A REMOTELY OPERATED, FIELD-DEPLOYABLE TRITIUM ANALYSIS SYSTEM FOR SURFACE AND GROUND WATER MEASUREMENT

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ABSTRACT. We have worked in collaboration with the Westinghouse Savannah River Company and Packard Instrument Company to develop a prototype instrument for remote, near real-time, *in situ* analysis of environmental levels of tritium in surface and ground water samples. The unit consists of a Packard Radiomatic™ 525TR liquid scintillation counter that has been modified to achieve lower detection levels through background reduction using Packard's After-Pulse Burst Discrimination electronics and an auxiliary $Bi_4Ge_3O_{12}$ scintillation detector guard. The system is fully programmable to enable remote calibration, sample collection, sample analysis, reconfiguration of operational mode and interrogation for analytical results.

INTRODUCTION

The environmental contamination resulting from decades of testing and manufacturing of nuclear materials for national defense purposes is a problem now confronting the United States. Nationwide, an estimated 2.6 MCi of fission products has been released to the environment by a trio of facilities that reprocess nuclear fuel from production reactors: the U.S. Department of Energy (DOE) sites located in Hanford, Washington, Aiken (Savannah River), South Carolina, and Oak Ridge, Tennessee (Bradley, Frank and Mikerin 1996).

At the Savannah River Site (SRS), a DOE facility responsible for the production of tritium and other special nuclear materials, there have been continuous low-level releases of an estimated 24 MCi of ^3H from operational and disposal processes and unplanned events, which have affected ground and surface waters (Murphy *et al.* 1991). As one of the most widespread and mobile contaminants found on site, ^3H poses a potential environmental hazard to populations in nearby communities and in the downstream Savannah River watershed. The management of the high- and low-level radioactive wastes resulting from SRS activities is a problem of ongoing concern, particularly in the face of continued cutbacks in the funding available to support the environmental monitoring programs necessary for tracking and control of the waste streams. For many years, the characterization of ^3H in ground and surface waters at the SRS has been conducted by manual sample collection and laboratory analysis. With present-day advancements in computers, remote communication technologies, and low-background radiation detection systems, the components are now in place for the development of remotely operated, field-deployable analytical systems for near real-time automated screening or monitoring of potential and existing contaminant plumes.

The Center for Applied Isotope Studies (CAIS) at the University of Georgia, in cooperation with the Westinghouse Savannah River Company (WSRC) and Packard Instrument Company, have developed a prototype unit for remote, near real-time, *in situ* analysis of ^3H in surface and ground water samples. The Field-Deployable Tritium Analysis System (FDTAS) is sufficiently sensitive to measure ^3H in water samples at environmental levels, and will be applied at the SRS for waste site and risk assessment, field monitoring and screening, and state and federal environmental compliance activities. The system is designed for remote control so that multi-site sampling, analysis, and data handling may be automated for on-demand or unattended operations (Fig. 1).

METHODS

The goal of the FDTAS development program was to design and test a field-deployable system capable of rapid and remote liquid scintillation (LS) measurement of ^3H in aqueous environmental

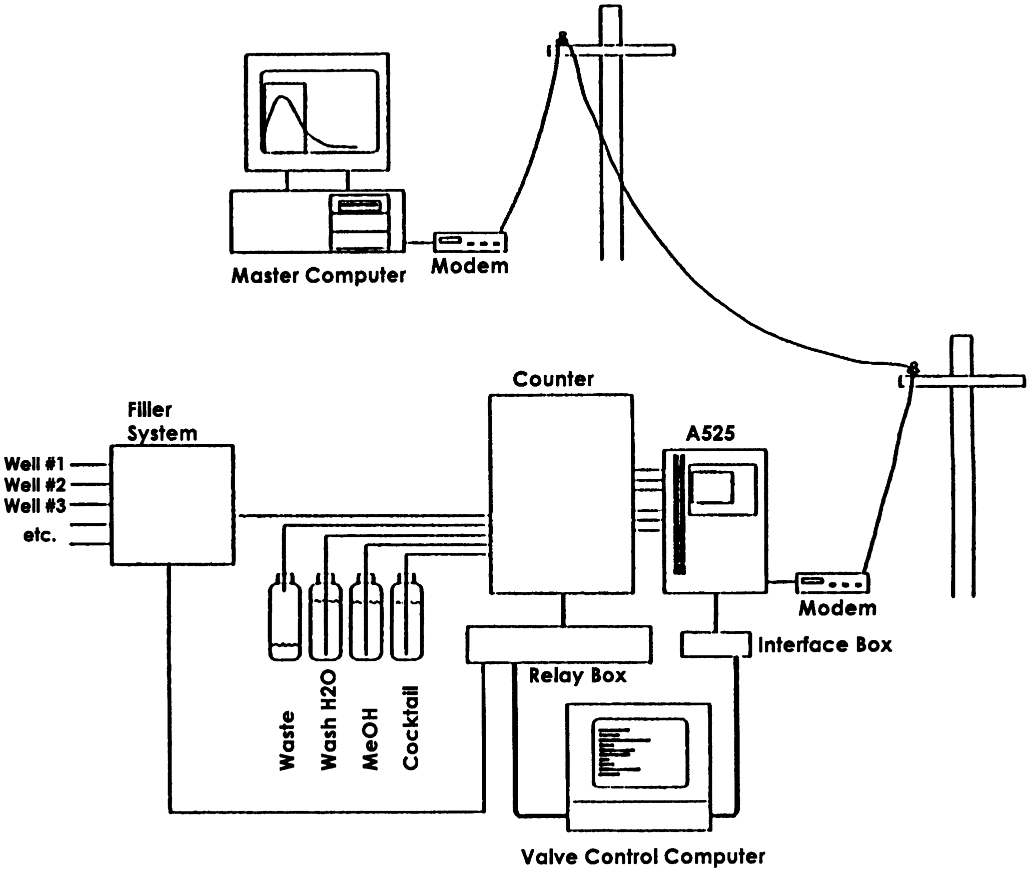

Fig. 1. Schematic diagram of the Field Deployable Tritium Analysis System

samples. Specifications for the system included a background of ≤3 counts per minute (cpm) and a counting efficiency of 25%, with a minimum detectable activity (MDA) of 10 Becquerels of radioactivity per liter (Bq L^{-1}) for a 100-min count. Sample standardization, between-sample cleansing, and two-way remote communication capabilities were of primary importance for system development. The FDTAS is designed to allow a laboratory-based operator to instruct a remote unit (or units) to collect and analyze a sample, to reconfigure its mode of operation, to conduct an internal calibration, or to be interrogated for analytical results. Direct-dial modems provide the telephone communication and control link between the laboratory computer and a remote LS counter.

The Packard Radiomatic™ 525TR LS counter (Fig. 2) was selected as the basic instrument for low-level ^3H measurement, but required a number of system modifications in order to meet the desired specifications. To accommodate measurement of the low levels of ^3H expected in aqueous environmental samples, the aqueous sample volume had to be increased to 5 mL and measured on a flow/stop (batch) basis rather than on the continuous basis for which the 525TR was originally designed. Background reduction was critical in achieving the lower detection levels specified, and was accomplished by adding lead shielding, redesigning the counting chamber and sample vial holder, and incorporating $Bi_4Ge_3O_{12}$ (bismuth germanate, or BGO) windows in the counting chamber, coupled with afterpulse counting electronics.

Fig. 2. Packard Radiomatic™ A 525TR liquid scintillation counter

The prototype FDTAS is now enclosed in a field-portable steel housing containing additional lead shielding (Fig. 3). The redesigned counting chamber is made of high-purity electrolytic copper that is coated on the interior with a white reflective paint to enhance photon collection by the photomultiplier tubes. The chamber accommodates a specially designed 10 mL flow-through quartz sample vial of 8 mm thickness and 40 mm diameter. A pair of BGO windows, 6 mm thick, are incorporated in the counting chamber for reduction of background, and bracket both sides of the quartz sample vial. The counting chamber, quartz sample vial and BGO window configurations were optimized through a series of tests that were conducted using optical modeling software.

To accommodate the need for multi-sample injection without sample cross-contamination, a flow/stop sample injection and system cleaning mechanism was assembled for the FDTAS using solenoid valves and metering pumps. This flow system assembly is controlled by a 24-channel relay accessory board and parallel digital I/O board in the valve control computer, permitting the collection of samples from multiple sites, with computer-automated sequencing of sample injection, cocktail mixing, sample counting, sample outflow and cell cleaning cycles.

A range of LS cocktails, including Packard Ultima Gold™ LLT, Ultima Gold™ AB, Ultima Gold™ XR, and Ultima Flo™ M were evaluated for use with the FDTAS. In addition to the parameters of background and counting efficiency, the viscosity of the cocktail also required consideration. Sample backgrounds were determined using a Packard Tri-Carb® 2550 LS counter. The cocktails were then spiked with a known ^3H activity and recounted to determine counting efficiencies. Ultima Gold LLT, normally a clear choice for ^3H measurement due to a low background and high counting efficiency, was found to be too viscous to be readily washed from the sample vial during the cleaning cycle. The two least viscous cocktails, Ultima Gold XR and Ultima Flo M, were found to have sig-

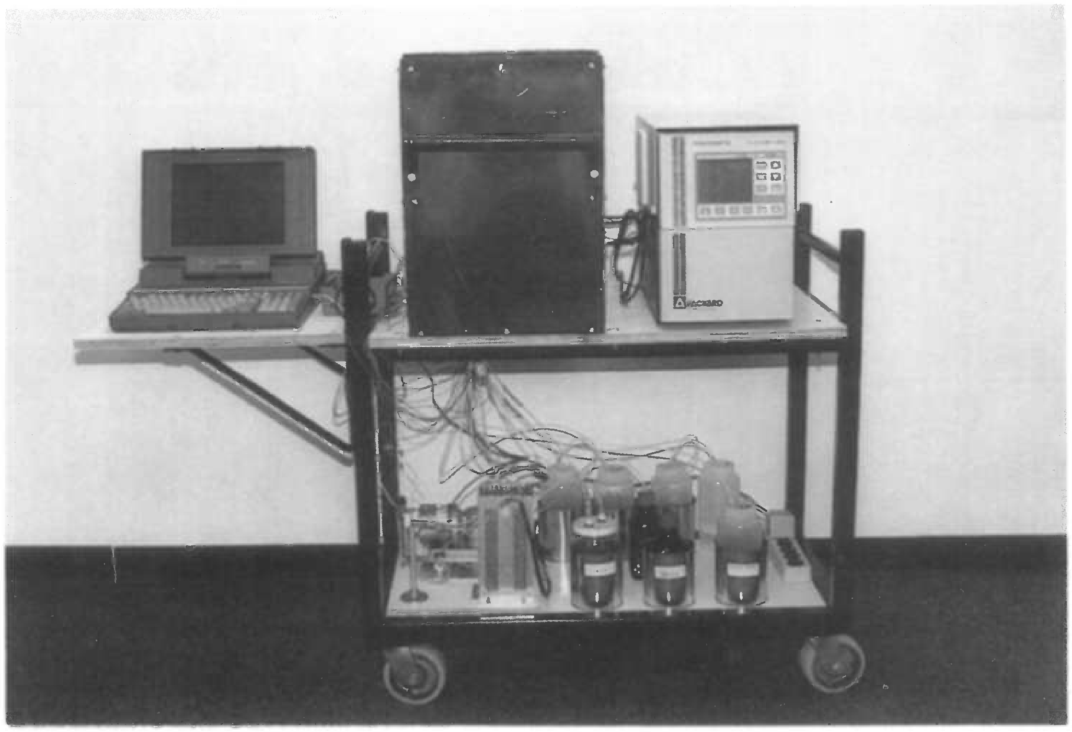

Fig. 3. FDTAS field-portable configuration

nificantly higher backgrounds and lower counting efficiencies when compared to Ultima Gold AB. Although Ultima Gold AB is a more viscous cocktail, it could nevertheless be washed fairly easily out of the sample vial, thus providing the best compromise for use with the FDTAS.

Using Ultima Gold AB, a series of mixtures having cocktail:water ratios of 40:60, 50:50, 60:40 and 70:30 were prepared and counted to determine background. The mixtures were then spiked with a known ^{3}H activity and recounted to determine counting efficiency. The 50:50 cocktail:water ratio was found to produce the best overall results for FDTAS ^{3}H measurement.

Several tests were also conducted to maximize the efficiency of the flow-cell cleaning cycle to eliminate cross-contamination of samples. Of the alcohol solvents tested, methanol was found to produce the best cleaning results. Three washes of pure methanol, followed by three washes of nontritiated water, serve to eliminate completely any sample "memory" in the system. The effectiveness of the cleaning cycle is demonstrated in Figure 4, which shows spectral data on sequentially measured samples including 1) the background on a nontritiated water sample; 2) a tritiated water standard (400 dpm); and 3) the background on a second nontritiated water sample following the methanol/water cleaning cycle.

RESULTS

The remote sampling and analysis capabilities were tested at the CAIS laboratory in March 1996. The master computer was set up in one laboratory and the LS counter and valve control computer were located in a separate laboratory. Using telephone/modem linkages, the sample collection/analysis/cleaning cycle was successfully operated from the remotely located master computer. A back-

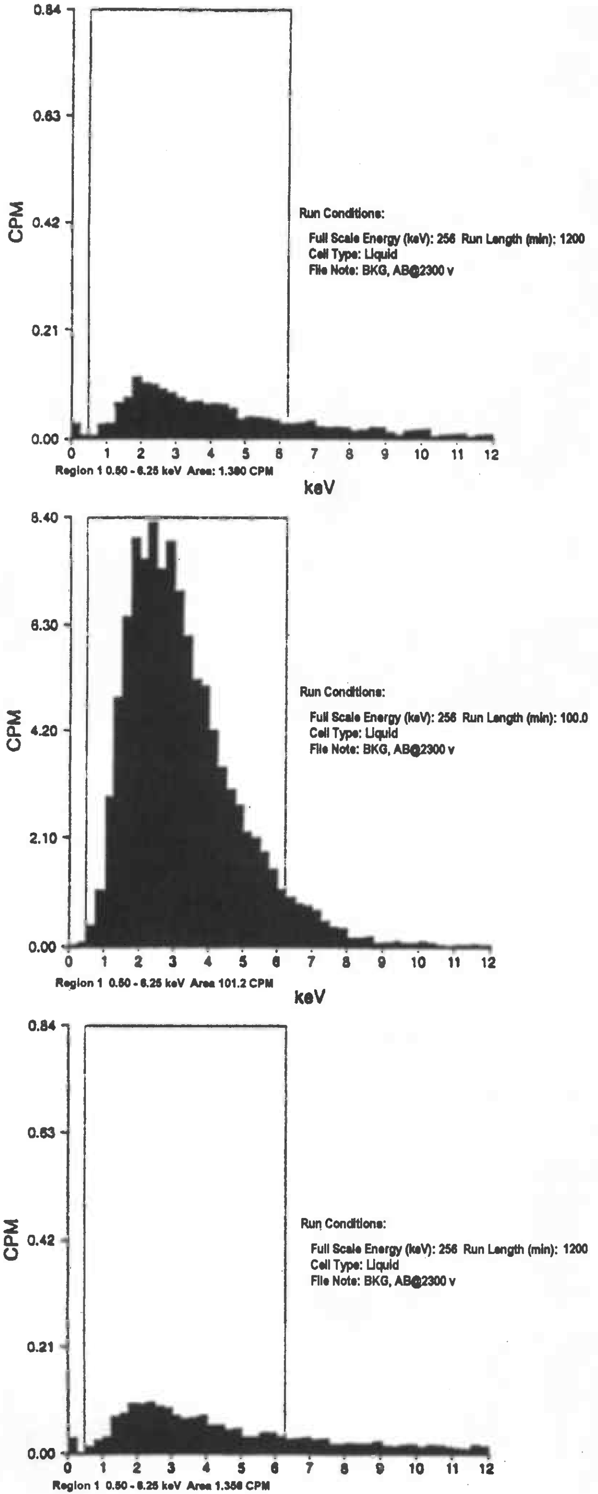

Fig. 4. Results of FDTAS cleaning cycle test

ground of 1.35 cpm and a counting efficiency of 24% were demonstrated in this laboratory setting, yielding an MDA of less than 7 Bq L^{-1} for a 100-min count and meeting the original specifications for the FDTAS.

A preliminary field test conducted on site at the SRS in May 1996 was similarly successful. The FDTAS field unit was placed in a temperature- and humidity-controlled enclosure mounted on a small trailer for ready mobility (Figs. 5 and 6). The test location selected for the FDTAS was "L" Lake, which was constructed in 1985 to receive the heated effluents of one of the five nuclear production reactors at the SRS. "L" Lake is located *ca.* 17 km from the central laboratory where CAIS and WSRC personnel set up the master computer. Remote communications between the laboratory and the field site were established using telephone and modem. Using the master computer, CAIS and WSRC personnel instructed the FDTAS to collect an aqueous sample, to acquire and analyze the sample spectrum, to evaluate background, and to empty and cleanse the sample vial in preparation for the next sample cycle. A background of 1.469 cpm and a counting efficiency of 23.9% were demonstrated in the field, which resulted in an MDA of 10.13 Bq L^{-1} for a 50-min count. An aliquot of the aqueous sample was brought back to the CAIS laboratory and measured using a Quantulus 1220™ LS counter. Figure 7 shows a ^3H spectrum collected with the FDTAS; Table 1 presents a comparison of field and laboratory measurement results, which are in very good agreement (Note: the laboratory sample was *ca.* twice the volume of the field sample, resulting in a lower MDA for the 50-min count).

Fig. 5. Trailer housing for FDTAS field unit

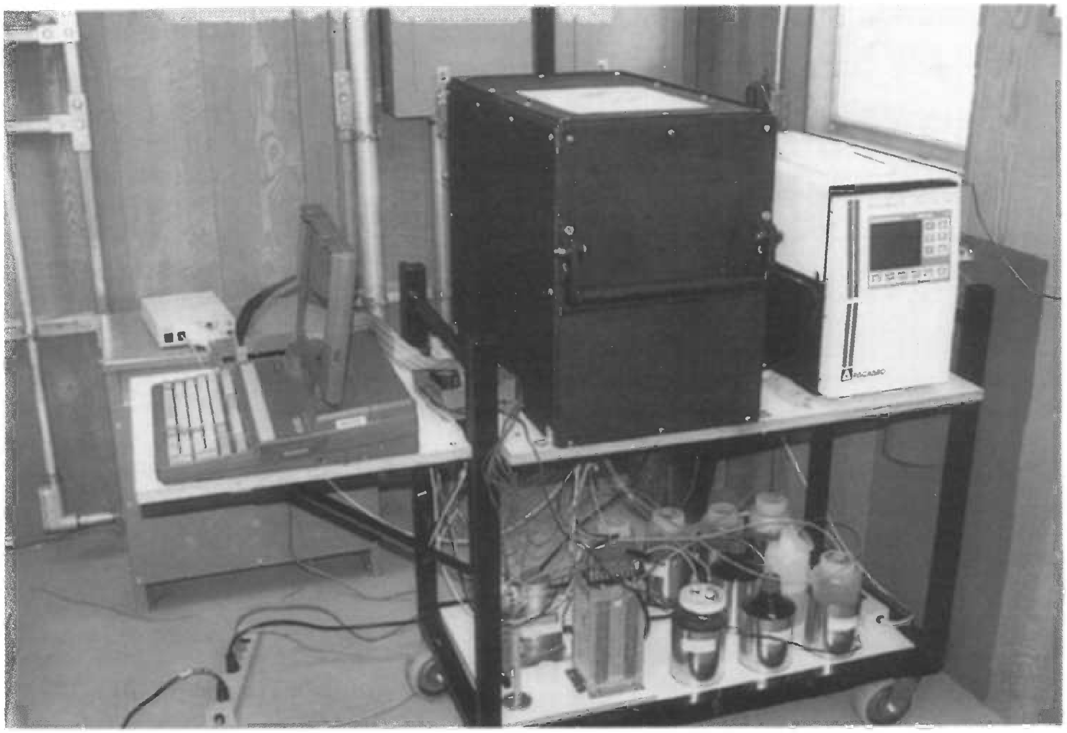

Fig. 6. FDTAS set up inside trailer

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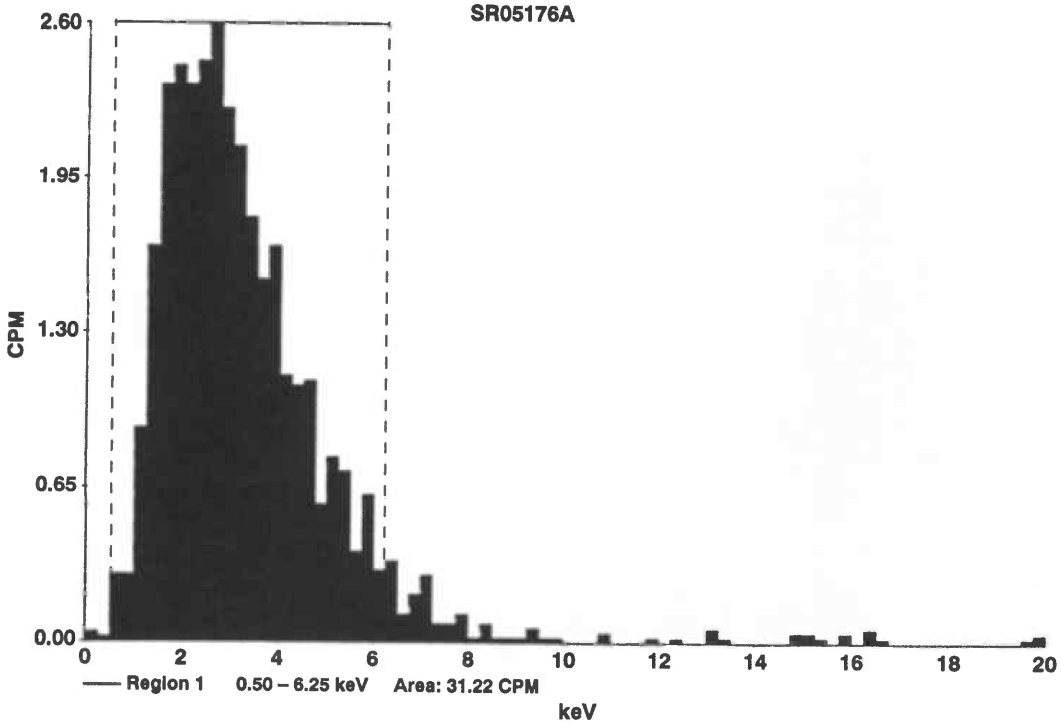

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Fig. 7. “L” lake ^3H spectra

TABLE 1. Field/Lab Data Comparison: Low-level ^3H Analysis Report, WSRC “L” Lake Water

Instrumentation	Sample description	MDA (Bq L^{-1})	Gross CPM	Background CPM	Counting efficiency	Activity (Bq L^{-1})
FDTAS	“L” Lake edge	10.13	31.22	1.469	0.239	377.47
Quantulus	“L” Lake edge	5.48	56.47	1.460	0.243	377.24

A second test was performed in June 1996 at an SRS site where groundwater was being treated for removal of trichloroethylene by gas stripping. A background of 1.5 cpm and a counting efficiency of 27% resulted in an MDA of 6.41 Bq L^{-1} for a 100-min count. As in the first SRS field test, an aliquot of sample was collected and measured in the laboratory using a Quantulus 1220 LS counter. A ^3H spectrum collected in the field using the FDTAS is presented in Figure 8, along with a comparison of field and laboratory measurements (Table 2), which are also in very good agreement. For these preliminary tests, the aqueous samples collected were introduced directly into the FDTAS following a filtration and cleanup process performed by WSRC personnel using EI ChroM resins.

The final months of the project were devoted to the development and integration of an external standardization capability and a fully automated multiport sample filtration system for the FDTAS. The CAIS directed its efforts toward the development of the external standard, which provides assurance

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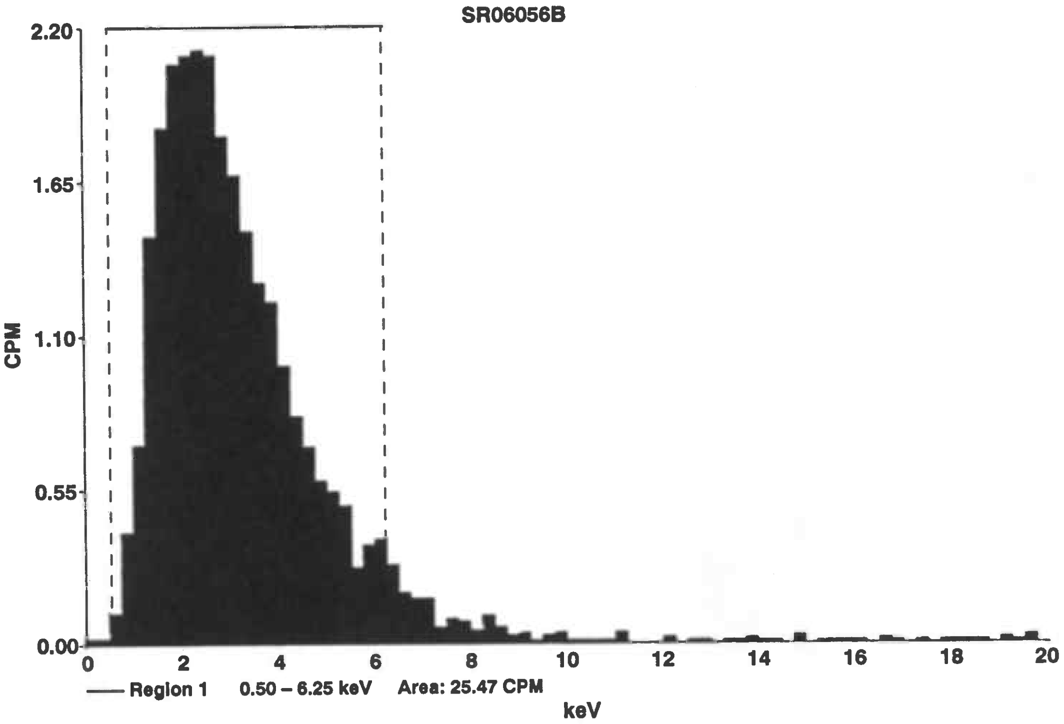

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Fig. 8. Stripper unit ^{3}H spectra

TABLE 2. Field/Lab Data Comparison: Low-level ^{3}H Analysis Report, WSRC Stripper Unit Water

Instrumentation	Sample description	MDA (Bq L^{-1})	Gross CPM	Background CPM	Counting efficiency	Activity (Bq L^{-1})
FDTAS	Stripper output–treated	6.41	25.47	1.50	0.27	269.02
Quantulus	Stripper output–treated	4.70	28.64	1.35	0.35	236.93

of sample quality with regard to both the quantity and the water/cocktail mix desired. Following a preliminary investigation of both ^{133}Ba and ^{241}Am as potential radiation sources for use in the external standard, the CAIS ordered an encapsulated ^{241}Am source. The lead shield and copper counting chamber of the FDTAS were then modified to accept the source and its custom-designed delivery system hardware. A series of tests were run to determine quench levels and ensure that the ^{241}Am source produced sufficient signal for use with the FDTAS.

WSRC personnel, in conjunction with Sampling Systems, Inc., of Houston, Texas, directed their efforts to the sample filtration system, which is designed to reduce quenching agents in the aqueous samples before they are delivered to the FDTAS for analysis (Fig. 9). This system permits remote sample acquisition from selected sample sites (*i.e.*, specific wells within a cluster of wells) following commands issued through the master computer, and results in more accurate ^{3}H measurements

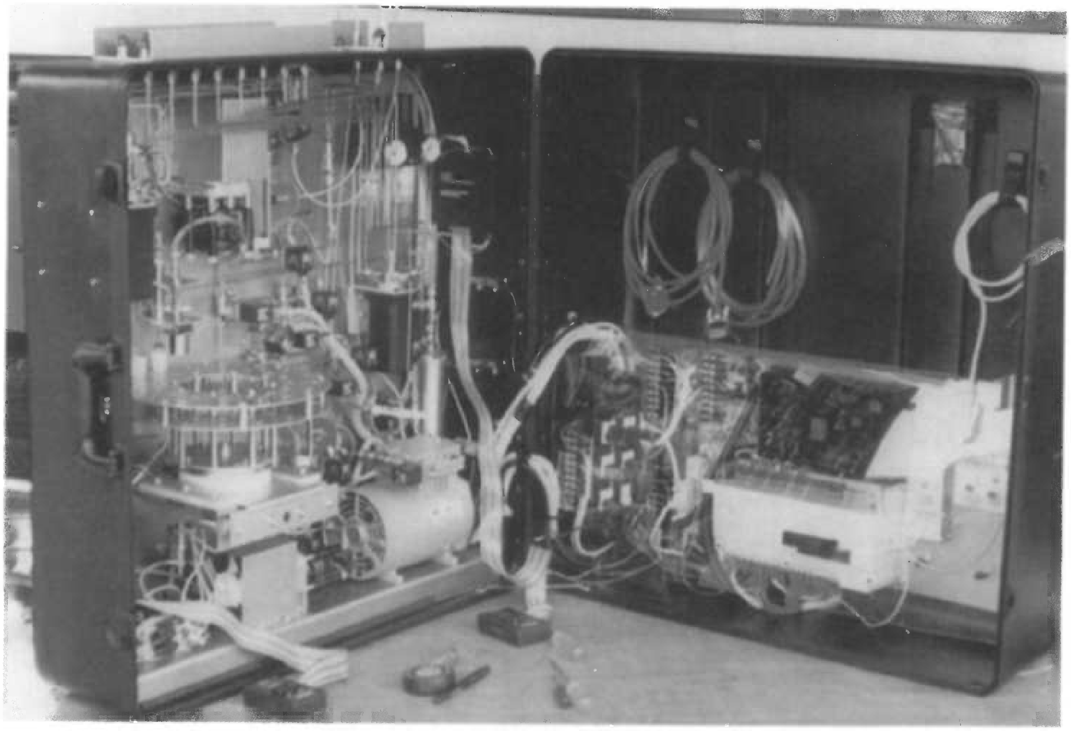

Fig. 9. Sample filtration and cleanup system

through the correction of quench problems related to LS counting of tritiated water. The FDTAS was integrated with the sample filtration system and its analytical capabilities demonstrated during a series of tests performed on-site at the SRS in November 1996.

CONCLUSION

The Field-Deployable Tritium Analysis System has promise of wide application to areas of public concern such as the U.S. DOE nuclear fuel reprocessing facilities, which require a highly sensitive level of monitoring that cannot presently be achieved on a cost-effective basis. By permitting collection and analysis of aqueous samples on a selective, on-demand, and rapid basis, the costly routine sample gathering and lengthy laboratory analytical processes now used to monitor ^{3}H in on-site wells and surface waters can be eliminated. The mobility and portability of the field unit, combined with the central laboratory operation of multiple units (Fig. 10), enables monitoring programs to be conducted with far fewer personnel than is now possible.

With some modification of the system electronics, the FDTAS also has the potential for use as a combined alpha/beta/gamma counter. An $\alpha/\beta/\gamma$ counting capability, coupled with the remote field operational parameters of the FDTAS, would have a broad range of applications in environmental monitoring. Current LS counters can effectively differentiate and measure α and β radiation through the use of pulse-shape analyses. The pulses of the BGO detectors, presently used to discriminate against γ events for background reduction, could conceivably be stored for a subsequent γ analysis. This capability would permit rapid and remote measurement of anthropogenically released radionuclides such as ^{226}Ra, ^{228}Ra, ^{89}Sr, ^{90}Sr, ^{90}Y, ^{137}Cs and ^{60}Co in addition to the measurement of ^{3}H in surface and ground water samples.

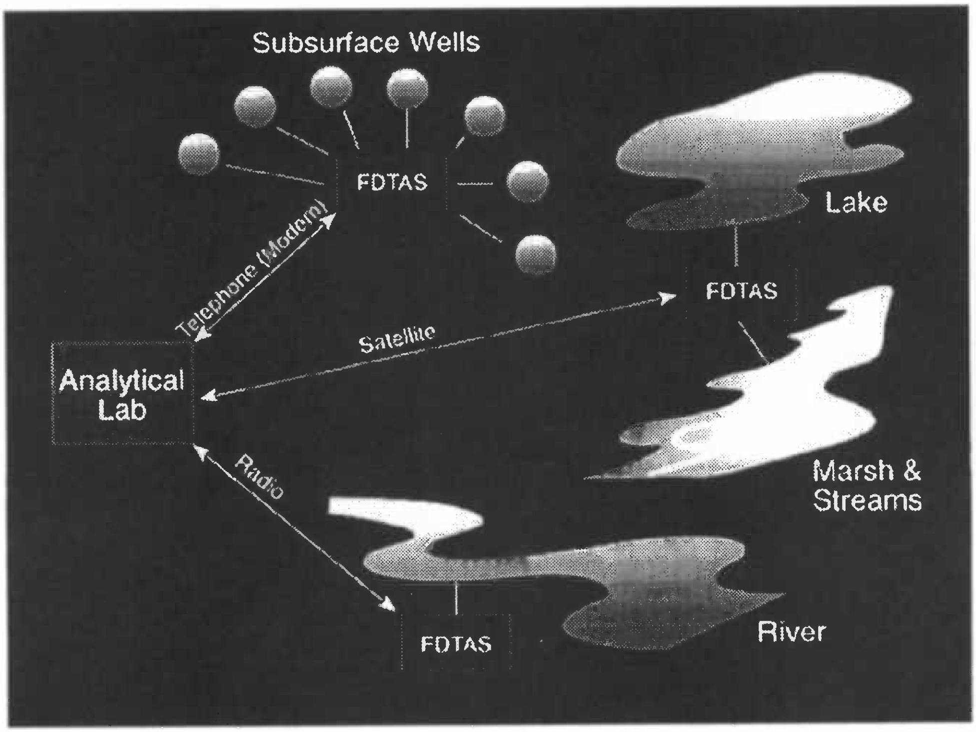

Fig. 10. Schematic diagram of possible FDTAS deployments

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