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# **Detection of radioisotope contamination in drinking water**

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## **Foreword**

The U.S. Environmental Protection Agency is charged by Congress with protecting the nation's air, water, and land resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, the Agency's Office of Research and Development provides data and science support that can be used to solve environmental problems and build the scientific knowledge base needed to manage our ecological resources wisely, to understand how pollutants affect our health, and to prevent or reduce environmental risks.

In September 2002, the Agency announced the formation of the National Homeland Security Research Center. The Center is part of the Office of Research and Development; it manages, coordinates, supports, and conducts a variety of research and technical assistance efforts. These efforts are designed to provide appropriate, affordable, effective, and validated technologies and methods for addressing risks posed by chemical, biological, and radiological terrorist attacks. Research focuses on enhancing our ability to detect, respond (through containment, mitigation, and response to public/media), and stabilize (through treatment and decontamination) in the event of such attacks.

The Center's team of scientists and engineers is dedicated to understanding the terrorist threat, communicating the risks, and mitigating the results of attacks. Guided by the roadmap set forth in the Agency's *Homeland Security Strategy*, the Center ensures rapid production and distribution of water security related research products.

The Center created the Water Infrastructure Protection Division to perform research in water protection areas including: Protection and Prevention, Detection, Containment, Decontamination and Water Treatment Mitigation, and Technology Testing and Evaluation. The detection research can be divided into two main categories: 1) support for contamination warning systems for timely detection of contamination events and 2) confirmation of events through sampling and analysis. This document focuses on online technologies for detection of radionuclides evaluated at the Agency's Test and Evaluation Facility in Cincinnati, Ohio and National Air and Radiation Environmental Laboratory in Montgomery, AL. Additional information on the Center and its research products can be found at <http://www.epa.gov/nhsrc>.

Jonathan Herrmann  
National Homeland Security Research Center, Director

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List of Acronyms

DHS	Department of Homeland Security
DI	Deionized
DPD	N,N-Diethyl-p-phenylenediamine
EPA	U.S. Environmental Protection Agency
IND	Improvised Nuclear Device
MCA	Multi-Channel Analyzer
MCL	Maximum Contaminant Level
NAREL	National Air and Radiation Environmental Laboratory
NDIR	Non-Dispersive Infrared
NHSRC	National Homeland Security Research Center
NIM	Nuclear Instrumentation Modules
ORIA	Office of Radiation and Indoor Air
ORP	Oxidation Reduction Potential
PAG	Protective Action Guideline
RDD	Radiological Dispersive Device
RERT	Radiological Emergency Response Team
SCADA	Supervisory Control and Data Acquisition
S/N	Signal to Noise
TA	Technical Associates
TOC	Total Organic Carbon
WDMP	Water Distribution Monitoring Panel

**List of Units**

Ci	Curie	KeV	kilo-electron volts	mV	millivolts
cpm	counts per minute	L	liter	NTU	nephelometric turbidity
cps	counts per second	Mg	milligrams	psi	pounds per square inch
ft	foot	mm	millimeters	μS	microsiemens
				sec	second

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## **Executive Summary**

This report presents data on detection of radionuclides in drinking water. Part I focuses on how water quality parameters (i.e. free chlorine, pH, etc) change when soluble surrogate radionuclide salts are introduced into drinking water. Cesium, cobalt and strontium chloride were injected into a pipe containing flowing tap water. An array of on-line sensors continuously monitored water quality in the pipe. Water quality changes caused by the salts were recorded. The results show that observing changes in water quality due to soluble radioisotope contamination is challenging. Part II presents data collected from an on-line sensor designed to detect radiation in water. The commercially available radiation detection system was challenged with a radionuclide (potassium-40) dissolved in water. The device did not detect the radioisotope solution at the Protective Action Guidelines (PAG) or Maximum Contaminant Levels (MCL) that were of interest. Furthermore, numerous operational and design issues were encountered that impair the usability of the device in the field.

The results presented in the two parts of this study show the effectiveness of detecting radionuclide contamination with on-line sensors using different detection principles. On-line sensors employing one or more parameters are used in drinking water distribution systems to monitor water quality. Exploring on-line sensors ability to detect contamination is important so they can be utilized beyond their traditional water quality monitoring role. In addition, on-line detection of radiation in water is desirable from the standpoint of contamination, but also to better quantify the amount of radiation in distributed water. The data presented in this report addresses both topics. Furthermore, these data shows where future work could be focused to improve online sensors detection technology for dissolved radionuclides.



## **Chapter 1 Detection of surrogate radioisotopes with on-line water quality sensors and the Hach Event Monitor Trigger System**

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## **Executive Summary**

Online water quality sensors were examined for their ability to detect cesium, cobalt and strontium salts injected into a model drinking water system. The pilot scale drinking water system consisted of a 1200 ft long, 3-inch diameter pipe with chlorinated tap water flowing through it at 1 ft/sec. Online sensors detecting free chlorine, conductivity, pH, oxidation-reduction potential (ORP), turbidity and total organic carbon (TOC) were operational throughout the study. A commercially available event detection system, the Hach Event Monitor<sup>®</sup> Trigger System, analyzed online water quality data continuously. Free chlorine, conductivity, pH, ORP and turbidity changed in the presence of cobalt chloride (1 and 10 mg/L), but only conductivity changed in response to the cesium chloride (5 and 10 mg/L) and strontium chloride (1 and 10 mg/L) injections. Event detection software detected an anomaly in water quality data only when cobalt chloride was injected at 10 mg/L. The inadequate performance of the online water quality sensors in these tests demonstrates the need for improved detection technology for radiochemical contamination of drinking water systems and the exploration of other sensor technologies.

## **Introduction**

The presence of radionuclides in drinking water distribution systems has historically occurred from low levels of naturally occurring radioisotopes. Accordingly, research attention has been paid to naturally occurring radioisotopes radium and uranium (Korshin et al., 2008; Valentine and Stearns, 1994). On the other hand, because chemical and biological contamination is due, in large part, to human activity, detection efforts have been ongoing. The result of these research priorities is that relatively little attention has been paid to real time detection of radioisotopes in drinking water as compared to the more common pollutants (Hall et al., 2007; McKenna et al., 2006; Murray, 2008; Szabo et al. 2008). However, with the potential for deliberate release of radioisotopes into drinking water in a terrorist incident, this lack of research focused on the variety of potentially harmful radionuclides has become problematic.

The lack of published literature regarding the real-time detection of radioactive agents in drinking water systems prompted this research. Experiments were designed to examine whether soluble cesium, cobalt and strontium can be detected in drinking water with standard water quality sensors. Non-radioactive salts acted as surrogates for cesium-137 (Cs-137), cobalt-60 (Co-60) and strontium-90 (Sr-90). A 1200 ft long, 3-inch fiberglass lined iron pipe outfitted with online water quality sensors acted as the experimental system for detection studies. Data from the water quality sensors was analyzed by commercially available event detection software that is designed to detect anomalies in water quality data and alert the user to those anomalies.

## **Materials and Methods**

### *Online Water Quality Monitoring Experimental Design*

Flow in a drinking water distribution pipe was simulated using a once-through, or single pass, pipe at the U.S. EPA Test and Evaluation (T&E) Facility in Cincinnati, Ohio. The pipe is made of 1200 ft of 3-inch diameter, fiberglass lined ductile iron. Flow was 22 gallons per minute, which yields an average velocity of 1 ft/sec and results in turbulent flow ( $Re \sim 26,000$ ) in the relatively smooth pipe.

Tap water from the Greater Cincinnati Water Works (GCWW) distribution system was pumped into a 750 gallon tank and then gravity fed into the single pass pipe. An air gap was maintained between the Greater Cincinnati Water Works system and the tank to ensure that contaminants did not backflow. Free chlorine was  $1.0 \pm 0.1$  mg/L, with temperature ranging from 10-30° C seasonally and turbidity was less than 0.1 NTU (nephelometric turbidity) throughout the year. Pressure was 10-12 psi inside the pipe. Twenty minute contaminant injections were performed by injecting 10 L of contaminant solution at 0.5 L/min directly into the pipe. Contaminant concentration in the pipe was varied by altering the concentration in the 10 L volume fed to the pipe. Contaminant solutions were made by dissolving CsCl (99+ %), CoCl<sub>2</sub> (97%) or SrCl<sub>2</sub> (99.99%) in 100 ml of deionized water and adding it to 9.9 L of Cincinnati tap water. Blank injections were performed by injecting 10 L of tap water with no contaminant present.

Online sensors measured water quality in the single pass pipe at 80 feet from the injection point via a slipstream. Water quality sensors, including those for free chlorine, total organic carbon (TOC), conductivity, pH and turbidity, were housed in a Hach Water Distribution Monitoring Panel (WDMP) (Hach Company, Loveland, Colorado). Free chlorine was measured using a Hach CL17 which uses an online version of the N,N-Diethyl-p-phenylenediamine (DPD) method (Clesceri et al., 1998). Conductivity and pH were measured using Hach GLI probes and turbidity was measured with a Hach 1720D Turbidimeter. Total organic carbon was measured with a Hach astroTOC UV total organic carbon analyzer, which employs UV/persulfate oxidation and a non-dispersive infrared (NDIR) detector. Oxidation reduction potential (ORP) was measured with a YSI 6920 multi-probe sonde (YSI, Inc., Yellow Springs, Ohio). All online sensors were operated and maintained according to manufacturer recommendations. Further detail is given in Hall et al. (2007).

Data from the online sensors (except ORP) was analyzed in real time by a Hach Event Monitor<sup>®</sup> Trigger System (Hach Co., Loveland, CO). This device alerts the user to unusual changes in water quality, which eliminates the need to visually observe the incoming water quality data in real-time. The event monitor utilizes a proprietary algorithm which simultaneously evaluates signals from all of the water quality parameters (except ORP). The algorithm generates a result called a “trigger” value, which can change as water quality input signals change. When water quality baselines are stable, the trigger value is close to zero. The event detection system also includes a function that alerts the user when the trigger crosses a pre-set threshold. In practice, the trigger value is based on the water quality variability at each monitored location. Typical alarm thresholds at the T&E facility are set at 0.5 or 1.0 depending on the variability of the water quality, which is usually low.

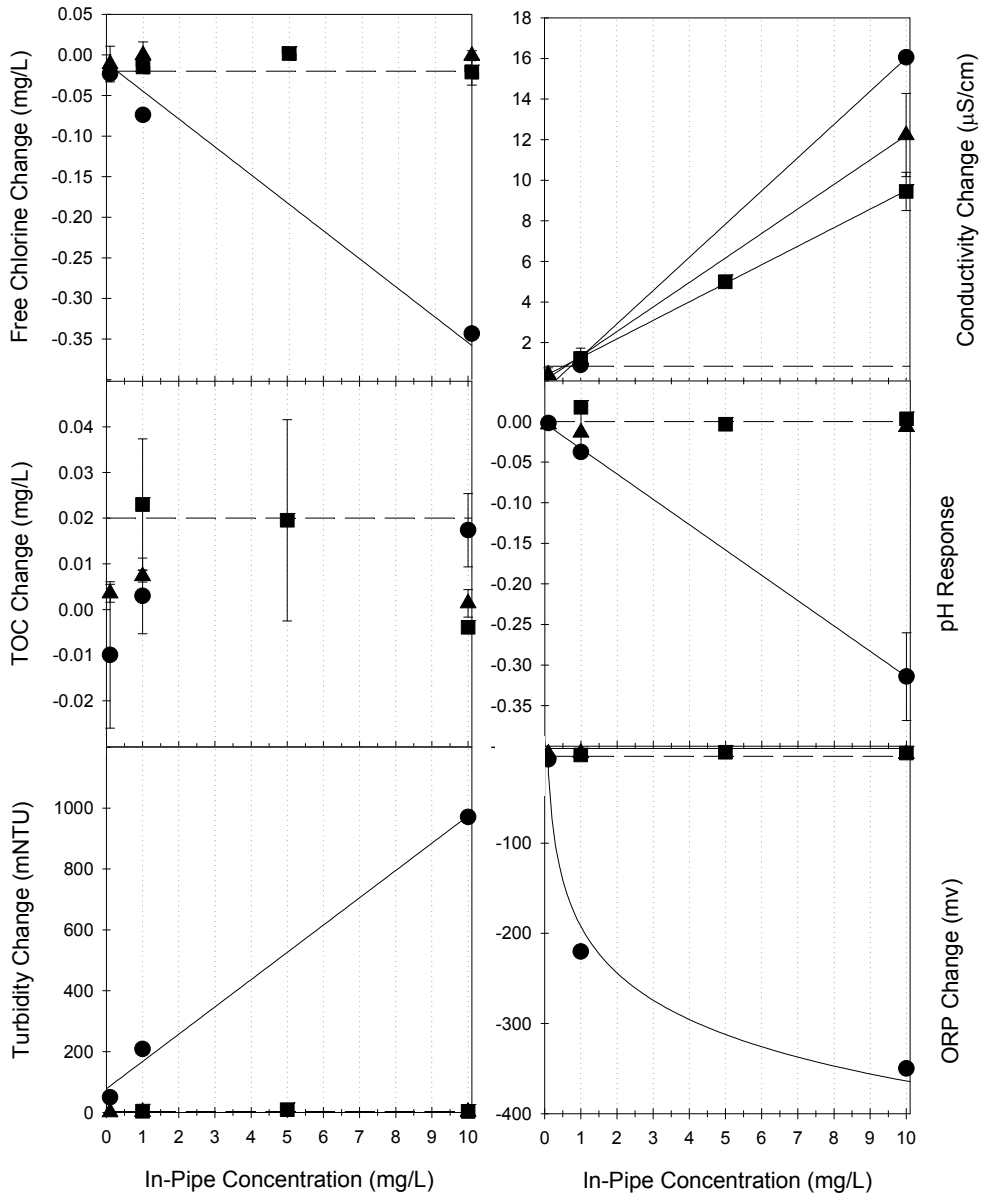
#### *Data Analyses*

Water quality sensor response experiments were conducted in triplicate. Data points in Figure 1-1 are the mean of those experiments while bars on data points represent standard deviation. Results are plotted as the change in sensor value from 1 hour of stable baseline to the peak sensor response after contamination. Statistical analysis of the sensor response data was performed using SigmaStat software (Systat Software Inc., Point Richmond, California). A paired, two tailed Student’s t-test ( $\alpha=0.01$ ) was used to determine whether there was a significant difference between the sensor change from baseline for the blank injection and for the contaminant injections. Results are presented in Table 1-1. All data sets passed the normality test in SigmaStat<sup>®</sup> (Systat Software, Inc., Chicago, Illinois). Results in Table 1-2 are the sensor response (from baseline to peak) as well as signal to noise ratio (S/N). S/N is the sensor response divided by the standard deviation of the baseline period.

## **Results**

### *Water Quality Sensor Response*

Figure 1-1 shows the change in water quality parameters resulting from contamination with cesium chloride, cobalt chloride and strontium chloride. Figure 1-1 includes data from the blank injection (tap water with no contaminant present), which shows any sensor response due to the act of injection (e.g. dilution). Conductivity was the only



sensor that

Figure 1-1 Water quality sensor response to cesium chloride (■), cobalt chloride (●) and strontium chloride (▲) and blank injection (---). Response is the difference between the peak changes during injection from a stable pre-injection baseline. Bars represent standard deviation from three experiments.

[acronyms and units: ORP, oxidation reduction potential; mg/L, milligrams per liter; mNTU, micronephelometric turbidity; mv, millivolts; µS, microsiemens; TOC, total organic carbon]

provided a meaningful visual response to cesium chloride at 5 and 10 mg/L and strontium chloride at 1 and 10 mg/L. Table 1-1 shows that all of these responses were statistically significant. Conductivity response to 0.1 mg/L strontium chloride and 1 mg/L cesium chloride were less than or equal to the blank injection change. TOC did not respond to any contaminant at any concentration level, which was expected since they are all inorganic salts.

A visible precipitate formed upon injection of cobalt chloride into chlorinated Cincinnati tap water, which elicited the turbidity response. Table 1-1 shows that the turbidity change was significant at 1 and 10 mg/L cobalt chloride, and was greater than the baseline response at 0.1 mg/L. The pH decrease is likely due to hydroxide ions being removed from solution through the formation of cobaltous hydroxide precipitate, which forms when a cobalt salt is added to basic solution, especially in the presence of hypochlorite (Figlarz et al., 1974; Windholz and Budavari, 1983). The t-test results show that the pH response was not significant at 10 mg/L cobalt chloride ( $P=0.012$ ), but there is a discernable visual change in pH 0.04 and 0.3 units at 1 and 10 mg/L, respectively. ORP decreased due to the drop in free chlorine, which reduced the oxidative strength of the water. Results were significant at 1 and 10 mg/l cobalt chloride ( $P<0.001$ ). Conductivity response for cobalt chloride was similar to cesium chloride and strontium chloride except that the increase with concentration was steeper. Results were significant at 10 mg/L ( $P<0.001$ ).

Table 1-1 T-test Results for Sensor Response Experiments

Contaminant	ppm	Water Quality Parameter					
		Free Chlorine	Total Organic Carbon	Turbidity	Specific Conductivity	pH	Oxidation Reduction Potential
Cesium Chloride	1	0.316	0.204	0.069	0.135	0.079	0.168
	5	0.905	0.348	0.023	0.003	0.423	0.348
	10	0.014	0.743	0.020	0.002	0.055	0.826
Cobalt Chloride	0.1	0.105	0.632	0.012	0.979	0.199	0.056
	1	0.028	0.204	0.001	0.086	0.071	<0.001
	10	< 0.000	0.004	<0.001	<0.001	0.012	< 0.001
Strontium Chloride	0.1	0.105	0.481	0.837	0.504	0.535	0.661
	1	0.427	0.177	0.543	0.004	0.785	0.572
	10	0.802	0.543	0.591	0.006	0.756	0.463

\*Significant results are highlighted ( $\alpha=0.01$ )

Table 1-2 shows the change of the Event Monitor<sup>®</sup> trigger above the pre-injection baseline due to injection of the salts. Baseline trigger mean over the test period was  $0.5 \pm 0.41$  ( $n=10,938$ ), but 98% of the data points were less than 0.1. The large standard deviation compared to the mean is due to periods of higher trigger value resulting from changes in water quality not related to contaminant injection, but possibly due to operational issues (i.e. sensor power failure, loss of connectivity to the event monitor). The trigger rose by 3.09 above the baseline after cobalt chloride was injected at 10 mg/L. All other injection resulted in an increase of 0.04-0.11. Signal to noise ratio was higher than the blank injection for cobalt chloride, but was comparable to the blank injection for cesium and strontium chloride.

Table 1-2 Event Monitor Trigger Response to Contamination

In-Pipe Concentration (mg/L)	Cesium Chloride	Cobalt Chloride	Strontium Chloride
	<u>Change from baseline</u> Signal to noise		
0.1	$\frac{0.05}{2.8}$	$\frac{0.08}{3.7}$	$\frac{0.05}{2.9}$
1.0		$\frac{0.11}{6.2}$	$\frac{0.05}{1.7}$
5.0	$\frac{0.05}{2.8}$		
10	$\frac{0.04}{1.0}$	$\frac{3.09}{159.7}$	$\frac{0.05}{1.5}$
Blank (Tap water only)	$\frac{0.03}{1.5}$		

## Discussion

### *Water quality sensor response*

Cesium, cobalt and strontium chloride salts changed the parameter measured by at least one water quality sensor when injected into the T&E facility experimental pipe system. Conductivity was the only parameter that yielded a significant response to cesium chloride (5 mg/L), cobalt chloride (10 mg/L) and strontium chloride (1 mg/L) injections. Conductivity is a stable water quality parameter in the experimental pipe systems used at the T&E facility when contaminants are absent (Hall et al., 2007; Szabo et al., 2008). Baseline relative standard deviation was less than 0.2% for conductivity throughout the study. Background conductivity is stable since much of the Greater Cincinnati Water Works system is fed by two distinct treatment plants, and mixing of various water qualities, which could increase background variability, is uncommon. The changes displayed in Figure 1-1 are calculated by subtracting the peak response observed during injection from the mean baseline water quality value during the 1 hour prior to contaminant injection. If variation in background conductivity increases, the change caused by contaminant injection would be less compared to the mean baseline value.

Free chlorine, pH, turbidity and ORP responded to the injection of cobalt chloride along with conductivity. This is encouraging since a cobalt precipitate formed by a reaction with free chlorine persists on the corroded iron infrastructure surfaces in drinking water (Szabo et al., 2009). The number and type of water quality sensors that drinking water utilities deploy in the field varies by utility, but free chlorine, pH and turbidity are commonly used by many utilities (Hall et al., 2007). However, 1 mg/L cobalt chloride was the lowest concentration that generated a statistically significant sensor response compared to the baseline injection.

### *Alarms due to water quality changes*



Many of the water quality changes shown in Figure 1-1 were significant in the current experimental design since the baseline water quality was stable, and it was known when the contaminant passed the sensor. In contrast, drinking water quality can be temporally and spatially variable in real distribution systems. Consequently, differentiating water quality changes resulting from contamination and from either natural variability or equipment malfunction can be challenging. Continuous observation of numerous water quality data streams by plant operators is not a feasible monitoring option, so event detection algorithms should ideally achieve this task (Dawsey et al., 2006; Hart and McKenna, 2009; McKenna et al., 2006).

The results in Table 1-2 show that the trigger value changes generated by the Event Monitor<sup>®</sup> algorithm were larger for contaminant injections than for the blank injection. An alarm was generated by the event monitor trigger when cobalt chloride was injected at 10 mg/L. The trigger rose by 3.09 from a pre-injection baseline value that ranged from 0.01-0.09. This surpassed the alarm value of 0.5 or 1.0 typically used at the T&E facility. Trigger changes at lower concentrations of cobalt chloride and cesium and strontium chloride ranged from 0.04 to 0.11. In addition to not rising above the 0.5 threshold, these changes were lower than some of the non-contamination events that raised the event monitor trigger during testing. For example, during the 18 day period in which the contamination experiments took place, there were 13 events that caused the trigger to quickly rise above 0.2 (the highest baseline value observed of 0.9, plus the highest cobalt chloride change of 0.11). Ten of those events were due to non-contamination changes in water quality such as spikes in TOC, free chlorine or turbidity that lasted less than 10 minutes and raised the trigger to 0.26-1.11. Three of the events lasted between 26-76 minutes and raised the trigger to 2.2-18.8. One of these events was due to changes in water quality, one was caused by a reagent running out in a TOC analyzer and one occurred because of an unknown sensor malfunction during which the TOC reading dropped to zero.

The trigger value changes resulting from surrogate radioisotope salt contamination were smaller than the trigger changes that came from normal variation in water quality or equipment operational issues. The rationale for setting a trigger value at 0.5-1.0 at the T&E facility was that this value range minimized alarms, but allowed for observation of water quality changes that could indicate contamination. Setting the trigger below the changes in Table 1-2 would result in multiple alarms per day based on the water quality changes observed in the T&E facility experimental pipe system (the exception is cobalt chloride at 10 mg/L). The event monitor is equipped with a visual display that shows the water quality values in near real time, and some of the water quality changes in Figure 1-1 were clearly larger than the baseline even though they did not generate an alarm. However, the Hach Event Monitor<sup>®</sup> is designed such that the visual display is located with the water quality sensors. Should numerous sensor stations be deployed in a water distribution system, the visual display might not be continuously monitored by a human being.

## **Conclusions**

Injection of cesium chloride, cobalt chloride, and strontium chloride changed on-line water quality at 1 mg/L and above in the experimental drinking water pipe system at the T&E facility. Conductivity responded to cesium at 5 and 10 mg/L and strontium chloride at 1 and 10 mg/L. Conductivity also responded to cobalt chloride at 10 mg/L. Additionally, cobalt chloride caused significant changes in free chlorine, turbidity, ORP at 10 mg/L, with turbidity and ORP responding at 1 mg/L. The Hach Event Monitor Trigger System did not produce an alarm in response to injections of cesium chloride or strontium chloride at concentrations up to 10 mg/L, but it did produce an alarm for cobalt chloride at 10 mg/L. The results show that testing and development of commercially available event detection algorithms both sensitive enough to detect anomalous changes in drinking water quality and discriminating enough to filter out false positives are needed. In addition, research is needed to develop sensors that can detect radiation in water with greater sensitivity and specificity. Part II of this report addresses this subject by discussing detection of ionizing radiation in water with a commercially available on-line instrument.

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**Chapter 2 : Testing of an In-line Radiation Monitoring System for  
Drinking Water: A Summary of the Tests and Responses of  
Technical Associates SSS-33-5FT Radiation Monitoring System**

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## **Executive Summary**

The ability of the SSS-33-5FT Radiation Monitoring System (Technical Associates, Canoga Park, California) to detect radiation in water in real time was tested. A series of aqueous radioisotope exposure experiments were designed using tritium, potassium-40, ruthenium-103, and americium-241. Background radiation measured by the SSS-33-5FT in the testing laboratory was higher than levels detected by laboratory-based alpha, beta and gamma sensors. The reason for the high readings was not uncovered, but this could make real-time detection difficult to use in the field and could decrease the sensitivity of the instrument. Exposure test results showed that the instrument did not detect radioisotopes in water at the Protective Action Guidelines (PAG) developed by the Department of Homeland Security (DHS) or Maximum Contaminant Levels (MCL) published by EPA. Furthermore, when exposed to three single isotope sealed sources, the device consistently detected activities lower than their true values.

The inability to detect radiation in water at levels of concern might be due to instrument design issues. For example, it appeared that the radioactive solutions were unable to travel close enough to the gamma radiation detector for it to be effective. Design issues also extend to the usability of the device. Maintenance and cleaning of instrument plumbing, in-line filters and detectors was not user-friendly and this might impact the longevity of the instrument in the field. Design changes could produce an effective radiation monitoring system for drinking water supply systems. Several off the shelf gamma radiation detectors (i.e. multi-channel analyzers) are readily available, although alpha and beta detection systems will require more research. In summary, experimental results and equipment design observations presented in this report can contribute to the design of more effective on-line radiation detection systems for drinking water.

## **Introduction**

Limited information exists on the performance of commercially available on-line equipment designed to detect radiation in water. In recent years, drinking water utilities have deployed on-line water quality sensors which monitor parameters such as free/total chlorine, pH, turbidity, conductivity, temperature and total organic carbon (TOC) (Hall et al., 2007). The dual purpose of these on-line sensors is (1) to better understand water quality so that it could be improved and (2) to enhance security by detecting unusual changes in water quality that might indicate contamination. Although a great deal of data has been generated about the field performance of on-line water quality sensors in drinking water distribution systems, previous research has not evaluated radiation detectors.

There are existing methods for detecting radiation such as scintillation and gamma spectroscopy. A key performance concern has been the ability to detect radioactive material in water at Maximum Contaminant Levels (MCL) (40CFR§141.25; EPA, 2009; EPA, 2002) or Protective Action Guidelines (PAG) (73 FR 45029). To obtain usable results at these levels, off-line detection often takes both hours of preparative chemistry and hours of counting the prepared samples with a lab-based radiation detector. Drinking water utilities, however, require the ability to monitor water quality on-line in real time or near real time so that they can respond to and mitigate contamination.

Few commercially available on-line radiation detection instruments are available for water (EPA, 2010). One such instrument is the Technical Associates Nuclear Instruments, Inc. (TA) Model SSS-55-3FT, hereafter also referred to as the TA unit or TA device. The instrument measures gamma emissions using sodium iodide detectors, alpha-beta emissions using an anthracene scintillation detector, and beta emissions via a plastic scintillation detector. EPA, through its Office of Research and Development and National Homeland Security Research Center (NHSRC), purchased a TA unit. Working with EPA's National Air and Radiation Environmental Laboratory (NAREL), EPA formulated a plan to test the ability of the TA unit to detect radioisotopes in water. The test plan was designed to investigate two objectives:

1. Determine if the TA SSS-55-3FT on-line radiation detector is able to detect radiological contamination, at what minimum radioactivity level and how that level compares to the MCL and PAG
2. Evaluate the TA device in terms of its usability for drinking water system operators, and determine if it could fulfill drinking water utilities' needs

## **Methodology**

### *Radioisotope Selection*

Radioisotopes of concern have been identified through examination of potential homeland threat scenarios by the various federal agencies including, among others, Department of Defense, Department of Homeland Security, Nuclear Regulatory Commission, and U.S. EPA. Radionuclides listed in Table 2-1 encompass the three major types of radiation emission (alpha, beta, and gamma).

Table 2-1 Radionuclides Proposed for Testing

Radionuclide	MCL (pCi/L)	PAG (pCi/L)
H-3	20,000	3,900,000
Co-60	100	26,100
Sr-90	8	5,250
Cs-137	200	13,800
Ra-226	5	500
U Total (234, 235, 238)	20.1	3,000
Am-241	15	320

MCL, maximum contaminant level; PAG, protective action guide; pCi/L, picocurie/liter

The MCLs listed in Table 2-1 were likely too low for the current on line detection equipment, but they provide a well known standard for comparison. Less restrictive PAGs, which correspond to a 500mrem /yr dose level, were also used for comparison. The radionuclides in Table 2-1 were deemed problematic to use at NAREL for purposes of evaluating the TA device for several reasons. First, some of these radionuclides have potential for contaminating the environmental laboratory. Second, there exists a potential radiation exposure hazard to personnel in the lab. Finally, disposal is a problem due to long half lives or high specific activity. As a result, surrogate radioisotopes with similar properties to the isotopes of concern, but with minimal hazard to lab operations and personnel, were selected (Table 2-2). The surrogate radionuclides provided a rigorous test for any radioactive measurement system, covering the range of energies needed for testing and radioactive emissions of concern.

Table 2-3 compares the isotopes of interest (Table 2-1) and their surrogates in terms of their decay mode, primary energy, and measurement technique typically used in a drinking water matrix.

Table 2-2 Radionuclides and Surrogates Selected by NAREL<sup>a</sup> for Testing

Radionuclide	Surrogate Nuclide	Reason
H-3	None	Low emission energy of beta is unable to be duplicated.
Co-60	K-40	K-40 is a naturally occurring isotope that is easier to dispose of it. It is not considered licensed material.
Sr-90	Ru-103	Ru-103 has a shorter half-life (40 days vs. 27.7 years). This makes it easier to dispose of by allowing for decay of the material over time (1 year). Sr-90 emits only beta energy and requires isotopic chemistry. Ru-103 can be measured by gamma spectroscopy without any chemistry requirements.
Cs-137	Ru-103	Ru-103 has a shorter half-life (40 days vs. 5.3 years). Allowing decay of the waste for a year makes it relatively easy to dispose of Ru-103.
Ra-226	Am-241	Ra-226 is an alpha emitter that requires isotopic chemistry for analysis. Due to interference from U-235, it is not able to be clearly identify with gamma spectroscopy
U Total (234,235,238)	Am-241	These are Alpha emitters that require alpha isotopic chemistry for analysis. U-235 is not able to be clearly identify with gamma spectroscopy
Am-241	None	Alpha emitter has clear gamma emission that allows for quantitative

		determinations eliminating need for isotopic chemistry
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<sup>a</sup>National Air and Radiation Environmental Laboratory

Table 2-3 Comparison of Surrogate Radionuclides Used in Study and Radionuclides of Interest

Radionuclide(s) of Interest	Type of Emission	Typical Measurement Technique	Alpha Average Energy (keV) <sup>a</sup>	Beta Maximum Energy (keV)	Beta Average Energy (keV)	Gamma Energy (keV) / (Yield)
<b>Surrogate Radionuclide</b>						
H-3 (no surrogate)	Beta only	Liquid Scintillation		18	6	
Co-60	Beta, Gamma	Gamma Spectrometry		1478	94	1173 / (98%) 1332 / (99%)
<b>K-40</b>	<b>Beta, Gamma</b>	<b>Gamma Spectrometry</b>		<b>1332</b>	<b>540</b>	<b>1461 / (11%)</b>
Sr-90	Beta only	Gas Proportional Counting		544	200	
Cs-137	Beta, Gamma	Gamma Spectrometry		1167	195	661.6 (85%)
<b>Ru-103</b>	<b>Beta, Gamma</b>	<b>Gamma Spectrometry</b>		<b>210</b>	<b>62</b>	<b>497 / (88%)</b>
Ra-226	Alpha, Gamma	Alpha Scintillation Counting	4771			186 / (4%)
U Total (234, 235, 238)	Alpha, Gamma	Alpha Spectrometry	4500			185 / (3%) U-235
<b>Am-241</b>	<b>Alpha, Gamma</b>	<b>Alpha or Gamma Spectrometry</b>	<b>5480</b>			<b>59.5 / (35%)</b>

<sup>a</sup>KeV, Kilo-electron volt

*Experimental Approach for Objective 1*



The first objective, to determine to the ability of the TA device’s ability to detect radiological contamination and at what minimum radioactivity level, was investigated using the bench-scale assembly presented in Figure 2-1. Detectors were removed from the TA device housing and mounted separately on a bench top so that each detector could be tested individually. Tap water was introduced into the sensor assembly and background radiation levels were established. The system was then drained and test solutions with various concentrations, listed in Table 2-4, were introduced into the assembly one at a time. Flow was stopped once the solution was in contact with the detector. Detector readings were recorded prior to, during, and after 10 minutes of solution introduction. The test assembly was then drained and rinsed with tap water, and the system’s readouts noted to ensure removal of the test solutions.

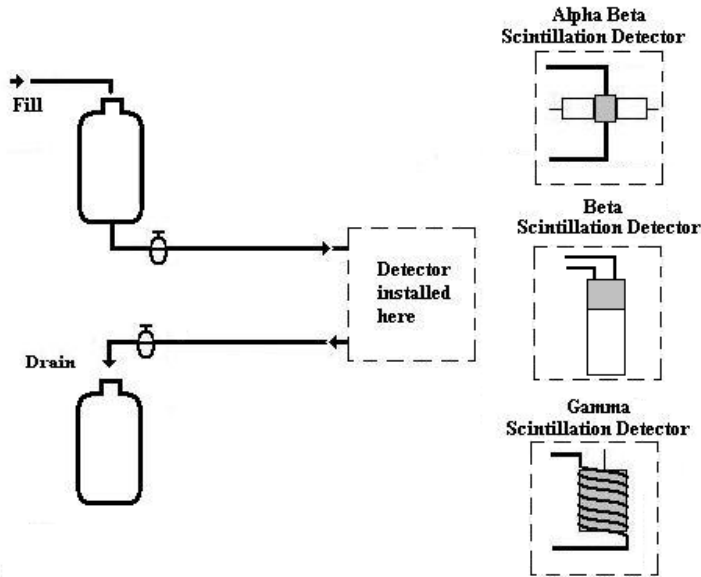


Figure 2-1 First test of study: Individual detectors component flow path.

Table 2-4 Target Concentrations of Prepared Test Solutions

Radioisotope	Tested Parameter	Concentration 1 (pCi/L)	Concentration 2 (pCi/L)	Concentration 3 (pCi/L)
Am -241	Alpha / Gamma	30, 000	15,000	1,500
H -3	Tritium	10,000,000	100, 000	10, 000
K - 40	Beta / Gamma	30,000	15,000	3,000
Ru - 103	Beta / Gamma	15,000	5,000	1,000

pCi/L, picocuries/liter

A second trial was performed in the same manner as the first set of experiments, except that the detectors were positioned in their original, manufacturer’s flow path (Figure 2-2).

Tests were performed with the 30,000 pCi/L potassium-40 solution (Table 2-4) circulating through the system.

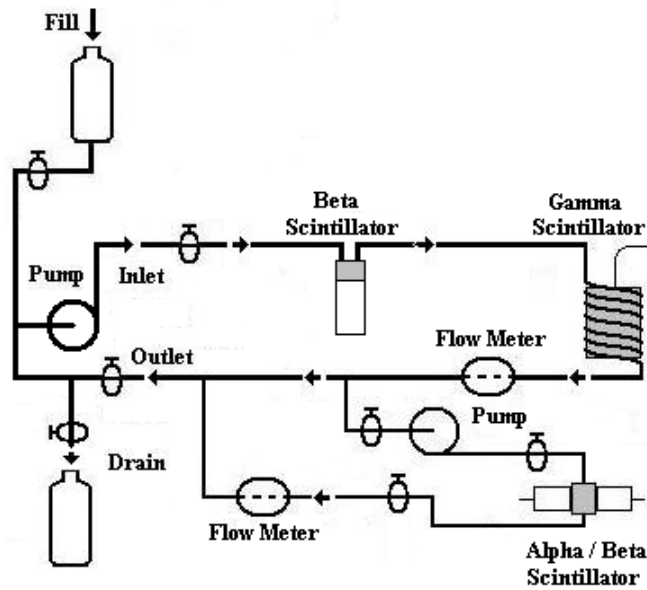


Figure 2-2 Second test of study: SSS-33-5FT integrated flow path.

The manufacturer specifications (Table 2-5) list detectable radionuclide concentrations in water for specific sample observation times. The results generated from alpha, beta, and gamma detectors during the exposure experiments will be compared to the manufacturer's stated values.

Table 2-5 Technical Associates Model SSS-33-5FT Sensitivity Specifications and Detection Method

Measurement Time of Observation	Sensitivity Range (pCi/L)	Top of Range (pCi/L)	Sensor / Method Used
<u>Gross Alpha</u> 2 min 24 hour	100,000 10,000	20,000,000	Crushed Anthracene Scintillation Bed
<u>Gross Beta</u> 2 min	100,000	100,000,000	Plastic Scintillator

24 hour	50,000		
<u>Gamma</u> 2 min 24 hour	10,000 500	100,000,000	Sodium Iodide Scintillation Crystal
<u>Tritium</u> (No time Frame Identified)	20,000	1,000,000	Crushed Anthracene Scintillation Bed

Technical Associates 5/22/2007

### *Approach for Addressing Objective 2*

Objective 1 required start-up, operation, and disassembly of the instrument. Experience gained from these experiments provided direct information regarding Objective 2, namely calibration, operational testing, ease of use, and maintenance. Experiences with the TA device were evaluated in the context of the duties of drinking water plant operator or maintenance personnel who would be responsible for the device in the field. These evaluations were based on discussion among the EPA investigators, as well as some questions raised with the vendor. Accordingly, the conclusions are subjective and are highly dependent on the individual water utility's actual requirements or expectations, which cannot clearly be predicted. These conclusions and observations provided the information for meeting the requirements of Objective 2.

### **Test Data, Results, Analysis, and Discussion**

#### *Observations about Instrument Set-up, Operating Parameters and Calibration*

Prior to the start of the testing, the vendor requested access to the delivered instrument to update the computer software. This allowed EPA investigators to observe the set-up of the instrument and the vendor's testing during the upgrade period. One resulting observation was that the readouts of the system were in counts per second (cps) or counts per minute (cpm). The use of these units is very acceptable to a health physicist or someone with a radiation protection background, but they would not be useful to a water utility. Based on water quality regulations, utilities use concentration-based units such as parts per million (mg/L) or parts per billion (ug/L) to express non-radioactive contaminant concentrations and pCi/L for radioactive contaminants. This practical consideration was conveyed to the vendor, who changed the computer software for the operational readouts to correspond to units of pCi/L.

In order to provide an operational readout in pCi/L, conversion factors are necessary to convert counts received by a detector to the activity of radioactive material. The determination of the conversion factors requires accurate knowledge of the detector efficiency, based on a known standard. The vendor supplied a list of conversion factors (Attachment 1) that stated how the instrument converted readouts for the detectors from cps to pCi/L. These conversion factors were based upon various isotopes. EPA investigators noted that the information provided did not show results for any specific aqueous solutions developed by the vendor. When asked about this, the vendor indicated that they based their conversion factors (from cps to activity) on the use of sealed, solid radioactive sources placed alongside the detectors, or from inference from other technical data not provided.

However, geometry and sample matrix are key elements of any determination of conversion factors. In order to account for effects such as detector geometry and matrix effects of the water, conversion factors would have to be based on measurements obtained by placing known radioactive sources concentrations in the water circulating around the detector, much the same as the tests described for Objective 1. From these aqueous sources, calibration factors based on efficiency would allow for conversion into useful readouts. Indeed, the lack of complete and decisive calibration information by the vendor was a significant problem. For instance, when EPA placed the solid, sealed sources alongside the gamma detectors, similar to what the vendor did to determine the conversion factors, the detector did yield a response. Although this illustrates a response to a known gamma source, it does not show the detector's response to the gamma source in the matrix (water) or in the same geometrical configuration as the sample.

In addition to the vendor-supplied conversion factors for the gamma detectors resulting in potentially inaccurate readings, there was also no vendor-supplied test data for the response of the beta and the alpha/tritium detectors. According to the vendor, these were set in the factory. The vendor was uncertain as to the methodology and frequency of field calibration at the time of the testing. The vendor suggested that the instrument could be removed from site and sent back to the vendor's facility for a minimum of one week for recalibration every year. Providing for removal, shipping, and reinstallation time, a utility could be without the instrument for potentially two weeks or longer and incur shipping expense.

Another observation by EPA investigators regarding the functionality of the instrument for a water utility involved a set-point alarm function that the user can set. The vendor supplied data that showed the response of the TA device to specific isotopes (Co-60, Sr-90, Ra-226, and H-3), but it was unclear if the data was generated by exposing the system to solutions with known radionuclide concentration. However, the instrumentation did not demonstrate by design or operation an ability to identify the radioisotope that caused an alarm or spike in counts. Thus, there is no means of relating the alarm function to the specific radioisotope that caused the alarm, except for tritium. For other isotopes, the alarms denote that a set level of alpha, beta or gamma radiation has been surpassed. It might be assumed that the user would know that a high alarm equates to an activity concentration greater than a given value, and that further investigation is required such as sending a sample to a laboratory. Thus, this type of alarm function might be useful to utilities in some circumstances.

From the standpoint of practical usability, however, the alarm set point is hidden in an administrator's screen and is not user-friendly. Also, in the event of an alarm, there was no means of obtaining a sample from the system that could be sent to a lab for analysis. An ISCO<sup>®</sup> sampler is an option provided by the vendor for the unit and connections are on the exterior of the instrument. However, there was no indication that the sampler could be controlled from the operator's computer interface or that an alarm situation would automatically trigger a sample to be taken. This function was not tested further.

### *Background Measurements*

Ideally, during operation, the instrument should be measuring the normal radioactive level of the water, or the background level. If the source of the water is constant during a given period of time (hour, day or week) the amount of radioactive material should have minimal fluctuations. However, this is not typical of radioactive material measurements and small variations are expected, for example, because of contributions to the background from radioactivity other than originating from the water. The gamma instrument would be the most influenced by the location of the instrument from building materials and personnel. Regardless, as long as the signal from the gamma detector is fairly stable, it might be possible for a water utility to determine that an excursion from “normal” has occurred. However, the background gamma reading as measured by the TA device never stabilized during the testing. Table 2-6 lists the results of the various background counts taken during the tests for the various radiation types and energy ranges.

Table 2-6 Instrument Background and Readings in Counts per Second

Date / Time	3/30/09 09:45	3/30/09 10:05	3/31/09 10:45	4/1/09 09:30	4/1/09 12:25	4/1/09 13:30	Variation mean/uncertainty at 1 sigma
Detector							
Gross Alpha	0.000	0.026	0.021	0.029	0.11	0.023	0.035 / 0.038
Gross Beta	3132.21*	1709.89*	239.32	330.12	318.40	1443.35	808.216 / 575.117 <sup>#</sup>
Tritium	0.00	0.13	0.12	0.18	0.071	0.14	0.109 / 0.064
High Gamma	7.14	6.28	5.15	7.28	6.12	6.10	6.345 / 0.781
Mid Gamma	5.60	4.01	3.51	6.89	3.93	3.87	4.635 / 1.323
Low Gamma	37.42	32.68	29.64	38.22	31.75	32.85	33.760 / 3.355

\* - prior to dark adapting detector

# - dark adapted values only used

The data in Table 2-6 relate to the performance of the TA device in terms of both background response and variation in the measured background. It is worthwhile to further analyze the results in Table 2-6 by comparing them to the typical behavior of radiation detection devices.

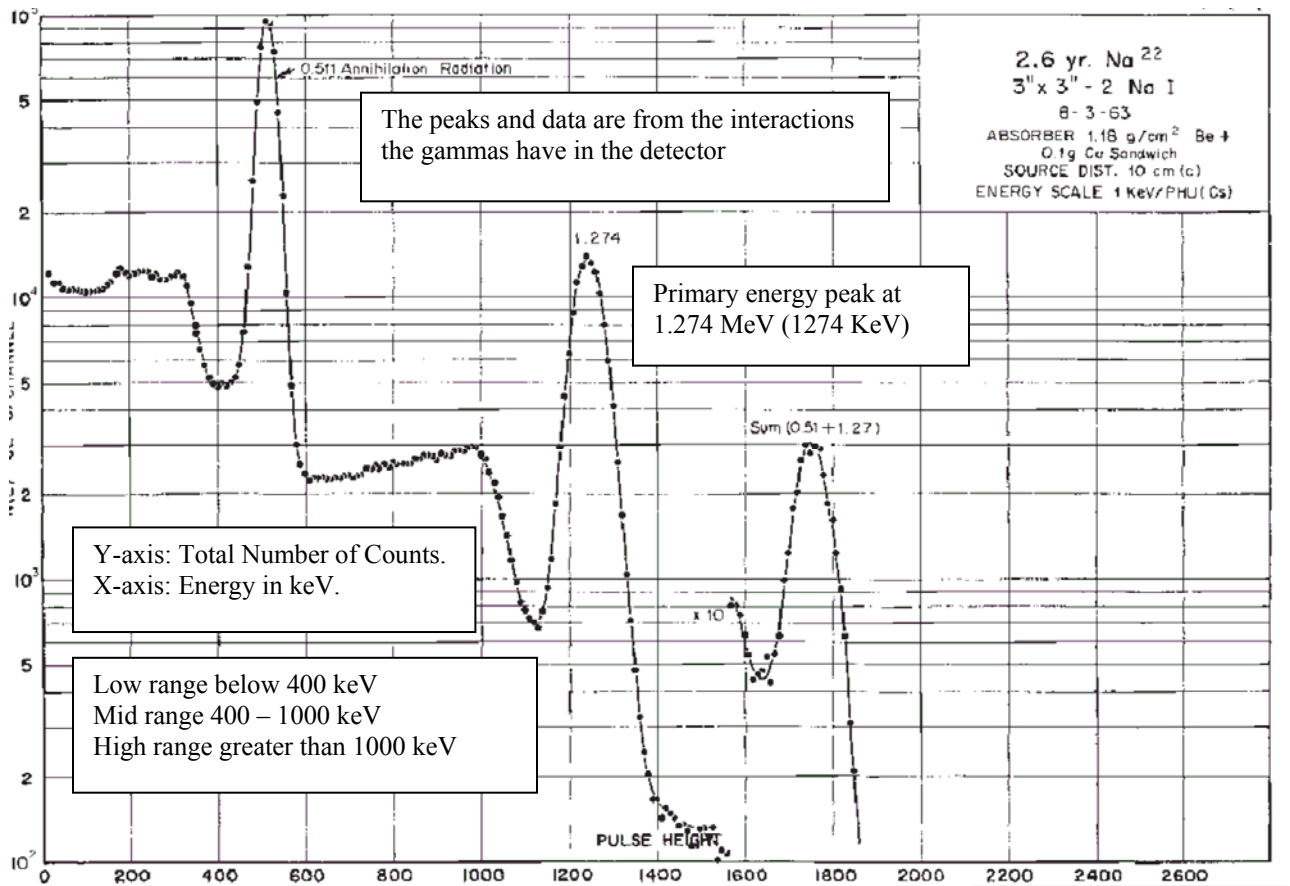


Figure 2-3 Spectrum of sodium-22 using a 3 × 3 inch NaI gamma detector (Heath, 1964).

It is standard practice that the background of typical laboratory instruments should be measured successively at least twice a day for a month to allow for proper statistics to be developed for operation. Typically, the background for a stationary radiation detector in a laboratory setting is measured at least 20 times and a statistical mean determined from which control charts are derived. Background levels in the counting room at NAREL for similar equipment might serve as a reference point for comparing the TA unit’s results in Table 2-6. For similar equipment at NAREL, background alpha counts for 100 minutes measure at 0.000583 cps and the uncertainty is lower by a factor of 10. Beta counts typically are measured at 0.50 to 0.67 cps with a corresponding uncertainty of 0.013 cps. Clearly, both these typical alpha and beta results are of a different order of magnitude than Table 2-6.

Gamma counts are harder to relate to laboratory response, and NAREL does not measure cps in a given range of energies. As a means of discussing the gamma results in Table 2-6, consider that gamma counts for a one microcurie source at a mean efficiency of 1 % would be 37 cps. When these gamma detection instruments are measuring background, it would be reasonable to expect a sub-numerical count rate (i.e., 0.06 cps). Thus, the TA unit’s results in Table 2-6 are far above what would be expected to be background. Further, EPA investigators measured a 2 year old calibration standard containing

approximately 50,000 pCi of activity with an independent 3 × 3 inch NaI detector connected to a multi-channel analyzer owned by NAREL. The result was a 100 cps reading, which is two to three times the value of the summation of gamma readings for the background in Table 2-6 (38-52 cps). This suggests that the background measured by the TA device was close to the radiation produced by a 50,000 pCi source, but the background should have been orders of magnitude less. Water utilities might experience difficulty in interpreting this kind of result.

#### *Gamma Detector Tests*

The response of the gamma detector was initially checked using the vendor-supplied point sources, and the TA device response was evaluated by comparing it to a standard laboratory instrument response, as well as to the theoretically correct response. The supplied gamma check sources consist of three single isotope sealed sources (Sodium-22, Barium-133, and Cesium-137) mounted at the end of an aluminum rod. The sources were introduced as described by the vendor by placing them in near proximity to the active detector area. The introduction of the gamma check sources resulted in signal output, but the results varied over the time measured, especially in the mid gamma energy channel (Table 2-7).

The comparison to the check source was not encouraging because the gamma results did not correspond to the known activity of the sources. The responses that should have been displayed by the instrumentation are based on the decay energies and frequency of emission (yield) for given radionuclides. For example, sodium-22 decays, emitting a single gamma at 1274 keV with a yield of 99.94% (only 6 out of 10,000 decays are not counted). The gamma spectrum showing the resulting energies seen by a sodium iodide detector indicates readings in every energy level below this energy. Therefore, the mid range energy channels should have had readings as well. This is illustrated in Figure 2-3, which shows a spectrum obtained using a NaI detector similar to the one used by the TA instrument. The x-axis is the energy measured in keV and the y-axis is the total number of counts.

Table 2-7 Measured Gamma Detector Responses to Check Sources in pCi/L

pCi / L Elapsed Time (Minutes)	Isotope							
	Ba-133		Cs-137			Na-22		
	Activity pCi / L							
	1,000,000		1,000,000			5,700,000		
	Range of the Detector							
	Low	High and Mid	Low	Mid	High	Low	Mid	High
2	16,310	0	17,394	98	0	71,101	0	25,287
4	33,763	0	19,988	111	0	83,556	2,872	15,260
6	35,393	0	19,690	108	0	78,337	0	20,346
8	35,360	0	10,719	121	0	86,539	10,015	10,663
10	35,442	0	19,304	111	0	78,650	0	20,231
12	35,261	0	20,149	102	0	72,043	0	26,163
14	36,450	0	10,686	127	0	72,086	0	26,145
16	35,341	0	19,397	109	0	78,562	0	20,189
18	36,379	0	10,956	119	0	78,507	0	20,091
20	36,527	0	24,292	103	0	78,768	0	19,880
22	35,379	0	10,963	118	0	86,389	10,646	10,486
24	36,484	0	19,597	107	0	79,523	0	19,345
26	33,493	0	20,411	109	0	84,173	3646	14,576
28	36,392	0	10,823	117	0	79,086	0	19,652
30	35,320	0	19,813	117	0	72,547	0	25,708
<b>Average</b>	<b>34,220</b>	<b>0</b>	<b>16,945</b>	<b>112</b>	<b>0</b>	<b>78,658</b>	<b>1,812</b>	<b>19,601</b>



The actual energy values corresponding to the low, mid, and high range of the detector were not stated in the user's manual. According to the manufacturer, the TA device sums all the counts received in a given energy range to determine activity. However, it is not clear what the energy ranges are since the manufacturer has not stated a cutoff point, and this makes it difficult to determine if the response of the TA device is appropriate. A further difficulty arises in comparing the TA device output with standard laboratory equipment, because in normal operation, a gamma spectrum analyzer would use only the maximum peaks to determine the activity. For example, in Figure 2-3, Na-22 has two prominent peaks at 511 keV and 1274 keV, and also shows the summation of these two peaks.

To better understand the meaning of the output from the TA device, let us consider the spectrum shown in Figure 2-3, for Na-22 (one of the check sources). Also, for this discussion, assume that the low energy range is below 400 keV, mid-range from 400 to 1000 keV, and high range 1000 keV and higher. Using this assumption, one can use the count rate across the spectrum to estimate the values each detector would achieve for this particular sample. In the low range, a series of counts start at 5,000 counts at 400 keV to approximately 15,000 counts at 0 keV. The mid range has a peak at 511 keV that rises to approximately 100,000 counts then drops to a plateau of approximately 2500 counts. The high range drops from the plateau to about 700 counts and then rises to a peak at the primary gamma emission of 1274 keV with a value of approximately 15,000 counts.

The low range has approximately 9 million counts in total, the mid range 6 million counts, and the high 1.6 million, or a ratio of about 10 : 6 : 2 for low : mid : high, respectively. While the conversion from counts to picocuries is dependent upon the calibration, this estimated ratio can be used to infer if the TA devices response to its check source is appropriate as a function of ratio of the gamma energies. Table 2-7 shows that the Na-22 source does not have this expected response, in terms of the ratios between average values of the energy ranges (10 : 0.025 : 0.25).

The cesium radionuclide, which has a primary decay energy of 662 keV, should have had counts in the mid range totaling approximately 80% of the counts in the low range and no counts in the high range. This is inconsistent with the results for cesium in Table 2-7. However, the barium-133 counts were consistent with expectations. Barium had counts only in the low range, consistent with the nuclides decay energy of 355 keV.

These ratios are not only dependent on choice of energy ranges, but might be influenced by the actual detector. Together, these factors, along with the results in Table 2-7, lead to great uncertainty in interpreting the output from the TA device for Na-22. To aid in determining the validity of the results, the upper and lower discriminator settings of the nuclear instrument modules used to determine the detector ranges were repositioned to full open and full closed respectively, but the results did not change.

The results in Table 2-8 show that the TA device response did not match expected activities of the single isotope sealed gamma check sources, which were supplied by the manufacturer. In Table 2-8, the known gamma ray count rate (cps) of the sealed sources

(low, mid and high gamma) is multiplied by the Cal Factor, which was supplied by TA. Cal Factor is short for Calibration Factor, and the product of these values count rate (cps) and the Cal Factor yields the activity (pCi/L). The bottom line of the low, mid and high gamma sections of Table 2-8 list the actual activity measured by the TA device. The measured gamma activity is consistently lower than the expected values of the check sources. The discrepancy between the measured activity and the check sources indicates that the Cal Factors are incorrect or the TA device is not counting the check sources properly.

Table 2-8 Technical Associate Test Data for Gamma Spectrum Detector

Isotope	Na-22	Cs-137	Ba-133
Low Gamma cps <sup>1</sup>	5700	1400	4500
Cal Factor <sup>2</sup>	N/A <sup>3</sup>	N/A <sup>3</sup>	505.0
Gamma cps × Cal Factor (1×2) pCi/L	N/A	N/A	2,272,500
Average Measured pCi/L <sup>4</sup>	78,658	16,945	34,220
Mid Gamma cps <sup>1</sup>	3400	1650	60
Cal Factor <sup>2</sup>	313.6	2220.0	N/A <sup>3</sup>
Gamma cps × Cal Factor (1×2) pCi/L	1,066,240	3,663,000	N/A
Average Measured pCi/L <sup>4</sup>	1,812	112	0
High Gamma cps <sup>1</sup>	1900	40	30
Cal Factor <sup>2</sup>	561.0	N/A <sup>3</sup>	N/A <sup>3</sup>
Gamma cps × Cal Factor (1×2) pCi/L	1,065,900	N/A	N/A
Average Measured pCi/L <sup>4</sup>	19,601	0	0

Acronyms: cps, counts per second; Cal Factor, calibration factor for sensor; pCi/L, picocurie/liter; N/A, data not available

1 - Data obtained from Attachment 1, page 4

2 - Data obtained from Attachment 1, page 5

3 - The data was not listed on Attachment 1 and was unavailable at the time of testing

4 – Conversion of measured response by the detector from the calibration source during the test:  
Measured × Cal Factor

Even with inconsistencies, calibration problems, and other issues, the device might still be able to indicate that radioactive emissions from the water have changed, though it would not quantify the true activity of the agent. Therefore, to investigate this practical application, a solution of K-40 was prepared containing an activity of 31,500 pCi/L. The potassium-40 solution used during testing resembles the sodium-22 calibration source in the types and energy of the respective gamma decay emissions (K-40 = 1460 keV versus Na-22 = 1274 keV). The main difference is that K-40 emits gamma radiation in 10.7% of its decays while sodium-22 has gamma emissions for 99.94% of all decay events. The best expected response, based on results for Na-22 source testing and calculations, was a reading of 435 pCi/L in the low level gamma detector. The high level gamma detector should have had a result of 108 pCi/L. The measured result of the K-40 solution was 0.0

pCi/L for the high and mid gamma, and 1 pCi/L for Low Gamma. Resetting the high gamma discriminators to their full open positions, an average high gamma reading of 436 pCi/L was measured after 2 minutes, with a maximum reading of 577 pCi/L before testing was stopped. The activity reading had started to drop after the maximum reading was obtained, even though the source solution was in continuous contact with the detector.

This data resulting from the experiments with the K-40 solution does correlate with the calculated data from the measured activity of the standards, but it is not the true activity of the solution, and it does not correspond properly to the provided calibration data. In addition, the same potassium solution had beta detector readings of minus 612 cps and the decay of K-40 is 100% beta emissions at a high energy level. Based on these results, which suggest limited practical application for water utilities, testing of the other potassium solutions (lower concentrations), the americium solutions, and ruthenium solutions was not performed.

#### *Investigation into gamma response characteristics*

In an attempt to understand why the gamma responses were inappropriate and to gain insight that might inform future detector design for the device, the TA device was carefully examined. The inappropriate response of the primary gamma detector was hypothesized to be linked to geometry and calibration, therefore, our examination focused on these parameters. This detector is housed in a shield in which Tygon<sup>®</sup> tubing (Saint-Gobain Performance Plastics, Beaverton, Michigan) is looped to form a cylinder that surrounds the detector as seen in Figures 2-4 and Figure 2-5. The detector has an active end in which a 3 × 3 inch sodium iodide scintillation crystal resides. That crystal is optically connected to a photomultiplier tube which converts the light impulses produced into an electrical current used by the instrument to produce the readout. Figure 2-6 illustrates the relationship between the crystal and the sample. There is a large air gap between the detector and the sample and the vast majority of the sample resides away from the active area of the crystal. This would seem to be a poor geometry for sample analysis. Figure 2-7 shows that in laboratory instruments, the sample would be placed as close as practical around the active area of the crystal.

So, as hypothesized, geometry does affect the detector sensitivity, and it also potentially affects the calibration response factor. Namely, the operational check sources provided by the vendor are positioned so that they are along the active area of the detector and within 1 mm of the detector casing (Figure 2-6). This is not at all similar to the geometry between the water sample and the detector, which might explain why the calibration response factor might not be leading to appropriate results.



Figure 2-4 Gamma detector sample loop sitting above shield cylinder.

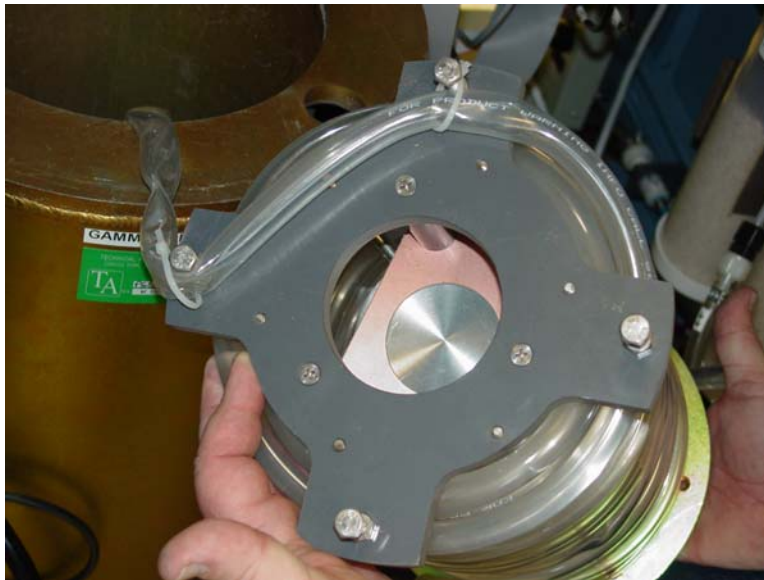


Figure 2-5 Gamma detector viewed from bottom showing detector mounting.

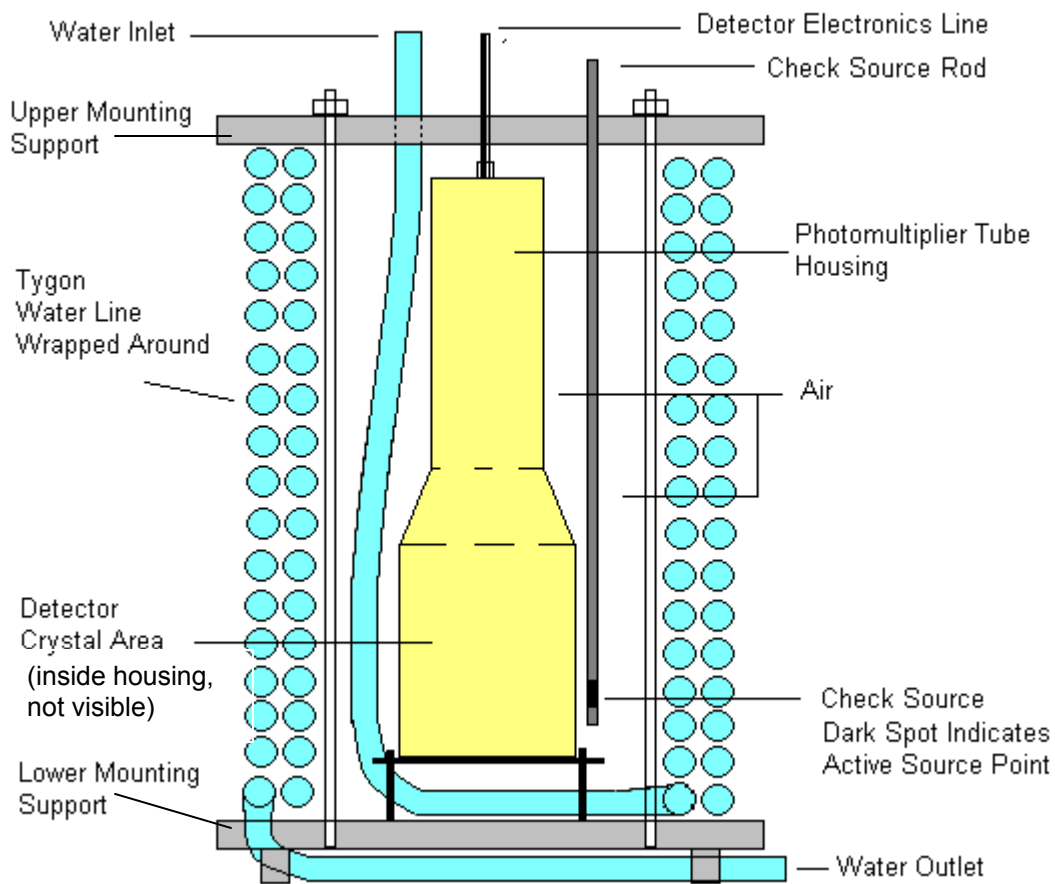


Figure 2-6 Gamma detector configuration in SSS-33-5FT.

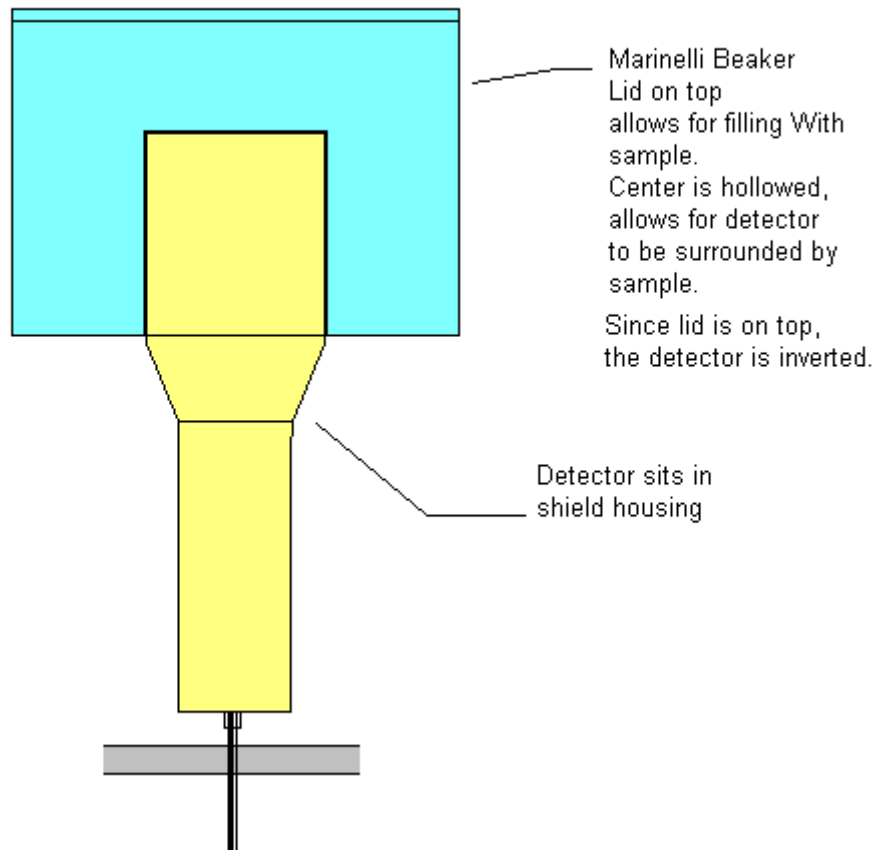


Figure 2-7 Typical laboratory configuration for sample analyzed with a NaI gamma detector.

In summary, and in keeping with the discussion above, a review of the instrument documentation and discussions with the manufacturer indicated that their in-house testing did not use liquid sources applied to the detector in an “as used” configuration, but employed static solid sources or bench scale tests. The sources employed by the manufacturer were in the microcurie range. Use of the static sources during the testing produced a positive response by the gamma detector. However, the manufacturer failed to provide a correlation of the comparability of the source check to the operational efficiency of the detectors. This analysis shows that the instrument did not perform per the manufacturer’s stated capabilities, and casts doubt on its ability to detect gamma radiological contamination in a drinking water system.

Subsequent to these tests, two portable sodium iodide meters used at NAREL were tested on the potassium and americium test solutions. The detectors and their instrumentation were able to detect and identify the isotopes in minutes without variation in signal response. The tests of these detectors are discussed in greater detail in the “Alternate Detector Design” section.

### *Beta Detector Tests*

The beta scintillation detector failed to measure activities above background. Introduction of the 15,000 pCi/L K-40 solution resulted in no activity measured on the beta detector. As shown in Table 2-3, K-40 has a high maximum energy beta, 1332 keV, and high average beta energy of 540 keV. K-40 is more energetic compared to the other isotopes selected for testing and had the best chance to provide a reading in the beta detector when weighed against the decay values of the other radionuclides to be used in the test.

To fully evaluate the detector, the discriminator settings were changed to allow for maximum signal input, and the result was a reading of 1309 pCi/L. Ramping the discriminator down to the factory setting resulted in immediate decrease to 0 pCi/L. The previous reading with open discriminator could be assumed to be a result of electronic noise coupled with signal from the interaction of radiation with the instrument and not a true reading. Since K-40 is an energetic beta emitter and the tests at this high level presented no activity measured, further testing was not performed.

These results might be explained by a combination of detector design and fundamental physics of beta particle detection in water. Although a plastic scintillation detector is routinely used in beta analysis, using it to detecting radiation in water is not as common. The typical sample measured with a plastic scintillator uses a planar detector and a dry sample. Water is a good attenuator of beta radiation. Beta particles similar to those in the measured samples have a travel range of 0.01 to 0.10 inches (0.25 to 2.5 mm) in water based on their maximum energies of up to 2000 keV, (Shleien, 1998). The amount of water in direct contact with the detector surface is smaller than the volume of sample in the detector, which is 430 mL. Detection works by water flowing across plates of the plastic scintillator. The distance between plates is approximately 0.16 in (4 mm), but this was estimated from the technical specifications. This indicates that a high energy beta particle would have the potential to interact with the scintillator within a maximum range of 2 mm. However, since the average beta energy is 150 to 500 keV, this range decreases to 0.25 to 1.3 mm in water.

This eliminates the average beta from interacting with the scintillator. Adding in flight and light losses, this theoretical efficiency would be diminished greatly in application. Typical beta particles traveling through air from a surface plated source result in efficiencies of 35% for a strontium-90 beta, with a 4 inch sample and a 5 inch detector of the same material coated with a zinc sulfide powder. Considering that beta energies in air are a hundred fold higher than in water, the relative efficiency of the detector to measure a beta emission through water could be as low as 0.35%.

### *Tritium and Alpha Detector Tests*

The final detector tested was also a scintillator. This detector uses anthracene, which is known for its high scintillation efficiency. The scintillation efficiency of anthracene (amount of light produced per unit of energy absorbed) allows for lower energy particles to interact producing a light pulse. This detector is employed in the measurement of tritium and alpha particles. Tritium is a beta emitter (Table 2-3) and it emits a very low

energy beta particle. The typical penetration of this beta particle in water is less than 0.007 inches or 0.002 mm (Shleien, 1998). The scintillation material is similar to sand and was placed into a small cylindrical container with two optical windows on either side held in place by a rim with an inlet and outlet port on opposite sides. This container holds less than 7 mL of water once it is filled with sand.

Injection of a tritium sample with activity of 10 million picocuries per liter yielded no response from the instrument. This might result from the anthracene volume being too thick relative to the range of light transmittal from interaction with the ionizing radiation. It should be noted that heavier charged particles, in this measurement alpha particles, have less scintillated light production than do beta particles. The true reason for the performance difficulties of this detector is unknown.

#### *Observations on System Design Relevant to Operational, Maintenance, and Decontamination*

Careful evaluation of the physical design of the instrument was performed to inform future detector design and facilitate evaluation of other detection systems. Below is a brief summary of numerous observations relevant to operational issues, including maintenance and decontamination. First, disassembly revealed numerous tubing diameters, connections and reduction fittings. This resulted in areas of trapped water that drained during disassembly. Many of the fittings were initially impractical to reach without cutting away retaining straps. Removal of tubing and fittings is common during repair and/or maintenance. If contaminated water was in the system, extensive contamination of the instrument housing and instrument components would have occurred. The subsequent total dismantling of the instrument and disposal or cleaning of the parts would lead to radioactive waste disposal issues.

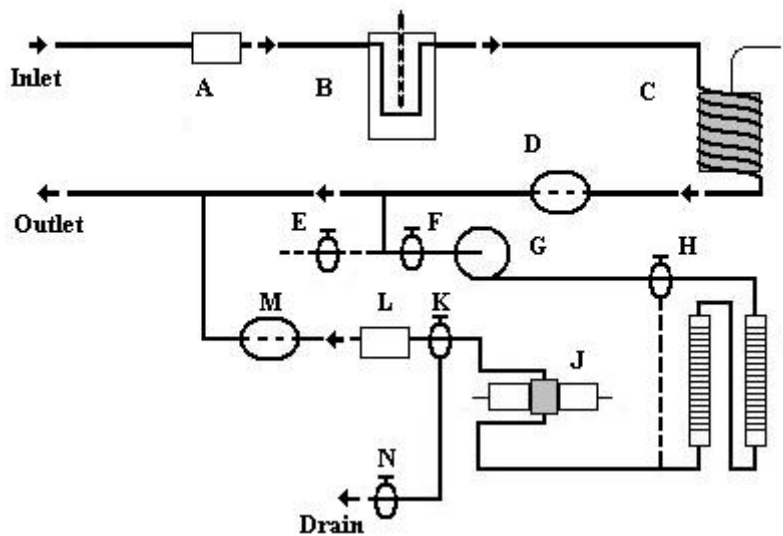
Observations related to detector operational factors were also collected. For instance, two of the detectors typically require the use of dark opaque tubing with a valve system or 90 degree joint in the flow path due to the light sensitivity and construction of the housing for the scintillation materials used. The use of the more common translucent tubing (as shown in Figure 2-4) can increase background counts beyond useful operating ranges. Another operationally relevant observation was that two detectors are housed in containers which act as shielding. These containers are a lead lined box for the alpha/tritium detector, and a steel cylinder capable of being filled with lead shot for the gamma detector. The need for such shielding in the latter is unclear, but might be necessary to reduce background from building materials. During testing, the presence of the high level solutions in the test room did little to alter background radiation readings on the instrument. Water is unshielded throughout the instrument for the remaining detectors (beta detector and the 2 × 2 inch NaI detectors co-located with the in-line filters).

The flow path of water through the instruments raises several questions and leads to several suggestions. The effects noted will impact the overall functionality of the system but will also vary based on the water supply and cannot be determined by these tests. Nonetheless, the implications merit further discussion. Figure 2-8 illustrates the flow



path of water through the system during normal operations. Water that enters the instrument passes through a UV disinfection unit. After exiting the system, water passes through an ion exchange unit and a filter unit. The goal of the UV unit is to prevent algae growth in the detectors. The filter unit is meant to prevent clogging, but this design has potential flaws. Plastic tubing, especially light transparent tubing, has been shown to allow for the growth of algae even with continuous water flow (Farshad, 2006). Water entering the detection system might not have been sufficiently disinfected to prevent algae growth. Thus, while the control of algae is important, the role of the UV in its present location is theoretically unclear, although this was not studied extensively.

Also related to the instrument design is that the low flow through the system coupled with the numerous bends and twists in the tubing might contribute to the degassing of the water supply, allowing vapor to be present in the tubing system, leading to potentially anomalous readings.



- A - Inlet Filter
- B - UV Disinfection System
- C - Sodium Iodide Gamma Spectroscopy Detector
- D - Main Flow Meter
- E - Tritium Calibration and Flush Water Inlet Isolation Valve
- F - Tritium Detection System Isolation Valve
- G - Metering Pump
- H - 3-way Port Valve
- I - De-iodization Cartridges
- J - Tritium Scintillation Detector
- K - 3-way Port Valve
- L - Tritium System Filter
- M - Secondary Flow Meter
- N - Tritium System Drain Isolation Valve

Figure 2-8 The SSS-33-5FT normal operational flow path.

The use of ion exchange resin in-line with the system is probably designed to remove all radionuclides except tritium which would likely be in the form of tritiated water.

However, in employing this strategy, it is important to remember that these units will remove dissolved and particulate material from the water, respectively. The effectiveness at removing radioactive material from source water by these technologies has been demonstrated and the results identified in regulation (40CFR 141.66(g)). The end result is that the radioactive material will simply be concentrated, increasing background radiation over time, thereby increasing the detection limit. This reduction in the sensitivity of the detection system is not desirable. This situation might be further exacerbated by the design of the TA unit places the sodium iodide gamma detectors are located next to the filter unit and ion exchange units. Thus, not only will the tritium detector be affected, but the results for other radionuclides will suffer as well.

#### *Alternate Detector Design*

As one goal of this study was careful physical and performance evaluation of the TA unit, the observations from the testing and evaluation of the TA unit lead to several suggestions for alternate detector designs. The first is related to the seemingly unfortunate placement of the ion exchanger and filter near the gamma detector. Interestingly, monitoring the activity of the ion exchange filter maintained in a plastic housing has some potential merit, although this would require further testing. If a portion of feed water was fed to the ion exchange column, the column activity would gradually increase over time. Conductivity monitoring of the resin outlet would indicate when it would require change out. However, a spike of radioactive material would result in a spike in the reading of the detector. Still, development of a proper alarm system based on rate of change might be a simple and effective monitoring technique.

A second observation relates to the potential use of solid state multi-channel analyzers (MCA) in water radiation monitors. MCAs employ sweeping algorithms to monitor 1000 to 4000 channels over a range of energy from 50 to 2000 keV, as opposed to the discrete energy windows in the TA unit design, also known as Nuclear Instrumentation Modules (NIM). MCAs allow identification of the incident photon by energy. Furthermore, with proper calibration and a library installed into a software program, MCAs can potentially quantify a measured radionuclide. This type of information would be more useful to a water supplier. Current MCAs also allow the incorporation of alarm set points when certain count rates are achieved, or, as in the case of the EPA RadNet system (<http://www.epa.gov/narel/radnet/>), when count rates in an energy region of interest are exceeded. [RadNet is a national network of monitoring stations that regularly collect air, precipitation, drinking water, and milk samples for analysis of radioactivity.] The RadNet system employs the same type of detector over a filter that is gathering particulates from the air. The detector software produces a spectrum that is divided into 10 energy regions. Each region is enabled with an alarm function, allowing the operator to check the data and analyze the data as necessary.

To perform a preliminary investigation of this approach with MCAs, the test solutions that emitted gamma radiation were analyzed using instrumentation employed by the NAREL Radiological Emergency Response Team (RERT). The two pieces of equipment

used included a 3 × 3 inch NaI detector and a 4 × 16 inch NaI detector each coupled to a Berkeley Nucleonics SAM-935 Spectral Analyzer (Berkeley Nucleonics Corporation 2955 Kerner Blvd., San Rafael, California).

Testing results from the use of available EPA RERT equipment gives the assumption that a gamma spectral analyzer can be developed for use by water companies to identify high level activity in water (10's to 100's of nanocuries per liter). The 3 × 3 inch RERT instrument was able to identify a rudimentary peak for Am241 at 25,000 pCi/L within 1 minute. The 4 × 16 inch RERT detector clearly identified the same Am-241 peak in the same time frame. Both instruments clearly identified the K-40 gamma peak after 1 minute. Both instruments were energy calibrated, but were not calibrated for the geometry of the samples measured (a 5 gallon carboy). Figures 2-9 and 2-10 show the detectors and how they were used. A 3 × 3 inch NaI detector coupled to a rudimentary multi-channel analyzer was also tested as shown in Figures 2-11 through 2-13. This instrument was calibrated for energy scaling of the output range and was also capable of identifying the K-40 peak within 2 minutes. Also, many of the multi-channel systems developed for use with hand-held gamma detectors have alarm functions that can be either pre-programmed or user programmed.

The results clearly indicate that a proper equipment design could be implemented in a straightforward manner. Also, a system could be developed to provide a system similar to RadNet that would allow transmittal of data that could provide basic information to water utilities and, in addition, provide a valuable asset for researchers. Data from gamma emitters alone could cover approximately 80% of the radionuclides of concern, but this does leave significant coverage gaps for some radionuclides of interest. There are several advantages to this approach, but two are the most important. First, a water system would be monitoring a stable background spectrum, as compared to RadNet's background spectrum rises over time as material is collected on the air filter. Radioactive material should not be accumulating in a water radiation monitoring system, so subtle changes in the spectrum would be easily recognizable. Second, a telepathy system, perhaps based on water utilities' existing SCADA (supervisory control and data acquisition) capabilities, could be implemented that would provide faster feedback into potential decisions for emergency response, as is currently done with RadNet. Electronically transmitting results would provide feedback within hours.



Figure 2-9 Encased 3 × 3 and 4 × 16 inch NaI detectors against 4 liter Am-241 sample.

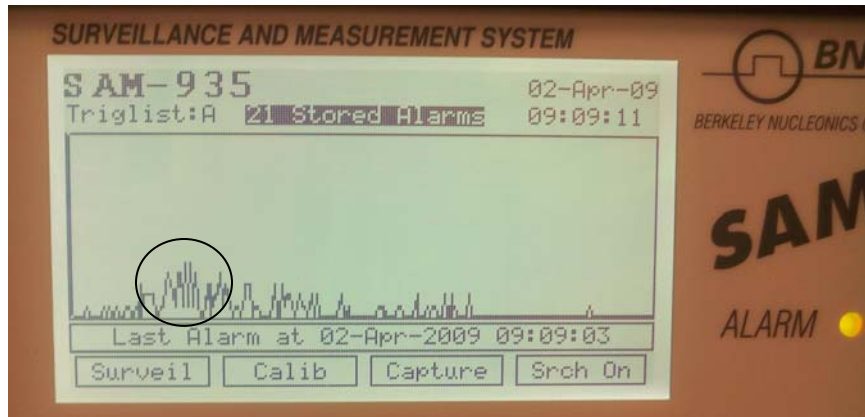


Figure 2-10 Readout of 3 × 3 inch detector showing peak at left from Am-241 after 2 minute acquisition.



Figure 2-11 Encased 3 × 3 inch detector setup in glass jar.



Figure 2-12 Encased 3 × 3 inch NaI detector submersed in 20 liter K-40 solution.

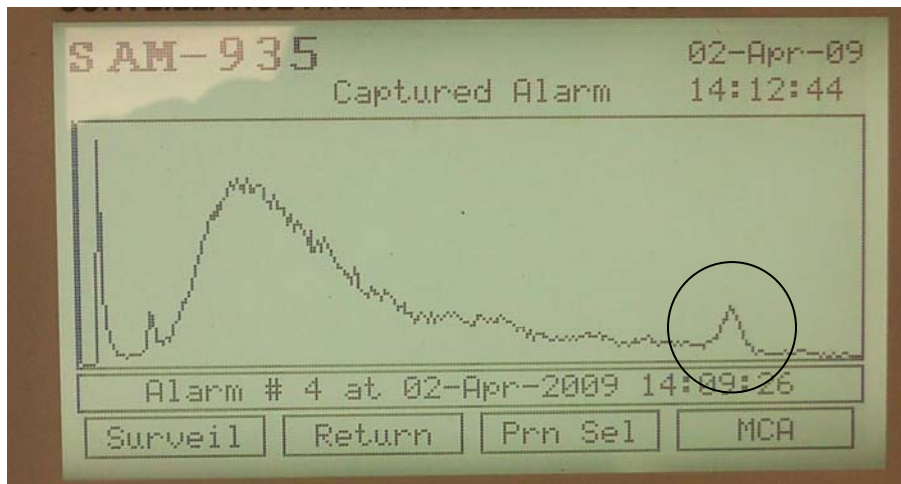


Figure 2-13 Encased 3 × 3 inch NaI detector submersed 10 minute readout showing K-40 (peak at far right).

The measurements of alpha and pure beta emitters present different problems. The use of anthracene in detection might theoretically work, but the functional design and construction of the actual detector needs to be examined before the technology is deployed. A larger volume of anthracene might be needed inside the detector, but one with a thinner overall depth to prevent internal absorption of the light photons produced. Also, different configurations of the plastic scintillator should be tested in the future for beta detection. Examination of the literature indicated that the Department of Energy tried to develop a plastic detector, but drew the plastic out in strands, much like spaghetti in a long container (Baldwin, 2003). Current literature has shown that a detector has been able to measure gamma emissions from palladium-103 at 2.6 mCi, (Sliski, 2006). This is a concentration 2600 times higher than what was used for the beta detection experiments. The gamma emissions of Pd-103 are mid range energies for gamma (356 – 583 keV), but have energies equivalent to the beta energies used in this experiment. Work done by Beddar (2006) at the University of Texas has demonstrated that plastic scintillators are extremely capable of measuring electrons in the million electron volt range. Tests have demonstrated that the measurement of high energy beta particles is possible at ranges as low as 40,000 picocuries per liter utilizing a liquid scintillator vial made of a plastic scintillation material (Ogata, 2007). This literature review shows the potential for development of a functional plastic scintillation instrument.

### **Conclusion and Summary**

Objective 1 of this study was to determine if the TA SSS-55-3FT on-line radiation detector able to detect radiological contamination and, if so, at what minimum radioactivity level. Testing of the SSS-33-5FT has shown that instrument would not detect radioisotope in water at the PAG or MCL. Test results did not match manufacturer detection claims.

Objective 2 of this study was to evaluate the TA device in terms of its usability for drinking water system operators, and determine if it could fulfill drinking water utilities' needs. The instrument was demonstrated to be difficult to use in a laboratory setting and, therefore, unsuitable for field applications like water treatment facilities. Examination of the physical design of the TA unit resulted in many concerns, but perhaps point the way towards an improved system. For instance, the industry has moved away from the Nuclear Instrumentation Modules, the technology used in the TA device for operation of gamma detectors, and has moved toward multi-channel analyzers (MCA), which have the ability to detect specific isotopes. Indeed, after being briefed on this suggestion, TA developers have incorporated an MCA into their latest model.

The results of this study suggest an effective radiation monitoring system can be developed for drinking water supply systems. Current attempts have failed to be properly engineered or tested prior to use of the instrument in the field. Gamma monitoring systems should be available relatively quickly with current off-the-shelf systems, but they should be tested prior to deployment despite their seeming promise. Alpha and beta monitoring systems, however, will require more extensive research and development. The system need not be complicated, either in design or operation, but concerns over calibration and usefulness of data output need to be addressed.

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## Attachments

- 1 Technical Associates SSS-33-5FT Test Data and Calibration Information