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Non-Destructive Bulk Soil Analysis for a Chlorinated Compound using Prompt Gamma-ray Neutron Activation Analysis

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A prompt gamma-ray neutron activation analysis (PGNAA) system was evaluated for the quantification of chlorinated compounds in soil. The system evaluation was divided into two phases. In phase one, the response of an n-type HPGe detector (20% relative efficiency) to point sources of ^{60}Co and ^{152}Eu was determined experimentally and used to calibrate an MCNP4a model of the detector. The refined MCNP4a detector model can predict the absolute peak detection efficiency within 7% in the energy range of 120 - 1400 keV. In phase two, a PGNAA system consisting of a light-water moderated ^{252}Cf (1.06 μg) neutron source, and the shielded and collimated HPGe detector was used to collect prompt gamma-ray spectra from Savannah River Site (SRS) soil spiked with chlorine. The experimental system response was used to calculate the minimum detectable concentration of chlorine in the SRS soil for a 1800 sec. irradiation as 2200 μg/g based on the analysis of the 788 keV gamma-ray. MCNP4a was used to predict the PGNAA system response, which was accomplished by analyzing the neutron and gamma ray transport components separately. In the energy range of 788 to 6110 keV, the MCNP4a predictions were generally within 60% of the calculated probability of detection of a prompt gamma ray based on the experimental data.

INTRODUCTION

Contamination of soils by chlorinated compounds poses a serious challenge at many industrial and commercial sites. The widespread use of highly chlorinated compounds such as the degreasing agents trichloroethylene and tetrachloroethylene, polychlorinated biphenyls used in capacitors and transformers, and pesticides like dichlorodiphenyltrichloroethane (DDT), 1,2,3,4,10,10-hexachloro-1,4,4a,5,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene (aldrin), and 1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoidene (heptachlor), and their subsequent accidental or deliberate release to the environment has led to contamination of soils and groundwater throughout the United States. For the purpose of site characterization and remediation, quantification of the level of soil contamination is important. The choice of the optimal technique for a specific analytical need is a balance between required sensitivity and sample preparation/characterization time.

Prompt gamma-ray neutron activation analysis (PGNAA) is a non-destructive nuclear technique that can be used for identification and quantification of elements in gas, liquid, or solid samples. PGNAA has been used effectively in research reactor based analyses as well as non-reactor based measurements, but the primary use has been at nuclear research reactors because the high neutron flux results in greater sensitivity. The applicability of PGNAA to detect chlorine or chlorinated compound analysis was first shown in the early work of Henkelmann and Born (1973) which placed the PGNAA detection limit for chlorine at 0.05 μg/g for a reactor based facility in Munich; this detection limit is notably the best of those referenced. Anderson et al. (1981) placed the detection limit for chlorine in the range of 1 to 10 μg/g for a research reactor system at the University of Maryland, which is the same order of magnitude reported by Yonezawa (1997) for a reactor system in Japan.

The use of cold neutron facilities at research reactors has been studied as a method by which the effectiveness of the moderated neutron flux could be increased. Cold neutrons would have the capability of making laboratory based PGNAA a better analytical tool for sample characterization (Lindstrom et al., 1987). Cold neutron PGNAA facilities have been installed at a number of nuclear research reactors (e.g. Lindstrom et al., 1993; Clark et al., 1992; Wehring et al., 1997). It has been reported that if cold neutrons were used, the chlorine detection limit at the reactor based facility using a neutron guide could be as low as 0.5 μg/g (Yonezawa, 1993).

Non-reactor based PGNAA has been used in industrial process stream analysis as well as environmental field measurements. PGNAA has been investigated for analysis of major components of coal, ash and concrete (Duffey and Wiggins, 1986; Yaun et al. 1986). Examples of environmental measurements in the field include: rapid, in-situ quantification of Cd and Hg down to the μg/g level in water (Chung and Tseng, 1988) and quantitative analysis of elemental composition of the vadose zone (Evans et al. 1982; Frankle and Conaway, 1997). As a means towards increasing the sensitivity of the portable system, a “cold neutron irradiator” (CNI) was been designed utilizing a cryogenic moderator (Clark, 1994).

Monte Carlo methods have been used to model radiation detector response and PGNAA systems. Specialized as well as general Monte Carlo codes have been used to predict relative detector response (e.g. Clark et al, *1982*). Modeling of PGNAA necessitates a Monte Carlo code capable of modeling neutron and photon interactions and transport. A Monte Carlo code is valuable in minimizing the time spent on system calibration and optimization, and assessing detection limits (Yuan et al., 1987). Monte Carlo methods have also been used to aid in the analysis of the data from a PGNAA system (Shyu et al., 1993; Frankle and Conaway, 1997).

EXPERIMENTAL METHODS

The research presented is divided into two phases: experimental evaluation and modeling of: 1) the high purity germanium (HPGe) detector used in the PGNAA system and 2) the complete PGNAA system. Phase one involves experiments and simulations of the response of an n-type coaxial HPGe gamma-ray spectrometer to point sources. Phase two involves experiments and simulations of a PGNAA system for chlorinated compound detection in bulk soil samples utilizing the same detector evaluated and modeled in phase one.

A Canberra[1] n-type HPGe (20% relative efficiency) detector was evaluated by comparing experimental data and theoretical data obtained from Monte Carlo simulations. For the experiments, the gamma-ray spectra were obtained using point sources (0.74 μCi ^{60}Co and a 0.90 μCi ^{152}Eu) from Isotope Products Laboratories[2] which were each placed along the centerline of the detector at a distance of 25 cm from the detector endcap. The signals from the detector were processed by a spectroscopy amplifier and collected with a Canberra S100 multichannel analyzer system. Net peak areas were used to calculate the absolute peak detection efficiency, $e_{abs}^{peak}(E)$, as a function of energy for the given geometry.

The Monte Carlo modeling was accomplished using Monte Carlo N-Particle Transport Code System version 4a (MCNP4a) run on a PC platform. Using the physical dimensions supplied by the manufacturer, a MCNP4a model was created to simulate the HPGe detector response to the point sources. The model was run separately for each gamma-ray energy corresponding to ^{60}Co (1332 keV) and ^{152}Eu (121, 344, and 1408 keV) emitted from a simulated point source 25 cm from the face of the modeled detector. The gamma rays were simulated at an intensity of 100% and then subsequently multiplied by their branching ratios. MCNP4a simulations were compared to the experimental measurements.

A PGNAA system at the Savannah River Technology Center was evaluated experimentally and with a Monte Carlo model to determine minimum detectable chlorine concentration in soil. The PGNAA detection system consisted of a 1.06 μg ^{252}Cf neutron source with 0.03 m^3 of light water moderator surrounded with cadmium sheeting and four inches of lead shielding. A sample tube was constructed through the moderator box. The distance from the centerline of the sample tube to the ^{252}Cf source was 10.7 cm. The sample was placed in the sample tube horizontally at a distance of 21.3 cm. from the face of the n-type HPGe detector.

Calibration of the PGNAA system was performed using various amounts of chlorine (in the form of KCl) in a soil matrix. Four 250 mL spiked soil samples containing 0% (blank), 2.5%, 5%, and 7.5% chlorine by mass were created by adding the appropriate amounts of reagent grade KCl to Savannah River Site (SRS) soil. Samples were homogenized by shaking vigorously by hand. A sample containing approximately 48% chlorine by mass (100% KCl) was also created. PGNAA spectra of the spiked soil samples and the KCl sample were then collected for 1800 seconds.

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[1] Canberra Industries Inc., Meriden, CT

The Monte Carlo modeling of the complete PGNAA detection system was accomplished using MCNP4a. The system components (detector, moderator box, sample bottle, etc.), materials (lead, germanium, cadmium, air, soil, etc.), and geometries (sample source distance, sample detector distance, etc.) were modeled. The soil was modeled as 60% silicon, 20% iron, and 20% aluminum. The PGNAA modeling proceeded in two parts: 1) neutron transport/sample excitation and 2) gamma-ray transport. This was necessitated by a limitation in the MCNP4a code such that the same model could not simultaneously transport neutrons and predict the prompt gamma-ray energy deposition in the detector. The first part of the PGNAA model involved the neutron transport and neutron reaction in the sample. Source neutrons were modeled having the characteristic neutron energy spectrum of ^{252}Cf. The neutron flux in the sample material was recorded as a function of energy. The model then calculated the probability of a radiative capture reaction with chlorine per source neutron from ^{252}Cf per unit sample volume, $P_{n,\gamma}$, as follows:

$$P_{n,\gamma} = \frac{1}{n_s} \int_t \int_V \int_E \phi_n(E) \cdot \sigma(E) \cdot N_{Cl} \cdot dV \cdot dt \cdot dE \qquad (1)$$

where: n_s = number of neutrons started in the MCNP run,
E = neutron energy,
σ (E) = microscopic radiative capture cross-section of chlorine, in barns, as a function of energy,
N_{Cl} = atom density for chlorine in the sample in units of atom/barn·cm,
V = volume of sample in cm^3,
t = time in seconds,
ϕ_n (E) = energy dependent neutron flux in units of neutrons cm^2 per second.

The second part of the PGNAA model involved the gamma ray transport. Gamma rays of energies characteristic to chlorine prompt gamma rays were transported from the sample material to the detector using the optimized detector model where the absolute peak detection efficiency, $\varepsilon_{abs}^{peak}(E)$, was recorded as a function of energy. $\varepsilon_{abs}^{peak}(E)$ was computed by dividing the number of photons incident of a given energy that were detected by the number emitted from the source. The probability of a chlorine prompt gamma-ray of a given energy being detected per source neutron per unit sample volume, P(E), was then calculated by combining the results from parts one and two as follows:

$$P(E) = P_{n,\gamma} \cdot \varepsilon_{abs}^{peak}(E) \cdot f(E) \qquad (2)$$

where: f (E) = branching ratio for chlorine prompt gamma ray at each energy.

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[2] Isotope Products Laboratories, Burbank, CA 91504

The experimental value of P(E) is determined by the ratio of the number of net counts in a given gamma-ray peak to the product of the number of source neutrons emitted, the branching ratio for a given prompt gamma- ray energy and the sample volume.

The sensitivity of the PGNAA system to chlorine was calculated and reported in the form of a minimum detectable concentration (MDC). Minimum detectable concentration (MDC) is an *a priori* estimate of the detection level based on a specified probability of a false detection and non-detection. For a five-percent chance that a false conclusion will be reached regarding the presence or absence of the element of interest in the sample, Currie (1968) defined the quantity called the lower limit of detection (LLD) via the following equation:

$$\mathrm{LLD} = 2.71 + 4.65\sqrt{B} \qquad (3)$$

where: B = gross background counts.

Equation 3 was derived for data that is Gaussian distributed, and has the same background and sample count time. The MDC in units of percent chlorine (10^4 μg/g) is calculated from the LLD as follows:

$$\mathrm{MDC} = \frac{\mathrm{LLD}}{\mathrm{n} \cdot \mathrm{V}} \times \frac{1}{\mathrm{C}(\mathrm{E}_g)} \qquad (4)$$

where: $C(E_\gamma)$ = Calibration factor in units of probability of a count per source neutron per unit sample volume per percent chlorine

n = calculated number of source neutrons emitted from source during experiment

V = volume of the sample in units of cm^3.

$C(E_\gamma)$ can be determined experimentally from the data or from the numerical model results and is related to P(E), via the following relationship:

$$\mathrm{P(E)} = \mathrm{a}\ \mathrm{C}(\mathrm{E}_g)^{\mathrm{b}}$$

where a and b are fitting parameters.

RESULTS AND DISCUSSION

A spectrum of the ^{60}Co point source was taken for the purpose of evaluating the ability of a MCNP4a Monte Carlo model to predict the observed detector response. The experimental absolute peak detection efficiency for the 1332 keV gamma ray was 2.19×10^{-4} for this geometry. An MCNP4a simulation of this experiment using the actual physical dimensions of the HPGe crystal resulted in a predicted absolute peak detection efficiency of 2.66×10^{-4} , which is 22% higher than the experimental result. Such disagreement is not unprecedented. Sanchez et al. (1991) and Clouvis et al. (1998) both found that theoretical results based on manufacturer's specifications of the detector size could not predict experimental results. This disagreement can be attributed to non-ideal charge collection in the germanium crystal. Sanchez et al. (1991) removed a 0.8-mm layer of germanium and added a 0.5-mm layer of aluminum around the crystal in order to compensate for the differences between the theoretical and experimental results. Clouvis et al. (1998) found it necessary to remove a 2 mm layer of germanium from the active volume to more accurately model their crystal and reconcile the results of their modeling efforts to actual experimental data.

Figure 1 summarizes the results of a series of MCNP4a models used to determine the effect of increasing thickness of inactive germanium, i.e. reductions in active volume while maintaining total volume, on the predicted absolute peak detection efficiency. For the n-type detector used in this study, the inactive germanium was modeled on the cathode side of the detector. Using this data set, the inactive germanium thickness of greater than 2.25 mm would be needed to match the experimental and theoretical results.

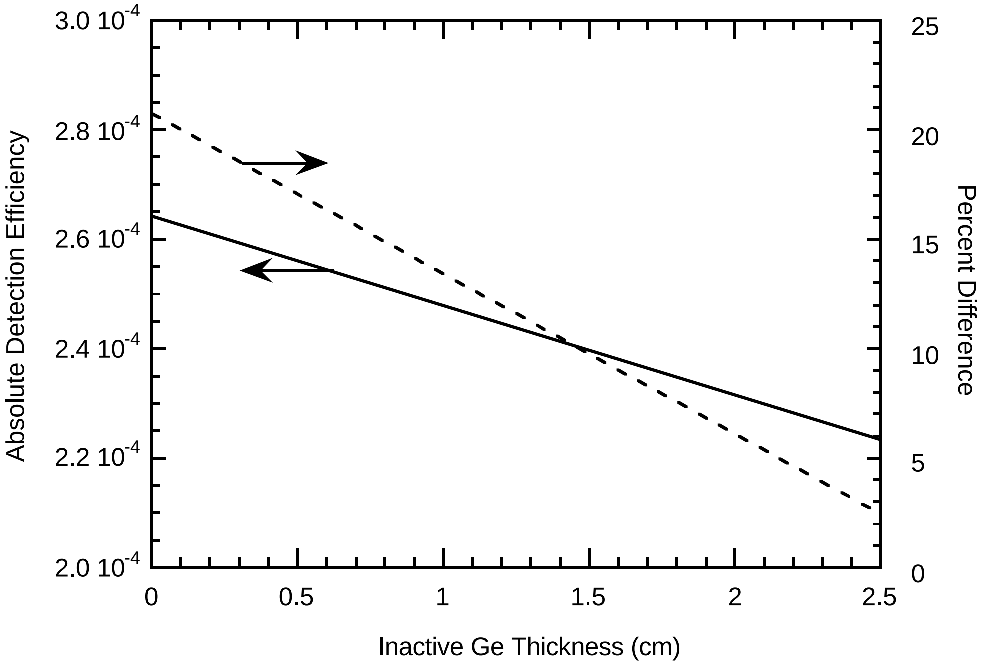

Figure 1. MCNP4a model results of ^{60}Co peak (1.332 MeV) absolute detection efficiency with various inactive germanium thicknesses.

A spectrum of the ^{152}Eu point source was acquired to evaluate the ability of the MCNP4a Monte Carlo model to predict the HPGe detector response over a range of energies and to better predict the inactive germanium thickness. The ratio of absolute peak detection efficiencies was experimentally determined to be, $e_{abs}^{peak}(121\ keV)/e_{abs}^{peak}(1408\ keV) = 11.04$ and $e_{abs}^{peak}(344\ keV)/e_{abs}^{peak}(1408\ keV) = 4.58$. The ^{152}Eu gamma rays were modeled individually at an intensity of unity, then multiplied by the branching ratios, 0.284, 0.266, and 0.208, for the 121, 344, and 1408 keV gamma-rays, respectively, to determine the absolute peak detection efficiency as a function of energy. The ^{152}Eu data indicates that the model generally under-predicted the actual experimental value, which is contrary to the ^{60}Co absolute peak detection efficiency results where the model over-predicted the experimental value. The effect of increasing inactive germanium thickness on the ratio of absolute gamma-ray detection efficiencies was explored through a series of simulations and is summarized in Figure 2. As the inactive germanium thickness was varied from 0 to 2 mm, $e_{abs}^{peak}(121\ keV)/e_{abs}^{peak}(1408\ keV)$ increased from 10.80 to 11.93 (2.2% to –8.1% difference). Over the same range of inactive germanium thickness, $e_{abs}^{peak}(344\ keV)/e_{abs}^{peak}(1408\ keV)$ increased from 4.75 to 4.92 (–3.7% to –7.4% difference). Optimizing the model between improvements in the absolute detection efficiency and the ratio of absolute detection efficiencies over an energy range led to an inactive germanium thickness of 1.25 mm being modeled on the cathode of the coaxial n-type HPGe detector. The use of this inactive germanium thickness resulted in a percent difference in the ^{60}Co absolute detection efficiency of 5% and the ^{152}Eu absolute detection efficiency ratios of -4.4% for 121 keV / 1408 keV and -6.6% for 344 keV / 1408 keV.

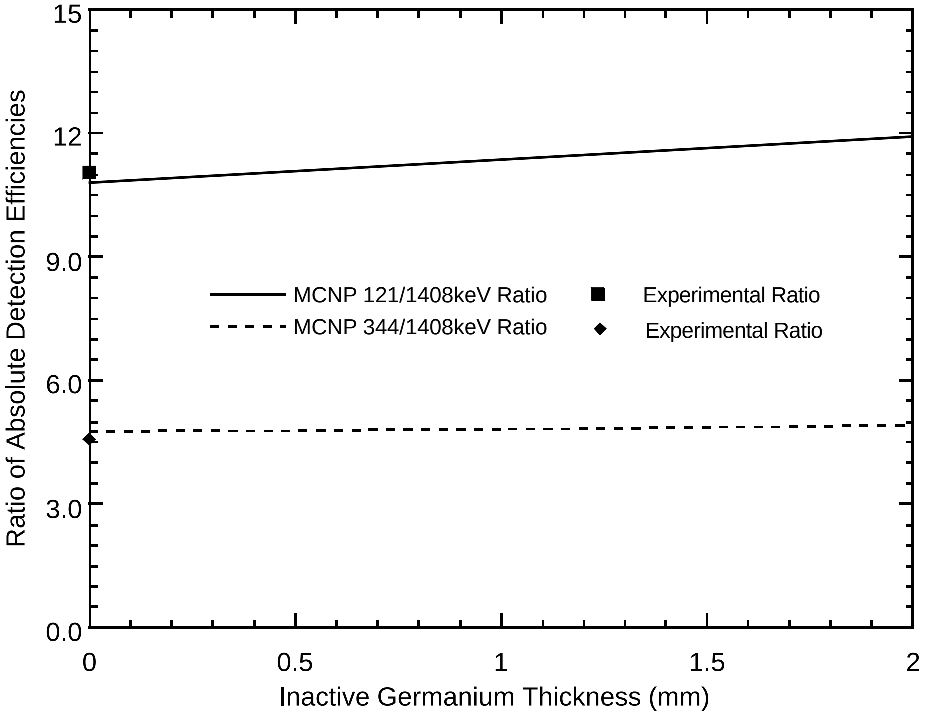

Figure 2. ^{152}Eu ratio of absolute detection efficiencies as a function of inactive germanium thickness.

The HPGe detector was used in the SRS PGNAA system to gather prompt gamma-ray energy spectra of SRS soils spiked at different chlorine concentrations (0%, 2.5%, 7.5%, and 48%) and this data was compared with the MCNP4a model. The samples were each counted for 1800 sec using the PGNAA facility and the net peak areas at E_γ = 788, 1164, 1950, 1959, and 6110 keV (corresponding to some of the major chlorine prompt gamma-ray energies) were recorded. The unspiked soil sample was used to correct the net peak areas of the spiked samples to obtain the net count rate due to the added chlorine. This data was used to calculate the experimental value of $e_{abs}^{peak}(E)$. The MCNP4a model was used to predict $P_{n,\gamma}$ and $e_{abs}^{peak}(E)$ which were used to calculate the theoretical P(E), which was compared with the experimental value. The resultant theoretical and experimental probabilities are plotted in Figure 3 as a function of percent chlorine added to the sample. MCNP4a was also used to model two additional soil samples of 15 and 30% chlorine by mass. These samples were not run experimentally, however the model results are included in the power fits of the theoretical data (see Figure 3).

Both the experimental data and the model predictions indicate a non-linear response between P(E) and added chlorine concentrations. However, the model does not correctly predict the degree of non-linearity. The non-linearity in both the experimental and modeled data suggests that some mechanism in neutron/sample interaction acts as a limiting factor. Neutron competition between sample constituents, leading to self-shielding, is a mechanism that may be responsible for the non-linear response of the detection system to increased levels of chlorine in the sample. The sample is composed of other elements such as iron, which do not have as high a cross-section as chlorine, but are present in the sample in appreciable quantities.

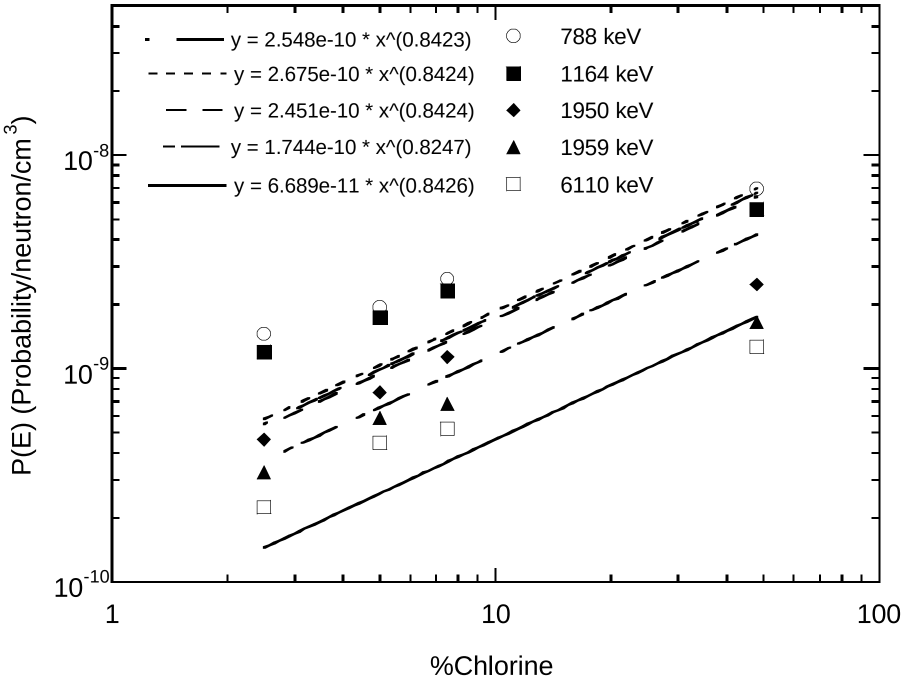

Figure 3. MCNP4a predictions versus experimental results for P(E) as a function of chlorine concentration for an 1800-second count time.

Two distinct trends between the experimental and theoretical results as a function of chlorine concentration are shown in Figure 3. The 1950 and 1959 keV data indicates that the ability of the model to predict the absolute experimental P(E) diminishes over the range of 2.5 to 48% chlorine. The 788, 1164, and 6110 keV data indicates that the ability of the model to predict the absolute experimental P(E) improves over the same range. It can be seen in Figure 3 that initially the model over-predicts the 1950 and 1959 keV data while under-predicting the 788, 1164, and 6110 keV data. These trends do not indicate an inconsistency in the experimental data, but rather an incomplete theoretical model of the detector and/or the experimental set-up.

The accuracy of the model is quite good when taken in context with the limitations and approximations in the modeling process. In the model, some neutron and photon interactions are simplified to increase the efficiency of the simulation. Neutron interaction in the germanium detector and the resulting production of prompt gamma-ray radiation is ignored in the model leading to a "lower than observed" background level. Photons were run singly with no allowance for influence of a Compton continuum from higher energy gamma rays such as the 2223 keV Hydrogen line, which also leads to lower than observed background levels. MCNP4a accumulated a probability of detection from the emission of a million simulated gamma rays instead of the tens of millions of gamma rays emitted in the experimental system. Finally, soil samples were modeled with approximate concentrations of major constituents only and complicated geometries such as the sample bottle were approximated by simple shapes such as cylinders. However, the model results agreed with the experiment to 60% with the exception of 1950 keV in the 48% chlorine sample.

The minimum detectable chlorine concentration was calculated using the experimental results from PGNAA analysis of an SRS soil blank and the experimentally determined calibration factor, $C(E_\gamma)$.

The MDC in units of μg/g are dependent on the prompt gamma-ray energy and are tabulated in Table 1.

Table 1. MDC of Chlorine for the SRS PGNAA facility for an 1800-sec. acquisition time.

Energy (keV)	MDC (μg/g)
788	2200
1164	3000
1950	5200
1959	7900
6110	49100

As predicted, the MDC for an 1800 second acquisition time were significantly higher than those reported in the literature for a reactor based facility. Increased sensitivity could be achieved with increased neutron flux, the use of cold neutrons, and/or an increase in count time.

CONCLUSIONS

A Canberra N-type (20%) HPGe detector was successfully modeled using the Monte Carlo code MCNP4a. Optimization of the detector model led to predictions within 5% of experimentally determined ^{60}Co absolute peak detection efficiency at 1332 keV and a percent difference in the ^{152}Eu absolute peak detection efficiency ratios for 121 keV / 1408 keV and 344 keV / 1408 keV of -4.4% and -6.6%, respectively.

A small PGNAA system, constructed around a light water moderated ^{252}Cf, was used to demonstrate the detection of chlorine in bulk soil samples. Minimum detectable concentrations were calculated for the PGNAA system and ranged from 2200 to 49100 μg/g depending on the gamma-ray energy for an 1800-sec. analysis time. The Monte Carlo code MCNP4a was used successfully to model and predict the non-linear response of the actual PGNAA system, providing a proof-of-principle that this code can be employed to predict PGNAA system response.

The MCNP4a model was generally able to predict the PGNAA system response within 60% of the experimental data.

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