MEASUREMENT OF AND CALIBRATION FOR GAMMA
SPECTROSCOPY OF MISSISSIPPI RIVER WATER

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 Nuclear Engineering

by

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B.S., Louisiana State University, 1975

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A special thank you to my parents for their support throughout my long university career.
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ABSTRACT

Natural sources and industrial operations are known to introduce radioactive materials into the Mississippi River. Public health implications of these materials have prompted studies from which limited information on gross activities and specific radionuclide inventories have been obtained. The current investigation was undertaken to extend qualitative and quantitative knowledge of specific radionuclides in river water through the application of gamma-photon scintillation spectrometry. Raw river-water samples, collected from the intake pipe at a Jefferson Parish water treatment plant, were counted as one-liter aliquots in a Marinnelli beaker for 12 hours with a 4-inch by 4-inch NaI(Tl) crystal housed in a large graded-steel shield. Net peak areas from multichannel analyzer data for gamma energies of interest were obtained by subtraction of 12-hour background counts.

A novel technique for establishing counting yield as a function of energy was developed to obtain both energy baseline and quantitative information from sample gamma spectra. This technique involved activation of selected target nuclides by Californium-252 neutrons, with the product radionuclides first being quantitated by point-source counting, and then dissolved for counting as one-liter samples in the Marinnelli beaker to provide empirical counting-yield data. Only naturally-occurring radionuclides were detected, with Potassium-40 being the major contributor; up to 90 picocuries per liter of this radionuclide were observed. Suggested improvements in methodology
include use of large-volume semiconductor detectors and longer counting times to obtain greater specificity and sensitivity.
CHAPTER I

INTRODUCTION

Radionuclides encountered in the Mississippi River have many origins, both natural and technological, with technological sources introducing synthetic as well as naturally radioactive materials. Among the naturally-occurring radionuclides are uranium, thorium, radium, and their daughters; tritium; carbon-14; and potassium-40. These enter the river through such processes as soil runoff and subsurface extraction, through industrial and agricultural operations resulting in direct plant effluents (aluminum and phosphate manufacturers), through leaching of landfills (manufacturing by-products and power plants), and through drainage runoffs from fields containing phosphate fertilizer (up to 400 parts per million uranium). Synthetic radionuclides include, among others, tritium, iodine-125, iodine-131, and cesium-137 which enter the river through atmospheric processes (fallout and washout), technological operations (power plants and research laboratories), and release from hospitals with nuclear medicine facilities. The contributive interrelationships of these various sources are diagrammed in Figure 1. It should be emphasized that dynamic biological, physical, and chemical interactions may alter or prevent radiochemical equilibria among radioactive parent-daughter decay chains.
FIGURE 1

Interrelationships of Various Natural and Technological Inputs of Radionuclides in the Mississippi River

Virgin-land sources
(U, Th, Ra, $^{14}$C, $^3$H, $^{40}$K, etc.)

- soil runoff
- erosion

Subterranean sources
(U, Th, Ra, $^{40}$K, etc.)

- spring water

River silt
(U, Th, Ra, $^{40}$K, etc.)

- sorption
- leaching

Medical sources
(Ra, $^{125}$I, $^{131}$I, $^{99}$Tc, etc.)

- sanitary sewers

Agricultural sources
(U, $^{40}$K, $^{14}$C, etc.)

- runoff
- erosion

- Classical Manufacturing including Al and P0$_4$ fert. (U, Th, Ra, $^{40}$K etc.)

- plant effluent

- landfill runoff

- Nuclear industry ($^{137}$Cs, $^{90}$Sr, $^{57}$Co, $^{131}$I, $^3$H, etc.)

- plant effluent

- waste leaching

- Atmospheric processes ($^3$H, $^{14}$C, Rn, $^{137}$Cs, etc.)

- washout
- fallout

Note: U, Th, and Ra daughter products are implied by the term etc.
Many of these radionuclides have detectable gamma radiations, some of which are listed Tables 1 and 2. Because certain daughter products of the Th, U, and Ra decay chains are identical, care must be exercised in specifying the parent nuclides.

Previous studies of baseline radionuclide content have been concerned mainly with gross alpha and gross beta activity measurements.\textsuperscript{2, 3, 4, 5}

Only in certain instances (e.g., when federal limits have been exceeded\textsuperscript{6, 7, 8, 9, 10, 11}) have any attempts been made to identify certain nuclides. For example, when a high gross alpha count rate is observed, a specific radiochemical analysis is made to determine the \textsuperscript{226}Ra content in the sample. Specific radiochemical analysis for \textsuperscript{90}Sr is performed when a high gross beta count rate is detected. These two nuclides are of importance because their chemical properties parallel those of calcium, making them bone seekers; once lodged in the bone their high specific energy deposition rates may cause well defined detrimental effects.\textsuperscript{12}

When available literature is examined in detail, it becomes evident that there has been no definitive water-quality surveillance for radioactivity from which time-dependence or specific radionuclide concentrations could be determined. Several of the literature sources reported gross alpha and gross beta activities only once or twice per year,\textsuperscript{2, 3, 4, 5} with wide variations in reported values. For example, gross alpha data

\textsuperscript{12}
from sampling stations above New Orleans ranged from less than detectable (approximately 2 picocuries per liter – pCi/l),\(^3\) to over 6,000 pCi/l.\(^2\) Identification of nuclides other than \(^{226}\)Ra and \(^{90}\)Sr has not been undertaken, although the capability of detecting other radionuclides exists.\(^3\) Several possible industrial sources of \(^{226}\)Ra and \(^{238}\)U upstream from New Orleans have been inferred from available data,\(^2\) but as yet no direct identification has been made. The effects of river level change have not been studied, although it would seem that rainfall upstream from New Orleans could leach both man-made and natural radionuclides into the river. No mention of the possible removal or addition of radionuclides during the chemical or filtration processes of water treatment has been made in available literature, nor has the possibility been reported of the addition of \(^{40}\)K and radium daughter products to water during storage in concrete reservoirs.

It is evident that a systematic investigation of radionuclide content of Mississippi River water is needed. Currently at Louisiana State University there are several different radionuclide-determination studies that are in progress. Gross activity (alpha-plus-beta) and gross alpha activity are being measured using thin-window flow proportional and GM counters. An alpha spectrometer is utilized to identify the alpha emitters. Gross beta activity is ascertained by using the low background GM counter.
To augment the work currently underway, a partial radionuclide inventory is to be established by means of gamma spectrometry. In conjunction with qualitative spectrometry, a special quantitative analytical technique utilizing tracer materials activated by Californium-252 neutrons will be employed. The gamma-emitter inventory is to be established over a sufficient period of time so that ultimately seasonal or cyclic changes may be known, understood, and predicted.

River water obtained at a single sampling station at New Orleans will be examined. This locality is considered ideal for modeling radionuclide variations in the lower Mississippi River resulting from a variety of industrial and chemical process plants located immediately upstream. Additionally, this sampling station located at the Jefferson Parish Waterworks #1 supplies a large portion of New Orleans, Louisiana with potable water.
<table>
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<th>Gamma Ray Energy (MeV)</th>
<th>Alpha T 1/2</th>
<th>Beta T 1/2</th>
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</thead>
<tbody>
<tr>
<td>$^{238}\text{U}$</td>
<td>0.069</td>
<td>4.47 x 10$^8$y</td>
<td>24.1 d</td>
</tr>
<tr>
<td>$^{234}\text{Th}$</td>
<td>0.043</td>
<td></td>
<td>1.17 m</td>
</tr>
<tr>
<td>$^{234}\text{Pa}$</td>
<td>0.053</td>
<td>2.44 x 10$^8$y</td>
<td></td>
</tr>
<tr>
<td>$^{230}\text{Th}$</td>
<td>0.068</td>
<td>7.7 x 10$^4$y</td>
<td></td>
</tr>
<tr>
<td>$^{226}\text{Ra}$</td>
<td>0.068</td>
<td></td>
<td>1600 y</td>
</tr>
<tr>
<td>$^{222}\text{Rn}$</td>
<td>0.51</td>
<td></td>
<td>3.824 d</td>
</tr>
<tr>
<td>$^{218}\text{Po}$</td>
<td>3.05 m</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{218}\text{Pb}$</td>
<td>0.352, 0.295</td>
<td></td>
<td>26.8 m</td>
</tr>
<tr>
<td>$^{214}\text{Bi}$</td>
<td>0.609, 1.12, 1.76</td>
<td></td>
<td>19.8 m</td>
</tr>
<tr>
<td>$^{214}\text{Po}$</td>
<td>0.792</td>
<td></td>
<td>1.637 x 10$^{-4}$s</td>
</tr>
<tr>
<td>$^{210}\text{Pb}$</td>
<td>0.465</td>
<td></td>
<td>22.3 y</td>
</tr>
<tr>
<td>$^{210}\text{Bi}$</td>
<td></td>
<td></td>
<td>5.01 d</td>
</tr>
<tr>
<td>$^{210}\text{Po}$</td>
<td>0.802</td>
<td></td>
<td>138.4 d</td>
</tr>
<tr>
<td>$^{208}\text{Pb}$</td>
<td>stable</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(y\) - year
\(d\) - day
\(h\) - hour
\(m\) - minute
\(s\) - second

Chart of the Nuclides, General Electric, April 1972
# TABLE 2

Detectable Gamma Energies and Other Physical Characteristics of the Naturally Occurring Radionuclides Belonging to the Primary Thorium Decay Series and Other Selected Radionuclides

<table>
<thead>
<tr>
<th></th>
<th>Gamma Ray Energy (MeV)</th>
<th>Alpha T 1/2</th>
<th>Beta T 1/2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}\text{Th}$</td>
<td>0.059</td>
<td>1.4 x $10^{10}$ y</td>
<td></td>
</tr>
<tr>
<td>$^{228}\text{Ra}$</td>
<td>0.007</td>
<td></td>
<td>5.75 y</td>
</tr>
<tr>
<td>$^{228}\text{Ac}$</td>
<td>0.911, 0.969, 0.34</td>
<td></td>
<td>6.13 h</td>
</tr>
<tr>
<td>$^{228}\text{Th}$</td>
<td>0.084, 0.216</td>
<td>1.913 y</td>
<td></td>
</tr>
<tr>
<td>$^{224}\text{Ra}$</td>
<td>0.241</td>
<td>3.64 d</td>
<td></td>
</tr>
<tr>
<td>$^{220}\text{Rn}$</td>
<td>0.54</td>
<td>55.6 s</td>
<td></td>
</tr>
<tr>
<td>$^{216}\text{Po}$</td>
<td></td>
<td>0.15 s</td>
<td></td>
</tr>
<tr>
<td>$^{212}\text{Pb}$</td>
<td>0.239, 0.3</td>
<td></td>
<td>10.64 h</td>
</tr>
<tr>
<td>$^{212}\text{Bi}$</td>
<td>0.727</td>
<td></td>
<td>60.6 m</td>
</tr>
<tr>
<td>$^{212}\text{Po}$</td>
<td></td>
<td></td>
<td>3.0 x $10^{-7}$ s</td>
</tr>
<tr>
<td>$^{208}\text{Tl}$</td>
<td>2.615, 0.583, 0.511</td>
<td></td>
<td>3.054 m</td>
</tr>
<tr>
<td>$^{208}\text{Pb}$</td>
<td>stable</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>1.461</td>
<td></td>
<td>1.28 x $10^{3}$ y</td>
</tr>
<tr>
<td>$^{60}\text{Co}$</td>
<td>1.173, 1.332</td>
<td></td>
<td>5.272 y</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>0.662</td>
<td></td>
<td>30.1 y</td>
</tr>
<tr>
<td>$^{125}\text{I}$</td>
<td>0.036</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{131}\text{I}$</td>
<td>0.365</td>
<td></td>
<td>8.04 d</td>
</tr>
</tbody>
</table>

**Note:**
- $y$ - year
- $d$ - day
- $h$ - hour
- $m$ - minute
- $s$ - second
CHAPTER II
PRINCIPLES OF ENVIRONMENTAL COUNTING

Radionuclide concentrations in raw river water were
determined using a thallium-doped sodium iodide [NaI(Tl)] crystal.
This scintillation detector is preferred to other solid-state
detectors, specifically lithium-drifted germanium and lithium-
drifted silicon, because of its greater counting efficiency
(i.e., higher energy-absorption coefficient). This efficiency
becomes important when dealing with very low levels of radiation,
which would make counting times for semi-conductor detectors
unacceptably long.

Mathematical solutions to problems involving absolute counting
with energy-dependent gamma detectors have generally assumed one
of two geometries: (a) a point source and a volume-distributed
detector,\textsuperscript{13} or (b) a point detector and a volume-distributed
source\textsuperscript{14} because of the simplifications implicit in such
assumptions. These mathematical simplifications can not be
applied to a finite-volume detector immediately adjacent to a
volume-distributed source, although a semi-empirical solution
for a right-cylinder source mounted atop a right-cylinder detector
has been developed.\textsuperscript{15}

When using a Marinelli beaker to contain the sample for
counting, geometrical considerations are far more complicated
than for point and volume-distributed sources (see Figure 2).
If a specific point p in the sample volume is selected, and is
connected linearly to a point $p^1$ in the crystal volume, such that the path length within the sample is $x_s$ and the path length within the crystal is $x_c$, and only a small solid angle $\omega$ is allowed for emission of gamma radiation along this path, then

$$P_r = \text{Probability of a gamma photon being emitted in the direction of the crystal,}$$

$$= \frac{\omega}{4\pi};$$

$$P_s = \text{Probability of a gamma photon escaping from the sample into the crystal,}$$

$$= e^{-\mu_s x_s};$$

$$P_c = \text{Probability of a gamma photon interacting in the crystal}$$

$$= 1 - e^{-\mu_c x_c};$$

in which $e$ is the base of the natural logarithm, and $\mu_s$ and $\mu_c$ are, respectively, the energy absorption coefficients for the sample and the crystal. These individual probability statements may be combined to obtain an inclusive probability, $P(p,p^1)$, that a count will be recorded for a gamma ray emitted in the sample volume:

$$P(p,p^1) = P_r \times P_s \times P_c,$$

$$= k \omega \left( e^{-\mu_s x_s} \right) \left( 1 - e^{-\mu_c x_c} \right),$$
with $k$ representing a unique efficiency constant for the electronic system coupled to the detector. In order for the total counting yield $Y$ to be computed, it would be necessary to integrate over all points within the sample volume and all points in the crystal volume:

$$Y = \int_{\text{all } p} \int_{\text{all } p^1} P(p,p^1) \, dp$$

Functionally, $\mu_s$ and $\mu_c$ are energy dependent, as is $k$ for all practical systems, while $x_c$ and $x_s$ may be written in either polar or cylindrical coordinate terms. The coordinate dependence of the two path-length quantities implies transcendental functions which are not readily amenable to analytical solution.

An empirical, rather than analytical, solution to the overall detector-sample system was therefore deemed necessary. This solution was based upon estimations of several values: (a) the probability that a gamma ray from a point source will be incident on the detector, known as the geometry factor, $G$; (b) the peak-to-total ratio, $P$, which describes the fraction of available information stored as a useful full-energy peak; and (c) the total absorption efficiency, $T$, which includes self-absorption of the water sample in the Marinelli beaker. These three factors, along with the system operating constant, $k$, describe the total probability that a gamma photon will produce a count in the digital printout. The net counts, $C$ (gross photopeak area counts minus
background counts), divided by the total detection probability will yield the integral number of gamma disintegrations, \( N \), from the entire sample at a specific energy, as described by the following equation

\[
N = \frac{(C)}{(G)(F)(T)(k)}.
\]

As an indication of the practicality of this quantitative technique, an estimate of the counting yield for \(^{40}\text{K}\) can be made

\(^{40}\text{K}\) is 0.0119\% of total \( K \)

The 1/2 is \((1.28 \times 10^{3}) \text{y} \) \((8.766 \times 10^{3} \text{h/y}) = 1.12205 \times 10^{13}\text{h}\)

\[1 \text{ pCi} = 1.332 \times 10^{2} \text{dph} = \frac{(6.93 \times 10^{-1})}{(1.12205 \times 10^{13})} N\]

\[
N = \text{no. atoms} \quad ^{40}\text{K/\ell} = \frac{(1.332 \times 10^{2})(1.12205 \times 10^{13})}{(6.93 \times 10^{-1})}
\]

\[
N_{k} = \text{no. atoms} \quad K/\ell = \frac{(1.332 \times 10^{2})(1.12205 \times 10^{13})}{(6.93 \times 10^{-1})(1.19 \times 10^{-4})}
\]

weight of \( K = \frac{(N_{k})(39.1)}{(6.023 \times 10^{23})} \text{g}\)

\[
w = \frac{(1.332 \times 10^{2})(1.12205 \times 10^{13})(3.91 \times 10^{1})}{(6.93 \times 10^{-1})(1.19 \times 10^{-4})(6.023 \times 10^{23})}
\]
\[ w = 1.176 \times 10^{-3} \text{ g K/pCi} \]

This weight of potassium corresponds to a total of 1598 disintegrations in 12 hours. However, only 11\% of \(^{40}\text{K}\) disintegrations lead to emission of the 1.46 MeV gamma photon. Therefore, 1.176 mg of total K will yield 176 potentially countable gammas in 12 hours.

If a geometry factor (G) of 40\% is assumed, a peak-to-total ratio (P), of 0.4, the total absorption efficiency (T) of 0.2 and \(k = 1\), for the 1.46 MeV gamma in a 4" x 4" NaI(Tl) crystal, then the expected count rate will be

\[ 1.76 \times 10^2 \times (0.4)(0.4)(0.2)(1) = 5.632 \text{ cts/12 hours} \]

for 1 pCi of \(^{40}\text{K}\), or 1.176 mg total K. This corresponds to

\[ \frac{5.632}{1.176} = 4.79 \text{ cts/12 hrs/mg K}, \]
FIGURE 2
Point-source Volume-detector Geometry Versus Volume-source

Volume-detector Geometry of the Marinelli Configuration

A. Ideal Geometry

B. Marinelli-Beaker Geometric Parameters
CHAPTER III

EXPERIMENTAL DESIGN AND PROCEDURE

From September, 1975 through June, 1976 one-gallon samples of raw river water have been collected weekly at Jefferson Parish Waterworks #1 in New Orleans, LA. On several occasions one-gallon samples were also collected at various stages of the purification process. These samples were stored in clean polyethylene bottles that had contained only distilled water before use. To limit cross-contamination effects from plateout of sediment, the containers were not reused. The samples were than stored in a cool, dark room to limit biological growth.

One-liter samples were examined by gamma spectrometry at the low-background laboratory of the Louisiana Division of Radiation Control. The gamma spectrometry system consisted of a 4" x 4" NaI(Tl) crystal encased in a commercially available low-background graded-steel shield. The detector was connected to a standard multi-channel analyzer (MCA) and printout (see Figure 3).

Sample preparation was initiated by vigorously shaking the raw river water container for approximately 30 seconds. This shaking resuspended most of the sediment and material plateout on the bottom and sides of the container. One liter of this sample was transferred into a Marinelli beaker, which was then placed over the NaI(Tl) crystal. The counting configuration of
the Marinelli beaker allowed the same thickness of sample on the sides and top of the crystal. The sample was then counted for 12 hours. This time period was considered optimum for balancing counting requirements with statistically significant information. After each 12-hour count the Marinelli beaker was washed to remove sediment and plateout contamination, and rinsed with distilled water.

Digital information was obtained from the MCA printout, permitting both qualitative and quantitative determination of the gamma emitters present. The digital information was then plotted on semi-log paper for ease of interpretation. Peak energies were ascertained by cross reference to a calibration analysis as described below.

An energy calibration was made after each series of samples was analyzed. This calibration used standard sources of $^{133}$Ba, $^{137}$Cs, and $^{60}$Co for comparison of peak positions. The energies represented by these standards gave good coverage over the range of gamma energies emitted from the natural radionuclides expected to occur in the Mississippi River.

Quantitative analysis was made in the following manner: known quantities of selected water-soluble standard materials were activated in a known flux of neutrons from a Californium-252 source, and the calculated activity produced was measured by counting as a point source above a 3" x 3" NaI(Tl) crystal at the Louisiana State University Nuclear Science Center. The activated
sample was then dissolved in one liter of distilled water and counted in the Marinelli beaker on the Division of Radiation Controls' 4" x 4" NaI(Tl) crystal. Various amounts of a radionuclide as well as different nuclides that have differing gamma ray energies throughout the expected range of natural radionuclides were also produced and counted. Several 12-hour background counts were taken routinely throughout the sample counting period. The net peak area of a specific energy gamma ray was obtained by summation of the peak area counts of the raw river water sample and subtracting from this total the average background peak area counts. This net peak area, expressed in counts, was then converted to disintegration rate by cross-reference to a factor obtained from the known activity of an activated standard of a specified energy. The above procedure accounted for both the complex geometry of the counting configuration and the varying detector efficiencies for the wide range of gamma ray energies.
FIGURE 3
Experimental Design and Equipment

Harshaw 4" x 4" NaI(Tl) crystal
Harshaw preamp
Gamma Products Low-background Graded-steel Shield
Harrison D.C. Power Supply
Technical Measurements Corporation MCA Model 401-D
Technical Measurements Corporation Resolver-Integrator
Technical Measurements Corporation Printer
Ortec Research Amplifier Model 450
CHAPTER IV

RESULTS

Figure 4 is a chronological presentation of gross alpha activities and gross alpha-plus-beta activities determined for individual sampling dates. This figure was used for selecting samples which exhibited either low, medium or high activities for further inspection by gamma spectroscopy. These three typical spectra are presented in Figures 6, 7, and 8, with a background spectrum included as Figure 5.

Table 3 is the compilation of background peak-area counts for four of the most important gamma-photon peaks: $^{40}$K, $^{208}$Tl (a thorium daughter), $^{214}$Pb and $^{214}$Bi (both uranium and radium daughters). Two of these background counts were taken with an empty Marinelli beaker in place over the detector, while the third count was with the Marinelli beaker filled with one liter of distilled water. Because there is no significant difference in counts between the empty and filled beaker, all counts were included in computing averages. The background counts were taken at the beginning and end of the sample counting period and gave fairly consistent peak area counts. These peak area counts were averaged, and this averaged count was subtracted from the selected sample counts to yield net peak-area counts for specific radionuclides, as represented by Table 4.
Quantitative standard information (Table 5) was plotted to establish detector efficiencies for various gamma energies (Figure 9). By comparing Figure 9 and information in Table 4, it is possible to measure selected radionuclide concentrations quantitatively.
FIGURE 5
Background Count Spectrum, June 11, 1976

4" x 4" NaI(Tl)
12 hour count
10 keV/channel

Counts

10^3

10^2

0 50 100 150 200 250

Channel Number
FIGURE 6

High Count Sample Spectrum, October 8, 1975

4" x 4" NaI(Tl)
12 hour count
10 keV/channel
FIGURE 7
Medium Count Sample Spectrum, September 24, 1975

4" x 4" NaI(Tl)
12 hour count
10 keV/channel

Counts

10^3

10^2

0 50 100 150 200 250

Channel Number
FIGURE 8
Low Count Sample Spectrum, October 1, 1975

4" x 4" NaI(Tl)
12 hour count
10 keV/channel
TABLE 3

Background Counts on Selected Radionuclides in the Marinelli Configuration

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$\gamma$ energy (MeV)</th>
<th>Background counts*</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(empty beaker)</td>
<td>(empty beaker)</td>
<td>(with water)</td>
<td></td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>1.46</td>
<td>13,794</td>
<td>13,323</td>
<td>13,866</td>
<td>13,661</td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>2.61</td>
<td>3,201</td>
<td>3,033</td>
<td>3,160</td>
<td>3,131</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>1.76</td>
<td>6,913</td>
<td>6,190</td>
<td>6,666</td>
<td>6,590</td>
</tr>
<tr>
<td>$^{214}$Pb</td>
<td>0.35</td>
<td>22,088</td>
<td>21,717</td>
<td>22,358</td>
<td>22,054</td>
</tr>
</tbody>
</table>

* Counts are summations of gross peak area counts obtained from the digital printout.
TABLE 4

Quantitative Information on Selected Radionuclides for Representative Samples

<table>
<thead>
<tr>
<th>Sample Date</th>
<th>9/24/75</th>
<th>10/1/75</th>
<th>10/8/75</th>
<th>10/15/75</th>
<th>11/12/75</th>
<th>1/28/76</th>
<th>3/24/76</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>$^{40}$K</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Peak Area Counts</td>
<td>14,044</td>
<td>13,791</td>
<td>14,262</td>
<td>14,111</td>
<td>13,915</td>
<td>13,968</td>
<td>14,114</td>
</tr>
<tr>
<td>Net Peak Counts</td>
<td>384</td>
<td>130</td>
<td>601</td>
<td>450</td>
<td>254</td>
<td>307</td>
<td>453</td>
</tr>
<tr>
<td>Quantity pCi/l</td>
<td>61.1</td>
<td>20.7</td>
<td>95.6</td>
<td>71.6</td>
<td>40.4</td>
<td>49.0</td>
<td>72.1</td>
</tr>
<tr>
<td><strong>$^{208}$Tl</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Peak Area Counts</td>
<td>3,164</td>
<td>3,166</td>
<td>3,169</td>
<td>3,029</td>
<td>3,132</td>
<td>3,140</td>
<td>3,220</td>
</tr>
<tr>
<td>Net Peak Counts</td>
<td>33</td>
<td>35</td>
<td>38</td>
<td>*</td>
<td>1</td>
<td>9</td>
<td>89</td>
</tr>
<tr>
<td>Quantity pCi/l</td>
<td>3.9</td>
<td>4.2</td>
<td>4.6</td>
<td>*</td>
<td>0.1</td>
<td>1.1</td>
<td>10.7</td>
</tr>
<tr>
<td><strong>$^{214}$Bi</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Peak Area Counts</td>
<td>6,851</td>
<td>6,818</td>
<td>6,972</td>
<td>6,835</td>
<td>7,128</td>
<td>6,836</td>
<td>7,012</td>
</tr>
<tr>
<td>Net Peak Counts</td>
<td>261</td>
<td>228</td>
<td>382</td>
<td>245</td>
<td>538</td>
<td>246</td>
<td>422</td>
</tr>
<tr>
<td>Quantity pCi/l</td>
<td>57.3</td>
<td>50.0</td>
<td>83.8</td>
<td>53.8</td>
<td>118.1</td>
<td>54.0</td>
<td>92.6</td>
</tr>
<tr>
<td><strong>$^{214}$Pb</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Peak Area Counts</td>
<td>23,203</td>
<td>22,764</td>
<td>22,896</td>
<td>21,977</td>
<td>22,016</td>
<td>21,816</td>
<td>22,300</td>
</tr>
<tr>
<td>Net Peak Counts</td>
<td>1,149</td>
<td>710</td>
<td>842</td>
<td>*</td>
<td>*</td>
<td>*</td>
<td>246</td>
</tr>
<tr>
<td>Quantity pCi/l</td>
<td>9.3</td>
<td>5.7</td>
<td>6.8</td>
<td>*</td>
<td>*</td>
<td>*</td>
<td>2.0</td>
</tr>
</tbody>
</table>

* Denotes less than background
### TABLE 5

**Quantitative Standards**

<table>
<thead>
<tr>
<th>Element</th>
<th>Compound</th>
<th>Weight of Element</th>
<th>Calculated Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>(NH₄)(₂O₇)</td>
<td>1.0 g</td>
<td>10 nCi</td>
</tr>
<tr>
<td>Mn</td>
<td>metal</td>
<td>1 mg</td>
<td>31.2 nCi</td>
</tr>
<tr>
<td>Na</td>
<td>Na₂SO₄</td>
<td>0.1 g</td>
<td>10 nCi</td>
</tr>
<tr>
<td>K</td>
<td>KCl</td>
<td>2.07 g</td>
<td>900 pCi</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Element</th>
<th>$\gamma$ Energy (MeV)</th>
<th>Actual Activity</th>
<th>Net Peak Area Counts</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr</td>
<td>0.32</td>
<td>10 nCi</td>
<td>3,245</td>
</tr>
<tr>
<td>Mn</td>
<td>0.84</td>
<td>27.3 nCi</td>
<td>22,229</td>
</tr>
<tr>
<td></td>
<td>1.81</td>
<td>27.3 nCi</td>
<td>2,760</td>
</tr>
<tr>
<td></td>
<td>2.11</td>
<td>27.3 nCi</td>
<td>1,372</td>
</tr>
<tr>
<td>Na</td>
<td>1.37</td>
<td>9.77 nCi</td>
<td>1,953</td>
</tr>
<tr>
<td></td>
<td>2.75</td>
<td>9.77 nCi</td>
<td>1,109</td>
</tr>
<tr>
<td>K</td>
<td>1.46</td>
<td>900 pCi</td>
<td>5,658</td>
</tr>
</tbody>
</table>
FIGURE 9

Detector Efficiency vs Gamma Photon Energy
CHAPTER V
CONCLUSIONS AND RECOMMENDATIONS

Gamma spectroscopy of Mississippi River water using a large volume NaI(Tl) detector is one method of determining both qualitative and quantitative information on gamma-emitting radionuclides. Obvious disadvantages include:

a. poor sensitivity compared to gas filled detectors,
b. very long counting times,
c. empirical determination of detector system efficiency,
d. no direct relationship between daughter-parent quantities,
e. the tedium associated with data reduction,
f. sedimentation during counting times resulting in a constantly changing counting geometry,
g. the need for a large volume of sample,
h. and a high background which produces large errors in calculated activities.

Among the advantages of scintillation gamma spectrometry are:

a. the ability to identify and quantify gamma emitters,
b. high sensitivity compared to other solid-state detectors
c. the ability to use large sample volumes,

d. and the possibility of simultaneous

identification of large numbers of

radionuclides.

Using Californium-252 for activation of carefully selected
nuclides has proved to be a useful technique for producing
calibration standards for determining detector efficiencies
at various gamma photon energies for complex counting geometries.

After analysis of the river water data several recommendations
become evident to aid in the determination of radionuclide
concentrations as listed below:

a. longer counts should be made to obtain

more statistically significant quantitative

information,

b. sampling collection procedures should be

standardized such that outside contamination

factors be minimized,

c. more radionuclides should be activated for

the determination of detector efficiency

over the energy range of interest,

d. and stirrers should be employed to keep

sediments in solution during the counting
time.
Implementation of these recommendations should improve the quality of information obtainable from important environmental samples including reduction of minimum detectable limits obtained during the current research.
REFERENCES


VITA

Orren W. Williams was born in Berkeley, California, on January 18, 1948. He traveled extensively throughout his childhood, attending many schools in various states and graduated in 1966 from Gonzales High School in Gonzales, Louisiana.

After attending Louisiana State University for three semesters he enlisted in the U. S. Navy and served from February 1968 until December 1971. During his enlistment he was trained as an Aviation Fire Control Technician and attained the rank of Petty Officer 2nd Class (E-5).

Upon completion of his military obligation he re-entered Louisiana State University and obtained his Bachelors of Science degree in Engineering Science (Nuclear Option) in May of 1975. He then enrolled in the Graduate Program at Louisiana State University's Nuclear Science Center leading to the Master of Science degree in Nuclear Engineering, for which he is now a candidate.