High Sensitivity Measurements of Ultra-Low Amounts of Radioactivity in the Environment

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Abstract

Since the first water sample was taken from the Savannah River on July 26, 1951, continuing technological advances in high sensitivity measurement of ultra-low amounts of radioactivity are being made at the Savannah River Site (SRS). Notable achievements in this nuclear technology area have been recognized locally, nationally, and internationally. During the "Cold War" peak nuclear material production period, the capability achieved in high sensitivity radioactivity measurement technology demonstrated the resolve of the Savannah River Site to be good stewards of the environment. This is further demonstrated by the extremely low doses of radiation received by the surrounding population from Site operations, which are far below that from the natural environment.

Continued achievements in high sensitivity ultra-low radioactive measurement technology since the Cold War have created, in addition to site emergency response, new missions in nonproliferation international safeguards, national security against terrorism, nuclear smuggling, and state and local law enforcement.

It is these achievements at SRS in high sensitivity ultra-low measurement capabilities that make for a cleaner, safer, and more secure nation and world.

In the Beginning

The development of measuring ultra-low amounts of radioactivity in the environment began following the first water sample collected from the Savannah River in early 1951 by a core of Du Pont health physicists led by Bill Reinig. This was the first pre-operation environmental survey ever conducted prior to the construction and operation of a U.S. nuclear production or commercial facility (Bebbington 1990).

In conjunction with the Philadelphia Academy of Science and under the direction of Dr. Ruth Patrick of the Department of Limnology, river and the surrounding land environment monitoring stations were established out to 100 miles of the Savannah River Plant (SRP as it was then known) to routinely collect samples for radioactivity measurement. During the next one and one-half years (June 1, 1951 to January 1, 1953), about 6600 environmental samples were collected and analyzed.

By today's standards, only crude technologies existed in 1951 to measure radioactivity. These technologies included the analytical wet chemical separation of elements, light emitting phosphors (ZnS) for measuring alpha activity, and Geiger Mueller counters using a counting scalar to measure beta-gamma activity. By knowing the element separated, the radioactive isotope could be closely identified. Using a series of thin aluminum plates of known thickness (mass) placed between a separated element and the counter, an absorption curve could be produced to determine the energy of the activity and the radioactive isotope from its discrete beta energy emitted. This very tedious method required many technical analysts and rooms of counting equipment to determine the very low amounts of background radioactivity, which are naturally occurring in nearly all materials including the human body.

The Methods Chemistry and Radiation Physics Groups, composed of several chemistry and engineering scientists, were formed in the

Health Physics Department to improve upon these measurement methods to be more efficient, cost-effective, and become more sensitive to meet a rapidly growing concern of the not only local but the entire American public of the hazards of nuclear radiation. The arrival of atmospheric fallout from the first Soviet Union nuclear weapons tests and the increase in U.S. atmospheric testing demanded more advanced high sensitivity radioactivity measurement technologies. Such advances were necessary to maintain the health of our SRP and other Atomic Energy Commission (AEC) workers, U.S. citizens, and the world population and to monitor the nuclear threat posed by the former Soviet Union and other potential nuclear weapon proliferators.

Early Developments

Early high sensitivity measurements of ultra-low amounts of radioactivity used gamma spectrometry where gamma rays emitted by many radioactive isotopes are identified by the energy of the gamma ray or rays they emit. This is done by measuring the energy deposited in a scintillator such as sodium iodide (NaI) in crystal form and emitted as light energy converted to electrical pulses, which register the energy over a nominal scale of 10 to 3000 kilovolts. The spectrum made up of these peaks from a sample will identify all of the radioisotopes present that emit gamma radioactivity without having to perform wet chemistry quantitative elemental analysis. This method provided SRP environmental monitoring with a cost-effective and sensitive method to analyze thousands of samples relatively quickly.

By combining better methods to shield against background radiation from the natural environment, the first whole body counter was constructed as a room using 12-inch thick pre-World War II battleship armor plate. In 1959, a room was constructed as a box with a 5-ton steel door and lined with lead and copper plate to reduce low energy x-rays produced by cosmic interaction with the steel (Winn et al. 1986). The use of pre-World War II armor plate was to eliminate low background radioactivity due to fallout following the initial atmospheric testing of the atomic bomb. This state-of-the-art facility was used until the late eighties as the premier whole body ultra-low measurement technology for trace radioactivity that was naturally occurring or ingested by SRP employees.

This whole body measurement technology was also used in bioassay research studies to determine where radioactive isotopes would locate in the human body and how long they would remain to contribute to a person's lifetime dose. Such research contributed to the location and half-life of I-131 in the body, the half-life of Cs-137 in the body and its dependence on age (3-60 years of age) and gender (male or female), and the location and dose as determined using chest counting technology developments of plutonium inhaled and deposited in the lung.

With the development of liquid scintillation low energy beta measurement, tritium analysis for bioassay was simplified, from several days per sample to less than a day for 50-100 samples. SRP Health Protection scientists introduced the plastic bottle counting technology for bioassay tritium analysis, reducing the background and cost of quartz bottles while increasing sensitivity.

Field and laboratory concentration techniques were developed to achieve greater sensitivity and increase environmental monitoring sample throughput (see Figure 1). Large volumes of rain, river, and stream water were directly concentrated in the field and passed through specially designed ion exchange columns or liquid extractors, which increased the measurement of various radionuclides factors of 100-10,000 fold rather than using a 1 liter of water evaporation method. High volume air samplers were developed to concentrate particulate in air for volumes greater than 10 cubic meters per minute to track trace concentrations of atmospheric radionuclides. This technology was used in the late 1950s and 1960s to monitor atmospheric testing and its impact on the environ-

Figure 1. Early development of high sensitivity environmental collection and detection expertise

ment to empirically demonstrate the good stewardship of SRP in the protection of the environment and surrounding population. Following the atmospheric testing moratorium between the U.S. and Russia, this technology was used to detect (in the local CSRA area) atmospheric tests by the Chinese and the nuclear accident at Chernobyl. Ion exchange concentration of milk samples in the late 1950s and early 1960s was used to detect trace levels of I-131, Cs-137, and Sr-90 concentrations from fallout. The technique was so sensitive that the quality of milk could determine, by the naturally occurring K-40 content, who was diluting their milk with water. Another early technology was developed to increase detection sensitivity by improving quantity and geometry of the sample for direct counting by NaI gamma spectrometry, eliminating time consuming and expensive chemical separation techniques. In this technique, vegetation is compacted and dried up to 110°C to avoid loosing volatile iodine, which is measured.

Fallout was definitely a fortuitous benefactor to the development of the early sensitive measurement technologies for low-level amounts of radioactivity in the environment. Fallout provided the radionuclide tracers used in measurement research to achieve ultra low levels. Releases from SRP in the 1950s were extremely low and well below the AEC radioactivity release guidelines of the time.

Significant Achievements

Many significant achievements in the high sensitivity measurement of ultra-low amounts of radioactivity not only in the environment but in analytical measurement of radioactive and non-radioactive compounds, elements, and isotopes were developed at SRP. The further development of ultra-low background facilities to measure ultra-trace amounts of radioactivity resulted in an ultra-low level atmospheric gas measurement counting facility for tritium molecular forms released to the atmosphere (gas, oxide, organic) and various noble gases resulting from nuclear fission (weapons testing and commercial power reactors). The sensitivity levels now achieved are well below the 1 picocurie per cubic meter of air (less than 1 part in 1,000,000,000,000).

These achievements have also resulted in the construction of the only U.S. specifically designed underground ultra-low background underground radioactivity measurement facility (see Figure 2). The facility built in 1982 is a 9′ x 12′ x 7′ steel box constructed from 4-inch thick pre-World War II armorplate from the aircraft carrier Antiedim buried 50 feet underground surrounded by 4-6 feet of highly pure dense specular hematite ore. The facility contains no additional natural or man-made materials that could contribute radioactivity to the background. The facility is entered from a tunnel beginning 20′ underground from an above ground clean room facility where all of the information from the detectors within the underground shield is read out. This one-of-a-kind facility is used to support special site programs and work for other federal agencies. For example, NASA uses this facility to measure trace radionuclides in materials returned from space. The Department of Defense for Nuclear Safeguards also contracts to use this facility. The detection sensitivity of this facility is equivalent to less than one part in 1 trillion or the ability to find one penny in the U.S. national debt, about 5 trillion circa 2000.

The Tracking Atmospheric Radioactive Contaminants (TRAC) vehicle (see Figure 3) is another noteworthy achievement for measuring trace radioactive materials released to the atmosphere. The vehicle, built in 1982, is a single-body 22-ton vehicle with a chassis-mounted atmospheric sample collection and radiation real-time monitoring laboratory. Originally built for emergency response for unplanned SRS reactor releases of radioactivity, it now serves as a high sensitivity remote radiation measurements research and development laboratory and is available for emergency response to the Site and other federal agencies.

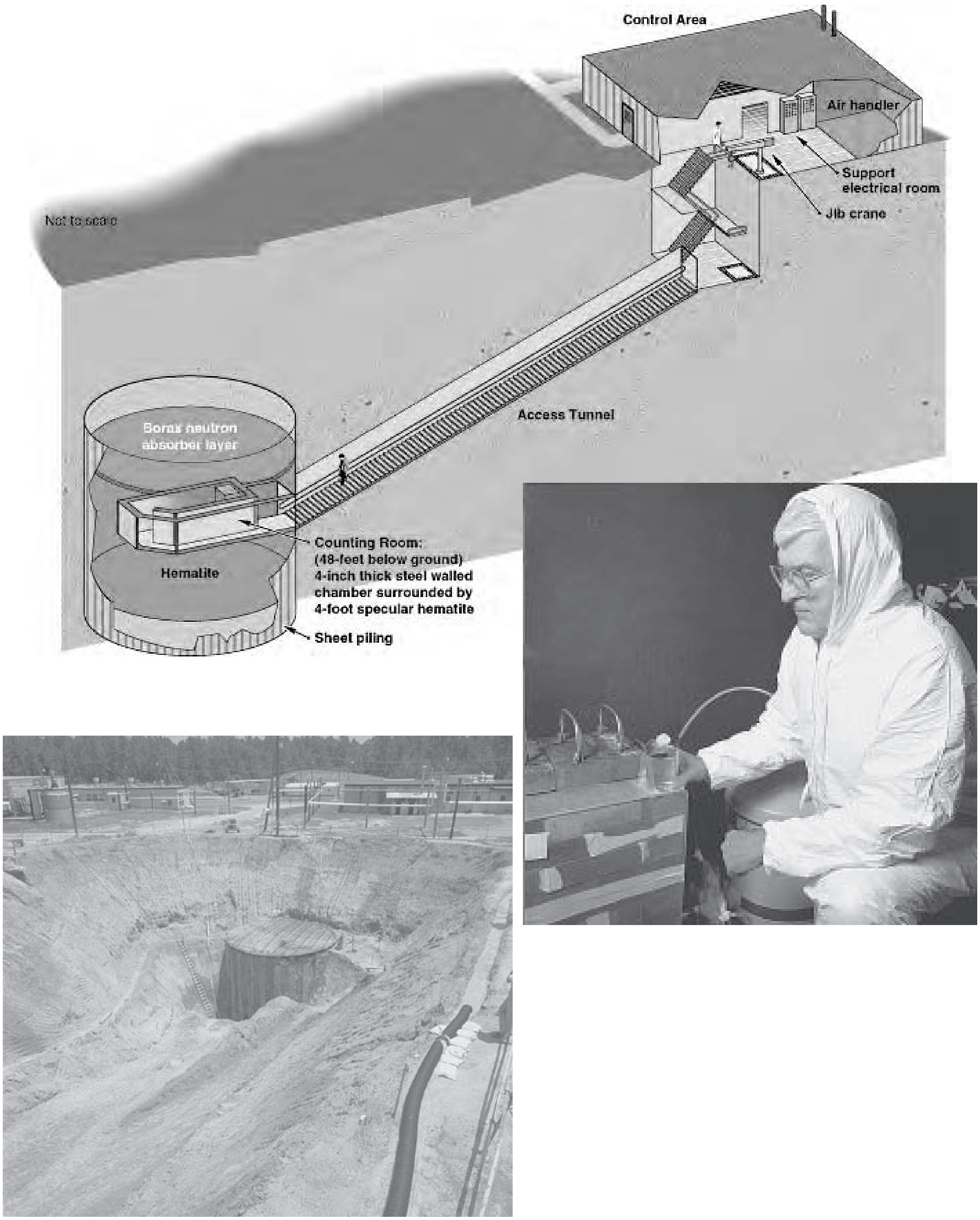
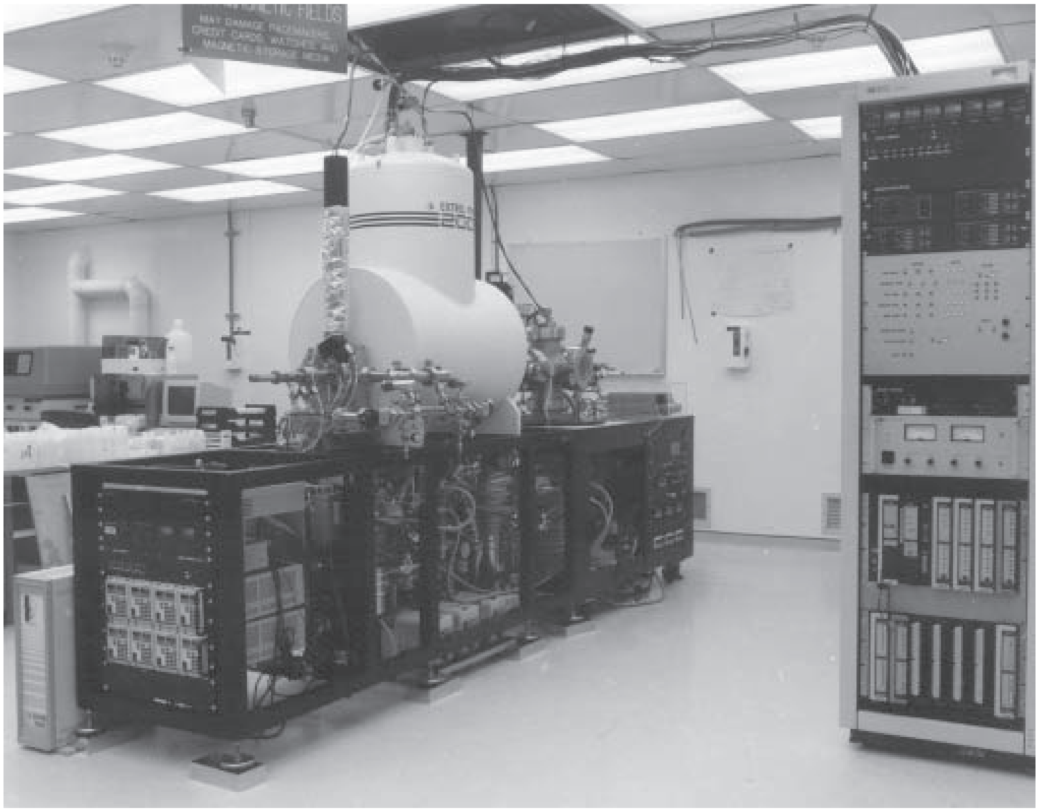


Figure 2. Highly sensitive counting and measurement—Underground Counting Facility

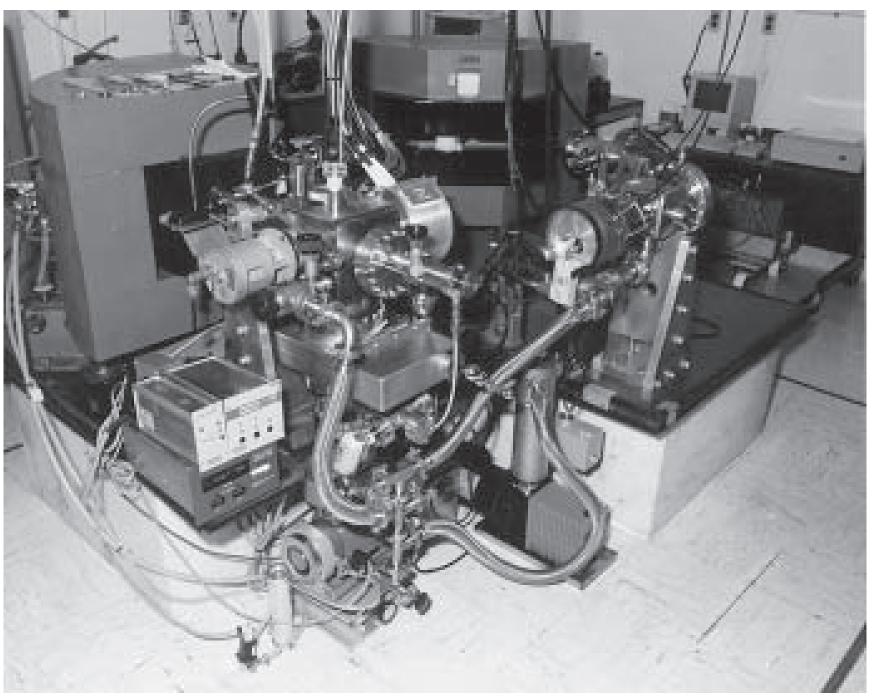
Figure 3. TRAC vehicle

The facility has a real-time gamma plume monitor for measuring a radioactive gaseous cloud, a high-volume 50-cubic-meter-per-minute atmospheric particulate filter system with real-time radioiodine and particulate radioactivity measurement detectors, and alpha surface barrier detectors to monitor actinides off-line. Detectors are also available to determine neutron activity levels at remote distances from a suspect source; when stationary, a portable high resolution Germanium detector for field sampling, and a low background liquid scintillation detector for off-line tritium oxide measurements in air or water. The vehicle is guided by geo-positioning satellite information system (GPS) and has local and distant radio communication. The vehicle can also be supplied with and periodically updated in real time with atmospheric plume transport maps through the SRTC Weather Center digital communication modem to aid in the location of radioactivity releases.

The measurement of ultra-low amounts of radioactivity by radiation measurement requires the emission of radiation by radioactive decay. Based upon their specific activity (curies per gram), many radioisotopes decay very slowly. Small amounts of these isotopes require weeks to measure by radiation measurement. SRS developed a highly sensitive mass spectrometric laboratory capable of measuring the actinides (uranium and plutonium) in a variety of environmental and bioassay samples that would require months by radiation measurement. In addition, this high sensitivity thermal ionization mass spectrometry technology measures the minor isotopes of uranium and plutonium that cannot be measured routinely by alpha spectrometry and identifies the source and age of the material, greatly assisting in contamination and unknown source investigations. The high sensitivity thermal ionization, time of flight secondary ion and x-ray fluorescence mass spectrometric laboratory (see Figure 4) has been greatly expanded to include developments in identifying the chemical signatures and morphology of individual micron size particles tracing them to their source and origin of formation. This forensic laboratory achievement has been recognized and routinely used for international nuclear safeguards, nuclear



Fourier Transform Mass Spectrometer (FTMS)



Thermal Ionization Mass Spectrometer (TIMS)

Figure 4. Forensic Science

smuggling, and law enforcement forensics investigations.

Numerous advances at SRS in the development of field and real-time radionuclide sampling, real-time radiation monitoring, and thermal remote sensing related to the measurement of small amounts of radioactivity in the environment have achieved national and international recognition (see Figure 5). Field sampling for atmospheric tritium chemical forms, streams and river water transport concentration of trace radionuclides, and the electrostatic high volume portable sampling of atmospheric radioactive aerosols developed at the SRS are widely used in international safeguards and national security programs.

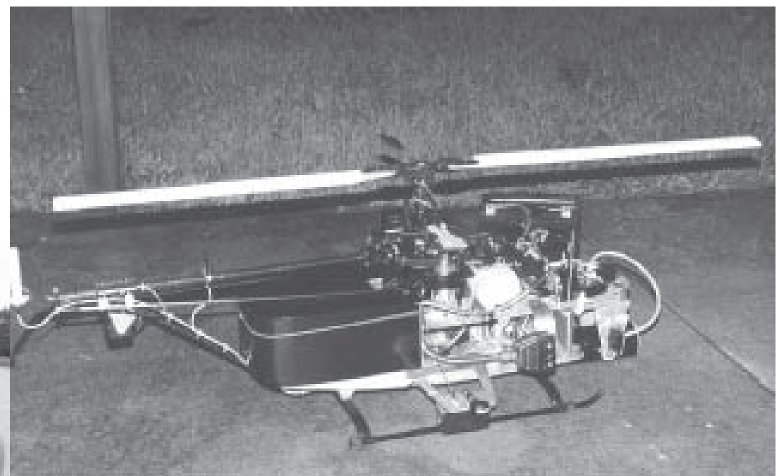
The high sensitivity measurement of small amounts of radioactivity in the environment is closely linked to the capability to locate in real-time sampling points and high probability of areas for collection and detection. This information is necessary to locate the source and measure the impact of the radioactive release on the environment and the surrounding population. SRS has developed one of the best meteorological and aqueous transport and dispersion real-time and forecasting centers in the nation (see Figure 6). Beginning in the 1950s, SRP used basic meteorological parameters, including wind direction, speed, barometric pressure, precipitation, temperature, and a standard relational graph to track unplanned releases. Today, an advanced three-dimensional model is generated from real-time meteorology data collected from 9 on-site towers and national and international meteorological data, updated every 3 hours. The SRS Weather Information and Display System (WIND) is recognized by the Department of Energy (DOE) and other federal agencies as a focal point for local and regional emergency response and in national and international nonproliferation safeguards and security circles. This achievement is responsible for the excellent emergency response capability and demonstrated good stewardship of the environment by SRS.

In addition to atmospheric transport, SRS has also achieved recognition for developing remote thermal sensing technology from U.S. multispectral satellites (see Figure 7). SRS is recognized by DOE as the ground truth (empirical ground measurement of temperature and meteorology for thermal sensing calibration) center for its newest multispectral thermal imaging (MTI) satellite to be launched in February 2000. SRS is recognized for its creative and innovative development of highly accurate measurement of the earth's surface temperatures. Such measurements are important to quickly determine with resolution on the order of square feet the source and magnitude of environmental pollution and natural disaster destruction over large areas of the earth's surface.

Several analytical high sensitivity measurement technology achievements include the use of C Reactor for real-time measurement of natural uranium and trace elements in environmental samples by real-time neutron activation coupled to prompt gamma, delayed neutron, and decay gamma emission. This was a nationally recognized analytical feat, which measured 100,000 samples over a 5-year period, for the National Uranium Resource Program to determine uranium deposits in the U.S. by taking environmental samples every 10 miles square. This amazing feat has never been duplicated. Because of expensive operating costs of the National Neutron Activation Facility in C Area, it was dismantled.

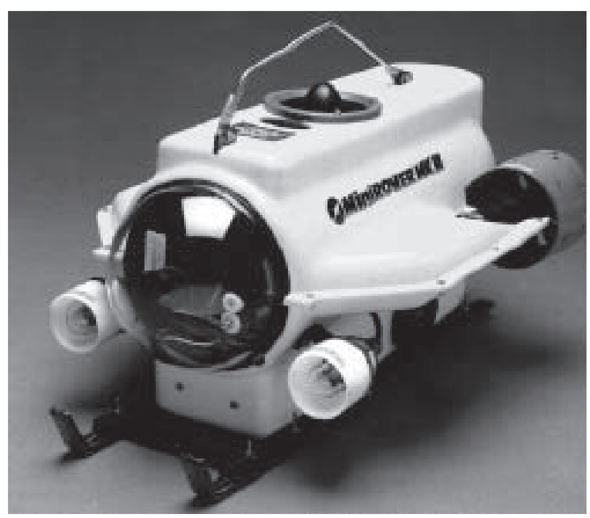
Another analytical high sensitivity measurement facility is the californium activation facility located in the Savannah River Technology Facility (SRTC). Although not the largest, it was one of the first such facilities and still remains in operation today. The facility houses a 1-Mg source of californium-252 in a large in-ground tank of light water and for shielding used an air pressure loading and removal "rabbit" (named for instant entry and exit of a small capsule upon initiation). The sample is housed in a small capsule called a rabbit, which after a selected time of irradiation by the

Borehole monitoring



Unmanned aerial vehicle

Underwater detector



Submersible

Figure 5. Unique field measurement systems

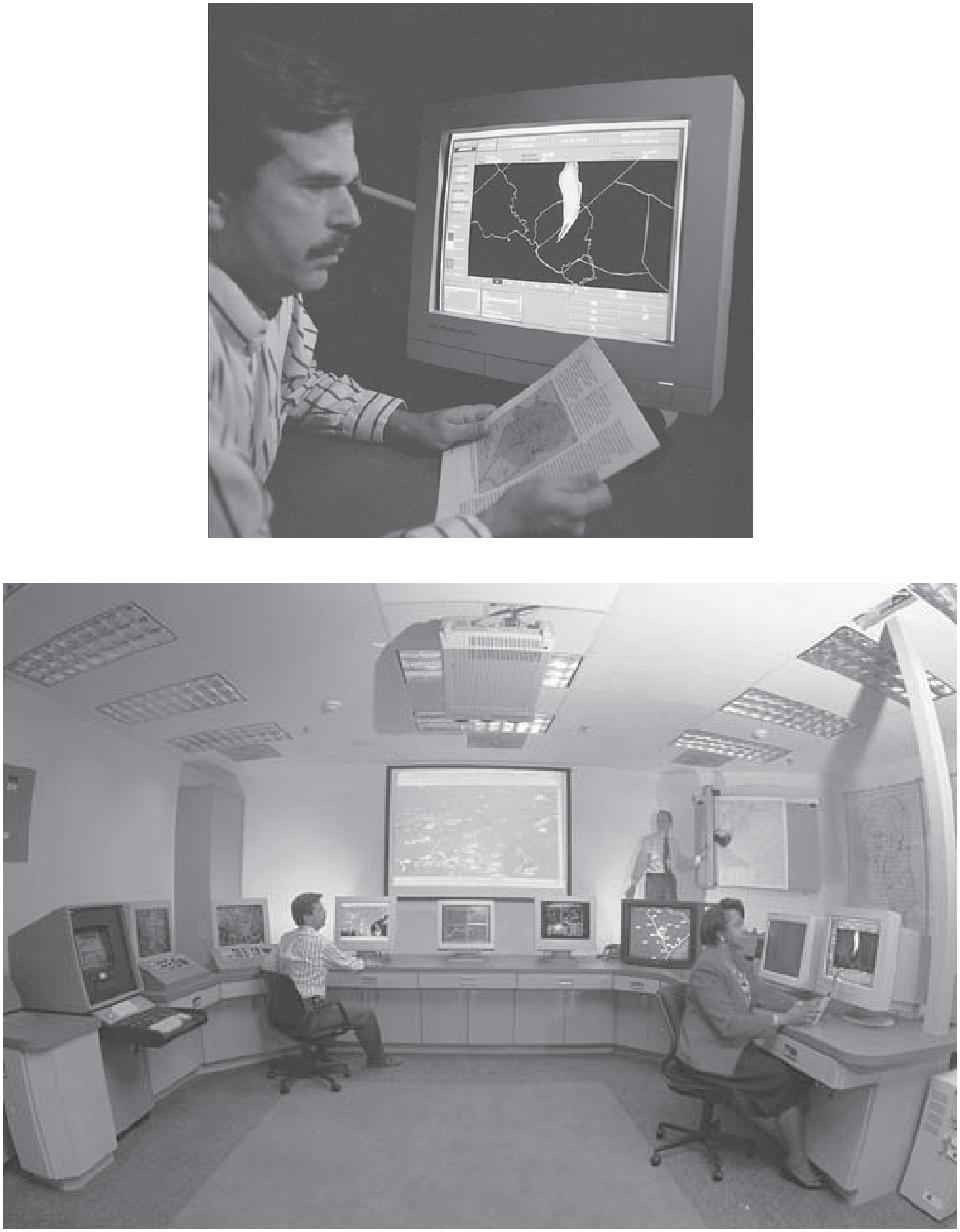


Figure 6. Meteorology

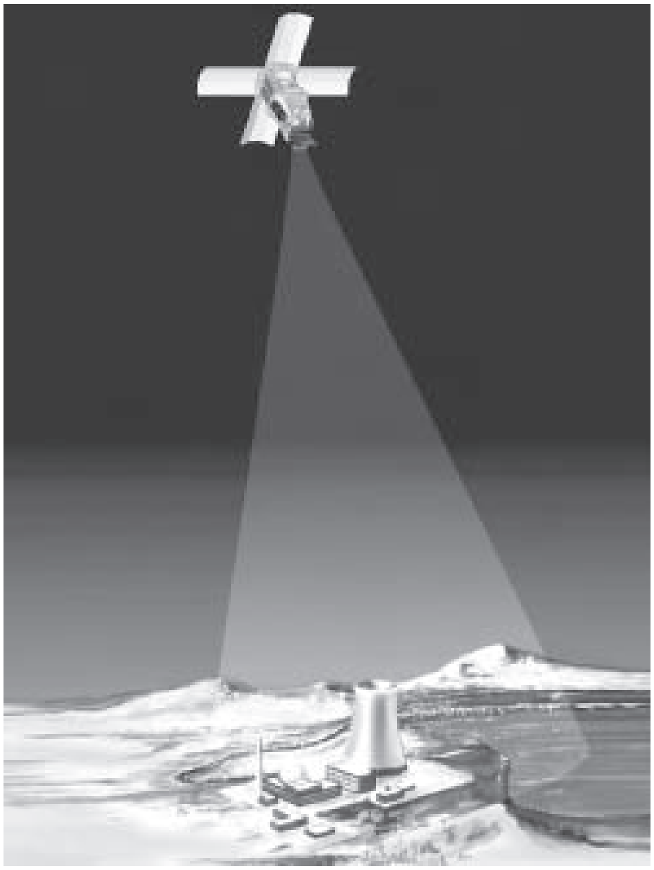


Figure 7. Remote sensing thermal imagery analysis

neutron source, is removed and counted immediately on a gamma spectrometric counting system. Californium-252 is a spontaneous fissioning isotope, providing an intense neutron source for activating stable isotopes and greatly increasing their analysis sensitivity through high sensitivity radiation measurement.

Missions in the 21st Century

Over the past 50 years since the first Savannah River water sample was collected, the innovative and creative development of high sensitivity measurement technology of small amounts of radioactivity has developed into a nationally and internationally recognized laboratory for ultra-trace radiation detection and measurement. Numerous health physicists, scientists, and engineers have contributed to this SRS technological achievement. Many are now retired, and a few remain; but, the creativity is carried on by new dedicated technologists. The technology of high sensitivity ultra-trace measurement of small amounts of radioactivity continues to grow and find new missions beyond the "Cold War" into new areas of nuclear nonproliferation, international safeguards, national security, and law enforcement.

International safeguards and nonproliferation efforts find SRS scientists at work in Iraq and other countries around the world conducting nuclear-related inspections (see Figure 8). These measurement technologies have also earned SRS/SRTC recognition as a nuclear forensic laboratory in support of the FBI in nuclear-related crimes such as international smuggling of nuclear materials and national nuclear terrorism. Although non-nuclear, the recognition of the Cold War high sensitivity technolo-

Figure 8. International nuclear safeguards: inspections and monitoring regimes to detect undeclared nuclear activities.

Figure 9. Regional law enforcement support (submersible vehicle)

gies has created a law enforcement mission to adapt these technologies to support South Carolina and Georgia state and local law enforcement agencies (see Figure 9).

Although the mission of high sensitivity measurement of ultra-low amounts of radioactivity in the environment may not be as large as nuclear materials production at SRS, it remains, even after 50 years and the end of the Cold War, important to the security of our site, nation, and the world. This technology continues to bring the most creative and innovative scientists and engineers to SRS, achieve recognition for SRS and SRTC, and continues to successfully provide the technology for a cleaner, safety, more secure nation and world.

Acknowledgments

It is impossible to acknowledge all the health physicists, scientists, and engineers who have contributed and those who are now contributing to this technological achievement. However, since the 1950s and the initial Methods Chemistry and Physics Methods Groups, there are early SRS health physics pioneers that deserve mentioning. They are C.M. Patterson, Bill Reinig, Ed Albenesius, Mac MacClaren, Paul K'burg, Jim Johnson, Al Boulogne, Henry Horton, Clarice Ashley, Eric Geiger, Harry Butler, Jack Hoy, Charlie Wright, Walt Marter, Ray Harvey, Gene Morris, Marshall Sanders, D.I. Ross, Ed Whitley, Sally Tucker, Bill Jacobsen, Bill Spell, Sally Leight, Worth Dalton, Dick Hawkins, Todd Crawford, Bob Taylor, Bill McMillan, Ed Miller, Ken MacMurdo, Larry Heinrich, and John Clark.

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