute efficiency depends on source geometry, including source-to-detector distance, so efficiency was measured at each distance planned for iodine cartridge measurements. Because of the low activity of Ba-133 and to reduce uncertainty in efficiency measurements, long counting times were used for the barium cartridge. These times were 180,000 seconds at 1.55 m, 72,000 seconds at 1 m, 18,000 seconds at 0.5 m, and 2,200 seconds at 0.25 m, to achieve approximately 2.75x10^5 counts in the energy window. Only one face of the barium cartridge was counted, with the same face used for all efficiency measurements. Absolute peak efficiency was calculated as:

$$\varepsilon_{\text{peak, abs}} = \frac{R}{AY}$$  \hspace{1cm} [2.1]

where A is the known activity (Bq) of barium in the cartridge, Y is the yield of the 356 keV gammas per decay, and R is the measured count rate (sec^{-1}).
The correction factor for self-attenuation by AgZ in the cartridge was determined by making a transmission measurement through a new unused AgZ cartridge using the Ba-133 cartridge as the source. The new AgZ cartridge was placed at the measurement location of 1 m from the detector, with the barium cartridge placed against it on the far side from the detector; the two cartridges were placed face to face. Counts were acquired for 72,000 sec and the count rate in the energy window was recorded. The AgZ cartridge was then removed and count acquisition was repeated for the Ba-133 cartridge alone. The attenuation correction factor was calculated as

\[ f = e^{\mu_{\text{eff}} L} = \frac{R_{\text{without AgZ cartridge}}}{R_{\text{with AgZ cartridge}}} \]  

where \( R_{\text{..}} \) are the respective count rates; note that the effective attenuation coefficient \( \mu_{\text{eff}} \) of AgZ and the cartridge thickness \( L \) are not explicitly determined. Correction for detector efficiency was not needed because the barium cartridge was in the same location for both counts.

The attenuation measurement was repeated at 0.5 m to determine if source-to-detector distance affected the determination of the attenuation correction factor.

2.2.3. Iodine measurements

For conjugate counting acquisitions of the I-131 cartridge, the cartridge face being counted was placed at distances of 1.55 m, 1m, 0.5m, and 0.25m from the edge of the detector. Longer distances were used when the source had high activity, to minimize detector deadtime; as the I-131 decayed, the cartridge was moved progressively closer to the detector. The acquisition time was 1800 seconds for each face of the cartridge. The same measurement time was used at every distance. The net count rate, as reported by the MAESTRO software, based on counts in the energy window was recorded for the measurement of each face of the cartridge.
2.2.4. Calculation of Activity

The activity of I-131 in the cartridge was calculated from the count rates of the two faces of the cartridge, with corrections for yield, efficiency, and self-attenuation, as

\[ A = \sqrt{R_1 R_2} \cdot \frac{f}{\epsilon_{\text{peak,abs}} Y} \]  

[2.3]

where \( A \) is the activity (Bq) in the cartridge and the \( R_i \) are the count rates from each face of the cartridge; \( \epsilon_{\text{peak,abs}} \) is the peak absolute efficiency for the detector (see Eqn. 2.1), the radiation yield is \( Y \), and the attenuation correction factor is \( f \) (see Eqn. 2.2).

2.3. SPECT Imaging

2.3.1. SPECT System

To determine if SPECT imaging with a pinhole collimator can provide adequate image quality to visualize the activity distribution in an AgZ filter cartridge, we acquired projection images of the I-131 cartridges (see section 2.1.2), then reconstructed the projections into tomographic images. One camera of a Symbia T series SPECT-CT imaging system (Siemens Medical Systems), equipped with a pinhole collimator, was used to collect the projection images. This camera model comprised a 3/8” NaI detector with a quoted intrinsic spatial resolution of \( \leq 3.8 \) mm FWHM and energy resolution <10%. The pinhole collimator had a 4mm aperture; the quoted sensitivity of this collimator was 67 cpm/µCi with a system spatial resolution of 7.6 mm, both measured at 10 cm from the pinhole. [Siemens Medical Solutions] Projections were reconstructed with an interactive reconstruction algorithm described in section 2.3.4.

2.3.2. Rotary motion system

SPECT systems typically acquire projection data by rotating the detectors around the object (cf. Figure 1.2). When using the pinhole collimator, however, the Symbia system does not
allow the gantry to rotate because of weight balance issues. Consequently, we required a setup to rotate the object in front of the pinhole collimator, mimicking detector rotation.

![Image showing components of the rotary motion system](image.png)

**Figure 2.6.** Photo collage showing the components of the rotary motion system [A]. Close-up views of the cartridge holder [C,D], capillary holder [E], rotary stage on optical table [B].

We assembled a rotary motion system to hold the cartridge and rotate it in defined steps. The components of this motion system are shown in Figure 2.6, which included a cartridge holder (fabricated in-house), a motorized rotary stage (B4836TS-ZR, Velmex Inc.), an optical breadboard (“TD” series, Newport), a motor controller (NF90, Velmex Inc.), and a software interface running on a Windows XP laptop. The cartridge holder was an acrylic plate (65mm x 65mm) mounted with a nylon screw on a 13-mm diameter by 17-cm acrylic shaft; for center-of-rotation measurements, the plate was replaced on the shaft with a capillary source holder, an acrylic piece with a 1.5-mm diameter by 4.5-mm deep hole to hold a glass capillary tube (22-362-574, Fisherbrand). The shaft fastened to an acrylic baseplate, which then attached to the face of the rotary stage. The rotary stage was mounted on its side to the optical breadboard using an
angle bracket. The motor controller provided power to the stage’s stepper motor, as well as communicating with the laptop via USB. The software interface (Figure 2.7) was written in labVIEW (v.13.0f2, National Instruments) using library code provided with the motor controller; this software allowed the stage to be rotated in either direction by a user-specified angle and logged each motion command.

![LabVIEW Software Interface](Figure2.7.png)

Figure 2.7. Screenshot of the labVIEW software interface

2.3.3. Acquisition setup

The setup for image acquisition is shown in Figure 2.8. The optical breadboard holding the rotary stage and acrylic mounting components was squared to the end of the patient table. Lateral adjustments of the breadboard’s position on the table were used to manually align the stage’s axis of rotation over the pinhole. The motor controller was connected to the stage, as well
as to the laptop. The Symbia T system’s hand-controller was used to put the patient table at a height of 14.5 cm and the detector at a radius of 5 cm, as reported on the gantry control screen; this put the center of rotation at a distance of 73 mm above the pinhole. The patient bed was moved in or out of the gantry as necessary to center the cartridge or capillary source above the pinhole.

For center of rotation measurements, a capillary tube containing a 20-mm length of I-131 was inserted into its holder and secured with a piece of tape; this holder placed the capillary tube at a radius of 25 mm from the rotation axis.

Figure 2.8. Photograph of pinhole SPECT imaging setup, showing the cartridge mounted to the rotary stage assembly. The cartridge was held above the pinhole collimator and gamma camera. The rotary stage assembly was positioned at the end of the patient table. The camera above the cartridge was not used.

For cartridge imaging, the cartridge was taped to its holder plate. The cartridge holder and the capillary source holder could be exchanged without moving the rest of the rotary motion system.
2.3.4. Image Acquisition

Because the rotary motion system was manually positioned above the collimator, a center of rotation measurement was obtained for each imaging session; if the rotation axis was not centered above the pinhole, a center of rotation offset correction must be incorporated during image reconstruction. To obtain this data, the capillary source holder was mounted onto the rotary stage. Projection images were acquired at 20° intervals over an angular range of 360°; the acquisition time was 300 sec per projection. A non-linear least-squares fit of the projection images to a modified sine function was used to determine the lateral position of the rotation axis relative to the pinhole and the image pixel matrix, as described in Appendix A.

To image the cartridge, the cartridge holder was mounted onto the rotary stage. Projection images were acquired at 6° intervals over 360°, with an acquisition time of 30 sec. The acquired pixel size was 1.48x1.48 mm, the matrix size was 128x128, and the zoom was 3.2x. The start time and current angle was logged for each projection, although decay correction per projection was not performed because the overall imaging time was much less than the half-life of I-131.

2.3.5. Image Reconstruction

Tomographic image reconstruction was performed from the pinhole projection images using an algorithm developed by Dey. [Dey 2012, Dey 2011, Bhusal 2011] This iterative reconstruction algorithm was an ordered-subset-expectation-maximization (OSEM) method. It provided resolution recovery for pinhole aperture size. The data was reconstructed using 3 subsets and 65 iterations into a 100x100x100 grid of 1x1x1 mm³ voxels. No attenuation correction was performed, because of the relatively small size of a cartridge; if desired, attenuation maps could be measured by computed tomography imaging.
The slices containing the filter papers were visually assessed for image quality, particularly the impact of limited spatial resolution and Poisson statistical noise for resolving the small features of the I-131 spot patterns. Slices across acquisitions, where the I-131 decayed appreciably, were used to estimate the projection acquisition time that would be required to achieve comparable imaging quality, when imaging an actual field air sampling cartridge that is expected to have very low activity.
Chapter 3. Results and Discussion

3.1. Cartridge Production

The I-131 spiked filter papers are shown in Figure 3.1 for the first cartridge and Figure 3.2 for the second cartridge. The assembled AgZ cartridge is shown in Figure 3.4. Once sealed, the cartridges were not reopened until after conjugate counting and SPECT imaging were completed.

![Pattern 1 Pattern 2a Pattern 2b](image)

Figure 3.1. Filter papers inserted into cartridge 1

To measure the I-131 on each filter paper in cartridge 1, after all data acquisitions were finished and the source had decayed to a low level, the filter papers were removed from the cartridge and counted with a well-characterized HPGe system (2002 C, Canberra). Based on the calibration and efficiency of the HPGe system, the activity was measured and used to plot a decay curve.
The activity placed on all three filter papers for the first cartridge was determined to be 202 µCi.

The amount of activity on each separate filter paper in cartridge 2, based on the volume and number of drops along with the activity concentration of the original iodine solution, was calculated to be 0.38 mCi, for a total of 1.11 mCi of activity in cartridge 2.

These activity measurements were later verified using a dose calibration system (Capintec, 55tW). When the activity on these filter papers had decayed to much lower activity after the
conjugate counting the filter papers were also counted using the HPGe detector. The activity obtained by this verification was lower than the activity obtained by the dose calibrator due to possible loss of iodine and so was not used in subsequent equations.

![Photograph of iodine cartridge. The AgZ and filter papers were interleaved as shown in Figure 2.2 and associated text.](image)

Figure 3.4. Photograph of iodine cartridge. The AgZ and filter papers were interleaved as shown in Figure 2.2 and associated text.

Figure 3.5 shows the assembled Ba-133 cartridge, including the secondary containment provided by the sealed plastic bag. Over time, a small amount of Ba-133 leaked from the cartridge, likely from gaps in the silicone sealant, as evidenced by the green tint that had been added to the solution. This loss was slow, and not appreciable over the time of this study.
3.2. Conjugate Counting

The measurements of peak absolute efficiency for the conjugate counting system are given in Table 3.1, as measured with the barium phantom. As expected, efficiency increased as the distance to the detector decreased. Because the barium cartridge is not a point-like source, the efficiency did fall off as rapidly as the $1/r^2$ trend with distance expected from a point source.

Table 3.1. Table summarizing parameters and results for efficiency measurements. The 2.11 μCi Ba-133 source had an emission rate of 48403.4 gammas/sec for the 356 keV peak. Error is based on Poisson counting statistics.

<table>
<thead>
<tr>
<th>Distance [m]</th>
<th>Counting time [s]</th>
<th>Net counts in ROI</th>
<th>Efficiency $\varepsilon_{\text{peak,abs}}$</th>
<th>Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5</td>
<td>180,000</td>
<td>388800</td>
<td>4.46E-05</td>
<td>9.28E-07</td>
</tr>
<tr>
<td>1</td>
<td>72,000</td>
<td>328320</td>
<td>9.42E-05</td>
<td>1.80E-06</td>
</tr>
<tr>
<td>0.5</td>
<td>18,000</td>
<td>274680</td>
<td>3.15E-04</td>
<td>5.51E-06</td>
</tr>
<tr>
<td>0.25</td>
<td>2,200</td>
<td>104544</td>
<td>9.82E-04</td>
<td>1.06E-05</td>
</tr>
</tbody>
</table>

For the first iodine cartridge, Figure 3.6 plots activities calculated from measured count rates from the front and back faces, as well as from the conjugate mean count rate from both
faces, acquired over a span of more than 7 half-lives of I-131. The activity was calculated using the count rates from side 1 and side 2 separately, and from the conjugate mean of the count rates of the two sides. The activity calculations for the separate sides included efficiency but did not include an attenuation correction factor. The line superimposed on the graph shows the expected decay curve of I-131, calculated from the activity measured with the HPGe detector. The conjugate counting results generally bracket the exponential decay curve, while the single-face results fall consistently below the curve. The average percent difference from theoretical for the conjugate counting method was 8.8%; the percent difference for counting the front and back of the cartridge were 28.4% and 25.1%, respectively. These differences are statistically significant at the 95% confidence level (p < 0.001). The contribution of Poisson counting statistics to the relative uncertainty in each data point was small, no more than 2.6%; the major contributor was the uncertainty in the I-131 activity that was placed on the filter papers. Overall, the error bars were on the same order as the size of the plotting symbols.

It is evident that the conjugate counting method was more accurate than counting of either face alone. This improved performance was the result of being able to correct the conjugate mean measurement for total attenuation.
One could conceivably improve the single-face estimates by including an attenuation correction factor, based on an assumed average pathlength of radiation through the AgZ cartridge. However, without knowing the nominal activity distribution of the cartridge, the “correct” average pathlength to use is unknown.

![Graph showing activity decay over time](image)

**Figure 3.6.** Comparison of the activity in the iodine cartridge calculated from conjugate counting vs. counting either face separately. The solid line is theoretical decay curve for I-131, based on activity measured with HPGe. The error bars are not pictured; the maximum error did not exceed 2.56%.

For the second I-131 cartridge containing 556 mCi (at the time of counting), counting measurements with the cartridge at a distance of 1.55 m yielded activity estimates of $463.20 \pm 11.90 \mu\text{Ci}$ from one face, $386.71 \pm 9.95 \mu\text{Ci}$ from the other face, and $562.18 \pm 14.51 \mu\text{Ci}$ for the conjugate mean. The percent differences from the expected activity were $16.72\%$, $30.47\%$, and $-1.07\%$, respectively. This measurement confirmed the results from the first
cartridge, that conjugate counting is significantly more accurate than counting of either face alone.

3.3. SPECT Imaging

Pre reconstructed images from the SPECT system are shown in Figures 3.7 and 3.8. Figure 3.7 shows the images from the parallel hole collimator, which shows a heavy star artifact that indicated significant gamma penetration through the lead of the collimator. Figure 3.8 shows the images from the parallel hole collimator, which are much more magnified. Reconstructed images from the SPECT system are shown in Figure 3.9. Images obtained from the SPECT imaging showed a distinct pattern that matched the pattern placed onto the filter papers. The spots were distinct from each other to a few millimeters, even when using a poor reconstruction algorithm.
Figure 3.7. Images from the parallel hole collimator. The scale is set to max counts on the black and min counts on the white.
Figure 3.8. Images from the pinhole collimator. The scale is set to max counts on the black and min counts on the white.
The activity in the cartridge during the 8-17 imaging was sufficient to identify the pattern on the filter papers but was not enough to determine the pattern accurately during the 8-24 and 8-31 imaging.

Figure 3.9. Comparison of image reconstruction for each filter paper and each date of imaging to the patterns inserted. Each image is auto corrected individually. The scale for each date had black as the zero value and the max value at 140,000 [8/17], 65,000 [8/24], and 24,000 [8/31] counts for the white.

The three largest spots are apparent in each image and the six smallest dots can be identified as spots of little to no activity.
The layout of each filter paper in the cartridge is also apparent, which shows confidence that this method can determine whether a cartridge is front loaded or homogenous. The resolution is high enough to show the 6 mm spots resolved, the 5 mm spots less so, the 4 mm faintly, and the 3 mm not at all. The 8-24 and 8-31 image sets show a significant loss of image quality due to lower counts, therefore the 8-17 image set is the target for achievable, useful image quality. To get image quality comparable to the 8-17 images, using a max activity of 10 μCi, an imaging time of 1500 s is required. For a total of 60 projections, this corresponds to a 25 hour acquisition time. Decay correction should be utilized for each projection during this acquisition due to the long time of imaging, although we did not use these corrections for our imaging.
Chapter 4. Summary and Conclusions

4.1. Cartridge Production

We produced two different types of cartridges for this study, one with drops of radiiodine on a filter paper and one with liquid barium in water. Both were used in the conjugate counting section but only the iodine cartridge was used in the imaging section. Overall, the cartridge fabrication process was manageable and the cartridges could be used for multiple measurements; however, some improvements are recommended for the future.

Opening the cartridge was straightforward by cutting off the top with a Dremel tool, but the process was time-consuming; either more practice at hand cutting or the development of cutting jig would likely lead to better consistency in separating the top from the cartridge. When putting the cartridge back together, several layers of vinyl adhesive tape was required for adequate adhesion; a more permanent solution such as cyanoacrylate glue would hold better, but would have precluded the removal of the filter papers for assaying in the dose calibrator or HPGe system, after conjugate counting and imaging were finished.

The filter paper production had only minor problems. In particular, the iodine solution obtained from the vendor had a relatively low concentration, making it difficult to put all of the iodine activity onto the filter papers without a large amount of droplet spread. A higher concentration solution would simplify the fabrication of the filter papers, especially those to be imaged.

To make the barium cartridge, a small hole drilled in the cartridge’s side would have been adequate to remove the AgZ and put in the barium solution; such a small hole would have been easier to seal, and the secondary containment of the vacuum seal bag would have been
unnecessary. Alternatively, a shrink-wrap type bag around the cartridge would have been less cumbersome than the vacuum seal bag.

4.2. Conjugate Counting

We found measures of the activity inside the cartridge using two count rates from each side of the cartridge. The average percent difference from the measured activity of both faces of the cartridge was much higher than the average difference of the conjugate counts. The conjugate count rate had the greatest agreement with the true activity in the cartridge and showed that using this method produces a more accurate reading of the activity in the cartridge. Using a similar geometry of the cartridge with a barium cartridge is reasonable and can be used for calibration and efficiency measurements outside of the field. Counting both sides of the cartridge should not be a burden in the field and the calibration should be done beforehand. It is clear that this method provides an accurate assessment of the activity within a cartridge without extra effort in the field, using calibration data acquired beforehand.

4.3. SPECT Imaging

We found that the patterns placed on the filter cartridges could be identified using the 8-17 image set. This set could also identify the different depths of each filter paper inside the cartridge. Using this method is adequate to obtain images with resolution on the order of 6 mm, and adequate to differentiate front loaded vs. homogenous distribution. Imaging a cartridge with assumed 10 µCi activity would require 25h to replicate our methods which seems manageable, considering this would not be done for every single cartridge.

4.4. Future Work

A simple continuation of this study could do attenuation and efficiency measurements for various distances and detectors to provide a table for use in calibration. Making a real cartridge
in the lab was not within our abilities but if we could obtain an actual field sampled cartridge, we would be able to image it using our methods. Making a real world cartridge in the lab would be useful to show a realistic distribution with a known activity where we could control the amount of activity in the cartridge. We could also use a different isotope of iodine in the lab, such as I-123, for better image quality. One could use this method to determine the effect of various collection parameters on the distribution of iodine. Implementing some improvements in barium cartridge production could give a more permanent, robust implementation using a solidified barium material rather than a liquid. This cartridge would be useful not only for the conjugate counting but also for other field calibration, leading to a better calibration because of similar geometry. These measurements could be repeated on cartridges with a wide variety of numbers and locations of filter papers to investigate the robustness of the conjugate counting methods versus source distribution. Optimizing the acquisition and reconstruction of the images obtained during the SPECT imaging for better resolution could also be a future line of work.
Appendix. Supplemental Equations

Fig. A1. Geometry used to determine center of rotation (COR) for pinhole SPECT imaging. The radioisotope source was located at $(x,z)$, rotating with radius $r$ about a rotation axis located at $(x_c,b+a)$. The pinhole was located at $(x_p,b)$, projecting the source to $(x_s,0)$ at the detector plane. The distance from pinhole to detector plane was $b$, while the distance from the pinhole to the rotation axis was $a$.

COR (Center of Rotation) calculation

- Analysis performed in IDL (v. 8.7.1, Harris Geospatial Solutions, Inc.)
- The projection images were loaded from the DICOM files
- All rows of each projection image summed to reduce noise, creating a 1D profile in $x$-direction for each projection
- Non-linear least-squares fit using Gaussfit routine was done to determine the center (peak location) of each profile.
- The set of center locations were fit to a sinusoidal function (details below and see Fig. A1) using the non-linear least-squares Curvefit routine, which called a custom IDL function that implemented the sinusoidal fitting function and its partial derivatives.
  - The fitting function had 5 fitting parameters: $r$, $a$, $b$, $x_c$, $x_p$ (cf. Fig. A1). The $r$, $b$, and $x_p$ parameters were held fixed, allowing only $a$ and $x_c$ to vary. Sample result is shown in Fig. A2
  - The fitted value $x_c$ was the COR offset specified to the SPECT pinhole reconstruction algorithm.
Derivation of fitting function for pinhole COR measurement:

Referring to Fig. A1, the source was located at \((x, z)\), at a radius \(r\) and an angle \(q\) on the path of rotation; the rotation axis was \((x_c, b+a)\). The pinhole was at \((x_p, b)\). The projected location of the source on the detector was \((x_a, 0)\). For any source location, the source, the pinhole, and the projected location must be collinear, satisfying

\[ \begin{vmatrix} x & z & 1 \\ x_p & b & 1 \\ x_a & 0 & 1 \end{vmatrix} = 0 \]

Solving for \(x_a\) gives

\[ x_a(\theta; r, a, b, x_c, x_p) = \frac{-b(r \cos \theta + x_c) + x_p(r \sin \theta + b + a)}{r \sin \theta + a}. \]
This is the fitting function to be applied to the measured COR projection images, allowing $x_c$ to be determined.

The curvefitting routine requires the partial derivatives of the fitting function for each fit parameter. The partial derivatives are

\[
\frac{dx_a(\theta; r, a, b, x_c, x_p)}{dr} = b \cdot \frac{-a \cos \theta + (x_c - x_p) \sin \theta}{(r \sin \theta + a)^2}
\]

\[
\frac{dx_a(\theta; r, a, b, x_c, x_p)}{da} = b \cdot \frac{(r \cos \theta + x_c) - x_p}{(r \sin \theta + a)^2}
\]

\[
\frac{dx_a(\theta; r, a, b, x_c, x_p)}{db} = \frac{-(r \cos \theta + x_c) + x_p}{r \sin \theta + a}
\]

\[
\frac{dx_a(\theta; r, a, b, x_c, x_p)}{dx_c} = \frac{-b}{r \sin \theta + a}
\]

\[
\frac{dx_a(\theta; r, a, b, x_c, x_p)}{dx_p} = 1 + \frac{b}{r \sin \theta + a}
\]

The fitting function and partial derivatives were encoded into an IDL procedure for use by the non-linear least squares curvefitting routine:

```idl
; CALLING SEQUENCE:
;   pinholecorprojection, theta, params, f [, pder]
; INPUTS:
;   theta:  Values for source location, theta, in radians
;   params: Array containing the fitting parameters
;  input order for params[... ] is [r, a, b, xc, xp]
;  r = radius of rotation of line source
;  a = distance from pinhole to rotation axis
;  b = distance from detector to pinhole
;  xc = center of rotation (relative to image coordinates)
;  xp = center of pinhole (relative to image coordinates)
; OUTPUTS:
;   f:  Calculated value of the function at each theta(i)
;   params: Array containing updated values of the fitting parameters
;
; OPTIONAL OUTPUT PARAMETERS:
;   pder: An array of the size (N_ELEMENTS(theta), 5) that contains the partial derivatives. pder(i, j) represents the derivative at the i'th point with respect to j'th parameter.
; PROCEDURE:
;   f(theta) = ( -b*(r*COS(theta)+xc) + xp*(r*SIN(theta)+a+b) ) / ( r*SIN(theta) + a )
PRO PinholeCORprojection, theta, params, f, pder
ON_ERROR, 2
  ;Return to caller if an error occurs
  cz = cos(theta)
  sz = sin(theta)
  pp = params[0]*cz + params[3]
  qq = params[0]*sz + params[1]
  rr = qq + params[2]
```
\[ f = \left( -1 \cdot \text{params}[2] \cdot pp + \text{params}[4] \cdot rr \right) / qq \]

IF N_PARAMS(0) LT 4 THEN RETURN ; NEED PARTIALS?

pder = FLTARR(N_ELEMENTS(theta),5) ; YES, COMPUTE PARTIALS

\[
pder[*,0] = \left( -1 \cdot \text{params}[1] \cdot a[2] \cdot cz + \text{params}[2] \cdot sz \cdot ( \text{params}[3] - \text{params}[4]) \right) / qq^2
\]

\[
pder[*,1] = \left( \text{params}[2] \cdot pp - \text{params}[4] \cdot \text{params}[2] \right) / qq^2
\]

\[
pder[*,2] = \left( -1 \cdot pp + \text{params}[4] \right) / qq
\]

\[
pder[*,3] = -1 \cdot \text{params}[2] / qq
\]

\[
pder[*,4] = rr / qq
\]

RETURN

END
References


Louisiana Department of Environmental Quality, Title 33. Part 15: Radiation Protection.


Vita

Daniel Joseph DiMarco was born in Marrero in 1994. Daniel grew up in Marrero, Louisiana and attended Brother Martin High School. He graduated from Brother Martin in 2012 and attended Louisiana State University, where he received his Bachelor of Science degree in Physics. After graduating from LSU in 2017, he matriculated into the Health Physics Master of Science Program at LSU. Following graduation from the health physics program in the fall of 2019, he will join the workforce as a health physicist.