Environmental Sampling in Water for Verification Purposes

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Materials released to the environment from natural or manmade sources are transported by several processes including atmospheric conditions, hydrologic systems, ground water and terrestrial movement. Of these, river systems, off-shore ocean currents, and atmospheric plumes are the major contributors to long range transport of effluents from industrial sources including nuclear-related activities. River systems, as opposed to atmospheric transport, channel the effluent substances not only from direct input from source aqueous outfalls, but also from watershed drainage carrying atmospheric-released effluents deposited on the terrain surfaces through rain or dry deposition. This channeling effect strengthens the signal whereas atmospheric dispersion weakens the signal. Deposition, absorption in sediments, and biological concentration in river systems can enhance the effluent signal and also retain the signal for some time, thereby reducing the need to provide continuous sampling.

The collection of hydrological samples such as water, collodial material, suspended and bottom sediments and blots can provide an effective signature of certain nuclear activities such as reactor operation and nuclear fuel reprocessing. Optimized environmental sampling methodology combined with ultra-sensitive analytical methods are used to determine extremely low levels of radioisotope signatures. Using hydrologic sampling as a nuclear activity verification tool requires understanding the aquatic behavior of radionuclides, sample collection methodology, hydrologic sample operational planning, high sensitivity sample analysis and data interpretation.

Since some radionuclides remain in solution and others tend to concentrate on sediments and biota, it is important to select a variety of collection techniques in order to completely cover as well as enhance the concentration of key radionuclides. For example, tritium will remain soluble and rapidly dilute with increasing volumes of water from river tributaries. Iodine will concentrate in water plants and cesium in clay sediments. Plutonium will be disproportioned between the water and collodial and suspended sediments

Sample collection for plutonium, uranium and gamma-emitting radionuclides involves high volume ion exchange water filters, sediment cores and aquatic vegetation. Grab water samples are collected for tritium, trace elements and organics. Physical measurements are also made, such as water conductivity, temperature and sample location by Global Positioning System (GPS),

Hydrological operational planning is very important in order to determine the terrain, identify possible nuclear operational areas, identify drainage basins and estimate river characteristics. This is necessary to select appropriate sampling points to maximize collection of the radionuclide signal and optimize collection logistics such as access. Sample chain-of-custody and the recording of collection information is very important to reconstruct the sample collection operation and provide for the accurate interpretation of the analysis data.

The most significant and critical part of the hydrological sampling program is the high sensitivity analytical detection and measurement technology. These technologies are continuously undergoing developments to improve sensitivity, accuracy and precision, and cost effectiveness. All samples are initially nondestructively analyzed by high sensitivity gamma spectrometry in an underground ultra low background counting facility with >150% efficient (relative to sodium iodide) high purity germanium detectors. The ion exchange filter, sediment and biota samples are then prepared for chemical separation of uranium and plutonium and other actinides or beta fission products upon request. Following chemical separation, high sensitivity thermal ionization mass spectrometry for isotopic analysis is performed on the actinides. Grab water samples are converted to hydrogen gas and analyzed for tritium by ultra low background proportional counting. Also, total uranium analysis is done by laser x-ray fluorescence and stable elements of interest by inductively coupled plasma mass spectrometry. Sensitivities of femto and attocurie levels are achieved.

These hydrological sampling and analysis techniques for verification purposes have been demonstrated during several field trials and are one element of the on-going monitoring program in Iraq. As part of the IAEA 93+2 trials and with the approval and assistance of the Swedish Nuclear Power Inspectorate, hydrological sampling and analysis technology was used to demonstrate its effectiveness in establishing a radionuclide and stable element baseline near major nuclear facilities in Sweden. The first field trials were conducted in September 1993 at locations near five of their nuclear fuel fabrication, nuclear research and nuclear power reactor facilities. Water, sediment and biota samples were collected from locations at distances up to 25 km from the facilities for radionuclide and trace metal measurements.

Results from the Field Test program show that nuclear reactors discharging effluent to coastal waters can be detected by the presence of activation products in waters, sediments, and biota near and up to 25km from the facility outfall depending on local conditions. Isotope ratios derived from fission products such as $^{134}Cs/^{137}Cs$ indicate waste handling resulting from fuel handling operations at several nuclear power and research sites. Plutonium mass spectrometric measurements show the presence of high burnup plutonium (power reactor ratios) near a major research facility. Slightly perturbed uranium isotopic abundances indicate the type of fuel and irradiation near the same research facility. This data could indicate the presence of some type of spent fuel characterization studies. Tritium measurements when used in the context of regional concentrations showed the

increased presence of the ternary fission product around a major research facility. Trace element measurements indicate that zirconium may have potential for detecting reactor operations (corrosion products).

The hydrological element of the ongoing monitoring program in Iraq that was begun in September 1992 has continued on a bi-annual basis since that time. The program was designed to establish a radionuclide composition baseline in the major watersheds of Iraq. This continuing monitoring program provides the capability to detect changes resulting from aqueous effluent of nuclear-related facilities that would indicate a renewed effort by Iraq to develop its nuclear weapons program. The focus of the monitoring program is to determine the presence of radionuclides that are indicative of nuclear operations and to qualitatively quantify their amounts.

A total of about 55 sites have been sampled within eight watershed regions in Iraq. A water ion exchange filter sampler is used to concentrate the radionuclides dissolved and present as collodial material in the water. A 5-10 cm sediment core sample is taken that consists of silt/clay which is preferred to sand. The preferred biota types are green algae and submerged aquatic or surface water plants such as water hyacinths, if available. A 500 ml water sample is also collected along with the measured water quality and temperature at each location. It takes approximately 1 hour to conduct a collection at any given location. Transport between most locations is effectively carried out by helicopter to minimize time.

High sensitivity gamma-ray spectrometer analyses of the hydrological samples have not detected to date the presence of anthropogenic fission or activation products with the exception of I-131, Cs-134 and Cs-137. The iodine-131 was detected in biological samples that concentrated the I-131 from waste water discharged to the river systems. The presence of only short-lived I-131 has been confirmed as being used for the treatment of thyroid cancer. Cesium-137 and some Cs-134 has been detected in water and sediment samples. The ratio of $^{134}Cs/^{137}Cs$ is indicative of fallout from the Chernobyl accident that occurred in April 1986. Uranium isotopic measurements are typical of naturally-occurring abundances with some radiogenic fractionation observed in surface and ground waters. The plutonium isotopic distributions in sediment and water filters are at or near expected distributions that occur in worldwide nuclear weapons fallout background.

These results demonstrate the effective capability of these environmental collection and analysis methods for the detection and interpretation of nuclear-related operations.