

# AUTOMATIC GAMMA SPECTROSCOPIC WATER MONITOR

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## ABSTRACT

Many nuclear facilities have continuous or batch discharges of very low level radioactive liquids to the environment. Many other facilities downstream withdraw this water for use as drinking water or process water. A continuous water monitor has been developed to constantly measure and provide a defensible permanent high quality record that these releases are at acceptable levels. It could also provide water users with proof that their intake water radioactivity is at acceptable levels. The Automatic Water Monitor generates these nuclide specific results utilizing high sensitivity gamma spectroscopy.

The Automatic Water Monitor is capable of achieving detection levels of 0.2-0.3 Bq L<sup>-1</sup> (5-8 pCi L<sup>-1</sup>) of common nuclides (<sup>137</sup>Cs, <sup>60</sup>Co, <sup>131</sup>I). This very low detection limit is achieved by combining a large sample volume (45 L), low level shielding (5-15 cm Fe), and a large NaI detector (5 cm diameter x 30 cm).

The unique gamma spectroscopy software uses a library directed peak search with background erosion spectrum pre-processing. This allows high quality gamma spectroscopy to be performed on the NaI spectra. The use of gamma spectroscopy, rather than gross counting, allows the Water Monitor to accurately quantify and generate alarms based on the specific nuclides of interest to the site, and to separate the response of naturally occurring Radium, Thorium, and Potassium.

## INTRODUCTION

Current regulations and practices require that the radioisotope concentrations in fluids being released to rivers, released to sewers, or used by the public for drinking water, etc., be limited. A record of the effluent must be kept in accordance with the state and federal regulations and as is prudent for legal reasons. Discrete samples could be collected and analyzed off line, but may be both difficult and expensive where there are multiple release points, and where the radioactivity varies significantly with time. A continuous record of the final release effluent is both an effective legal record, and perhaps a cost saving approach.

The maximum limits for release may be 4 or 5 orders of magnitude lower for unknown mixtures than for known mixtures of radionuclides (10CFR20, 1993).<sup>1</sup> Facilities which do not identify and quantify radioisotopes may

therefore be required to use release and concentration limits that are more restrictive than necessary, the result of which is an increase in processing costs. This could be avoided by better quantification and identification of radioisotopes in the fluids.

An on-line, automated spectroscopic water monitor has been developed to constantly measure and provide a defensible permanent high quality record that releases are at acceptable levels. Variability of environmental results due to changing temperature conditions has been overcome by providing detector temperature stabilization. Precision and bias test results indicate that the spectroscopic water monitor achieves acceptable results at gamma energies greater than 200 keV. The monitor is capable of achieving detection levels of 0.2-0.3 Bq L<sup>-1</sup> (5-8 pCi L<sup>-1</sup>) for common radionuclides such as <sup>137</sup>Cs, <sup>60</sup>Co and <sup>131</sup>I.

Figure 1 shows a hypothetical situation in which water monitoring is used by nuclear power plants, hospitals, universities and drinking water supply facilities. Facilities which release radionuclide to the river use water monitors to measure effluents. Facilities which take in water downstream of a release location may also use water monitors on their intake streams to confirm that the water is acceptable for use and to account for concentrations not released by their facility.

## DISCUSSION

### COMPARISON OF METHODS

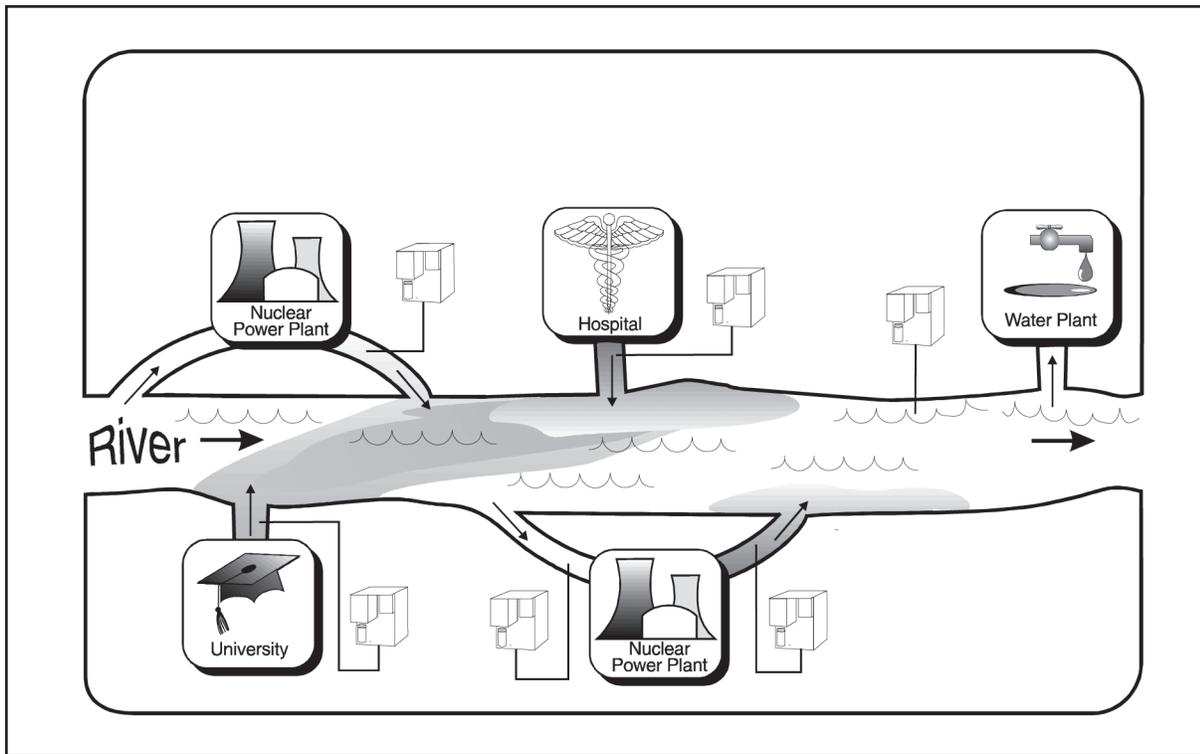
The current methods of identifying and quantifying radionuclides in fluid streams used most frequently are on-line gross gamma counting and grab sampling with subsequent laboratory gamma spectroscopy. Both of these methods have inherent limitations. On-line gamma spectroscopy can overcome most of these disadvantages.

#### On-line Gross Counters

Gross counters cannot distinguish environmental background radiation from that due to radionuclides of concern in the fluid stream being analyzed.

Interference from environmental radiation sources occurs both in ground water and surface water. Ground water can contain fairly high levels of radon prior to being aerated. Other sources of radionuclides in ground water

<sup>1</sup>10CFR20, Appendix B Table and Note 2



**Figure 1:** Typical Facilities using On-Line Spectroscopic Water Monitoring Systems

are leaching of uranium and thorium and their progeny from soil or seepage of surface water into the ground water (NCRP 77, NCRP 50, Klement, 1982). Surface water commonly contains radionuclides from ground water sources as well as those from wet deposition in rain and snow (NCRP 52, Villa and Meyer, 1983). Water in rain or fresh snow typically has elevated background levels of naturally occurring radionuclides from radon washout and  $^7\text{Be}$  from cosmic spallation (NCRP 94).

Sources of external gamma radiation such as tanks filled with radwaste may also increase the instrument background and give false positive readings. A high but stable background can be subtracted, but it may then be difficult to get MDAs low enough to see small amounts of radionuclides and to verify that levels are below limits.

Variable background levels can be from changing plant conditions, or from changing environmental levels such as those due to wet radon deposition. This makes the background correction task much more difficult and subject to error and loss of sensitivity.

Gross counters must assume a particular mixture of radionuclides in order to determine the alert or alarm setpoint levels. The components of the mixture vary with time in most facilities. The sensitivity of gross gamma counters varies with gamma energy, and gamma abundance. Therefore, as the components of the mixture change, the alarm set point must also be changed. Set points may be generated using the most conservative mixture of radionuclides in order to assure that releases are equal to or lower than the allowable limits.

The drawback to being too conservative is that the detector can then have many false positive responses, which results in an unnecessary expenditure of time and money to solve a problem which doesn't really exist. Gross counters which have false positive results may require that a grab sample be obtained and analyzed using gamma spectroscopy to verify the results. This is expensive. Gross counters which have false negative results may result in exceeding procedural and/or legislative limits. This is also undesirable and expensive.

#### **Grab Samples and Laboratory Gamma Spectroscopy**

Grab sampling techniques followed by subsequent gamma spectroscopy at a laboratory are also used to record effluents. Since the data is obtained intermittently, the assumption must be made that the mixture of isotopes and levels of activity in the sample stream do not vary significantly over time. In most facilities this is not true. Therefore, it is possible to have a release above the action level for some time prior to analysis of the sample because of the lengthy time between obtaining a grab sample and performing the analysis. Or even worse, if the increase occurs between grab sampling times, it is possible that levels have been exceeded but never detected.

#### **On-line Spectroscopy**

On-line gamma spectroscopy is the best method available today for sampling fluid streams containing gamma emitters as it eliminates most of the problems associated with both gross counters and grab sampling methods.

On-line spectroscopic methods using pulse height analysis techniques can be used to distinguish between those radionuclides of interest in the fluid stream and others that may be present. Decision making statistics are further improved using background erosion techniques, which allow spectroscopic methods to separate the net peak area from the underlying Compton scatter. Increased background from external sources outside the water monitor shield can also be properly corrected, using the net peak area calculation technique of gamma spectroscopy. Due to the ability of spectroscopic systems to separate background radionuclides and external radioactivity from those radionuclides of interest in a sample stream, on-line spectroscopic methods are much less subject to false positive responses than gross gamma counters. Set points may be generated based on concentrations of individual isotopes when using on-line spectroscopic methods. Each nuclide can have its own acceptable concentration limit. Scaling factors are still necessary to account for those radionuclides which cannot be detected using gamma spectroscopic methods. However, gamma spectroscopy allows more accurate scaling factors, as they are a function of the actual measured radionuclides rather than a function of gross gamma counts.

On-line spectroscopy decreases the uncertainty of whether or not the mixture of radionuclides in a sample is representative of the total release, because on-line spectroscopy systems are essentially continuously sampling and measuring.

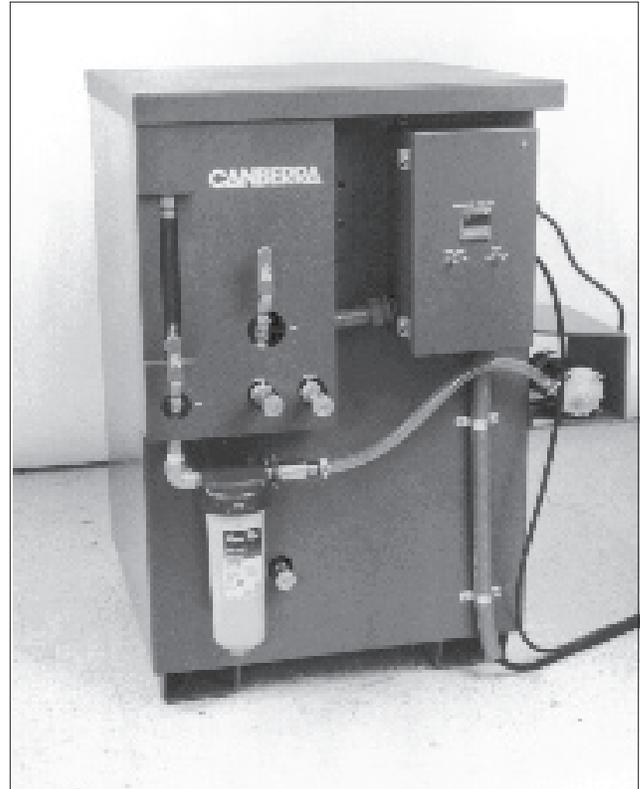
#### Cost of Analysis

On-line gross counting devices cost approximately \$12500 per year to purchase and maintain.<sup>2</sup> In addition, a facility performing gross counting will have false positive results due to interferences discussed before. Grab sampling will be necessary periodically to verify results, and whenever positives exist. If periodic analyses are done weekly and there is 1 false positive per week, the annual cost is estimated at \$20300.<sup>3</sup> Or, if the facility uses grab sampling only and collects only 5 grab samples per week. The annual cost would be \$19,500.

An on-line NaI spectroscopic monitor costs about \$22000<sup>4</sup> per year to own and operate. Therefore, the extra benefit of continuous on-line gamma spectroscopic analyses and documented defensible results can be obtained at little extra cost. If the laboratory analysis costs are higher, or if there are more samples taken, then the on-line spectroscopic systems costs can be much lower than the cost of gross gamma or grab sampling systems.

## DESCRIPTION OF THE ON-LINE SPECTROSCOPIC WATER MONITOR

The automatic gamma spectroscopic water monitor must meet field operation requirements; it must be rugged, reliable, accurate, and easy to use. Although initially designed for environmental water monitoring, the system may also be used for monitoring any type of fluid - liquid or gas.



**Figure 2:** Photograph of Spectroscopic Water Monitoring System

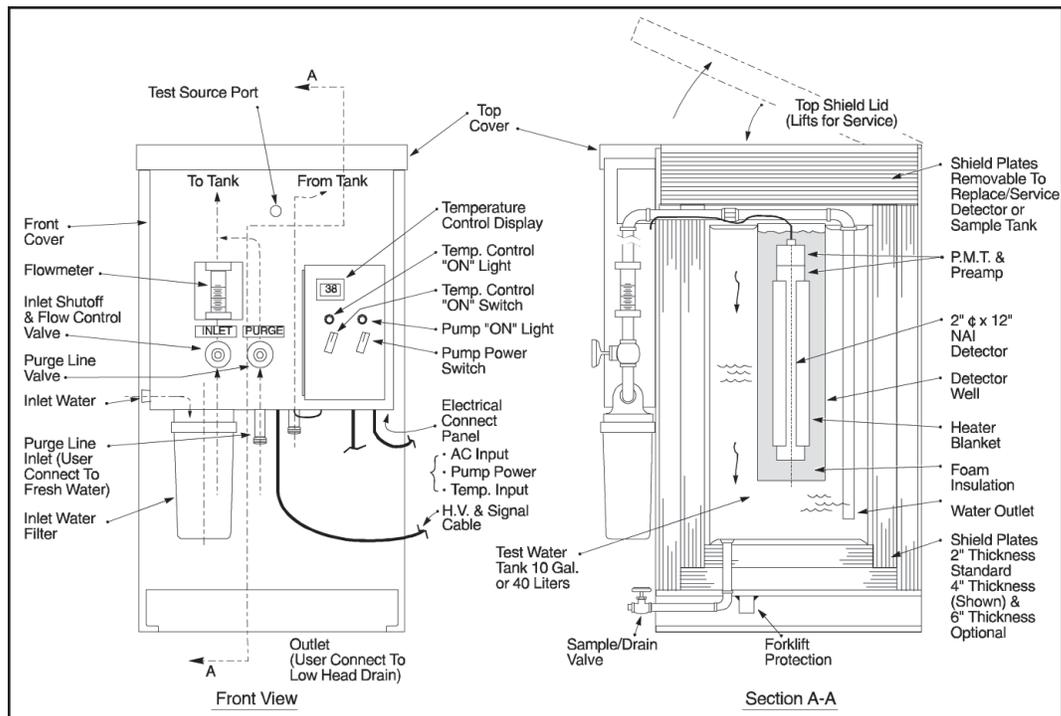
Each integrated water monitoring system includes:

- Temperature stabilized NaI detector
- Low background steel shielding (15 cm, 10 cm and 5 cm)
- Fluid handling and control components
- 45 L (11 gallon) sample container
- PC based MCA
- Automatic analysis software
- Quality Control software and check source
- Full factory calibration

<sup>2</sup>The cost of equipment and maintenance are based on equipment cost, amortized over 5 years. Labor is assumed to be 1 hour per week.

<sup>3</sup>Grab samples are assumed to take 1 technician hour to collect and prepare, at \$25/hr cost. Gamma spectroscopic analysis is assumed to cost \$50 per sample, whether performed internally or sent to a commercial lab.

<sup>4</sup>The cost of equipment and maintenance are based on equipment cost, amortized over 5 years. Labor is assumed to be 2 hours per week.



**Figure 3: Spectroscopic Water Monitoring System**

A large (5 cm diameter x 30 cm long) NaI(Tl) scintillation detector assembly provides high efficiency which allows detection of low levels of gamma emitting radionuclides present in the sample. The detector is thermally stabilized in order to minimize gain shifts due to variations in sample stream or room environment temperatures. Active thermal stabilization assures stable gain and consistent detector performance regardless of the variation in ambient or inlet water temperatures. The system is designed so that a Ge detector can be used in place of the NaI detector for those situations where NaI spectroscopy is inadequate (e.g. effluents from hospitals with extensive nuclear medicine facilities).

The 45 L (11 gallon) sample chamber has a staggered inlet and outlet to promote sample mixing within the sample chamber. The sample chamber is an annular configuration with the detector placed in the center, a geometry designed for maximum detector efficiency. The detector chamber and all plumbing are stainless steel to resist corrosion and to minimize contamination build-up. The sample chamber is built of pressure vessel components to withstand over 50 psi internal pressure.

The liquid handling system includes a centrifugal pump, filter, flow control valves, a flowmeter, and freshwater purge controls. The normal operating configuration of the system is with both the inlet and outlet valves open, counting a sample flowing through the chamber. The valves may also be shut and the sample counted in a grab sample (static) configuration.

The shield used for these analyses is 15 cm (6 inches) thick low background steel. The steel is from a special source known to contain zero <sup>60</sup>Co contamination.

The shield is designed with a Quality Control check source access port so that a technician may perform routine QC counts without taking the shield apart.

The system can be operated in an unattended completely automatic mode in which a continuous series of spectrum acquisitions and analyses are performed on a sample stream flowing through the sample chamber. The system can also be operated in attended modes, either using the standard default parameters, or in a fully manual mode with the operator selecting individual operation and analysis parameters (count time, analysis library, etc.).

The analysis software is an adaptation of Canberra's well documented and field proven ABACOS software (East, et al, 1982), which was designed to be an easy to use and flexible software package, and which is optimized for low level spectroscopy. The library driven peak search and Gaussian peak fitting routines have been shown to be essential for reliable detection of peaks near the Lower Limit of Detection (LLD). A full Quality Control program to document system performance is standard. The software also contains a variety of utilities to perform QA reviews, batch printing, batch data reviews, and to generate ASCII compatible output files.

## PERFORMANCE RESULTS

### TEMPERATURE STABILITY

NaI detection systems are subject to gain shifts with temperature changes. Normally, without thermal stabilization, there is commonly 6% gain shift for every 10 °C. The software is able to correctly identify peaks to within ± 2% of the energy calibration, therefore

temperature shifts of approximately 3.5 °C without thermal stabilization would cause the system to be out of energy calibration and would result in incorrect identification. Since the detector is surrounded by the water sample chamber, it will eventually equilibrate at the inlet water temperature, which can easily vary by 20 or 30 °C.

The range of the thermal stabilizer components is quite wide, however, NaI detection systems with normal construction can only tolerate continuous temperatures of up to 40 °C. Temperature tests were performed on the system in order to verify that the thermal stabilization system limited peak drift to less than ± 2% for a range of fluid temperatures of 0 °C to 40 °C. Tests were performed at 3 temperature ranges to prove that the detector stability could be controlled to within the acceptable range. These temperatures were 0 °C, 18 °C, and 40 °C.

From a power off condition, it took up to 4 hours for the detector to stabilize under worst case conditions, (0 °C water). Once the detectors were stabilized, and with power continuously applied, the maximum peak drift from 0 - 40°C was 1%, which is within the acceptable criterion of less than ± 2%. Figure 4 gives the results of the test.

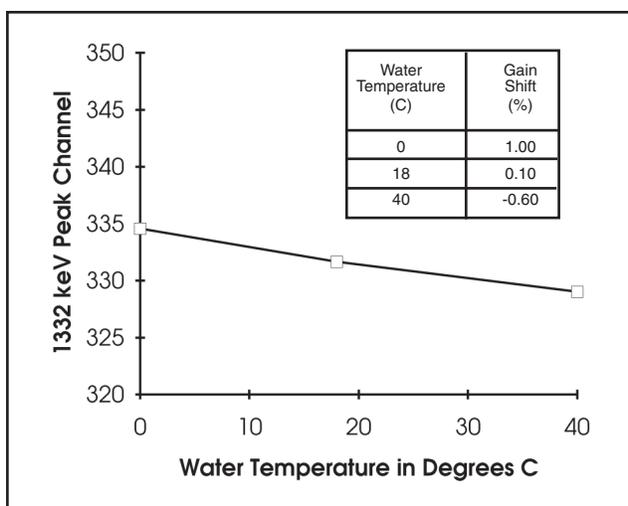


Figure 4: Temperature vs Detector Stability

## PRECISION AND BIAS

The efficiency calibration used by the Water Monitoring System generates a set of equations for determining efficiency as a function of peak energy.

The calibration source used was a NIST traceable source with gamma energies ranging from 88 keV to 1836 keV. The activities used are shown in Table 1.<sup>5</sup> The source was diluted into 45 L of distilled water and placed in the tank of the Water Monitoring System. Metallic non-radioactive chloride salts of each radioactive element in the source were also dissolved in the distilled water, to reduce the amount of plateout and maintain the source radionuclides in solution during calibration.

<sup>5</sup>North American Scientific, Inc. Model Cal3000 mixed gamma solution  
<sup>6</sup>ANSI-N-13.30, 1989

Table 1: Calibration Data

NUCLIDE	ENERGY (keV)	SOURCE ACTIVITY (kBq)	RELATIVE BIAS (%)	RELATIVE PRECISION (%)
Cd-109	88.03	51.93	not used	not used
Co-57	122.00	2.08	not used	not used
Te-123m	158.99	1.94	8.38	22.23
Cr-51	320.10	32.94	0.50	5.10
Sn-113	391.69	10.29	not detected	not detected
Sr-85	514.00	7.67	-4.92	1.71
Cs-137	661.65	9.34	-1.62	1.88
Y-88	898.00	16.98	-0.58	0.92
Co-60	1173.22	10.17	-4.44	1.87
Co-60	1332.49	10.17	3.26	3.37
Y-88	1836.08	16.98	0.31	0.85
Absolute Mean =			3.00	4.74

The efficiency calibration is a polynomial equation which is derived by the software from net peak countrates, using least squares fitting techniques (East, et al, 1982). The calibration source was counted for 5200 seconds. The three lowest energy peaks (88 keV <sup>109</sup>Cd, 122 keV <sup>57</sup>Co, and 159 keV <sup>123m</sup>Te) had poor statistics and were not suitable for calibration because their relative activities in this single mixed nuclide source were too low in the presence of the high Compton background from the higher energy sources. Lower energy radionuclide activities of the calibration source will be increased by a factor of 3-10 in order to obtain better low energy peak statistics for future calibrations. The 391 keV <sup>113</sup>Sn net peak area is lower than expected, due to precipitation of the element. This is generally known to be a problem with these sources. Figure 5 shows the efficiency calibration curve generated using peak data.

The calibration was verified by performing bias and precision tests. Data was accumulated using a series of five minute counts, of the calibration source; spectra were analyzed using the efficiency calibration being verified.

Bias values indicate the accuracy of the results, and indicate whether the calibrations are good calibrations. Average bias values were calculated using the following method:<sup>6</sup>

$$B_r = \frac{\sum_{i=1}^n [(A_{ri} - A_{ra})(A_{ra})^{-1}]}{n}$$

Where:

- $B_r$  is the average bias for radionuclide r
- $A_{ri}$  is the activity of radionuclide r reported by the spectroscopic water monitor for count number i
- $A_{ra}$  is the actual activity of radionuclide r in the source being counted
- n is the number of repeated counts performed

Values of  $B_r$  are averaged for all radionuclides used in the test, to determine the overall bias.

Precision values indicate the short term statistical stability of the system for the count time chosen. The count time used to test precision is the same as that recommended for routine counts, therefore results of the precision test give an indication of what variability is expected of a spectroscopic water monitoring system operating under routine field conditions.

Relative precision is calculated using the following method:<sup>7</sup>

$$S_r = \left[ \frac{\sum_{i=1}^n \left( \frac{A_{ri}}{A_r} - 1 \right)^2}{(n-1)} \right]^{1/2}$$

Where:

- $S_r$  is the relative precision for nuclide r
- $A_r$  is the average activity reported for nuclide r

Values of  $S_r$  are averaged for all radionuclides used in the test, to determine the overall relative precision.

Table 1 shows the results. The verification results indicate that the calibration is acceptable at energies greater than 500 keV, but should only be considered accurate to within 30% from 150 keV to 500 keV.

Monte Carlo techniques were also used to model the source and detector geometries, and to predict the results to further validate the calibration. The MCNP code (Briesmeister, 1991) was used. Results are shown in Figure 5. Monte Carlo projections show good agreement with the efficiency generated using the Water Monitor software program for energies of about 500 keV and above. The MCNP efficiency results reflect the correct low energy efficiency more accurately than the efficiency calculated using actual data; this is because of the poor statistics of low energy peaks which were due to low source activity.

### MDA

Minimum detectable activity (MDA) values were calculated to demonstrate the system performance under normal conditions of operation. The water monitoring system reports MDA values for library nuclides for which statistically significant peaks are not detected. The person setting up the water monitoring system may modify statistical parameters in the software for any confidence level desired.

An *a priori* count was performed on a representative water blank (tap water) which contains normal environmental levels of radium. The MDA values are generated using

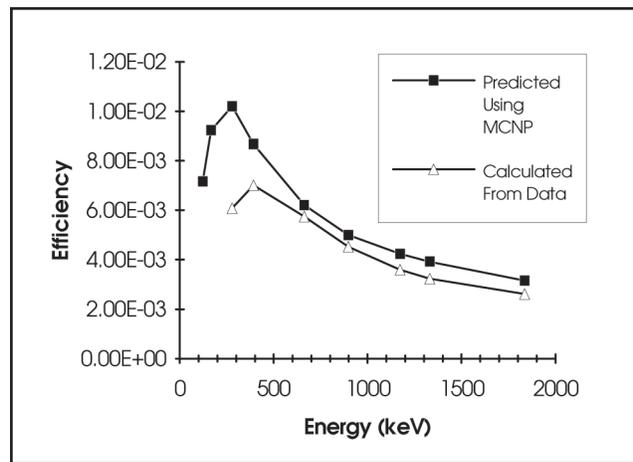


Figure 5: Predicted vs Calculated Efficiencies

a minute count time at the 95% confidence level, using a 5% Type I (false positive) error and a 5% Type II (false negative) error. The MDAs are calculated as follows:

$$MDA = 3 + 4.66 \frac{\sqrt{\text{Bkgr Counts Spanning Energy Range of Analysis}}}{\text{Efficiency} \times \text{Counting Time}}$$

MDA values for the Canberra on-line Water Monitoring System are shown in Figure 6.

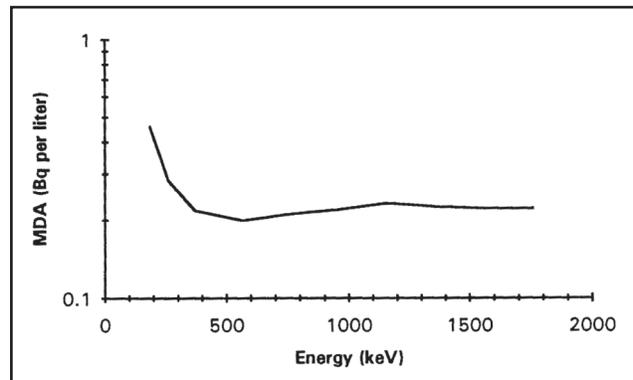


Figure 6: MDA Values for Spectroscopic Water Monitor

The MDA for gamma energies greater than 300 keV and for a gamma abundance of 100% is relatively constant, at approximately 0.2 Bq L<sup>-1</sup> (5 pCi L<sup>-1</sup>) for a 60 minute count.

<sup>7</sup>ANSI-N-13.30, 1989

## CONCLUSIONS

On-line spectroscopic methods have advantages over on-line gross counting and periodic grab sampling. On-line spectroscopic sampling eliminates the problems of variable background environments and variable sample stream concentrations. The sampling frequency for an automated on-line spectroscopic sampling system is essentially continuous, and uses less labor, thus decreasing the risk of intervals of undetected increased activity. Each nuclide can have its own acceptable release limit, so that changes in the sample stream radionuclide concentration do not require recalculations of alarm set points. Facilities performing on-line gross counting which have variable background levels of radionuclides or changing nuclide mixtures and facilities performing grab sampling can benefit by changing to on-line spectroscopy systems, because the results are more accurate and the quality of the data received is better.

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