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## Workshop on Signatures of Medical and Industrial Isotope Production— A Review

|               |               |              |
|---------------|---------------|--------------|
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February 2010

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PACIFIC NORTHWEST NATIONAL LABORATORY

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## Summary

On July 1-3, 2009, in Strassoldo, Italy, more than 70 professionals representing the medical isotope production and the international monitoring communities from 16 countries came together to discuss the impacts of medical isotope production on the international monitoring system at the Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP). The workshop was hosted and organized by PNNL.

Medical and industrial isotopes are fundamental tools used in science, medicine, and industry and hence large amounts of isotopes are produced every year at locations across the globe using a variety of means, and releasing detectable amounts of radioisotopes into the atmosphere.

At the same time, the scientific community has explored increasingly sensitive methods for detecting isotopes as part of nuclear treaty monitoring and verification and for other reasons. As a consequence, isotopes including those that are short-lived, are frequently detected in these advanced measurement systems usually well below levels of naturally occurring radioisotopes such as Rn-222 and its daughters.

WOSMIP presented the first opportunity for these two communities of people (medical isotope production and monitoring) to come together to discuss the impacts their missions have on each other. The workshop provided a forum to foster communication and build a stronger collaboration and information sharing between scientists. The workshop has resulted in a better understanding of the isotopic and chemical signatures created through isotope production mechanisms and the trace quantities of these isotopes that are detected in the environment.

The workshop was very successful with a number of positive outcomes.

## Symbols, Acronyms and/or Initialisms

|         |  |
|---------|--|
| ANSTO   | Australian Nuclear Science and Technology Organisation |
| ARIX    | Analyzer of Radioisotopes of Xenon                     |
| ARSA    | Automated Radioxenon Sampler/Analyzer                  |
| ATR     | Advanced Test Reactor                                  |
| BLIP    | Brookhaven LINAC Isotope Producer                      |
| CNEA    | Comisión Nacional de Energía Atómica                   |
| CRL     | Chalk River Laboratories                               |
| CTBT(O) | Comprehensive Nuclear-Test-Ban Treaty (Organization)   |
| HEU     | Highly-enriched uranium                                |
| HFIR    | High-Flux Isotope Reactor                              |
| HFR     | High-Flux Reactor                                      |
| IAEA    | International Atomic Energy Agency                     |
| IMS     | International Monitoring System                        |
| INVAP   | Investigación Aplicada                                 |
| IPF     | Isotope Production Facility                            |
| IRE     | Institute of Radioelements (Fleurus, Belgium)          |
| LEU     | Low-enriched uranium                                   |
| LINAC   | Linear Accelerator                                     |
| MURR    | University of Missouri Research Reactor                |
| NECSA   | Nuclear Energy Corporation of South Africa             |
| NPP     | Nuclear Power Plant                                    |
| NPT     | Non-Proliferation Treaty                               |

|          |   |
|----------|---|
| NRU      | National Research Universal reactor   |
| OPAL     | Open Pool Australian Lightwater reactor                                       |
| PINSTECH | Pakistan Institute of Nuclear Science and Technology                          |
| SAUNA    | Swedish Unattended Noble-gas Analyzer   |
| SPALAX   | Système de Prélèvements et d'Analyse en Ligne. d'Air pour quantifier le Xénon |
| USA      | United States of America  |
| WOSMIP   | Workshop on Signatures of Medical and Industrial Isotope Production           |

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## 1.0 Overview

The Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP) was held July 1-3, 2009 at Strassoldo, Italy. The meeting was intended to bring together experts from communities that would not normally interact –those producing and using medical radionuclides, and those monitoring the environment for associated emissions with concerns for nuclear security and proliferation. The meeting was expected to be an initial step towards improving the understanding of processes and nuclide signatures from their production to movement and detection in the environment. These are important issues because the prevalence of manmade nuclides is increasing in the environment, they are often detected in monitoring networks and at border crossings, and they impinge on treaty monitoring operations connected with the Comprehensive Nuclear-Test-Ban Treaty (CTBT), the Non-Proliferation Treaty (NPT), and Fissile Cut-off considerations, for example. A greater awareness and understanding of trends in nuclide production is therefore required – how they are produced, trapped, transported, used, and detected – and the WOSMIP meeting laid the groundwork for improved communication, cooperation and understanding between the disparate groups involved.

The information provided in this document was compiled from the presentations given at the workshop.

## 2.0 Medical applications of radioactive materials

Medical applications of radionuclides in diagnosis and treatment have grown exponentially over recent decades. Today some 10,000 medical centers worldwide use radionuclides in almost 90% of diagnostic procedures. In developed countries these procedures are applied to up to ~4% of the population annually, with therapeutic usage being about one tenth of this. This amounted to over 20 million procedures in the United States of America (USA) alone during 2008.

The various possible combinations of availability, half-life, decay mode, decay energy and ease of labeling of biomolecules provide a wide range of radionuclides that may be used in diagnosis and therapy. Isotopes used for diagnostic and radio-therapeutic applications are shown in **Table 1** and **Table 2** below.

**Table 1.** Isotopes used in diagnostic applications

| Beta/gamma emitters | Positron emitters | Auger-electron emitters |
|---------------------|-------------------|-------------------------|
| $^{131}\text{I}$    | $^{18}\text{F}$   | $^{111}\text{In}$       |
| $^{111}\text{In}$   | $^{11}\text{C}$   | $^{123}\text{I}$        |
| $^{201}\text{Tl}$   | $^{15}\text{O}$   | $^{125}\text{I}$        |
| $^{89}\text{Sr}$    | $^{13}\text{N}$   |                         |
| $^{103}\text{Pd}$   | $^{82}\text{Rb}$  |                         |
| $^{192}\text{Ir}$   | $^{68}\text{Ge}$  |                         |

| Beta/gamma emitters      | Positron emitters               | Auger-electron emitters |
|--------------------------|---------------------------------|-------------------------|
| $^{153}\text{Sm}$        | $^{60}\text{Cu}$                |                         |
| $^{166}\text{Ho}$        | $^{64}\text{Cu}$                |                         |
| $^{99\text{m}}\text{Tc}$ | $^{61}\text{Cu}$                |                         |
| $^{90}\text{Y}$          | $^{76}\text{Br}$                |                         |
| $^{175}\text{Yb}$        | $^{77}\text{Br}$                |                         |
| $^{166}\text{Dy}$        | $^{124}\text{I}$                |                         |
|                          | $^{94\text{m}}\text{Tc}$        |                         |
|                          | $^{86}\text{Y}$                 |                         |
|                          | $^{89}\text{Zr}$                |                         |
|                          | $^{66}\text{Ga}$                |                         |
|                          | $^{68}\text{Ge}/^{68}\text{Ga}$ |                         |
|                          | $^{30}\text{P}$                 |                         |
|                          | $^{34\text{m}}\text{Cl}$        |                         |

**Table 2.** Isotopes used in radio-therapeutic applications

| Beta Emitters     | Positron Emitters | Alpha Emitters    | Auger-Electron Emitters   |
|-------------------|-------------------|-------------------|---------------------------|
| $^{131}\text{I}$  | $^{64}\text{Cu}$  | $^{211}\text{At}$ | $^{77}\text{Br}$          |
| $^{89}\text{Sr}$  | $^{66}\text{Ga}$  | $^{223}\text{Ra}$ | $^{111}\text{In}$         |
| $^{153}\text{Sm}$ |                   | $^{225}\text{Ac}$ | $^{123}\text{I}$          |
| $^{166}\text{Ho}$ |                   | $^{149}\text{Tb}$ | $^{125}\text{I}$          |
| $^{90}\text{Y}$   |                   | $^{224}\text{Ra}$ | $^{67}\text{Ga}$          |
| $^{177}\text{Lu}$ |                   | $^{212}\text{Bi}$ | $^{201}\text{Tl}$         |
| $^{149}\text{Pm}$ |                   | $^{213}\text{Bi}$ | $^{51}\text{Cr}$          |
| $^{199}\text{Au}$ |                   | $^{227}\text{Th}$ | $^{140}\text{Nd}$         |
| $^{64}\text{Cu}$  |                   | $^{255}\text{Fm}$ | $^{195\text{m}}\text{Pt}$ |

| Beta Emitters             | Positron Emitters | Alpha Emitters | Auger-Electron Emitters |
|---------------------------|-------------------|----------------|-------------------------|
| $^{186}\text{Re}$         |                   |                |                         |
| $^{188}\text{Re}$         |                   |                |                         |
| $^{67}\text{Cu}$          |                   |                |                         |
| $^{117\text{m}}\text{Sn}$ |                   |                |                         |
| $^{32}\text{P}$           |                   |                |                         |
| $^{165}\text{Dy}$         |                   |                |                         |
| $^{105}\text{Rh}$         |                   |                |                         |
| $^{111}\text{Ag}$         |                   |                |                         |

With this many radioisotopes a challenge begins to arise as to how to label an ever-expanding range of biomolecules that allow targeting of diagnosis and treatment to specific sites in the body. Because massless amounts of the nuclides are used, treatments and diagnosis can be carried out without disrupting normal biochemical or metabolic processes.

The most commonly used radionuclide is  $^{99\text{m}}\text{Tc}$ , comprising ~80% of medical applications. It was used in 30 million procedures during 2006, comprising 75% of all nuclear medicine procedures. In 2007, portions of the German, US and Swedish populations examined with  $^{99\text{m}}\text{Tc}$  procedures were 4.4%, 3.9% and 1.2% respectively. On a world-wide basis radionuclide application obviously varies with the level of health care available, but at the highest level ~40,000,000 people were examined with  $^{99\text{m}}\text{Tc}$  procedures in 2000, involving a total application of 24,000 TBq, with global  $^{99\text{m}}\text{Tc}$  usage increasing at a rate of 4.5% per year.

Iodine-131 is another widely used radionuclide, with application in the treatment of hyperthyroidism (0.2 – 0.5 GBq per treatment) and thyroid cancer (1.8 – 9.2 GBq per treatment). In 1996 some 180,000 patients were treated for hyperthyroidism and 20,000 for thyroid cancer. The projected requirement for  $^{131}\text{I}$  in the USA for thyroid cancer treatment in 2009 is  $1.7 \times 10^5$  GBq. Application rates of  $^{131}\text{I}$  in other countries in recent years indicate the extent of its use: Argentina: 300 TBq/y; Bangladesh: 5 TBq/y; Chile: 15,000 patients per year requiring 15 TBq/y; India: 60 TBq/y; Thailand: 10,000 patients in 2008 requiring 15 TBq; Austria: 1 TBq in 2008; Australia: ~200 patients per year requiring 4-6 GBq per patient.

Medical uses of nuclides are becoming increasingly sophisticated. With new nuclides becoming available, and more importantly, better detector/imaging systems, the use of multiple nuclides concurrently is a possibility. The targeting agents used to link nuclides are also increasing in number and sophistication. Along with this burgeoning usage of medical radionuclides there is growing concern over the robustness of supply. The radionuclides are produced principally by neutron irradiation of precursors or nuclear fission in small nuclear reactors, in cyclotrons, or by accelerators, with most production being centered in a small number of large institutions. This creates an uncertainty of supply. In response, new facilities are

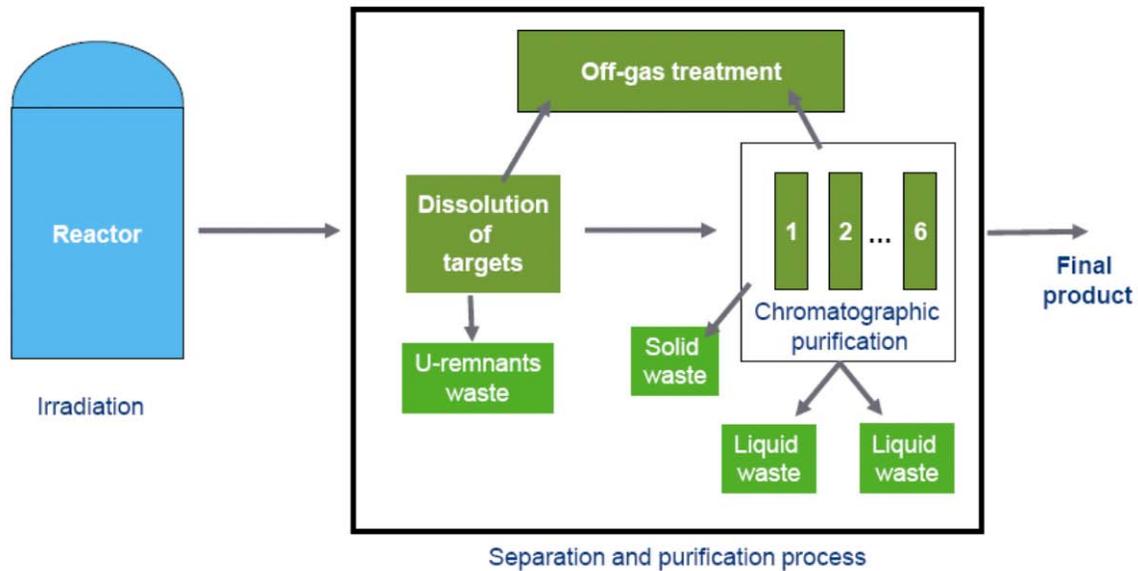
being set up in many countries, particularly for the production of  $^{99m}\text{Tc}$ . Concerns over the security of nuclear materials have increased proportionately.

## 3.0 Radionuclide production

### 3.1 Technetium-99m and Iodine-131

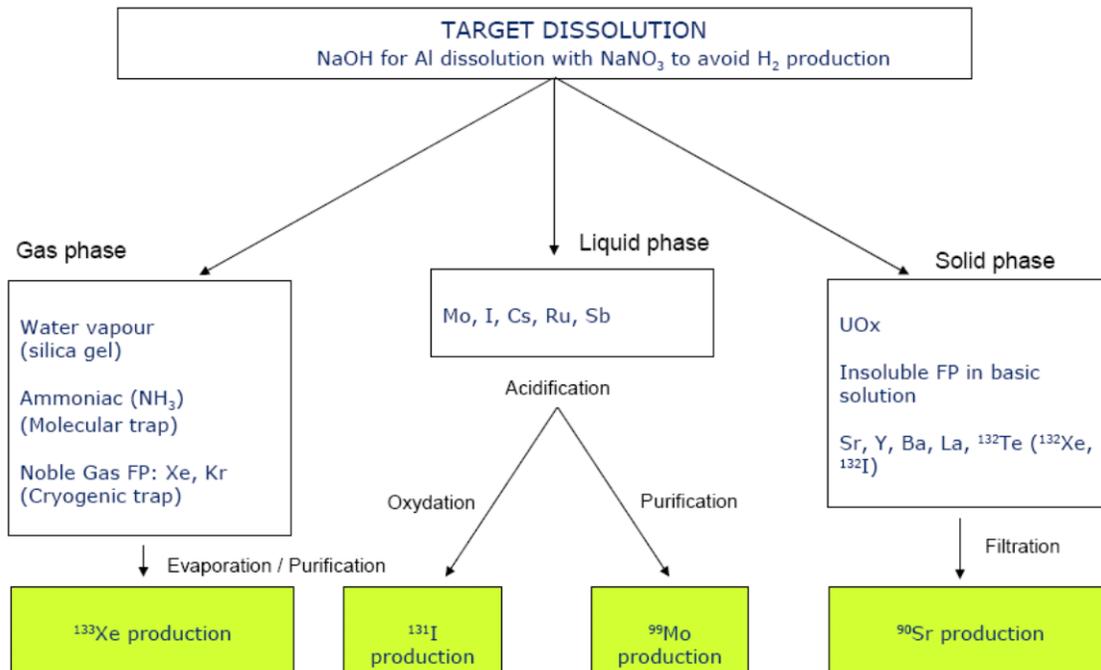
Neutron-induced fission of  $^{235}\text{U}$  is currently the most common method of producing  $^{99}\text{Mo}$ , the precursor of  $^{99m}\text{Tc}$ , and  $^{131}\text{I}$ . To meet global demand, several thousand TBq of  $^{99}\text{Mo}$  must be produced weekly, or 12,000 “six-day Curies” (6 dCi) per week. The production process involves highly-enriched uranium target (HEU- $\text{Al}_x$  dispersed in aluminum powder, with aluminum cladding); target irradiation; dissolution of target and fuel; recovery and purification of  $^{99}\text{Mo}$ ; and finally shipment of  $^{99}\text{Mo}$  to  $^{99m}\text{Tc}$  generator producers. As  $^{131}\text{I}$  is also a fission product, it may also be recovered during  $^{99}\text{Mo}$  production. Additionally, it is produced by neutron activation of  $^{130}\text{Te}$ , as at Chalk River, and as well as in Algeria, Bangladesh, Brazil, Chile, India, Iran, Kazakhstan, Pakistan, Poland, Syria, Uzbekistan and Vietnam. The basics of the  $^{99}\text{Mo}$  process are shown below.

### Mo-99 Process



**Figure 1.** Overview of molybdenum-99 production process

## Target processing



**Figure 2.** The typical target processing procedure, shown schematically (using the Fleurus procedure as an example)

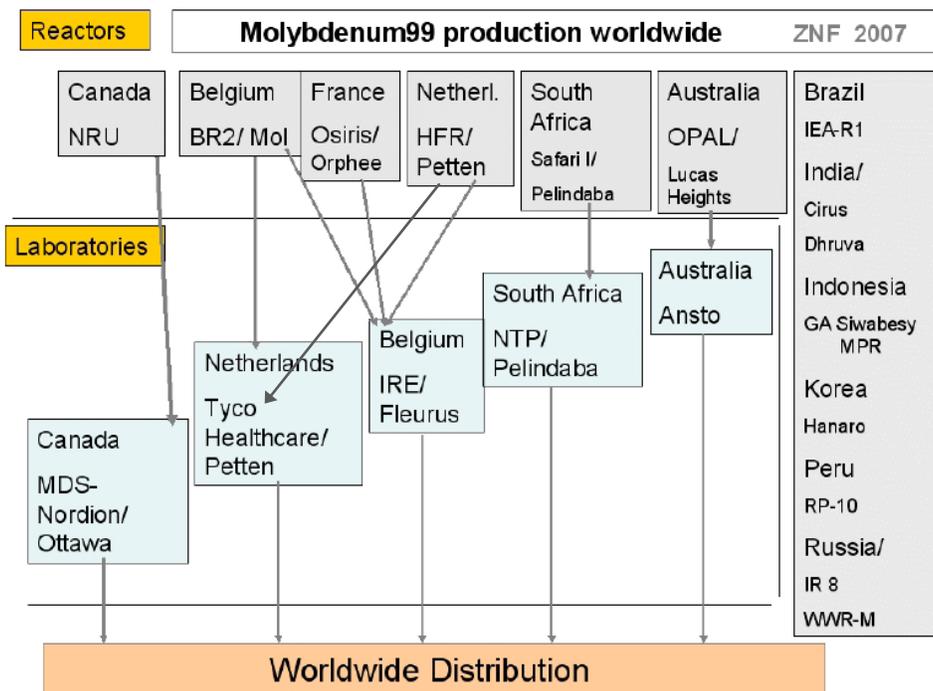
More than 95% of all  $^{99}\text{Mo}$  is produced by irradiation of HEU targets by four major suppliers:

- MDS Nordion, Canada, using 93% HEU irradiated in the National Research Universal (NRU) reactor at the AECL Chalk River Laboratories (CRL); to date providing 40% of the world supply of  $^{99}\text{Mo}$  at a rate of  $\sim 800$  TBq/week. Irradiation time is 10-20 days; after which the fuel is removed from the reactor, cooled for several hours, then transported to processing hot cell. Processing includes mechanical decladding, dissolution in  $\text{HNO}_3$ , separation on alumina columns that retain molybdenum which is then stripped with ammonium hydroxide solution. The “raw moly” product is shipped to Nordion for purification.
- Institute of Radioelements (IRE), Fleurus, Belgium, using 93% HEU irradiated in three European reactors – High Flux Reactor (HFR, Petten, Netherlands), BR-2 (Mol, Belgium), and Osiris (Saclay, France); supplying 10-30% of world demand at a rate of  $\sim 600$  TBq/week. After cooling, targets are transported to Fleurus; 30 hours after irradiation they are dissolved in  $\text{NaOH}/\text{NaNO}_3$  solution; xenon and krypton are released to off-gas. After filtration and acidification, molybdenum is recovered on alumina columns and purified on anion exchange and charcoal columns. The BR-2 reactor uses targets containing 4-5 g  $^{235}\text{U}$  with a neutron flux of  $2.5 \times 10^{14}$  n/cm<sup>2</sup>/s, and an irradiation time of 150 hours.
- Mallinckrodt Medical (Covidien), Netherlands, using 93% HEU irradiated at the HFR and BR-2 reactors, providing  $>25\%$  of demand at a rate of  $\sim 600$  TBq/week. Twenty to 30 hours after irradiation, the target is dissolved in  $\text{NaOH}$  solution. Xenon and krypton off-gases are collected in pre-evaluated tanks before their controlled release through charcoal beds.

Uranium precipitates as the diuranate and the alkali-insoluble fission products precipitate as hydroxides. The filtrate is purified by anion exchange after iodine is pre-separated on a floating bed of hydrated silver oxide. The molybdenum eluate is further purified. Uranium and alkali-insoluble fission products precipitate as hydroxides and the filtrate is purified by anion exchange which retains molybdenum and iodine, which are stripped separately and purified.

- The Nuclear Energy Corporation of South Africa (NECSA) NTP Radioisotopes, South Africa, using 45% HEU targets irradiated in the SAFARI-1 reactor; meeting 10-15% of demand at a rate of ~200 TBq/week.

The figure below shows a subset of the relationships.



**Figure 3.** Relationships between irradiating facilities and laboratories that purify the <sup>99</sup>Mo

Much of medical nuclide production utilizes research reactors, but no reactors are solely dedicated to medical nuclide production. This makes medical nuclide production one of many users for reactor time, and as such, does on occasion experience a disruption in production due to reactor shutdown for other programs.

There is a growing trend toward use of low-enriched uranium (LEU) caused partly by increasing concerns over the proliferation of HEU facilities worldwide. This process has been adopted by various smaller-scale producers:

1. Comisión Nacional de Energía Atómica (CNEA), Argentina, converted from HEU to LEU in 2002, with processing similar to that used by Covidien and others, producing 200 6dCi per week. In Argentina approximately 80% of all nuclear medicine procedures use <sup>99m</sup>Tc, with pulmonary

studies using aerosolized forms of  $^{99m}\text{Tc}$  (0.9 to 1.3 GBq per procedure) and with injections for brain, heart, liver, bones, and kidney scans (average roughly 0.45 GBq per procedure). CNEA also produces  $^{131}\text{I}$  by fission. Ezeiza Atomic Center produces  $^{99}\text{Mo}$  for  $^{99m}\text{Tc}$  generators from fission of LEU in RA3 reactor at rate of  $\sim 10$  TBq per week.

2. Australian Nuclear Science and Technology Organisation (ANSTO), currently starting with Argentinean Investigación Aplicada (INVAP) LEU targets for up to 2000 6 dCi per week
3. Egypt – Atomic Energy Authority of Egypt also using INVAP technology for small-scale production
4. Indonesia – Batan Teknologi currently produces 50 6dCi per week using the “Cintichem process” with HEU targets but is converting to LEU targets in January 2010.
5. Pakistan – Pakistan Institute of Nuclear Science and Technology (PINSTECH), is expected to come on-line soon using the German ROMOL-99 process from 2009, with LEU targets to produce 25 6dCi/week
6. MURR - University of Missouri Research Reactor, Columbia, is currently in the research and development stage with an LEU-modified “Cintichem” process

The USA relies on imported  $^{99}\text{Mo}$  to meet its needs, but there is a desire for production there by upgrading the University of Missouri Research Reactor (MURR) with LEU targets and fuel; developing a Babcock & Wilcox partnership with Covidien using an “aqueous homogenous reactor” (solution reactor) fueled by LEU; or with the Advanced Medical Isotope Corporation and the University of Missouri using a proprietary  $(\gamma, n)$  system.

### 3.2 Other radionuclides

In addition to nuclear fission, other techniques are employed in the production of medical radionuclides. Neutron-activation reactions in reactors are obviously important in this regard, but cyclotrons, linear accelerators, alpha-particle accelerators, and electron beam (x-ray) interactions are making a growing contribution.

The BR-2 reactor facility employs  $(n, \gamma)$  reactions to produce  $^{192}\text{Ir}$  from natural  $^{191}\text{Ir}$  discs,  $^{188}\text{W}/^{188}\text{Re}$  from enriched  $^{186}\text{W}$  discs,  $^{89}\text{Sr}$  from  $^{88}\text{SrCO}_3$  targets,  $^{186}\text{Re}$  and  $^{153}\text{Sm}$  from  $^{185}\text{Re}$  and  $^{152}\text{Sm}_2\text{O}_3$ ,  $^{177}\text{Lu}$  from  $^{176}\text{Lu}$ ,  $^{203}\text{Hg}$  from metallic Hg, and  $^{60}\text{Co}$  from  $^{59}\text{Co}$ . The Maria water/beryllium-moderated reactor in Poland (Radioisotope Centre POLATOM) is the main supplier of radiopharmaceuticals for Poland and also exports products of  $(n, \gamma)$  reactions producing  $^{32}\text{P}$ ,  $^{89}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{105}\text{Rh}$ ,  $^{117m}\text{Sn}$ ,  $^{131}\text{I}$ ,  $^{149}\text{Pm}$ ,  $^{153}\text{Sm}$ ,  $^{166}\text{Ho}$ ,  $^{177}\text{Lu}$  and  $^{186}\text{Re}$ . Scandium-47 is also produced in this facility, by the  $^{47}\text{Ti}(n,p)^{47}\text{Sc}$  or  $^{46}\text{Ca}(n,\gamma)^{47}\text{Ca}$  (with decay of  $^{47}\text{Ca}$ ) reactions.

Electron accelerators produce radionuclides through  $(\gamma, p)$  or  $(\gamma, n)$  reactions based on 25 MeV electron bremsstrahlung. Carrier-free  $^{155}\text{Tb}$ ,  $^{167}\text{Tm}$ ,  $^{111}\text{In}$  and  $^{123}\text{I}$  are produced through reactions  $^{156}\text{Dy}(\gamma, n)^{155}\text{Dy}/\text{EC}/\rightarrow^{155}\text{Tb}$ ;  $^{168}\text{Yb}(\gamma, n)^{167}\text{Yb}/\text{EC}/\rightarrow^{167}\text{Tm}$ ;  $^{112}\text{Sn}(\gamma, n)^{111}\text{Sn}(\text{EC}, \beta^+)\rightarrow^{111}\text{In}$  or  $^{112}\text{Sn}(\gamma, p)^{111}\text{In}$ ; and  $^{124}\text{Xe}(\gamma, n)^{123}\text{I}$ . Positron-active radionuclides  $^{15}\text{O}$ ,  $^{13}\text{N}$ ,  $^{11}\text{C}$ ,  $^{18}\text{F}$ ,  $^{30}\text{P}$ ,  $^{34m}\text{Cl}$  and  $^{38}\text{K}$  are also produced by  $(\gamma, n)$  reactions. There is also potential for production of  $^{99}\text{Mo}$  through the reaction  $^{100}\text{Mo}(\gamma, n)^{99}\text{Mo}$ .

The Austrian cyclotron facility (Seibersdorf) produces  $^{18}\text{F}$  and  $^{11}\text{C}$  through  $^{18}\text{O}(p,n)^{18}\text{F}$  and  $^{14}\text{N}(p,\alpha)^{11}\text{C}$ .

Within the USA various institutions produce a range of radionuclides: the High-Flux Isotope Reactor (HFIR, Oak Ridge, TN) uses HEU fuel elements to produce  $^{75}\text{Se}$ ,  $^{252}\text{Cf}$ ,  $^{188}\text{W}/^{188}\text{Re}$ ,  $^{63}\text{Ni}$  and  $^{75}\text{Se}$ ; the Advanced Test Reactor (ATR, near Idaho Falls, ID) produces  $^{60}\text{Co}$ ; The Brookhaven Linear Accelerator (LINAC) Isotope Producer (BLIP) produces  $^{68}\text{Ge}/^{68}\text{Ga}$ ,  $^{82}\text{Sr}/^{82}\text{Rb}$ , also  $^{65}\text{Zn}$ ,  $^{28}\text{Mg}$ ,  $^{52}\text{Fe}$ , and  $^{83}\text{Rb}$ ; the Isotope Production Facility (IPF) at Los Alamos employs a LINAC proton beam to produce  $^{68}\text{Ge}/^{68}\text{Ga}$ ,  $^{82}\text{Sr}/^{82}\text{Rb}$  and smaller amounts of  $^{26}\text{Al}$  and  $^{32}\text{Si}$ ; commercial cyclotrons accelerate charged hydrogen atoms (protons, deuterons) to energies up to 100 MeV for production of proton-rich nuclides including

- |                    |                    |                                   |                                   |                    |
|--------------------|--------------------|-----------------------------------|-----------------------------------|--------------------|
| • $^{18}\text{F}$  | • $^{13}\text{N}$  | • $^{67}\text{Ga}$                | • $^{111}\text{In}$               | • $^{89}\text{Zr}$ |
| • $^{82}\text{Sr}$ | • $^{76}\text{Br}$ | • $^{67}\text{Cu}$                | • $^{123}\text{I}$                |                    |
| • $^{64}\text{Cu}$ | • $^{77}\text{Br}$ | • $^{60}\text{Cu}$                | • $^{103}\text{Pd}$               |                    |
| • $^{67}\text{Cu}$ | • $^{124}\text{I}$ | • $^{61}\text{Cu}$                | • $^{32}\text{P}$                 |                    |
| • $^{15}\text{O}$  | • $^{86}\text{Y}$  | • $^{68}\text{Ge}/^{68}\text{Ga}$ | • $^{82}\text{Sr}/^{82}\text{Rb}$ |                    |
| • $^{11}\text{C}$  | • $^{66}\text{Ga}$ | • $^{82}\text{Sr}$                | • $^{201}\text{Tl}$               |                    |

The University of Missouri produces  $^{51}\text{Cr}$ ,  $^{192}\text{Ir}$ ,  $^{186}\text{Re}$ ,  $^{166}\text{Ho}$ ,  $^{32}\text{P}$ ,  $^{33}\text{P}$ ,  $^{153}\text{Sm}$ ,  $^{177}\text{Lu}$ ; and other universities have the capability to produce  $^{18}\text{F}$ ,  $^{111}\text{In}$ ,  $^{211}\text{At}$ ,  $^{64}\text{Cu}$ ,  $^{73}\text{As}$ ,  $^{77}\text{Br}$ ,  $^{124}\text{I}$ ,  $^{86}\text{Y}$ , and  $^{89}\text{Zr}$ . A number of important nuclides are needed in the U.S. that are not currently available in sufficient amounts and quality for special applications in medical research, applied clinical nuclear medicine, science, oil exploration, construction, homeland security, national security, and defense including  $^{241}\text{Am}$ ,  $^{252}\text{Cf}$ ,  $^{99}\text{Mo}$ ,  $^{225}\text{Ac}$ ,  $^{232}\text{U}$ ,  $^{153}\text{Gd}$ ,  $^{147}\text{Pm}$ ,  $^{67}\text{Cu}$ ,  $^{211}\text{At}$ ,  $^{89}\text{Zr}$ , and  $^{117\text{m}}\text{Sn}$ .

Bench-scale electronic devices are under development for achieving various high-energy nuclear reactions and isotope enrichment processes as a “next-generation” approach to nuclide production where nuclear reactors and cyclotrons are not available, are too complex, or are too expensive to acquire and operate. Proton accelerators; alpha accelerators; neutron generators; electron-beam x-ray systems; and stable isotope plasma separation systems have significant roles to play. The first U.S. 7-MeV proton linear accelerator for medical nuclide production is now operating and is producing  $^{18}\text{F}$  for regional hospitals, together with  $^{111}\text{In}$ ,  $^{123}\text{I}$ ,  $^{11}\text{C}$ ,  $^{13}\text{N}$ , and  $^{15}\text{O}$ ; alpha LINAC accelerating helium or deuterium to 40 MeV, and electron cyclotron resonant plasma source for helium ions are employed in production of nuclides such as  $^{117\text{m}}\text{Sn}$ ,  $^{225}\text{Ac}$ ,  $^{73}\text{As}$ ,  $^{55}\text{Fe}$ ,  $^{211}\text{At}$ ,  $^{109}\text{Cd}$ ,  $^{88}\text{Y}$ ,  $^{75}\text{Se}$ ,  $^{210}\text{Po}$ , and  $^{148}\text{Gd}$ . Neutron generators such as the Berkley coaxial D-T radiofrequency-driven plasma ion source cylindrical neutron generator provide reactions such as  $^2\text{H}$  on  $^9\text{Be} \rightarrow ^{10}\text{B} + \text{n}$ ; while electron beam accelerators, producing bremsstrahlung from 10-25 MeV electrons, are proposed for nuclide production through photo-fission of heavy elements,  $(\gamma, \text{n})$  reactions, photo-neutron activation, and  $(\text{n}, 2\text{n})$  reactions.

In terms of scale of production, this wide range of radionuclides is relatively insignificant compared with  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ , although the burgeoning demand will continue to stress the supply system. A crisis is currently looming with  $^{99}\text{Mo}$  supply due to the forthcoming closure of the Chalk River facility. A small leak of heavy-water moderator was discovered on 15 May 2009; a two-day search focused on a small area of corrosion near the base of the aluminum reactor vessel. A pinhole leak in the area of corrosion was determined to be the source of the moderator water; there were also ventilation losses of tritium, well within regulatory limits, due to the leak. The reactor has been defueled and the moderator drained to a minimum. A successful repair is anticipated to be likely, once the procedures have been adequately tested,

but the plant may still be permanently closed. The medical impact of such a closure would be felt severely worldwide, and pressure to develop other means of production is increasing.

In view of the increasing demand for medical radionuclides and the widening geographic regions in which they are applied, the International Atomic Energy Agency (IAEA) Coordinated Research Project aims to assist Member States in adopting the LEU Cintichem process or neutron activation technique, and to foster capacity building for local/regional  $^{99}\text{Mo}$  self-sufficiency. In the near future this is expected to bring online suppliers in Chile, Libya, Poland, Romania, and gel generators in Kazakhstan and Romania. There may also be a reactivation of historical production in Russia and Germany.

## 4.0 Environmental releases and their signatures

In terms of production volume,  $^{99}\text{Mo}$  is the dominant medical radio-nuclide and its production mechanism is, in turn, dominated by the use of nuclear reactors and fission reactions. This results in the unavoidable production of xenon isotopes, which are of significance to various environmental surveillance programs such as that involved in compliance verification of the CTBT. Radioxenon gases are a key indicator of whether or not an (underground) explosion is nuclear in nature. An understanding of release mechanisms and rates and isotope ratios is therefore critical in avoiding misinterpretation of monitoring data.

The global atmospheric radioxenon background is mainly determined by radiopharmaceutical facilities. In the Northern Hemisphere it is dominated by the Chalk River and Fleurus facilities and in the Southern Hemisphere by the Pelindaba facility in South Africa.

The particular nuclides of interest in CTBT verification are the xenon isotopes  $^{131\text{m}}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ ,  $^{133}\text{Xe}$  and  $^{135}\text{Xe}$ , with half-lives of 9 h - 11.9 d, as illustrated below:

**Table 3.** Approximate xenon releases incurred during  $^{99}\text{MO}$  and  $^{131}\text{I}$  production

| <b>Categorization List for Xenon Samples (CLXS)</b> |                  |   |   |                                    |                                     |
|---|------------------|---|---|------------------------------------|-------------------------------------|
| <b>Fission Product</b>                              | <b>Half-life</b> | <b>Primary <math>\gamma</math> Energy and Abundance</b> | <b>K-shell X-ray Emission and Abundance</b> | <b>Beta Spectrum and Abundance</b> | <b>K-Shell Conversion Electrons</b> |
| $^{131\text{m}}\text{Xe}$                           | 11.93 d          | 163.9 keV<br>(1.96%)                                    | 30 keV<br>(54.05 %)                         |                                    | 129 keV<br>(60.7%)                  |
| $^{133\text{m}}\text{Xe}$                           | 2.19 d           | 233.2 keV<br>(10.3%)                                    | 30 keV<br>(56.3)%                           |                                    |                                     |

|                   |        |                    |                   |   |                   |
|-------------------|--------|--------------------|-------------------|---|-------------------|
| $^{133}\text{Xe}$ | 5.25 d | 81.0 keV<br>(37%)  | 31 keV<br>(48.9%) | $E_{\text{max}}$ 356 keV<br>$E_{\text{avg}}$ 99 keV<br>(99%)  | 45 keV<br>(54.1%) |
| $^{135}\text{Xe}$ | 9.14 h | 249.8 keV<br>(90%) | 31 keV<br>(5.2 %) | $E_{\text{max}}$ 905 keV<br>$E_{\text{avg}}$ 300 keV<br>(97%) | 214 keV<br>(5.7%) |

Xenon releases incurred during fissionogenic  $^{99}\text{Mo}$  and  $^{131}\text{I}$  production are summarized below. Note that many other countries produce  $^{131}\text{I}$  by activation which will not contribute to the xenon release data, and that Argentina produces  $^{131}\text{I}$  via fission but on a significantly smaller scale than IRE and NTP.

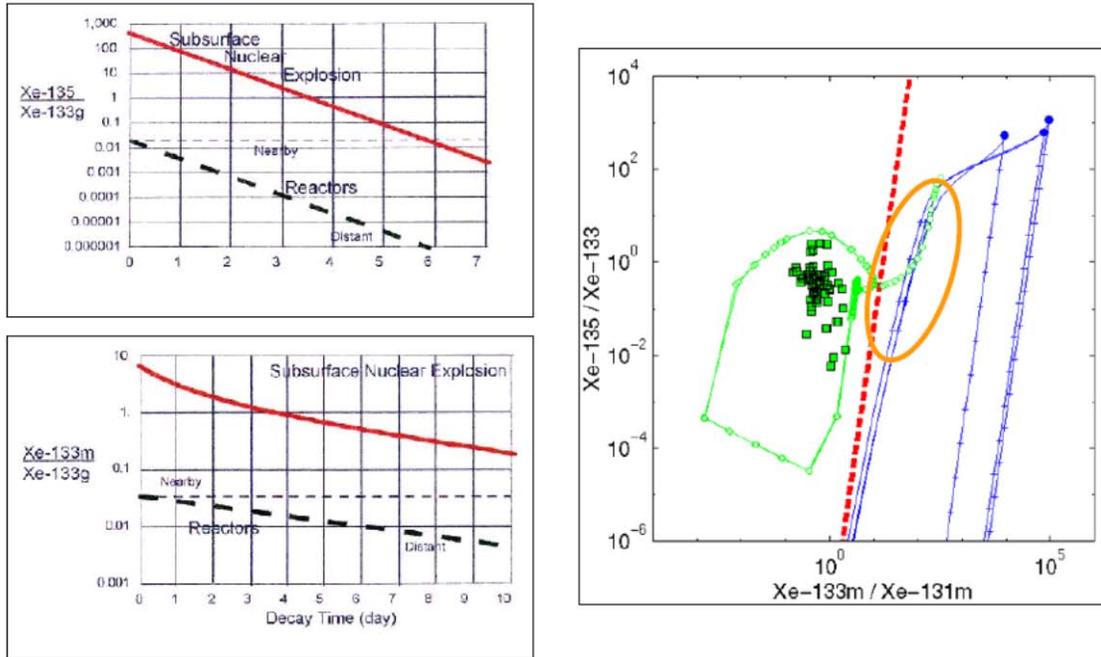
**Table 4.** Relevant xenon cumulative fission yields

| Producer        | Country      | Production       |                  | Average release/day [Bq] | Average release/year [Bq] |
|-----------------|--------------|------------------|------------------|--------------------------|---------------------------|
|                 |              | [%]              |                  |                          |                           |
|                 |              | <sup>99</sup> Mo | <sup>131</sup> I |                          |                           |
| MDS Nordion     | Canada       | 38               | none             | $1.6 \times 10^{13}$     | $6.0 \times 10^{15}$      |
| Tyco Healthcare | Netherlands  | 26               | none             | $2.5 \times 10^{09}$     | $7.3 \times 10^{11}$      |
| IRE             | Belgium      | 16               | 75               | $4.6 \times 10^{12}$     | $1.0 \times 10^{15}$      |
| NTP             | South Africa | 16               | 25               | $1.3 \times 10^{13}$     | $4.1 \times 10^{15}$      |
| Others          | Others       | 4                | minor            | -                        | -                         |

The estimated total global xenon emission incurred in meeting the required <sup>99m</sup>Tc output is of the order of 10 – 30 PBq.

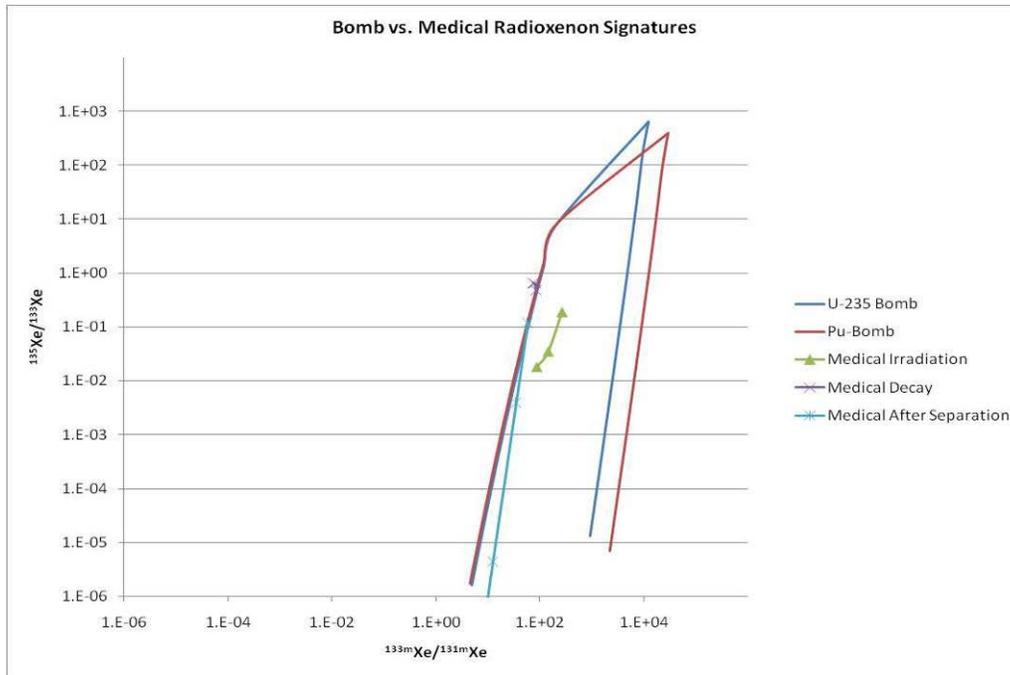
When monitoring for radionon in the atmosphere, it is important to be able to distinguish radionuclide sources by their signatures. The main known sources of radionon to the atmosphere include nuclear explosions, commercial reactors and medical radionuclide facilities.

Isotope ratios are used to discriminate between sources. The <sup>135</sup>Xe/<sup>133</sup>Xe ratio rapidly decreases and can be considered the “clock” of the sample. The <sup>131m</sup>Xe/<sup>133m</sup>Xe ratio allows the distinction, among other criteria, between civil applications and nuclear explosions. Xenon source signatures may thus be described in terms of the isotope ratios <sup>135</sup>Xe/<sup>133</sup>Xe and <sup>131m</sup>Xe/<sup>131m</sup>Xe, with signatures of weapons being defined in the ratio plot in **Figure 4**.



**Figure 4.** Isotopic ratios generated by subsurface nuclear explosions, and comparison with civil application. The right-hand plot shows the “weapons region” in blue and the civil region in green, but emissions from some medical isotope production facilities fall within the orange ellipse, on the “weapons” side of the Kalinowski Discrimination Line (dashed red)

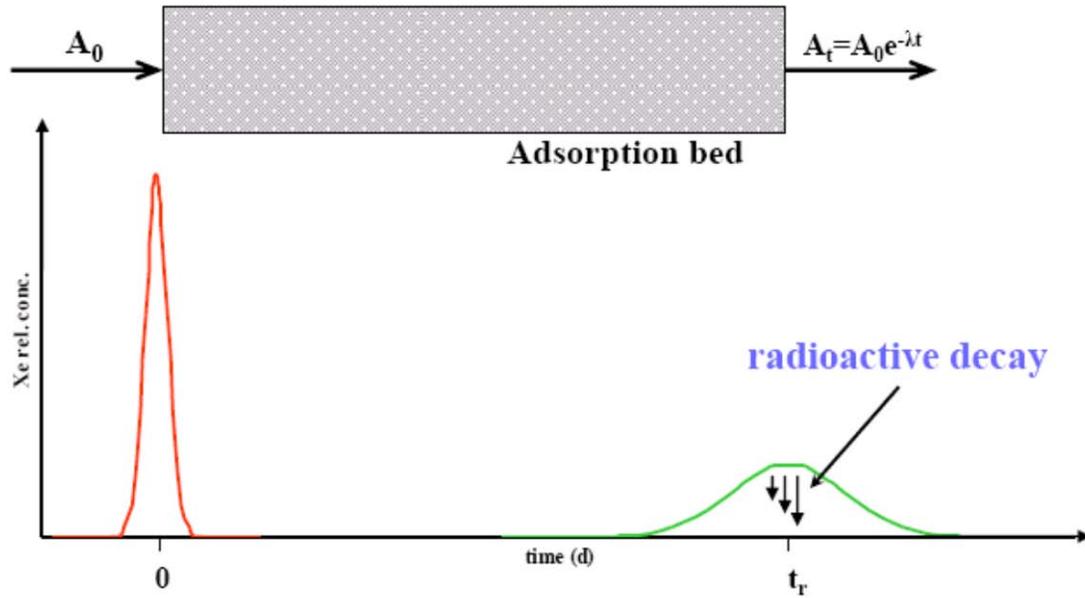
It seems that the distinction between weapon- and non-weapon-related releases is somewhat blurred for medical nuclide production facilities. Measurements appear on either side of the Kalinowski discrimination line, with emissions from medical radio-nuclide HEU-based production facilities sometimes falling within the “weapons zone” of the ratio plots shown in **Figure 5**.



**Figure 5.** Four-isotope xenon plots showing the ratios expected for various processes

Reduction of xenon emissions from the dominating HEU facilities, or at least changing their isotopic signature, is obviously of great importance to proliferation-related environmental monitoring operations. Modeling studies have shown that use of longer irradiation times and the use of emission holding tanks can shift the medical nuclide signature out of the weapons zone in the above plots. Holding tanks would also reduce emissions, as would the use of adsorption beds.

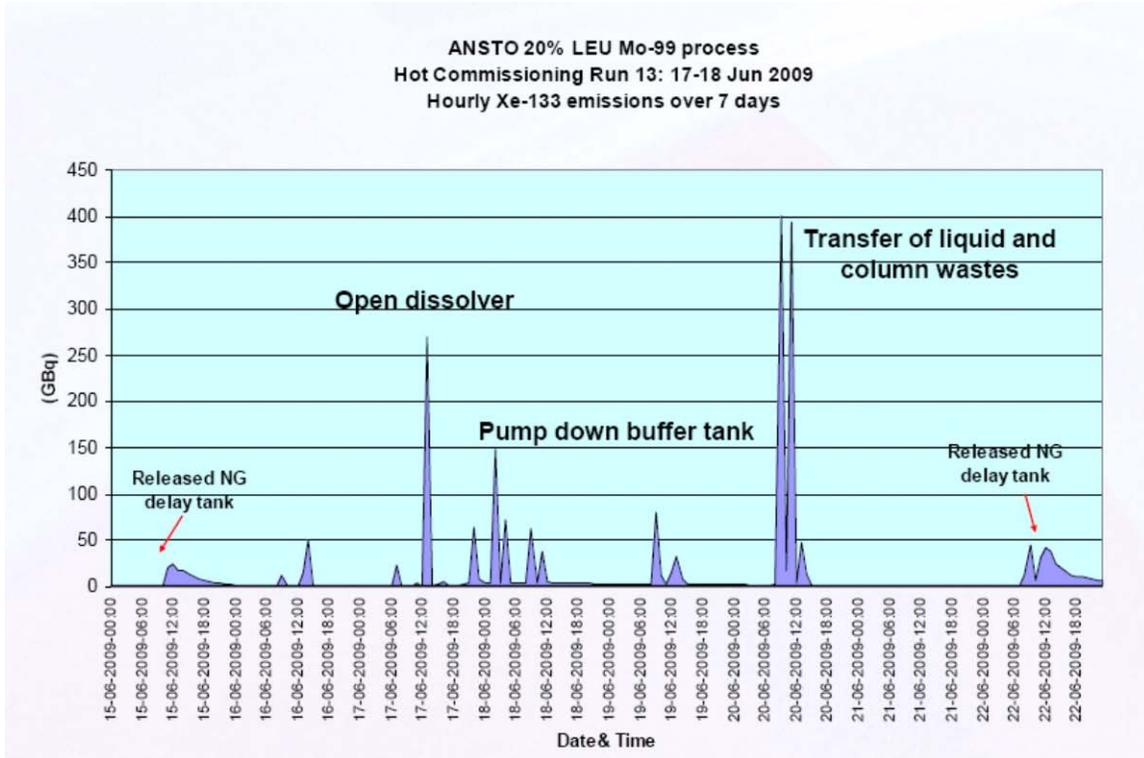
Adsorption beds containing activated carbon (relatively high adsorption coefficient, but impaired by moisture, and with a fire hazard); molecular sieves (lower adsorption coefficient but incombustible and able to be regenerated at high temperatures); and polymers (polycarbonates) could be used to introduce a decay period before gas release, which would lower the activity and also spread the release:



**Figure 6.** The effect of xenon concentration and release time by the addition of an adsorption bed

Other strategies to reduce emissions include reducing demand through longer use of  $^{99m}\text{Tc}$  sources in hospitals, using shorter irradiation times in the reactor, retaining exhaust air, and producing  $^{99}\text{Mo}$  by neutron activation of molybdenum rather than by fission.

Experience at ANSTO during hot commissioning runs with the new Open Pool Australian Lightwater (OPAL) reactor plant (projected annual emission: 280 TBq  $^{133}\text{Xe}$  per year) indicate emissions at various stages of the production cycle as described in the graphic below.



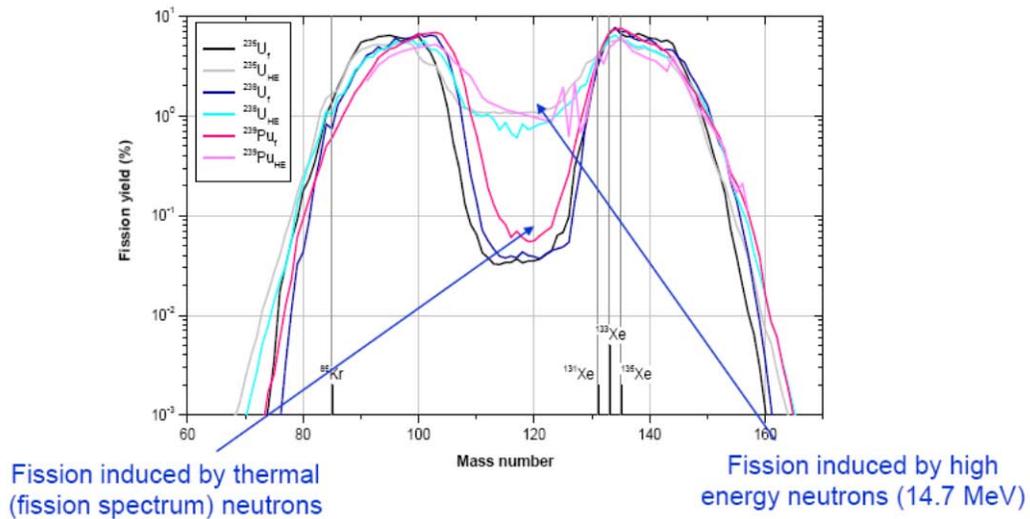
**Figure 7.** Xenon-133 emissions from ANSTO processing with time

Careful attention to releases at the various production stages, combined with use of adsorption columns could reduce emissions from such LEU plants as well.

## 5.0 Nuclear reactors, weapons and radioxenon

During fission of uranium or plutonium in a nuclear reactor, thermal (slow) neutrons are used, whereas during a nuclear explosion the fission is induced by fast neutrons. The full fission sequence in a device is finished within a microsecond. There is little time for complex activation buildup in a nuclear explosion, whereas there is sufficient time for production of many activation products in a nuclear reactor. These differences produce different radionuclide abundances. Since a nuclear blast produces different radionuclide abundances, nuclide ratios may be used for source identification. The ranges of fission products produced in various fission scenarios are depicted in **Figure 8**.

Fission yield in % for several nuclear explosion relevant nuclides  
 (Fission yield is a function of the fissioning nuclide and the incident neutron energy)



**Figure 8.** Fission yield as a percentage for several nuclides relevant to nuclear explosion

Cumulative fission yields of the four most prominent xenon isotopes range from <0.1 to 7.5% as indicated below, where f indicates fast neutron irradiation and he indicates high energy neutrons:

**Table 5.** Summary of statistics from radioxenon monitoring stations

| Fission Product    | Half-life | Time unit | <sup>235</sup> U <sub>f</sub> | <sup>235</sup> U <sub>he</sub> | <sup>238</sup> U <sub>f</sub> | <sup>238</sup> U <sub>he</sub> | <sup>239</sup> Pu <sub>f</sub> | <sup>239</sup> Pu <sub>he</sub> |
|--------------------|-----------|-----------|-------------------------------|--------------------------------|-------------------------------|--------------------------------|--------------------------------|---------------------------------|
| <sup>131m</sup> Xe | 11.934 d  | d         | 0.05                          | 0.06                           | 0.05                          | 0.06                           | 0.05                           | 0.07                            |
| <sup>133m</sup> Xe | 2.19 d    | d         | 0.19                          | 0.29                           | 0.19                          | 0.18                           | 0.24                           | 0.42                            |
| <sup>133</sup> Xe  | 5.243 d   | d         | 6.72                          | 5.53                           | 6.76                          | 6.02                           | 6.97                           | 4.86                            |
| <sup>135</sup> Xe  | 9.14 h    | h         | 6.6                           | 5.67                           | 6.97                          | 5.84                           | 7.54                           | 6.18                            |

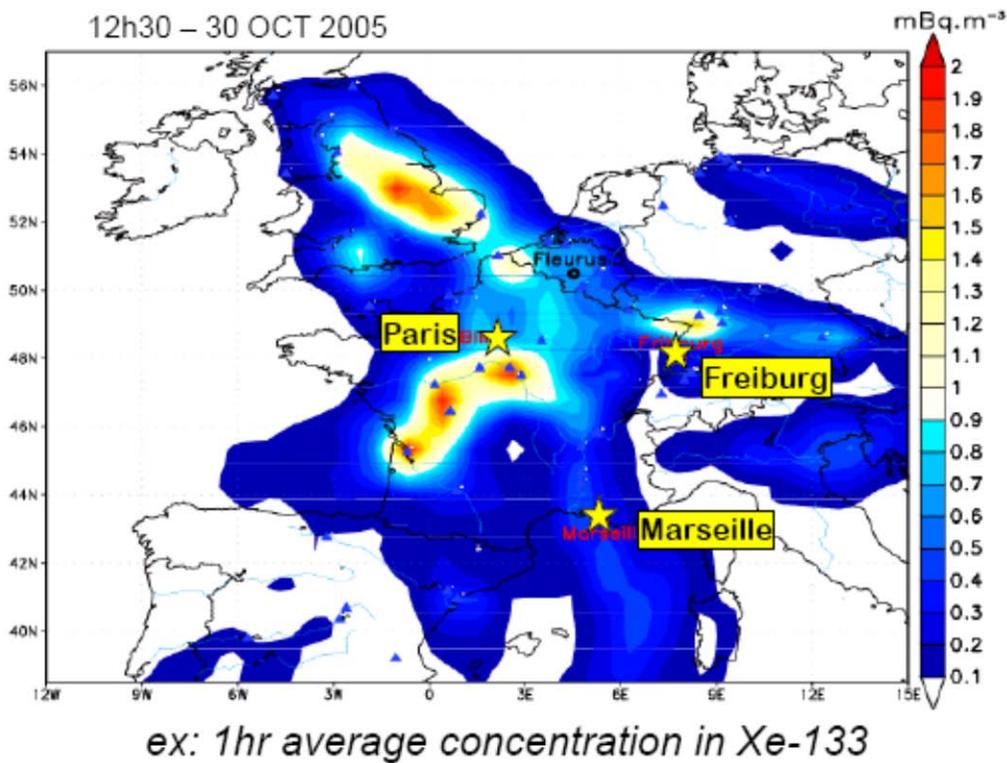
As <sup>133</sup>Xe has high a production rate (fission yield 4.86 – 6.97%) and a reasonably long half-life (5.2 d), it is the isotope most commonly observed in the atmosphere. Its high production rate is a result of a combination of anthropogenic and natural mechanisms. Xenon will be released into the environment by

- Unintentional release due to containment system failure
- Early venting due to the high pressure of the explosion and other dynamic effects pushing gas through cracks and fissures in the bedrock
- Venting due to opening of tunnels for recovery of test material
- Drilling of holes (operational releases)

- The sucking of gases from deposits in the walls of cracks and fissures by low-pressure weather systems.

Depending on the fission material ( $^{235}\text{U}$ ,  $^{233}\text{U}$  or  $^{239}\text{Pu}$ ),  $1.08$  to  $1.33 \times 10^{16}$  Bq are produced in a 1 kiloton nuclear explosion. The most likely scenario for clandestine nuclear weapon tests is that they would be conducted underground. In that case, xenon may be released to the atmosphere through (a) unintentional release due to failure of the containment system, (b) early venting due to the high pressure of the explosion and other dynamic effects pushing gas through cracks and fissures in the bedrock, (c) venting due to opening of tunnels for recovery of test material, (d) drilling of holes (operational releases), and (e) the sucking of gases from deposits in the walls of cracks and fissures by low-pressure weather systems.

Nuclear power plants (NPP), on the other hand, give a continuous  $^{133}\text{Xe}$  release of  $\sim 3$  GBq/reactor/day (which may be contrasted with Chalk River and Fleurus releases up to 17 and 5 TBq/d respectively). There are 131 reactors in Europe: Belgium (7), Germany (19), France (58), Netherlands (1), Spain (9), Switzerland (5), and United Kingdom (32). The result is an almost continuous baseline on the order of  $1 \text{ mBq/m}^3$  as measured in Paris, Freiburg and Marseille. The NPP background levels in Western Europe are depicted graphically below:



**Figure 9.** Background  $^{133}\text{Xe}$  levels from nuclear power plants in Western Europe

## 5.1 Measurements of radioxenon in the atmosphere

The main xenon decay modes that lead to its detection by beta-gamma coincidence systems are the decays of  $^{135}\text{Xe}$  to  $^{135}\text{Cs}$ ,  $^{133}\text{Xe}$  to  $^{133}\text{Cs}$ ,  $^{131\text{m}}\text{Xe}$  to  $^{131}\text{Xe}$  and  $^{133\text{m}}\text{Xe}$  to  $^{133}\text{Xe}$ , as indicated below:

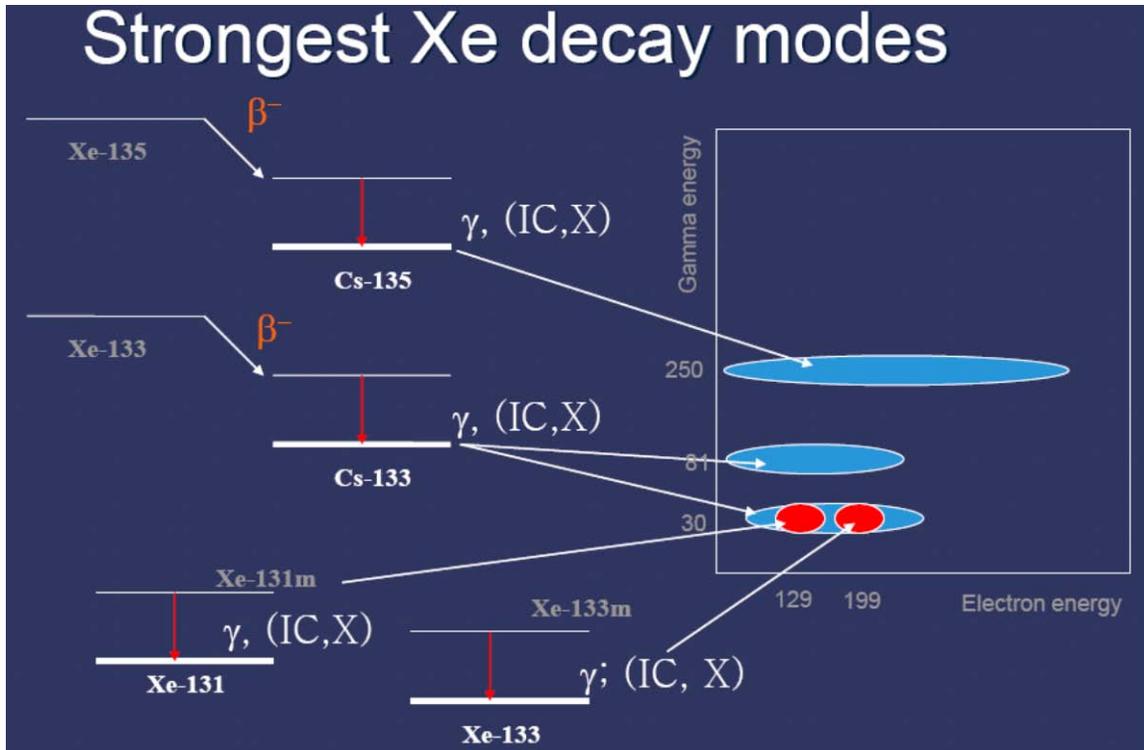


Figure 10. Xenon decay modes

These characteristics are utilized in the CTBT verification system.

## 5.2 The CTBTO International Monitoring System

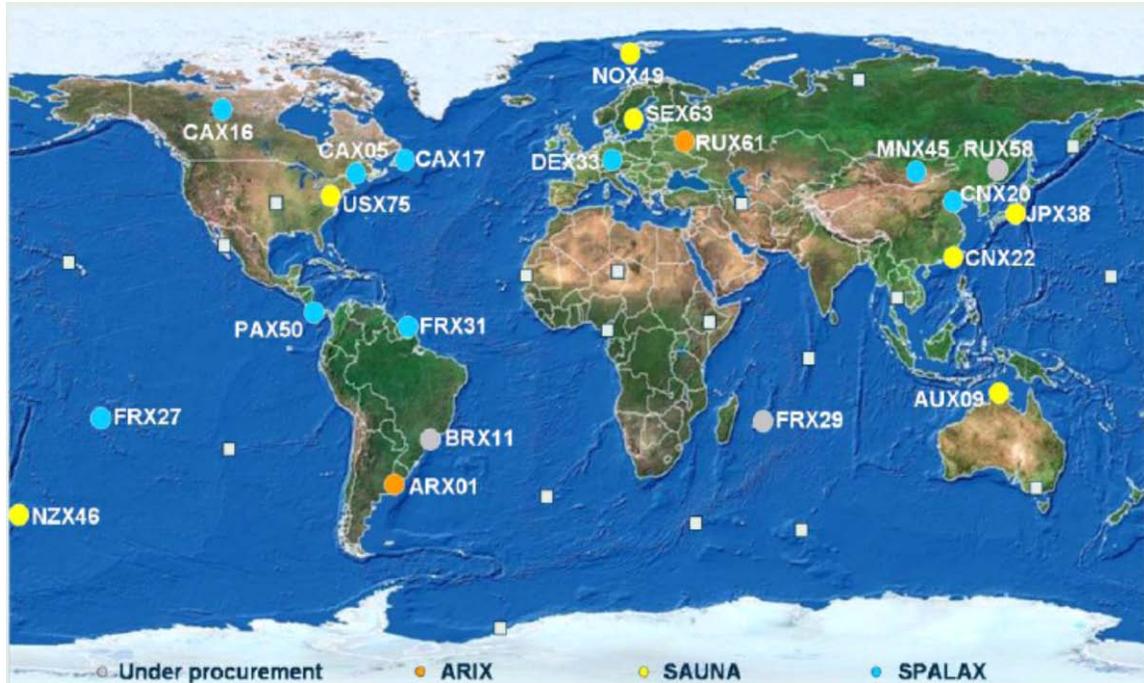
Leakage of xenon from underground test cavities is of prime interest in CTBT compliance verification. The CTBTO Preparatory Commission is currently establishing the CTBT verification regime. As part of this verification regime, the International Monitoring System (IMS) will consist of a network of 321 monitoring stations and 16 radionuclide laboratories. Among these stations, 80 are radionuclide monitoring stations, 40 of which have noble gas capability. Other stations are for seismic, hydro-acoustic or infrasound monitoring. The primary role of the radionuclide monitoring network is to provide unambiguous evidence of a nuclear explosion through the detection and identification of fission products. Among the technologies in IMS, radionuclide monitoring provides “forensic evidence” that the explosion is nuclear in nature. The network is designed to have the capability of 90% detection within approximately 14 days for a 1 kt nuclear explosion in the atmosphere or from venting by an underground or underwater detonation. One design criterion is that the minimum detectable concentration of  $^{133}\text{Xe}$  is to be  $< 1 \text{ mBq/m}^3$ .

Four different measurement systems have been developed:

- SPALAX (France): high-resolution gamma spectra
- ARSA (USA): two-dimensional beta-gamma coincidence spectra
- SAUNA (Sweden): two-dimensional beta-gamma coincidence spectra

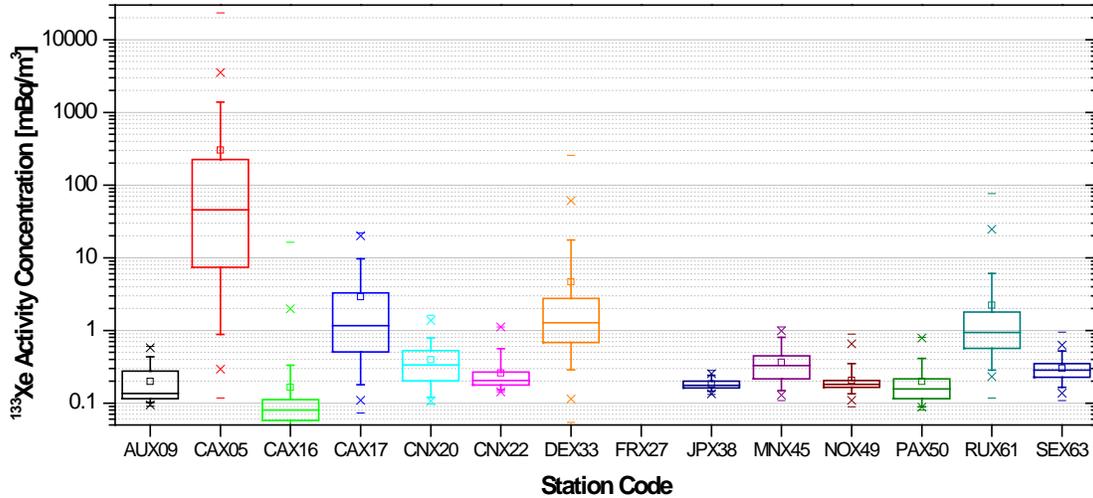
- ARIX (Russian Fed.): two-dimensional beta-gamma coincidence spectra

The monitoring network is shown below, indicating currently installed stations (colored) and those yet to be installed.



**Figure 11.** IMS noble-gas monitoring network

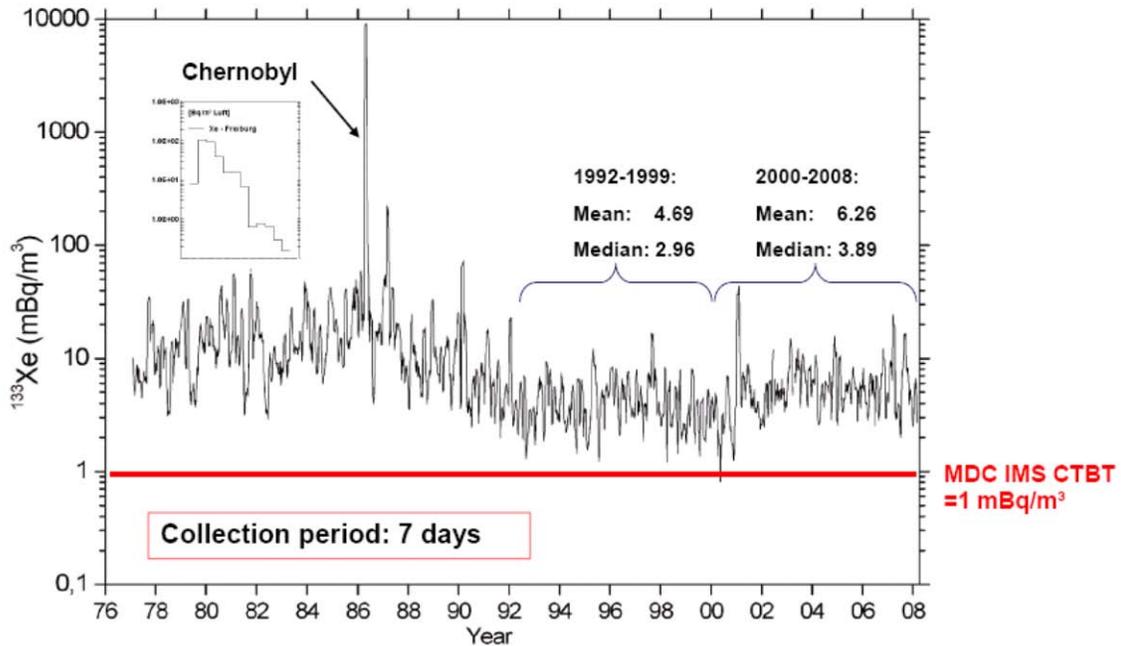
Atmospheric concentrations of  $^{133}\text{Xe}$  in 14 existing international monitoring system (IMS) noble-gas stations are summarized in the figure below, where stations with local influences can be clearly distinguished: stations CAX05 and CAX17 in Canada, influenced by CRL; and DEX33 and RUX61 influenced by local sources. Levels at locally influenced stations range from  $<1$  to  $>1000$   $\text{mBq/m}^3$ , while levels at the others are  $<1$   $\text{mBq/m}^3$ .



**Figure 12.** Atmospheric concentrations of  $^{133}\text{Xe}$  in 14 existing IMS noble-gas stations

A time-series of detections of  $^{133}\text{Xe}$  in Freiburg, Germany, is illustrated below for the period 1976 to 2008. The normal background level of  $\sim 2$  to  $\sim 80$  mBq/m<sup>3</sup> is clear, with a significant spike to 10,000 mBq/m<sup>3</sup> caused by the Chernobyl event.

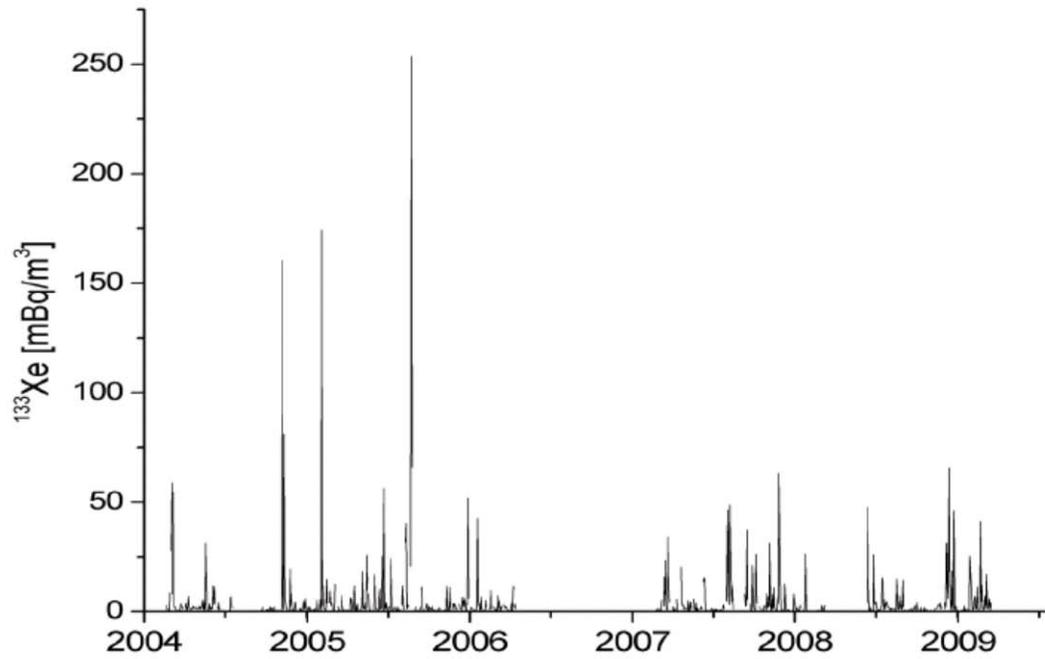
### $^{133}\text{Xe}$ Activity Concentration in Freiburg



**Figure 13.** Xenon-133 activity concentration in Freiburg from 1976-2008

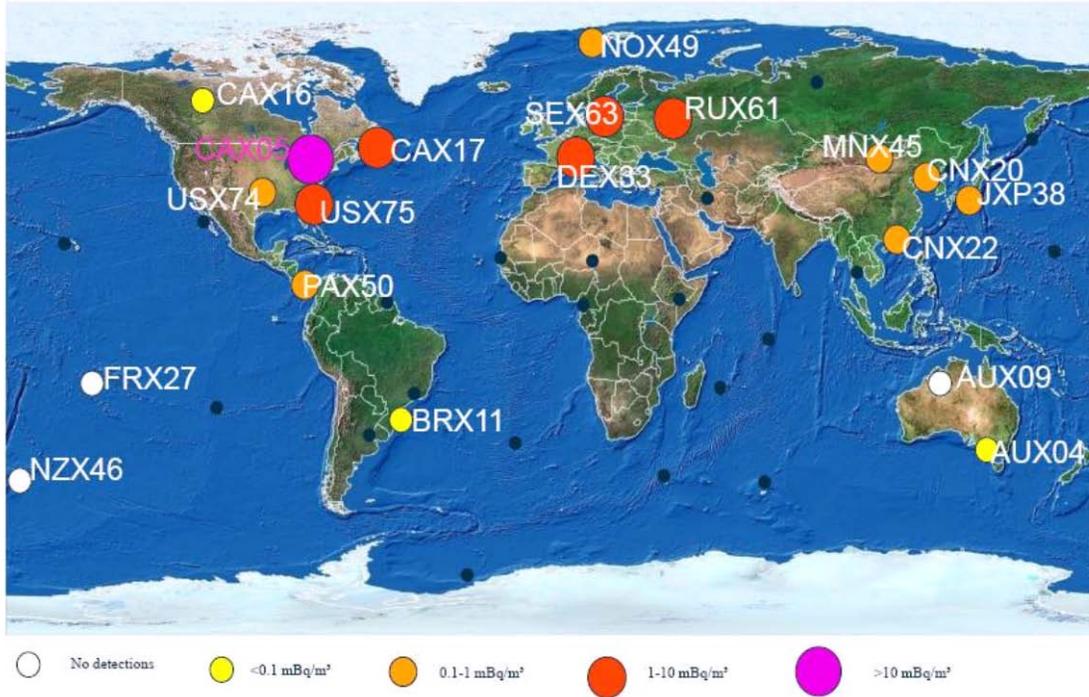
A similar but more detailed time series is shown for the period 2004 – 2009 at the IMS monitoring station, DEX33, located nearby at Schauinsland, **Figure 14.**

## Radioxenon time series at DEX 33



**Figure 14.** Xenon-133 activity concentration from 2004-2009 at DEX 33

The radioxenon detections are summarized graphically in the map below.



**Figure 15.** Map of radioxenon detections

### 5.3 The global radioxenon background

Detection of radioxenon is likely to be the only unambiguous proof of the nuclear nature of an underground explosion, but such an interpretation requires a thorough understanding of background xenon levels and the isotopic ratios arising from different production mechanisms. Over the last 10 to 15 years the new generation of radioxenon detection equipment mentioned above has been developed with 12-24 h time resolution, high sensitivity for  $^{133}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ ,  $^{131\text{m}}\text{Xe}$  and  $^{135}\text{Xe}$  (LC  $\sim 0.1 \text{ mBq/m}^3$ ), and automatic operation. The global deployment of these systems has resulted in a dramatically increased knowledge of the radioxenon background.

Results to date provide the statistics shown in **Table 6** and **Table 7** below.

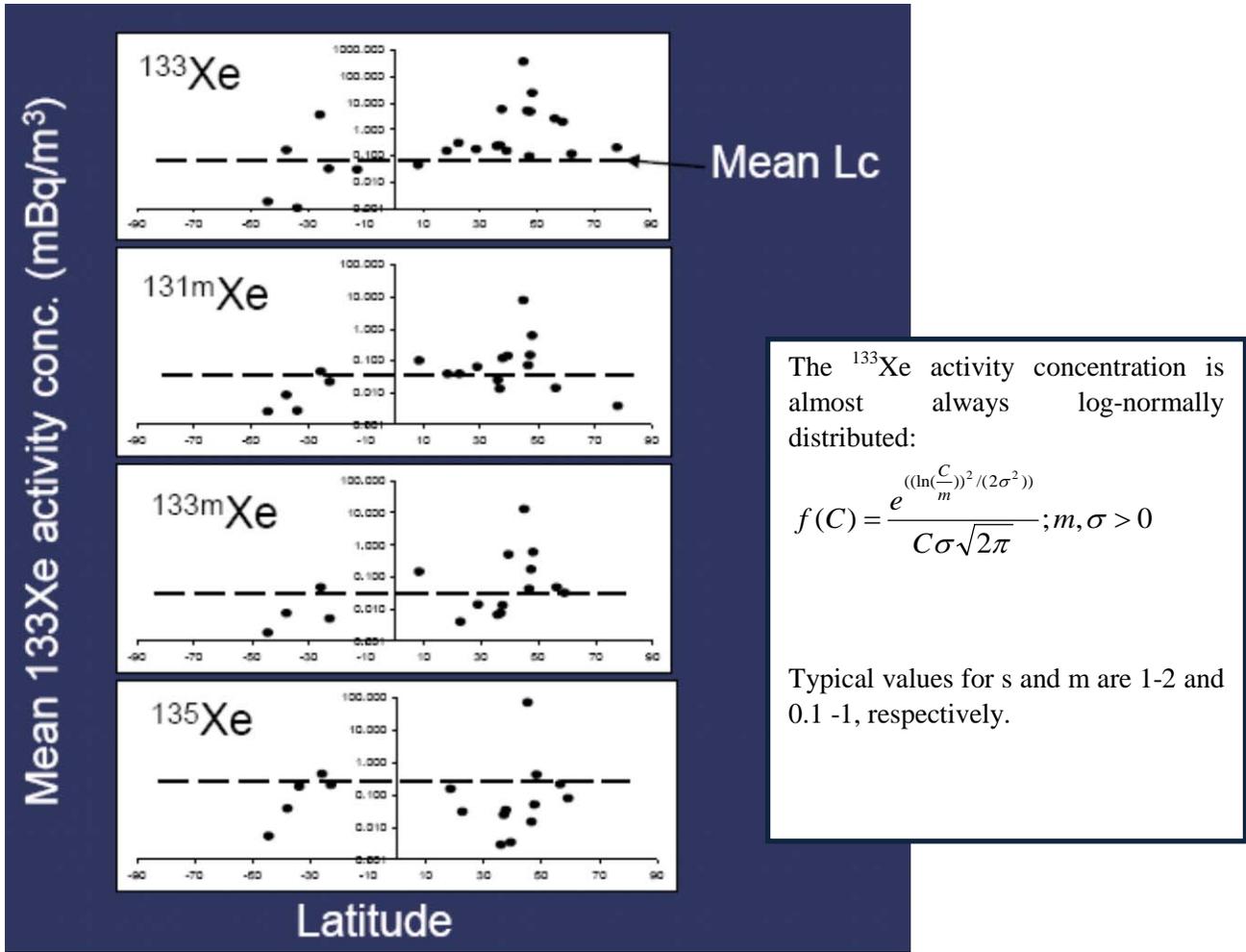
**Table 6.** Measurements of anthropogenic radioxenon and expected order of magnitude releases excluding Northern Hemisphere producers

| <b>Excluding data from stations in Ottawa and Paris</b> |                   |                           |                           |                   |
|---|-------------------|---------------------------|---------------------------|-------------------|
|   | $^{133}\text{Xe}$ | $^{131\text{m}}\text{Xe}$ | $^{133\text{m}}\text{Xe}$ | $^{135}\text{Xe}$ |
| Maximum (mBq/m <sup>3</sup> )                           | 257               | 5.7                       | 16                        | 72                |
| Fraction > 1 mBq/m <sup>3</sup>                         | 18%               | 0.4%                      | 0.7%                      | 0.5%              |

**Table 7.** Measurements of anthropogenic radioxenon and expected order of magnitude releases including Northern Hemisphere producers

| <b>Including Paris and Ottawa (influenced by CRL and Fleurus)</b> |                   |                           |                           |                   |
|---|-------------------|---------------------------|---------------------------|-------------------|
|   | $^{133}\text{Xe}$ | $^{131\text{m}}\text{Xe}$ | $^{133\text{m}}\text{Xe}$ | $^{135}\text{Xe}$ |
| Maximum (mBq/m <sup>3</sup> )                                     | 24500             | 236                       | 817                       | 4590              |
| Fraction > 1 mBq/m <sup>3</sup>                                   | 26%               | 4%                        | 4%                        | 4%                |

As shown in **Figure 16**, there is more xenon in the Northern than Southern Hemisphere.



**Figure 16.** Xenon activity concentration variations with latitude

Monitoring data also indicate the following:

- The majority of  $^{131\text{m}}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ , and  $^{135}\text{Xe}$  detection are at a few sites close to medical radionuclide production facilities
- A few detections of  $^{133\text{m}}\text{Xe}$ , and  $^{135}\text{Xe}$  due to releases from NPPs (shutdown, startup)
- Xenon-133 and  $^{131\text{m}}\text{Xe}$  can be detected almost everywhere
- Several pure  $^{131\text{m}}\text{Xe}$  detections have yet to be explained (“old xenon, hospitals?”)
- There have not been many verified detections from hospital releases

An attempt has been made to construct a global radioxenon map based on calculated yearly averages for 23 existing sites; modeled releases using known source terms to provide estimated averages for the full IMS complement of 80 sites; normalization of concentrations to measurements; and interpolation between stations. The resulting map is shown in **Figure 17** below; the influence of the major medical radionuclide production facilities is clear.

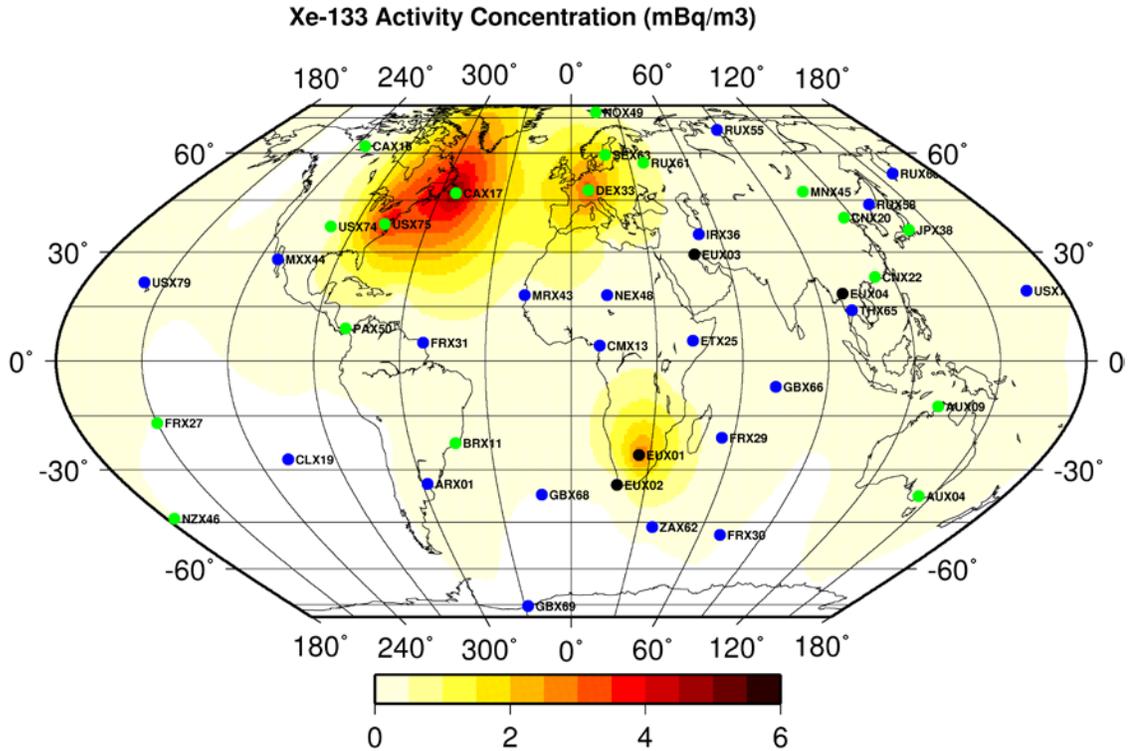


Figure 17. Global background radioxenon map

Anthropogenic radioxenon isotopes detected by the IMS are estimated to have been created mostly in medical radionuclide production facilities or in NPPs, as shown in Table 8:

Table 8. Approximate radioxenon releases from all sources

- The daily IMS noble gas measurements around the globe are influenced from anthropogenic sources, that disturb the signal we look for
- Anthropogenic radioxenon isotopes are created mostly in nuclear power plants or medical isotope production facilities

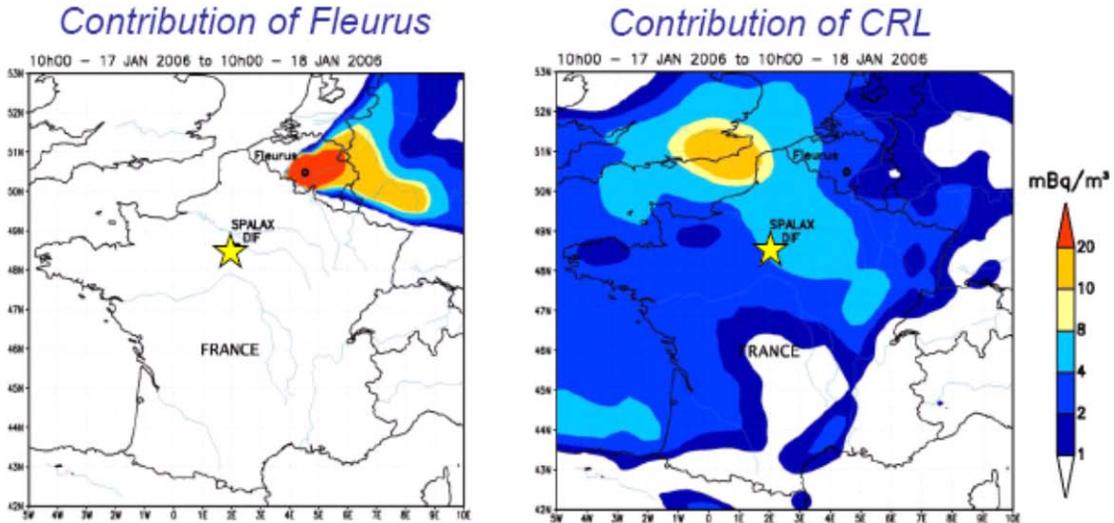
| Type of release                      | Major xenon isotopes released in the atmosphere                     | Typical order of magnitude of radioxenon release |
|--------------------------------------|---|--|
| Hospitals                            | $^{133}\text{Xe}$ and $^{131\text{m}}\text{Xe}$                     | $\sim 10^6$ Bq/d                                 |
| Nuclear Power Plants (NPP)           | $^{133}\text{Xe}$   | $\sim 10^9$ Bq/d                                 |
| Isotope Production Facilities (IPF)  | $^{133}\text{Xe}$ and $^{133\text{m}}\text{Xe}$                     | $\sim 10^{11} - \sim 10^{13}$ Bq/d               |
| 1 kton nuclear explosion underground | $^{133}\text{Xe}$ , $^{135}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ | 0 - $\sim 10^{15}$ Bq                            |
| 1 kton Nuclear explosion atmospheric | $^{133}\text{Xe}$ , $^{135}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ | $\sim 10^{16}$ Bq                                |

- Radiopharmaceutical plants produce 3-4 orders of magnitude higher radioxenon than nuclear power plants and 0-3 orders of magnitude lower than an underground 1 kiloton nuclear explosion
- Information of radiopharmaceutical production is crucial and therefore any effort to reach a comprehensive understanding of this major sources is very welcome (-> Tuma and Kalinowski)

On a daily basis, radiopharmaceutical plants produce two to four orders of magnitude more radioxenon than NPPs, and have daily releases several orders of magnitude lower than that produced in a 1 kt nuclear explosion.

## 5.4 Influence of medical radio-nuclide production facilities in Europe and beyond

The contributions of Fleurus and Chalk River to background  $^{133}\text{Xe}$  levels in Western Europe are highlighted for 17 and 18 January 2008 in **Figure 18** below:



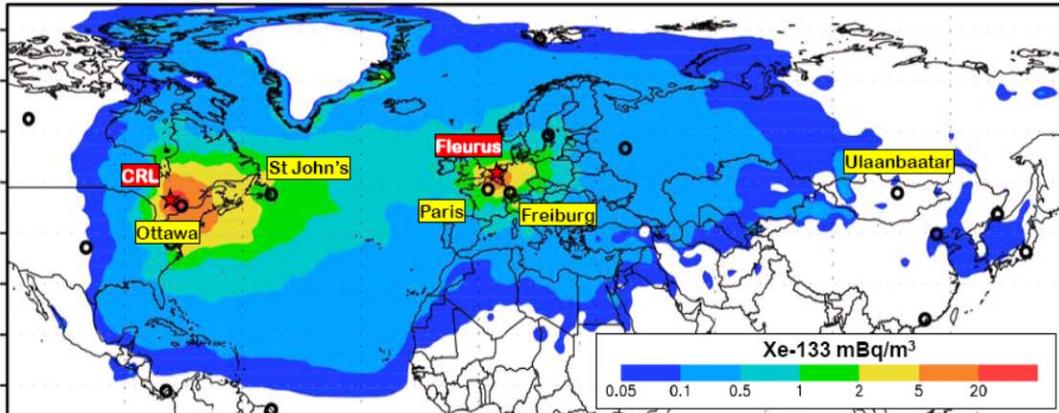
**Figure 18.** Contributions of Fleurus and Chalk River to background  $^{133}\text{Xe}$  levels in Western Europe

While in Western Europe NPPs generate a baseline of the order of a  $\sim 1$  to a few  $\text{mBq}/\text{m}^3$ ; CRL leads to average contribution similar to that of NPPs, but can lead to peaks up to a few tens of  $\text{mBq}/\text{m}^3$ ; Fleurus leads to major peaks of the order of tens to a few hundreds of  $\text{mBq}/\text{m}^3$  in Paris and Freiburg, and tens of  $\text{mBq}/\text{m}^3$  in Stockholm. Further east, contributions of CRL and Fleurus seem to be very weak (as measured in Mongolia and China). These effects are indicated graphically in **Figure 19**.



## Impact of Fleurus and CRL releases on European detections during 2008

- 2008 annual average concentration in Xe-133: sum of the contributions of CRL and Fleurus (including Fleurus shutdown)



- Because of the prevailing westerly wind direction, stations Ottawa, St John's, Paris and Freiburg are highly impacted by isotope production plants
- Weak influence in Asia (ex: Ulaanbaatar)

**Figure 19.** Impact of Fleurus and CRL releases on European detections in 2008

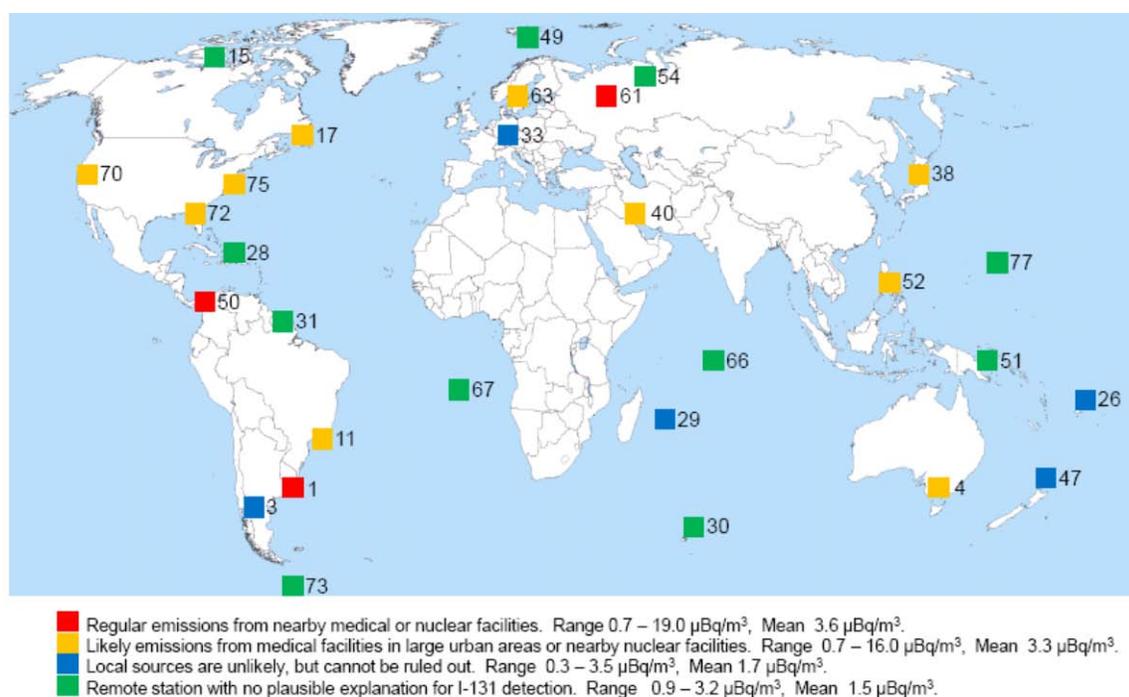
## 5.5 Detections of other radionuclides

The CTBT verification monitoring regime includes particulate radionuclides as well as noble gases. The particulate monitoring network is very sensitive, with a daily sampling of at least 12,000 m<sup>3</sup> of air, and a specified detection limit for <sup>140</sup>Ba of 30 μBq/m<sup>3</sup>. This sensitivity results in many detections of CTBT-relevant particulate nuclides each year. During 2008, for example, there were over 900 such detections reported by the International Data Centre. A wide range of nuclides has been reported in recent years including:

- <sup>76</sup>As
- <sup>155</sup>Eu
- <sup>86</sup>Rb
- <sup>91</sup>Y
- <sup>198</sup>Au
- <sup>131</sup>I
- <sup>103</sup>Ru
- <sup>65</sup>Zn
- <sup>115m</sup>Cd
- <sup>133</sup>I
- <sup>120</sup>Sb
- <sup>69m</sup>Zn
- <sup>144</sup>Ce
- <sup>42</sup>K
- <sup>122</sup>Sb
- <sup>95</sup>Zr
- <sup>58</sup>Co
- <sup>54</sup>Mn
- <sup>124</sup>Sb
- <sup>97</sup>Zr
- <sup>60</sup>Co
- <sup>24</sup>Na
- <sup>153</sup>Sm
- <sup>51</sup>Cr
- <sup>95</sup>Nb
- <sup>99m</sup>Tc
- <sup>136</sup>Cs
- <sup>112</sup>Pd
- <sup>129m</sup>Te
- <sup>137</sup>Cs
- <sup>84</sup>Rb
- <sup>88</sup>Y

Medical radionuclides are recognizable in this list, although some reported detections would inevitably have been false alarms generated through either spurious nuclide identification or false peak detection. The principle nuclides detected (>1% of detections) during 2008, when 55 of the projected 80 stations worldwide were in operation, were  $^{24}\text{Na}$  (46.9%),  $^{60}\text{Co}$  (17.8%),  $^{137}\text{Cs}$  (13.8%),  $^{131}\text{I}$  (6.0%),  $^{99\text{m}}\text{Tc}$  (4.7%),  $^{122}\text{Sb}$  (2.1%),  $^{88}\text{Y}$  (1.3%) and  $^{91}\text{Y}$  (1.3%). In addition, there were more detections at levels deemed “normal” for the station, and which were therefore not reported as being “anomalous;” this particularly applied to the commonly detected  $^{24}\text{Na}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$  and  $^{99\text{m}}\text{Tc}$ .

Iodine-131 is of particular interest for CTBT verification because as a particulate nuclide in the atmosphere it is easily captured and detected and, because of its volatility, it is released from underground as well as above-ground explosions. It is also, however, also a commonly used medical radionuclide so, like the xenon isotopes, it is widely detected by the IMS. Detections during the period mid 2005 to mid-2009 are shown graphically **Figure 20**.



**Figure 20.** Iodine-131 detections from 2005-2009

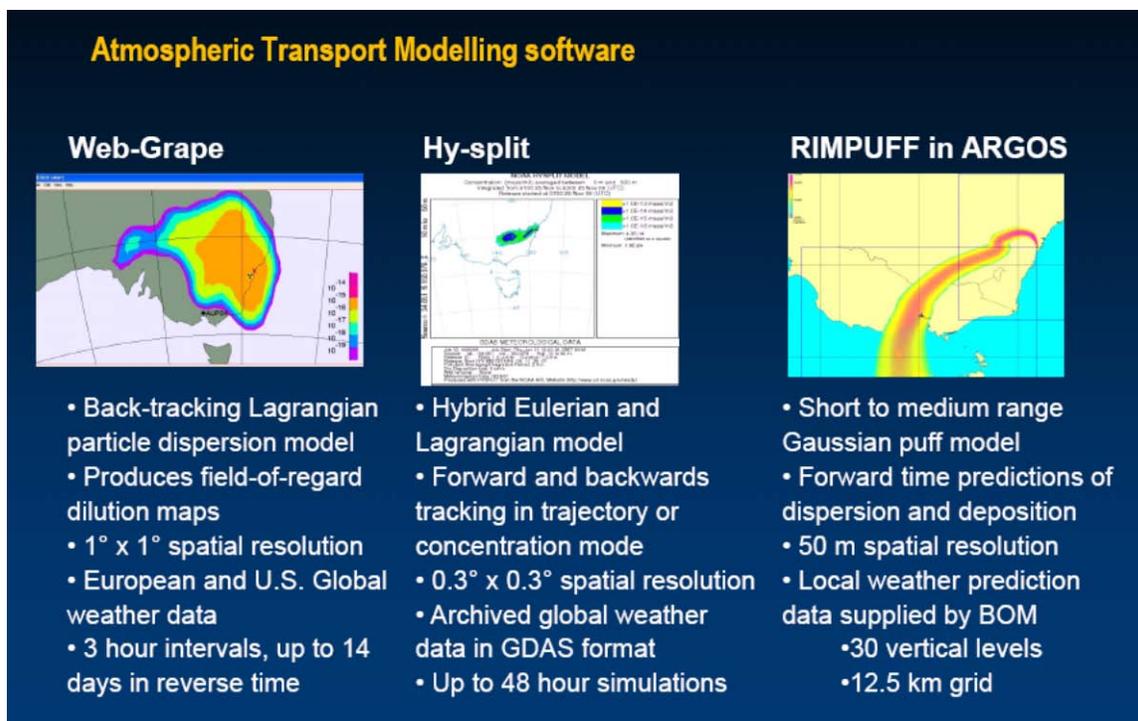
As with the xenon isotopes, an understanding of the apparent  $^{131}\text{I}$  has to be developed in order for unambiguous source attribution to be possible. The same applies to  $^{99\text{m}}\text{Tc}$ , although with its shorter half-life, it is less widespread, and the important determination is whether or not it is supported in the atmosphere by its  $^{99}\text{Mo}$  parent.

## 5.6 Atmospheric backtracking

Backtracking is clearly important in applying atmospheric radioactivity monitoring in treaty verification programs because while waveform (seismic, hydro-acoustic and infrasonic) technologies can geo-locate sources by relatively straightforward triangulation, the situation is much more complex for radionuclide

monitoring because the medium that carries the signals, Earth's atmosphere, is forever moving. Geolocation of radionuclide signals is vital, however, for fusing data from the various technologies in order to provide unambiguous source attribution and event characterization. An impressive range of tools for the application of atmospheric transport modeling in backtracking from detection to source has been developed in recent years, typified by the Web-Grape program developed by the CTBTO.

An example of such application is the recent attribution of xenon detections in Melbourne, Australia, to the ANSTO  $^{99}\text{Mo}$  facility during hot commissioning runs at the facility. Three separate modeling approaches were adopted, as illustrated below in **Figure 21**.



**Figure 21.** Three available atmospheric transport modeling software packages used to attribute detections in Melbourne to ANSTO

In this case, all backtracking methods were applied together with isotopic ratios to pinpoint the ANSTO source.

## 6.0 Conclusion

Medical radionuclides make contributions of inestimable value to medical practice, with applications in the majority of diagnostic procedures and also in therapy. The expanding range of biomolecules able to transport attached radionuclides to sites within the body without disrupting metabolic processes will enable further expansion of this exciting field of medicine. Along with this growing demand there is obviously a need for increased capacity for production and new technologies are being developed and applied worldwide. Most diagnostic procedures still rely on  $^{99\text{m}}\text{Tc}$ , however, and demand for this is increasing at  $\sim 5\%$  per year. Employment of HEU targets in reactors is currently the favored method of

production and 95% of the necessary  $^{99}\text{Mo}$  parent is produced by four major suppliers. This causes a fragility of supply and a crisis is looming with the announced plan to close the Chalk River facility which produces ~40% of the world supply. There is a movement away from HEU towards use of LEU and other technologies, and the IAEA is encouraging this through research projects aimed at ensuring regional self sufficiency. The number of medical radio-nuclide production facilities is increasing rapidly in response to these pressures.

Coincident with this growing demand and rate of production is a growing concern for nuclear security and proliferation. Accordingly, new treaties such as the CTBT have been opened for signature, and treaty compliance-verification monitoring is gaining momentum. Of particular concern in this regard are radioxenon emissions from nuclide production facilities. Indeed, existence of a global  $^{133}\text{Xe}$  background is largely due to the  $^{99}\text{Mo}$  production facilities. Yet the radioxenon is a highly sensitive tracer for detecting nuclear explosions, even underground explosions where only small fractions of the nuclear debris may be released into the environment. The four isotopes  $^{131\text{m}}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ ,  $^{133}\text{Xe}$  and  $^{135}\text{Xe}$  provide a means of distinguishing between civil and military emission sources through plots of isotopic ratios.

Emissions from nuclide-production facilities are variable and well below regulatory limits, but they are still regularly detected in the global IMS associated with CTBT verification, and high enough to complicate the interpretation and attribution of signals observed in monitoring networks. The CTBT radioxenon network currently under installation is highly sensitive with detection limits around  $0.1 \text{ mBq/m}^3$  and, depending on transport conditions and background, able to detect civil release signatures from sites thousands of kilometers away. Data from the IMS, coupled with sophisticated atmospheric transport modeling and backtracking capability has made great progress in understanding sources and background levels of radioxenon world-wide. Attention is focused on methods for distinguishing these civil signatures from nuclear-weapon-significant detections. Currently plots of isotope ratios  $^{133\text{m}}\text{Xe}/^{131\text{m}}\text{Xe}$  versus  $^{135}\text{Xe}/^{133}\text{Xe}$  are used to distinguish between most civil application and military sources. Several sampling campaigns have been shown to be consistent with atmospheric transport and production models and show that under the current conditions, effluents from nuclide-production facilities interfere with the current capability of nuclide detection systems, such as those in the IMS, targeted at the detection of nuclear explosions.

Signals from the  $^{99}\text{Mo}$  production facilities are ambiguous in that the current screening techniques do not successfully distinguish them from military applications. There are still many open questions and a need for information on the characteristics of major radioxenon sources and the effects of particular production pathways on xenon emissions. Techniques could be employed for altering the  $^{99}\text{Mo}$  signature, such as increasing irradiation times and introducing delay systems for gaseous emissions. Modeling expertise developed for treaty-verification purposes have been useful in indicating possibilities for this mitigation. Continued cooperation between the environmental monitoring and radiopharmaceutical production communities, with continued exchange of information, will improve this signature recognition dilemma.

Particulate radioactivity monitoring is also a major component of the IMS and there too medical radionuclides are included in the wide range of radionuclides detected, notably  $^{131}\text{I}$  and  $^{99\text{m}}\text{Tc}$ . More in-depth understanding of the sources of these nuclides and their behavior in the environment are also needed.

To date there has been a void in the communication and appreciation of problems between the environmental monitoring and nuclide production communities, and the WOSMIP workshop has opened

communication pathways. The monitoring community is gaining a better understanding of the complexities of the processes at nuclide production facilities, and the production community is gaining a better understanding of the impact their operations have on the monitoring systems and their goal of nuclear security improvement. Further collaboration and discussions between the monitoring and production communities are needed to continue the exchange of information and encourage advances in trapping technology and understanding of detections in monitoring systems. Such initiatives will help in addressing the dichotomy which exists between expanding production and improving monitoring sensitivity, with the ultimate aim of enabling unambiguous distinction between different nuclide signatures.

## Appendix A: WOSMIP Agenda



**WOSMIP2009** • Workshop on Signatures of Medical and  
Industrial Isotope Production • Pacific Northwest National Laboratory

**Castello di Strassoldo di Sopa • Udine, Friuli-Venezia Giulia • Italy**  
July 1-3, 2009

### Final Agenda

*Tuesday*, June 30, 2009 • Castello di Strassoldo

|       |                                       |  |
|-------|---------------------------------------|--|
| 1530h | Shuttle transfer from hotel to castle |  |
| 1600h | Registration and Reception            | Gabriella di Strassoldo, <i>Italy</i><br>Laura Wilhelm, <i>USA</i> |
| 2200h | Shuttle transfer from castle to hotel |  |

*Wednesday*, July 1, 2009 • Castello di Strassoldo

|   |  |   |
|---|--|---|
| 830h  | Shuttle transfer from hotel to castle                              |   |
| <b>INTRODUCTORY SESSION – CHAIR: TED BOWYER</b>                 |  |   |
| 900h  | Welcome  | Paul Saey<br><i>Vienna University of Technology, Austria</i>    |
|   | Introductions and Overview of WOSMIP                               | Ted Bowyer<br><i>Pacific Northwest National Laboratory, USA</i> |
| <b>OVERVIEW OF GLOBAL PRODUCTION – CHAIR: SURESH SRIVASTAVA</b> |  |   |
| 910h  | <b>KEYNOTE</b> – Overview of Mo-99 Production Throughout the World | George Vandegrift,<br><i>Argonne National Laboratory, USA</i>   |

|  |   |  |
|--|---|--|
| 950h   | <b>KEYNOTE</b> – Production and Use of Radioisotopes<br>and Their Influence on Environmental Radioactivity Monitoring | Natsean Ramamoorthy,<br><i>International Atomic Energy Agency,<br/>Austria</i>   |
| 1030h  | Use of Medical Isotopes   | Cathy Cutler,<br><i>University of Missouri, USA</i>                              |
| 1100h  | <b>Break</b>  |  |
| <b>PRODUCTION FACILITIES – CHAIR: JUDAH FRIESE</b>       |   |  |
| 1120h  | Production of Medical and Industrial Isotopes in the BR-2 High Flux Reactor   | Bernard Ponsard<br><i>BNRC, Belgium</i>  |
| 1150h  | Potential of Medium Flux Reactors to Produce Radionuclides for Therapy – Polish Experience                            | Renata Mikolajczak,<br><i>Radioisotope Centre POLATOM, Poland</i>                |
| 1220h  | Medical Isotopes Production on Electron Accelerators  | Yu.M. Tsipenyuk<br><i>PL Kapitza Institute for Physical Problems,<br/>Russia</i> |
| 1250h  | <b>Lunch</b>  |  |
| 1420h  | Radium Institute Experience in Medical Isotopes Production and Application  | LM Solin<br><i>VG Khlopin Radium Institute, Russia</i>                           |
| 1450h  | Production of Isotopes in the United States   | Darrell Fisher<br><i>Pacific Northwest National Laboratory, USA</i>              |
| 1520h  | Update of Chalk River Facility  | George Dolinar<br><i>Atomic Energy of Canada Limited, Canada</i>                 |
| 1550h  | <b>Break</b>  |  |
| <b>ISOTOPE RELEASES AND SCRUBBING – CHAIR: PAUL SAEY</b> |   |  |
| 1620h  | Safeguards Environmental Sampling Methods to Detect Mo-99 Production Signatures                                       | Randa Higgy<br><i>International Atomic Energy Agency,<br/>Austria</i>            |
| 1640h  | By-products in the Production of $^{18}\text{F}$ and $^{11}\text{C}$ with a GE PETtrace Cyclotron                     | Irene Schraick<br><i>Australian Research Council, Austria</i>                    |
| 1710h  | Noble Gas Emissions into the Atmosphere of a Fission Radioisotope Plant   | Eduardo Carranza   |

*Comision Nacional de Energia Atomica,  
Argentina*

1740h End of July 1

***EVENING SOCIAL EVENT***

1810H Walking tour of nearby Castello di Sotto and Park

*Gabriella di Strassoldo*

*Castello di Strassoldo, Italy*

1910h Drive or walk (10 min.) to local rural restaurant; typical food and wine tasting; shuttle transfer to hotel after dinner

## Distribution

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