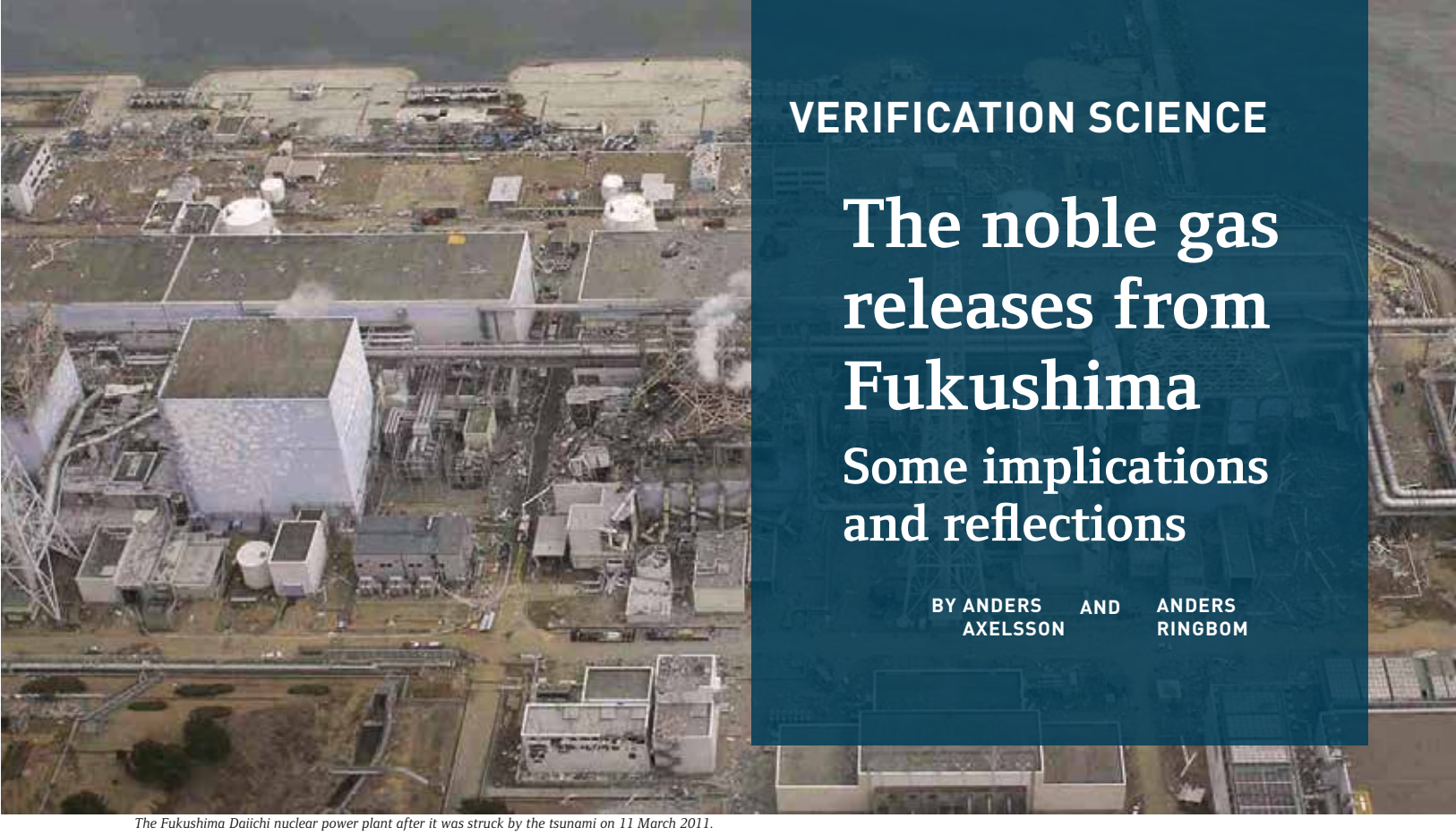


VERIFICATION SCIENCE

The noble gas releases from Fukushima

Some implications and reflections

BY ANDERS AXELSSON AND ANDERS RINGBOM

The Fukushima Daiichi nuclear power plant after it was struck by the tsunami on 11 March 2011. Photo courtesy of Air Photo Service Co. Ltd., Japan.

The tsunami caused by the large underwater earthquake close to the north-east coast of Japan on 11 March 2011 resulted in large releases of radionuclides from the Fukushima Daiichi nuclear power plant in the days that followed. Particle-borne radionuclides like Cs-137 and I-131, as well as noble gas isotopes like Xe-133 (see fact box on page 30) were released into the Earth's atmosphere. Based on measurements from the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization's International Monitoring System (IMS) in combination with atmospheric transport modelling (ATM)[1], several research groups have estimated that all of the xenon contained in the nuclear reactor was released. After a few weeks the entire northern hemisphere contained radioxenon concentrations about 1,000 times above the normal background level[2]. It should be pointed out that atmospheric radioxenon concentrations at that level are still insignificant from a human health perspective.

The estimated total release of Xe-133 exceeded 1019 Bq[3]. This is one of the largest radioxenon releases in history, even exceeding the release from Chernobyl in 1986. As a source of noble gases, the Fukushima accident corresponds to an atmospheric nuclear explosion of 1 megaton. The design criterion for the IMS is 1 kiloton , or 1,000 times less. Nevertheless, consideration of the Fukushima releases and their aftermath can prompt several interesting points of discussion regarding the design of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) monitoring network, the incorporated technologies, the use of the data and future lines of development for technology and data use. In this article we will address some of these issues, focusing in particular on the noble gas component of the IMS.

HOW NOBLE GASES DIFFER FROM OTHER RADIONUCLIDES

The key property that makes noble gases different from other radionuclide releases is the fact that they do not react chemically with surrounding material. This means, for instance, that they are difficult to contain in an underground nuclear test and will not be washed down by precipitation. The latter property was also well illustrated by the Fukushima accident, resulting in a very even distribution of Xe-133 observed by the IMS network a few weeks after the accident. All stations in the northern hemisphere measured very similar concentrations. This can

[1] Atmospheric Transport Modelling (ATM) is the calculation of the travel and dispersion of radionuclides released into the atmosphere, using meteorological data. This calculation can be performed in two ways:
- As backtracking ATM, which identifies the area from which radionuclides may have been released, calculated from the location where they were observed.
- As forward ATM, which identifies where radionuclides may travel from their known point of release.

[2] On a global scale, the background is dominated by releases from medical isotope production facilities, locally, the background can be strongly influenced by releases from nuclear reactors and from hospitals.

[3] A Becquerel (Bq) is the amount of radioactive material in which 1 atom decays every second.
1 milli-Becquerel (mBq) = 10**-3 Bq
1 micro-Becquerel (uBq) = 10**-6 Bq

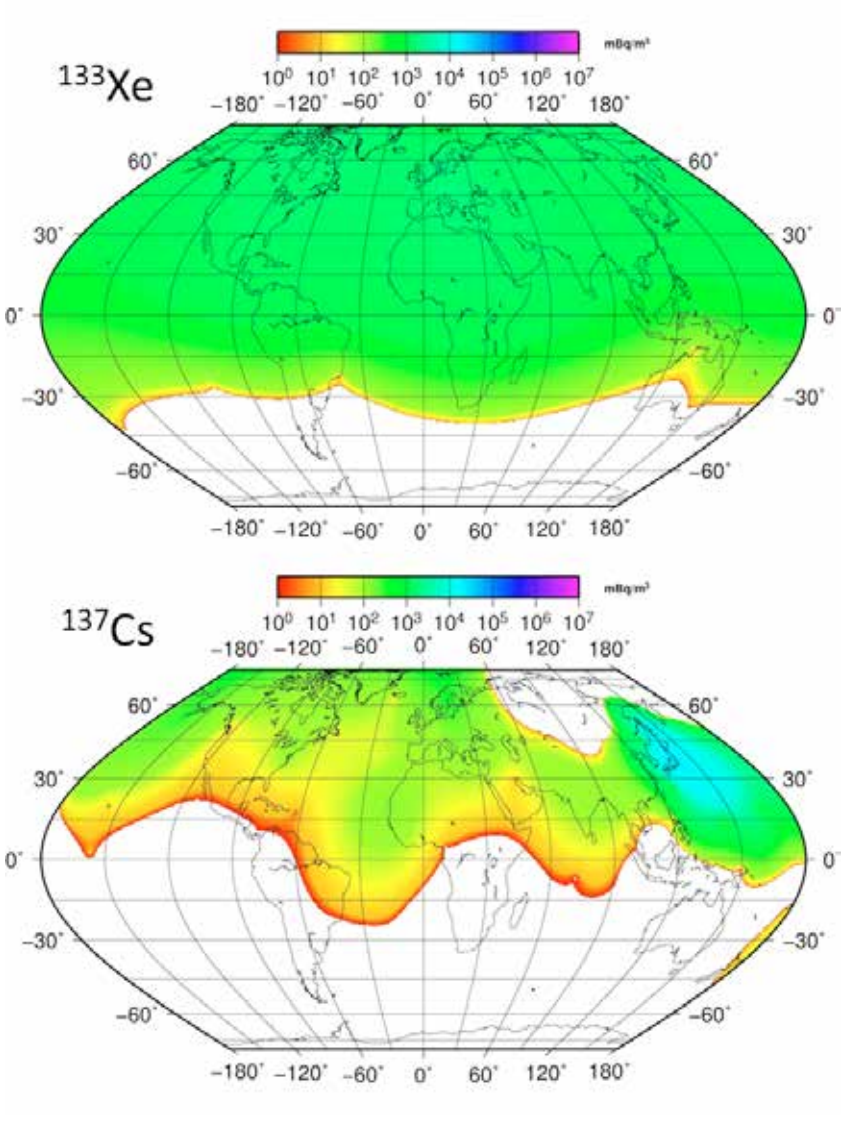


Figure 1:
Approximate distribution of the atmospheric activity concentration of the noble gas isotope 133Xe and the particle-borne radionuclide 137Cs on 30 March 2011, almost three weeks after the massive earthquake in Japan. The maps were obtained by interpolating data collected by the IMS radionuclide network. Note the homogenous distribution of xenon in the upper panel. The maps illustrate only the main features of the activity concentration, and should not be used to draw any quantitative conclusions for any specific geographic locations. In particular, the xenon concentration did not reach as far south as illustrated by the upper map.

be compared to the distribution of a particle-borne nuclide like Cs-137, which had a more inhomogeneous spatial activity distribution (see Figure 1).

KEY ROLE OF NOBLE GAS DETECTION SYSTEMS

Had the event occurred ten years earlier, before the IMS was equipped with noble gas detection systems, it would have been impossible to obtain a global picture of the noble gas releases from Fukushima. The development of the measurement systems that made this possible started some 15 years ago within the framework of the so-called International Noble Gas Experiment (INGE) which was set up in 1999 to test the measuring of radioactive noble gases in the atmosphere (see page 31). Of the 80 IMS radionuclide stations foreseen in the Treaty, 40 are to have additional noble gas detection capabilities. More than 75% of the noble gas network is now complete and equipped with these very sensitive measurement systems. A common way to represent the measurement capability of this network is to estimate the average number of stations that would detect a nominal explosion-size release of radionuclides from each point on the planet. This notion of coverage indeed captures an essential feature of the network. However, in judging how well the radionuclide network fulfills its part of the CTBT verification mission, one also needs to consider under which circumstances detections are most likely to be useful. These include possibilities for determining the source location and time of an event and the ability to distinguish it from detections that are not relevant to the CTBT, such as noble gas emissions by the radiopharmaceutical industry.

From this point of view, the releases from Fukushima constituted an enormous, if temporary, increase in background. To what extent was the effectiveness of the noble gas network hampered? The question is interesting both on a measurement level and on a data evaluation level. In fact, the noble gas detection systems responded very well to the impact of the Fukushima radiation releases. Apart from the very first samples measured at the IMS station in Takasaki, Japan, (the station located closest to the Fukushima Daiichi power plant which was hit by the largest concentrations of radionuclides), it was possible to use the data generated by the station to obtain accurate concentration values – even at such high levels – after appropriate corrections had been made.

On a measurement level, the strong global signature of the Fukushima releases reminds us of the need to overcome the so-called "memory effect" by which the measurement of a sample in systems using plastic scintillator detectors such as the SAUNA and ARIX systems (see page 31) containing a high amount of radioxenon activity will degrade the sensitivity of subsequent measurements for some time. Recent advances in the surface treatment

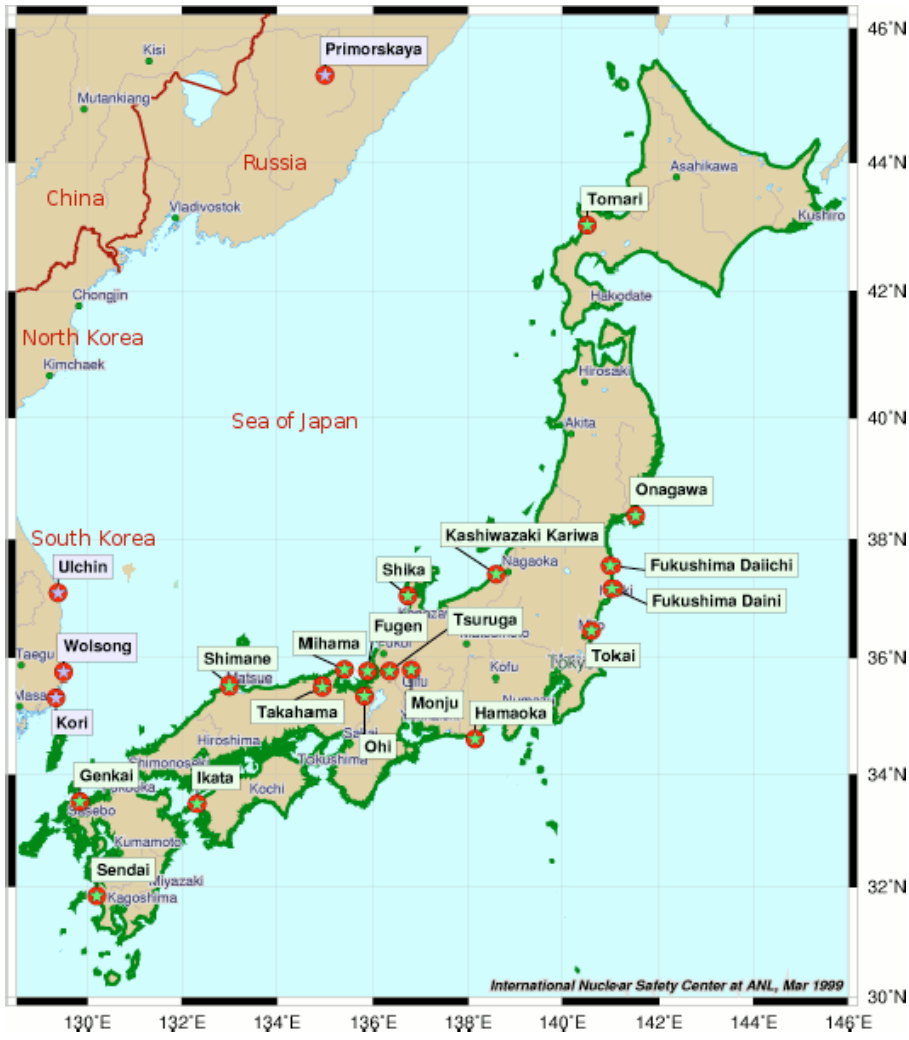
of detector cells (i.e. reducing the amount of radioactivity retained by the detector from previous measurements) promise to radically shorten or totally eliminate this recovery time following the measurement of a sample with a high concentration.

KNOWLEDGE OF RADIOXENON BACKGROUND LEVELS IS ESSENTIAL

On a network response level, the experience highlights the need to understand and discriminate against background from non-explosion radioxenon sources such as nuclear power plants and radio-pharmaceutical production facilities. The latter type of background source constitutes the greater problem, both in terms of released amounts (obviously, Fukushima is an exception) and, more seriously, released signatures: the short irradiations used in medical isotope production can produce radioxenon with isotopic ratios similar to those produced in a nuclear explosion. The solution lies partly in improved source location to screen out known background sources, and partly in mapping and understanding specific background sources as well as generic background in various regions of the world. The use of mobile or transportable radioxenon detection equipment in various campaigns around the world for this purpose has been highly successful and is expected to continue to yield valuable knowledge to improve background discrimination.

THE NEXT GENERATION OF DETECTION SYSTEMS

The experience that is being accumulated from operating a global network of sampling stations can now be used to design a next generation of detection systems, which will also be more specialized for the intended mission and incorporate a more holistic view of that mission. The overall task is to detect and identify nuclear explosions as part of a global network of monitoring stations incorporating a number of technologies. The optimal use of data from all relevant technologies embodies the concept of data fusion, which is increasingly discussed today. It is evident that while seismic signals would be the preferred primary source of location information pertaining to an event, the usefulness of radionuclide detections in demonstrating the nuclear character of that event depends on how well the source region of detected radionuclides can be defined.



Map of Japan showing the Fukushima Daiichi Nuclear Power Plant.

Obviously, the sensitivity for the detection of nuclides of interest is important. Source timing and source discrimination are typically obtained from ratios of concentrations measured in one nuclide to another. Thus, simultaneously detecting and quantifying two or more radioxenon isotopes will be considerably more valuable than the simple detection of only one. There are examples of detections of other radioxenon sources only a few weeks after the Fukushima accident, which could be identified despite high background concentrations. However, sensitivity is not the only parameter of interest. Given an ambient concentration of radionuclides around a station, the more air that is sampled, the better the detection sensitivity, but sampling more air takes more time. However, the time resolution of a measurement directly impacts the source location effectiveness of ATM. The 12- or 24-hour sampling periods of the systems deployed in the network may not in fact represent the optimum

ARIX noble gas system

balance between detection sensitivity and time resolution from the point of view of source location.

HOW TO AUGMENT NETWORK DENSITY

The most favourable case for using ATM to estimate a well-defined source region is when several network stations at different locations detect radionuclides from the same source event. However, there is no guarantee that this will be possible if the releases are from a nuclear explosion with a yield in the kiloton range. This is due to the current density of radioxenon measurement systems deployed by the IMS, even though the systems themselves exceed the initial sensitivity specifications. The operation of national monitoring stations contributing data to the CTBTO's International Data Centre (IDC) in Vienna and National Data Centres should be encouraged as a way to augment network density.

In the context of the Fukushima releases, it is important to remember that the IMS radionuclide network is not designed as a radiological emergency system. However, unlike national networks, the IMS network is a globally integrated radionuclide

FACT BOX

CAESIUM-137 (Cs-137):
Cs-137 has a half-life of 30.1 years. This is the most common radioactive form of caesium and is produced by nuclear fission. Cs-137 is one of the major radionuclides in spent nuclear fuel and radioactive wastes associated with the operation of nuclear reactors and fuel reprocessing plants. Large amounts of Cs-137 and other radioactive isotopes were released into the environment by atmospheric nuclear weapon tests between 1945 and 1980. Cs-137 did not occur in nature before nuclear weapon testing began.

IODINE-131 (I-131):
I-131 has a half-life of 8.0 days. I-131 is a radioactive isotope released into the environment mostly in gaseous form as a result of the atmospheric testing of nuclear weapons and accidents that have occurred at nuclear power plants (e.g. the Chernobyl nuclear power plant in 1986 and the Fukushima power plant in March 2011). It was a significant contributor to the effects on human health from atmospheric nuclear weapon testing and from the Chernobyl disaster.

XENON
is a chemical element in gaseous form. It is one of the noble gases which is inert and rarely reacts with other chemicals. Several of its radioactive isotopes, of which one of the isotopes is xenon-133 (Xe-133), are short-lived and typical of technological processes and are therefore measured to detect clandestine underground nuclear explosions.

BIOGRAPHICAL NOTES

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monitoring system, the only one of its kind. The global nature of the IMS network enabled the global tracking of radionuclides released in the Fukushima incident, and made the data available to the governments of all CTBTO Member States. The IMS network clearly has some special qualities which make the data valuable. The extent to which the data can be made public is governed by both practical and political constraints. On the practical side, only data which have been properly evaluated in the context of a possible radiological emergency should be released to the public. On the political side, the operation of the IMS is a collaborative endeavour among all Member States. It is in some ways an intrusive verification measure, with very sensitive monitoring stations located on the territory of a large number of States. In order to preserve long-term support for the CTBT verification regime and the continued availability of data, decisions on the handling of the monitoring data need to be made with due regard for the different perceptions that Member States may have of their national interests in this respect.

A WEALTH OF DATA HELPING TO INCREASE KNOWLEDGE

The build-up of the noble gas system component of the IMS has been very successful. New technology that many doubted would work 15 years ago has been developed and implemented in a short time. The new systems provide us with a vast amount of data that results in new knowledge every day. The tragic Fukushima accident reminded us how important the global detection network can be beyond its primary mission of verification. We now have to start gathering ideas on how to use this new knowledge in a broader context in order to fulfill the CTBT verification mission to the maximum extent possible, and to further investigate the possibilities that exist for using the system for other purposes as well.

THE DIFFERENT 'NOBLE GAS' SYSTEMS USED TO DETECT RADIONUCLIDES IN THE ATMOSPHERE

Radioactive xenon (radioxenon) isotopes are produced in abundance by a nuclear test. Four of them are used in CTBT monitoring: Xe-131m, Xe-133, Xe-133m, and Xe-135. They have a half-life of between 9.10 hours and 11.9 days (the half-life of any given nuclide is the time required for one half of the sample to decay). Although Xe-131m has the longest half-life of these four isotopes, its yield is the smallest. With a half-life of 5.2 days, xenon-133 (Xe-133) is in most circumstances the most abundant of the isotopes. With suitable equipment, it is therefore possible to detect radioxenon isotopes days or even weeks after their release and at great distances from their source.

The CTBTO uses three noble gas systems to detect radioxenon. Forty of the CTBTO's network of 80 radionuclide stations are being equipped with one of these special systems designed to detect noble gases:

- The Swedish Automatic Unit for Noble Gas Acquisition (SAUNA)
- Le Système de Prélèvements et d'Analyse en Ligne d'Air pour quantifier le Xénon (SPALAX)
- The Analyzer of Xenon Radioisotopes (ARIX). (See below for more information).

HOW RADIOXENON IS ISOLATED

The systems work by continuously and automatically separating xenon from ambient air using a purification device that contains charcoal. Contaminants such as dust as well as air constituents like oxygen, humidity, CO^2 and radon are all removed during this process. Radioactive levels in the isolated xenon are then measured in a radiation counting device. The resulting spectrum is sent to the International Data Centre (IDC) in Vienna on a daily basis for analysis. The noble gas systems that are currently deployed were developed in the late 1990s and have been tested under the International Noble Gas Experiment.

SPALAX noble gas system

SAUNA was developed by the Swedish Defence Research Agency and made commercially available in 2004. SAUNA samples up to $15m^3$ of air during a 12 hour sampling interval, thus producing two samples per day. The system uses a measurement technique called 'beta-gamma coincidence'.

ARIX was developed and commercialized by the Khlopin Radium Institute in the Russian Federation. ARIX samples up to $15m^3$ of ambient air and extracts xenon from it in 12-hour cycles This system produces two samples per day and also uses the beta-gamma coincidence technique.

SPALAX was developed by the French Commissariat à l'énergie atomique. This equipment samples up to $75m^3$ of air continuously in 24-hour cycles. At the end of each collection cycle and after final purification, the xenon gas is transferred into a High Purity Germanium detector counting system.

THE ROLE OF THE ANALYST

Analysts need to know which radionuclides occur naturally and which are man-made. It is imperative to know which radionuclides are generated by a nuclear explosion since xenon radionuclides also enter the atmosphere from other man-made sources. Analysts must also be familiar with the quantities and ratios in which these nuclides are produced during a nuclear explosion. Based on the analysis by National Data Centres and the IDC, Member States can then determine whether or not the sample suggests that a nuclear explosion has indeed taken place.