KEY TOOLS FOR NUCLEAR INSPECTIONS

ADVANCES IN ENVIRONMENTAL SAMPLING STRENGTHEN SAFEGUARDS

BY DAVID L. DONOHUE

For the international community, the summer of 1991 was a turning point of scientific discovery, one that set the stage for stronger nuclear safeguards. IAEA and United Nations and inspectors were combing the rubble of Iraq's nuclear installations looking for evidence of a secret programme to produce atomic bombs, something expressly forbidden by Iraq's ratification of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT). What the inspectors found that summer were minute traces of radioactive elements such as uranium and plutonium which helped them to piece together details of the programme, its scale, timetable and likely purpose.

Thus was opened a new chapter in the development of international nuclear safeguards. States in 1995 adopted measures for a strengthened safeguards system that authorize and equip inspectors to assure that any undeclared nuclear activities would not be overlooked.

The science behind this enhanced capability – since given the name "environmental sampling for safeguards," or ESS — relies on highly sensitive and selective analytical measurements to detect traces of nuclear materials collected in the "environment" of a known or suspected nuclear facility. Its application demonstrates the importance of having at hand an advanced suite of analytical and measurement techniques to address a significant world problem.

Some methods were used by IAEA inspectors during Iraq inspections in the early 1990s. Since then, a large amount of work has been carried out by the IAEA in collaboration with national and international experts to establish and implement ESS in all countries covered by comprehensive (NPT-type) safeguards agreements. The tools of ESS also will be applied by inspectors of the IAEA Iraq Action Team who resumed on site inspections in late November 2002 under a reinforced mandate of the Security Council. *(See box, page 18.)*

To acquire the capability, a number of tasks had to be done in parallel: development and validation of sampling methods, provision of certified clean sampling materials, establishment of a Class-100 clean facility for handling the samples without risk of cross contamination, coordination of a network of analytical laboratories (NWAL) with highly-specialized measurement capabilities, and the application of an air-tight quality assurance system to eliminate any doubts about the credibility of the results.

Today, IAEA inspectors take several hundred environmental samples per year in facilities all over the world. The laboratories of the IAEA and its NWAL perform thousands of measurements on such samples. The results of these measurements are compared to the declarations of the inspected facilities using sophisticated computer codes and statistical methods which search for significant discrepancies.

The analytical methods used for these samples are state-of-the-art in terms of sensitivity and selectivity for the elements or isotopes of interest, but newer and more powerful methods are constantly under development. A truly amazing amount of information can be obtained from the small amount of material collected, for example, in a sample that inspectors swipe on a 10 x 10 cm piece of cotton cloth. The results that can be obtained demonstrate the power of these methods.

Key Tools of ESS. IAEA capabilities for ESS reside in the Clean Laboratory for Safeguards, part of the Safeguards Analytical Laboratory near Vienna, Austria. The labs employ the following analytical techniques to screen

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1991 SUMMER OF DISCOVERY

More than 10 years ago, IAEA inspectors in Iraq uncovered a clandestine nuclear weapons programme. The discovery relied upon technologies and IAEA capabilities for analyzing samples of nuclear and radioactive materials, including environmental monitoring techniques. Inspectors took samples of materials within and near facilities, and swipes of dust that had collected on the surfaces of equipment.

■ Analysts surprisingly discovered evidence of uranium enrichment, specifically isotopically altered uranium which did not match any known declared materials. They also discovered extremely depleted uranium that could only have been produced with the electromagnetic separation technique, a method no longer known to be in use anywhere in the world. Further inspections and analyses led to Iraq's disclosure of the enrichment programme.

■ Analysis of other samples also disclosed undeclared irradiation of uranium to produce very small quantities of plutonium. Additionally uranium from three different ore bodies was discovered, including indigenous production as a byproduct from an Iraqi phosphate plant.

■ Environmental monitoring techniques were used from 1991 to 1998 to verify the accuracy of the final Iraqi declarations of its nuclear programme. By 1998, when inspections were ceased, the IAEA was able to map and neutralize the Iraqi programme.

The Iraq experience led to a strengthening of the IAEA safeguards system and the wider use of environmental sampling and analsysis. The techniques were introduced in 1996 as an inspection measure that can be applied under comprehensive safeguards agreements and more broadly under Additional Protocols to such agreements that enable Agency inspectors to verify both declared and undeclared nuclear materials and activities.

For a fuller account, see the IAEA Bulletin Vol. 34, No. 1 (1992), "Behind the Scenes: Scientific Analysis of Samples from Nuclear Inspections in Iraq", by David Donohue and R. Zeisler. Also see "Technologies for Detection of Emissions", chapter 3 of a 1995 report on nuclear safeguards by the former US Office of Technology Assessment.

and measure environmental samples:

■ *High-resolution gamma spectrometry (HRGS)* is used for the initial radiometric screening of samples when they are first received in Seibersdorf. This screening can be performed without removing the samples from their bags or bottles, thus further reducing the chances of cross contamination.

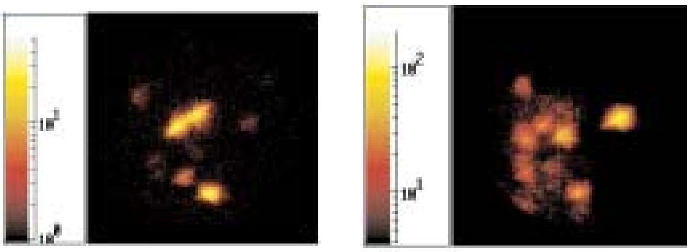
■ *Radioisotope or X-ray tube-excited X-Ray fluorescence (XRF) spectrometry* can detect sub-microgram amounts of uranium in environmental samples. This information is used to decide about the safe handling of the samples in a clean laboratory as well as in choosing the detailed analysis methods to be applied later.

■ *Scanning electron microscopy with electron-excited X-ray fluorescence spectrometry (SEM/XRF)* measures the elemental composition of micrometer-sized particles removed from environmental samples. In particular, the uranium/plutonium and the americium/plutonium ratios are of interest in samples taken inside glove boxes or hot cells.

■ *Thermal ionization mass spectrometry (TIMS) using ion-counting detection* is used to measure sub-nanogram amounts of uranium or plutonium following dissolution of the samples (so-called "bulk analysis"). Bulk analysis gives an average composition of the sample, regardless of the physical form of the elements present, and complements the information obtained from "particle analysis" methods such as SEM/XRF.

■ *Secondary ion mass spectrometry (SIMS)* is used to measure the isotopic composition of micrometer-sized particles. The isotopes uranium-235 and uranium-238 are of greatest interest because they reveal the enrichment of uranium, showing whether it is intended for use in reactor fuel or nuclear explosives. *(See figure on next page.)*

Particle Analysis of Uranium Isotopes



Images of uranium-235 and uranium-238 ions obtained by IAEA analysts using Secondary Ion Mass Spectrometry. The images show ions coming from an area 150 micrometers in diameter. Each bright spot is a single particle and the enrichment of all particles detected can be readily calculated by mathematically comparing the two images. *(Credit: IAEA)*

Extensive Experience. The IAEA has gained experience in the collection of most types of environmental samples: soil, sediment, water, vegetation and biota. However, these sample types are usually taken some distance from a known or suspected facility and the dilution effects can be large.

Since IAEA inspectors have access to the actual buildings on a nuclear site, it was decided to concentrate efforts on taking and analyzing surface swipe samples. Swipes have several advantages: they are small and compact (compared to a 1 kg soil or a 1 liter water sample); they facilitate the taking of multiple replicate subsamples at each location; the swipe medium can be chosen to have a low background of the elements of interest; and they are well adapted to sampling dust particles from horizontal surfaces and equipment inside buildings.

Two types of swipe sampling kits have been developed. Each contains the supplies which a trained inspector needs to take replicate swipes in one location -- gloves, bags, bottles, data sheets (with sampling instructions on the back), pen, etc. The Clean Laboratory produces these kits in a Class-100 clean area with sufficient quality assurance measures to prove that the swipes were not contaminated with actinide elements or radionuclides before use. Both the unused kits and the final samples are kept under the inspector's control at all times to prevent inadvertent cross contamination or tampering.

Although SAL and the Clean Laboratory are well equipped for making detailed analysis of environmental samples, it was necessary to establish a network of laboratories in IAEA Member States which can meet three basic needs: to apply analytical methods which the IAEA cannot afford to establish (such as fission-track analysis or accelerator mass spectrometry), to provide backup capacity for the IAEA laboratories in coping with peak sample loads and to provide parallel measurements on replicate subsamples to increase confidence in the accuracy of the results.

The Clean Laboratory also produces blank and "spiked" quality control samples which are sent "blind" along with the inspection samples to monitor the occurrence of false positive or negative results. Finally, SAL and the Clean Laboratory have demonstrated the serious commitment to quality by achieving certification under the ISO-9002/1994 quality assurance standard.

Examples of Analyses. Environmental monitoring deals with elements and isotopes which carry a unique "signature" of anthropogenic (human-made) processes such as isotope enrichment or neutron irradiation. In a fashion similar to forensic laboratories studying the evidence of a crime, analysts look for patterns which unambiguously point to a particular scenario -- what were the starting materials, what was the process which transformed them and when did this happen?

When the results agree with the facility's declarations, they provide additional assurance that no misuse has occurred. When a possible discrepancy is found, actions are taken to confirm it by, for instance, re-checking results by the reporting laboratory, analysis of archive samples or the taking of new samples. The data evaluator must be well informed about the limitations of the analysis methods and must be constantly on the lookout for misleading data caused by contamination, interferences or mistakes.

ADVANCED TOOLS SUPPORT INSPECTORS IN IRAQ

IAEA nuclear inspectors have a range of high-tech tools at their disposal for arms inspections in Iraq, which resumed in November 2002 after a four year absence. Since 1998, there have been significant improvements in technology.

Among the main tasks for IAEA inspectors will be to throw a detection system, that's like a net, over Iraq — a country about the size of France. Its mesh has to contain important evidence from slipping through it. The approach is known as a broad area search. Within a given net, target areas will be selected for closer inspection and fact-finding.

As they gather evidence, inspectors will use a broad array of technologies, such as hand-held radiation detectors and measurement instruments. Some small instruments are used to search for nuclear and radioactive materials known to be associated with weapons-making. Others, known as multi-channel analyzers, can identify specific radioactive elements in samples that inspectors collect for fuller analysis in laboratories.

Analysis of samples can determine "nuclear fingerprints", and reveal indicators of past and current activities in locations handling nuclear materials, particularly those associated with uranium conversion, fabrication, and enrichment. Determining such cases, however, requires expertise and the right equipment — the fingerprints of different isotopes, for example, can overlap, and an abundant constituent of one element can mask a rare one.

Reaching conclusions can be tricky, often requiring multiple dimensional analytical approaches. The IAEA has its own experts and facilities, through its Safeguards Analytical Laboratory in Austria, experienced in sample measurement and analysis, including hundreds of samples from the 1990s Iraq inspections. Fully operational since then is a "Clean Laboratory" equipped with highly sensitive instruments, including electron microscopes and mass spectrometers. Experts can precisely measure even tiny nanogram (one-billionth of a gram) particles and detect traces of nuclear materials collected in the environment of known or suspected nuclear facilities.

Multi-channel analyzers (MCAs). These standard and portable tools for IAEA inspectors register the energy emitted by a radioactive source. MCAs use software that reads the pattern of the energy output, matches it to a signature, and displays the result. One portable analyzer is designed for detection of gamma radiation from radioisotopes and the presence of neutrons for enhanced detection of plutonium, which is produced in a reactor by irradiating uranium-238. Unlike typical radiation detectors, the device can be used to search for and locate an unknown source of radiation, determine the relative dose rate, and isotopically identify the source. Results are displayed on a digital screen. Uranium and plutonium isotopes, for example, are a good indication of whether nuclear fuel has been reprocessed. Another type, a portable gamma spectrometer, is specifically designed to measure uranium and whether it has been enriched. It can perform accurate and rapid uranium verification measurements in laboratories, at facilities, or in other industrial environments. The ratio of certain isotopes can yield valuable information — for example, the type of enrichment that was used.

Alloy detectors. Faced with a yard filled with metal objects, the inspectors have use of another portable device, known as ALEX, short for the brand name "alloy expert". Nuclear activities use exotic steels and unusual elements such as zirconium. ALEX allows rapid identification in the field and gives the opportunity to intensify inspections when something important is discovered. For example, UF6 (uranium hexafluoride) is a highly

Photo: A variety of tools are used by nuclear inspectors in Iraq. (Credit: Calma/IAEA) corrosive material used in uranium enrichment. The special alloys required in its production apparatus would be quickly identified by ALEX. Technically, the device is an x-ray fluorescence spectrometer. It generates x-rays to penetrate the material being inspected. ALEX matches the response pattern of elements in the alloy to the x-rays against a library of information in its software and displays the results.

Environmental monitoring instruments. Monitoring water, air and vegetation will form other strands of the search net over Iraq. A nuclear weapons development programme, despite best efforts to conceal it, is likely to leave its fingerprints on nature. Water monitoring will be conducted across Iraq, using a system that draws raw water through a filter for one hour, the equivalent of testing a large volume of water. Laboratory analysis of the filter is able to find the most minute traces of materials in the water with pinpoint accuracy. Air sampling stations can be set up at various points across Iraq and samples of vegetation tested for tritium, an isotope of hydrogen. Finding tritium in waterways or the air strongly suggests reactor operations, for example.

Digital video surveillance sytems. Tamper proof and digital video systems are used for surveillance and monitoring at facilities. They could include factories where dual purpose activities could be conducted - for example, the potential use of machine tools to manufacture components for a nuclear programme. Data is fed into powerful computer systems that inspectors use to review and analyze images and related data.

Satellite imagery. For monitoring purposes, images obtained by commercial satellite can help inspectors track activities. A new generation of global satellite positioning (GPS) devices will make it easier for the inspectors to monitor the large country.

Inspection Database. Alongside the full suite of radiation-detection gear and other monitoring equipment, a key tool inspectors rely upon is the Action Team's confidential database. It contains comprehensive and exceptionally detailed information obtained from past inspections, Iraq declarations, defector disclosures, intelligence information, and other multiple sources about Iraq's nuclear-related activities. Inspectors can find out, for example, that a device has been moved from one side of a room to another, and learn why.

Many Keys to Success. The array of tools provide a powerful and complementary package of resources for inspectors that augment their own extensive knowledge and experience.

Ultimately, however, inspections in Iraq ride on factors beyond the inspectors' experience and toolbox. As IAEA Director General Mohamed ElBaradei has emphasized, success will depend on five interrelated prerequisites: (1) immediate and unfettered access to all locations and sites in Iraq and the full use of the authority provided to the inspecting organizations by the Security Council; (2) timely access to all sources of information, including all information available to States; (3) unified and full support by the Security Council throughout the inspection process; (4) the preservation of the integrity and impartiality of the inspection process, free from outside interference; and (5) active cooperation from Iraq, with a sustained demonstration of its stated willingness to be transparent, and to assist the inspecting organizations in fully carrying out their missions.

This report first appeared on the IAEA's WorldAtom pages at www.iaea.org. Visit the site for updates of IAEA inspections in Iraq and other topics.

Photo: Images from commercial satellite and digital monitoring systems are analyzed as part of IAEA inspector operations in Iraq. *(Credit: Calma/IAEA)*

■ *Fission and Activation Products.* Swipe samples can be taken inside hot cells where spent reactor fuel has been handled years before. It would be expected that the ratios of fission and activation products as measured by HRGS would show the effects of radioactive decay during this time. Of interest are radionuclides, such as cerium-144 and europium-154, which have half-lives in the range from less than one year to nearly nine years. The swipes are measured as received for up to 24 hours and the results for each isotope expressed as a ratio of activity compared to caesium-137.

■ Particles Containing Plutonium. Sample preparation for SEM examination of particles involves touching the surface of the swipe with an adhesive carbon disk. Particles are removed from the surface of the swipe and held in place by the electrically conducting adhesive. After placing the stub in the electron microscope, an automated search routine is started which looks for particles which display a high backscattered electron signal, associated with the presence of heavy elements.

Each particle identified in this way is measured with energy-dispersive x-ray fluorescence spectrometry (EDX) and the results stored for later inspection by the analyst. An automatic measurement session typically takes four to six hours and covers several square millimeters of the sample surface. After sorting and examining the EDX data, the analyst then selects a number of interesting particles for more detailed measurements using a wavelength-dispersive X-ray spectrometer (WDX). This is done to make precise measurements of actinide element ratios, such as uranium/plutonium or americium/plutonium, in each particle. The relative concentration of these elements in such a particle gives information about the irradiation history of the uranium fuel and reveals whether chemical separation operations have been carried out.

For such plutonium particles, the ingrowth of americium-241 is a measure of the time since the plutonium was last chemically purified. This "age dating" ability is useful in safeguarding shutdown or decommissioned facilities to detect if chemical separation operations have occurred recently. Evaluating such results takes into account the fact that the amount of plutonium in a one micrometer-diameter particle is only a few picograms (roughly 10^{10} atoms) and the X-ray fluorescence spectrometer can detect minor components such as americium which are in the low-femtogram range.

■ Uranium Enrichment Measurements in Particles. The advantages of SEM/XRF are the high elemental sensitivity and ability to screen and measure large numbers of particles. The main disadvantage, however, is that SEM/XRF does not give uranium or plutonium isotopic information, which can be of critical importance. For this purpose, analysts apply the technique of secondary ion mass spectrometry (SIMS), using an imaging detection system to produce spatially-resolved images for each isotope of interest. The "enrichment" of each particle is calculated by comparing the total accumulated counts for uranium-235 to those from uranium-238; the other isotopes are monitored to detect possible molecular interferences.

An automated measurement covering several square millimeters of area may involve over 200 fields, containing up to several thousand uranium particles. Once displayed on a scatter plot for analysis, the particles of highly enriched uranium can be easily identified. However, analysts must take into consideration the uncertainty of the measurement, which is dominated by the counting statistics error of the minor isotope (uranium-235 in most cases).

■ Measurement of Uranium and Plutonium. One of the most challenging measurements applied to environmental swipe samples is the so-called "bulk analysis". It involves dissolution of the entire sample, spiking with separated isotope tracers, chemical separation and purification of the uranium and plutonium fractions for measurement by thermal ionization mass spectrometry (TIMS). The isotopic spikes are quite pure and available with certified concentration values.

Swipe samples can be taken, for example, at a facility which fabricates high-enriched uranium (HEU) fuel elements for reactors. Samples clearly show the presence of plutonium. Samples also can show the highest concentration of HEU, thus suggesting a correlation between HEU and plutonium. This illustrates the sensitivity of the methods used.

Serving Global Aims. More than five years of routine implementation of ESS has allowed the IAEA to draw certain conclusions about the value of the sampling and analytical methods used. While ESS, like other analytical tools, is no panacea in its own right, the techniques yield critical information. Isotopic ratios are of primary importance because they reveal the history of nuclear material and the effects of enrichment or irradiation with neutrons. Ratios of fission or activation products to each other can be a useful way of determining the burn-up of reactor fuel and the time since discharge from the reactor. Age-dating techniques can also be used to check on the status of shut-down facilities.

It should be pointed out that the analytical methods used for ESS are directly applicable in other, somewhat related areas. One of these is the analysis of nuclear materials seized by customs officials or police in connection with illicit trafficking incidents, thus ushering in a new field - "Nuclear Forensic Analysis". *(See box, this page.)* As in the case of international safeguards and nuclear inspections in Iraq, the international community can benefit significantly through the application of scientific techniques to address serious global concerns. ❐

NUCLEAR FORENSICS & ILLICIT TRAFFICKING

Fingerprints and forensic analysis have played important roles in criminal law for well over a century. As science and technology have advanced, investigative methods have become more and more sophisticated, so that now specialists can extract genetic material from a single hair.

In the relatively new field of "nuclear forensics" -- which focuses on analyzing the nature, use and origin of nuclear materials — similar methods are now being applied to determine material characteristics with high degrees of accuracy. Just as with human fingerprinting, nuclear material can be identified, examined, and profiled. The determination of radioisotopes, isotopic and mass ratios, material age, impurity content, chemical form and physical parameters may reveal a "nuclear fingerprint" of the material. Test results, together with other evidence gathered, make it possible to trace the most minute quantities accurately. Analytical methods developed for these purposes are used in international safeguards as well as for nuclear forensics.

In today's changing world, the IAEA, together with the Institute for Transuranium Elements of the European Union's Joint Research Commission, is taking the lead in helping countries establish a system for improved response to cases of nuclear smuggling. A primary aim is to upgrade capabilities to accurately identify and characterize seized material. Studies are best carried out in a laboratory already operating highly sophisticated applications, in which both nuclear and non-nuclear materials -- from sealing wax, glass and paper to residual radionuclides -- can be analysed.

In October 2002, international experts in the field examined the latest developments at the IAEA International Conference on Advances in

Destructive and Non-Destructive Analysis for Environmental Monitoring and Nuclear Forensics in Karlsruhe, Germany. Participants included laboratory scientists and law enforcement officers who examined a range of issues, from gathering, protecting, and analyzing material to legal systems and requirements in different countries. Also on the agenda was the IAEA's role in collective efforts to combat nuclear trafficking, including assisting analytical laboratories and advising on the safe handling of samples seized in trafficking cases.

Discussions additionally covered technologies to identify the origin of materials seized in cases of nuclear trafficking. One main focus was on ways to further strengthen nuclear security strategies and cooperation between analytical laboratories. Plans include providing specialized international support for required analytical work.--*This report first appeared on the IAEA's WorldAtom site at www.iaea.org. Visit the site for more information.*

Photo: Samples can be analyzed for their "nuclear fingerprints". (Credit: IAEA)