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WOSMIP III - Workshop on Signatures of Medical and Industrial Isotope Production

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June 19 – 22, 2012

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Summary

Radioactive xenon isotopes are produced by uranium fission in nuclear reactors and in nuclear-weapon explosions. Four xenon isotopes are produced in high yield and have sufficiently long half-lives to be useful in environmental monitoring associated with verification of compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT): ^{131m}Xe, ^{133m}Xe, ¹³³Xe and ¹³⁵Xe. Xenon emissions from medical isotope production facilities are a major source of the global atmospheric xenon background, and may confuse monitoring results by reducing the clarity of source attribution. Comparing the activity ratios of ¹³⁵Xe/¹³³Xe and ^{133m}Xe/^{131m}Xe provides one possible means of discriminating detections of nuclear explosions from detections of radioxenon from other sources.

Technetium-99m is the most common isotope in the radiopharmaceuticals used in tens of millions of medical imaging procedures each year. Its precursor, ⁹⁹Mo, is most often produced through the fission of ²³⁵U in a reactor. Currently, the majority of the world's supply of ⁹⁹Mo is provided by five major producers, though aging facilities could cause shortfalls in supply from 2017 onwards. The fragility of supply has raised the demand for independence of supply in many countries, and new isotope production facilities are planned to be commissioned across the globe in the next decade. Some new producers are considering alternate ⁹⁹Mo production technologies, while many existing producers are also switching to reactors that use low-enriched uranium instead of highly enriched uranium.

Global environmental monitoring associated with the CTBT relies on the detection of both noble gasses and particulate radionuclides, inter alia. The sensitivity of the monitoring systems required for this mission is inevitably impaired by the xenon background arising from medical isotope production. Monitoring stations are proven to be sensitive to xenon emissions from production facilities, even over distances exceeding thousands of kilometers.

While discrimination between radioxenon sources through analytic methods (e.g. activity ratio comparison) is possible, the long-term solution probably depends on emission abatement and cleaner isotope-production technologies. Research into methods of reducing xenon emissions from isotope production facilities continues to build on previous work, with a particular focus on off-gas adsorber systems (activated charcoal and zeolites). New technologies, including homogeneous reactor systems with non-fission neutron sources, neutron activation, and accelerator systems, also hold promise for ⁹⁹Mo production with lower xenon emissions.

Acronyms and Abbreviations

ANSTO	Australian Nuclear Science and Technology Organisation
CTBT	Comprehensive Nuclear-Test-Ban Treaty
ENEA	Italian National Agency for New Technologies, Energy and Sustainable Economic Development
HEU	Highly enriched uranium
IMS	International Monitoring System
IPF	Isotope production facility
IRE	National Institute for Radioelements, Belgium
kt	kiloton
LEU	Low-enriched uranium
LINAC	Linear particle accelerator
NaI	sodium iodide
NPP	Nuclear power plant
NTP	NTP Radioisotopes Ltd, South Africa
Pu	Plutonium
SPALAX	French radioxenon monitoring system
SZ	Silver-exchanged zeolites
U	Uranium

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1.0 Introduction

Medical radioisotopes are well established as essential tools in diagnosis and treatment of an increasingly wide range of illnesses and conditions. As their range of application increases, worldwide demand is steadily rising, too. The supply of the most heavily used medical isotope, ^{99m}Tc, is limited by the small number of major isotope production facilities, though. About 90% of world demand for ^{99m}Tc comes from five producers. Furthermore, infrastructure associated with these facilities is aging and breakdowns are common. Coupled with this, an increased global concern for nuclear security has led not only to the conversion of reactors that use highly enriched uranium (HEU) to using low-enriched uranium (LEU), but also to the development of new technologies altogether. Many countries have begun pushing to develop their independence of supply.

An inevitable product of fission-based medical isotope production is the emission of radioactive xenon. As such, medical isotope production facilities have become a major contributor to a rise in the global background of radioxenon in the atmosphere. However, this conflicts with a growing need for environmental monitoring for nuclear nonproliferation purposes. In particular, verifying compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT) requires a global monitoring system capable of unequivocally determining whether detected events are nuclear in nature, and of attributing them to a particular source. Higher radioxenon background levels impede monitoring associated with the CTBT.

Recognizing the complexities in the supply and demand of medical isotopes, as well as the need for high quality data from sensitive environmental monitoring systems, three Workshops on the Signatures of Medical Isotope Production (WOSMIP) have been convened to consider solutions to this conflict.

The first meeting, WOSMIP I, July 2009, brought together scientists from the "producing" and "monitoring" communities in order to raise awareness within each group of the needs and concerns of the other. The meeting was successful in this, and also generated a common interest in the issues facing radiopharmaceutical production and global monitoring generally.

The second meeting, WOSMIP II, July 2011, continued the discussions with an increasingly technical focus on methods to reduce emissions from isotope production facilities. Research and development carried out since WOSMIP I in emission reduction, production techniques and environmental monitoring was considered, together with the impact of the Fukushima nuclear event on monitoring operations.

The third meeting, WOSMIP III, July 2012, is the subject of this report. The emphasis for WOSMIP III continued to be on a combination of emission abatement approaches, whether through modifications to

existing technologies or through the introduction of new technologies, with improvements in monitoring technologies and means of discriminating among civilian and military xenon sources. While round-table discussions occupied greater time than they had in previous meetings, a wide range of information was successfully presented in oral and poster presentations. These presentations are summarized herein.

The material included in this report was drawn from the thirty oral presentations at WOSMIP III. They have been condensed into a selection of figures and key points to illustrate what was considered to be the most relevant and instructive information. Unavoidably, some presentations have been highlighted more than others, but authorship for the report belongs to all presenters, because they all contributed to the success of the meeting. This report is intended to stimulate ongoing work while also, together with the reports of the earlier meetings, to maintain the threads of information flow and personal contacts, which have been growing since the first day of WOSMIP I. There is still much work to do; all problems have not been solved; the production/monitoring "conflict" remains to a large degree; but understanding is increasing, new technologies look promising, and improvements are being made. All these bode well for the combined futures of isotope production and environmental monitoring.

2.0 Introductory physics of Xe production and source attribution

Radioxenon, or the radioactive isotopes of the noble gas xenon (i.e. ^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe), is produced in all uranium fission reactions, whether in nuclear weapon explosions or in civilian nuclear reactors. As such, all sources of radioxenon, no matter how small, may be of interest for the detection of clandestine nuclear weapon tests. But, growing demand for radiopharmaceuticals, particularly ^{99m}Tc, has required continuous processing of uranium fission byproducts, which has resulted in releases of the associated radioxenon in quantities that can complicate explosion detection. The importance of xenon in both contexts brings two disparate groups of specialists in these fields together.

The first section of this report provides introductory material concerning the physics of radioxenon production. Processes occurring in nuclear-weapon explosions are depicted in Figure 2.1.





As indicated in Figure 2.1, approximately two-thirds of the radiation energy of a nuclear explosion is released residually, that is, in the form of delayed radiation from fission-product decay. This provides the basis for event detection using airborne radionuclide detection technologies, such as the detection of radioxenon isotopes through air sampling.

Two differences in how fission occurs in weapons explosions and in reactors distinguish the process and quantities in which radioxenon isotopes are produced. In a nuclear reactor, *thermal (slow) neutrons* are

involved in a steady fission reaction maintained for *long periods* ranging from days to months. In contrast, fission in a nuclear weapon involves *fast neutrons*, and the full fission sequence is completed *quickly*, on the order of microseconds. These differences change the time during which neutron activation reactions, which both produce and consume some radionuclides, can take place. For example, in a reactor, ¹³⁵Xe produced from U fission acts as a neutron absorber, or "poison," thereby slowing down and even stopping a chain reaction. This is because ¹³⁵Xe has a high cross-section for thermal neutrons $(2.65 \times 10^6 \text{ barns})$, through the absorption of which ¹³⁵Xe becomes stable ¹³⁶Xe. The shorter time during which ¹³⁵Xe can undergo neutron activation during a nuclear explosion, then, means that different nuclide isotope ratios will result from each of the two processes. Knowledge about these "signatures" of the two processes is applied to methods that distinguish among sources.

Fission yield curves (Figure 2.2) indicate the relative abundance of fission products in terms of atomic mass. Xenon isotopes with atomic mass between 131 and 135 are situated in the high-yield portion of such curves. Cumulative fission yields for the four xenon isotopes ^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe are shown in Table 2.1. "Cumulative yield" denotes the inclusion of xenon produced through decay of other fission products, as well as quantities produced directly by fission: ¹³¹Xe, for example, is produced both by fission and through the decay of another fission product, ¹³⁵Te.

Fission Product	Half-life	Time unit	²³⁵ U _f	²³⁵ U _{he}	²³⁸ U _f	²³⁸ U _{he}	²³⁹ Pu _f	²³⁹ Pu _h
^{131m} Xe	11.934	d	0.05	0.06	0.05	0.06	0.05	0.07
^{133m} Xe	2.19	d	0.19	0.29	0.19	0.18	0.24	0.42
¹³³ Xe	5.243	d	6.72	5.53	6.76	6.02	6.97	4.86
¹³⁵ Xe	9.14	h	6.6	5.67	6.97	5.84	7.54	6.18

Table 2.1. Cumulative fission yields for the four major radioxenon isotopes, induced by thermal neutrons (f) and high-energy neutrons (he).



Figure 2.2. Fission yield vs. atomic mass of fission products.

Following atmospheric detonations of nuclear weapons, most fission products condense and/or attach to aerosols in the atmosphere, forming particulate radionuclides. Noble gas fission products are chemically inert, however, and remain in the gas phase. They are therefore less likely to be washed out in rain, and in the case of underground explosions, noble gasses are more likely to escape than are particulate radionuclides.

Xenon-133, with its high production yield and fairly long half-life (Table 2.1), is the most prevalent isotope observed in environmental monitoring for nuclear explosions. Depending on the fission material (235 U, 233 U, or 239 Pu), a 1 kiloton (kt) nuclear explosion produces between 1.08×10^{16} and 1.33×10^{16} Bq of 133 Xe.

2.1 Radioxenons and environmental monitoring

Xenon isotopes can provide information on fission origins and are therefore useful, first, in distinguishing between civilian and non-civilian sources. Second, comparisons of the activity ratios ^{133m}Xe/^{131m}Xe and ^{133m}Xe/^{133m}Xe/^{133m}Xe have also been found to be useful in distinguishing between U and Pu fission. Radionuclides of interest in nuclear explosion monitoring, both particulate and noble gas, are shown in Figure 2.3, and characteristics of relevant xenon isotopes and isotopic ratios are summarized in Table 2.2.

Isotope	CTBT relevancy		Other source	
Xe-135	Fission product, 9.1h half time		Medical production, fresh fission product	
Xe-133	Fission product, 5.24d half time		Medical production, reactor releases	
Xe-133m	Fission product, 2.19d half time		Very short irradiation	
Xe-131m	Fission product, low fission yield, 11.9 d half time		Reactor releases, I-131 production	
Isotope ratio	CTBT relevancy	Civilia	an sources	
Xe-135/133	High ratio indicates very fresh fission, ratio >5 detected rarely by civilian sources	Reactor as Xe- cross s	ctor nearly always with low ratio e-135 has high neutron capture s section.	
Xe-133m/131m	High ratio indicates short irradiation time, ratio >2 is rarely seen by civilian sources	Reactor long ti and lo	or fuel is typically irradiated ime, this increases Xe-131m wers the ratio quickly under 1	

Table 2.2. Characteristics of Xe isotopes and their ratios.

Key radionuclides: the traces that indicate nuclear test * Xe-135 and Xe-133 are most abundant in a 1 kt nuclear explosion, with activities increasing after a couple of days due to formation of beta-decay chain of precursors (Table 1). • The use of several radionuclides are preferred (Table 2). These radionuclides are considered relevant as nuclear test indicators because their presence or their mutual ratios can be used to discriminate other possible sources of radioactivity. Table 2. Radionuclides relevant as nuclear test indicators. Table 1. Most abundant radionuclides after a 1 kt explosion. 64 d 1-day decay 3-day decay 10-day decay 30-day decay 35 d 13.6% Ru-103 Xe-135 11.8% Mo-99 176 9.6% / Xe-133 12.2% 2.75 d I-133 7.8% Rh-105 8.3% Ba-140 7.9% Ce-141 10.7% 39 d 6% Xe-133 7.3% 1-131 7.0% Ba-140 9.6% 1.008.4 Zr-97 8 d 4% Te-132 6.5% Zr-95 Rh-105 6.9% Mo-99 6.3% 3.3 d 44% Co-143 6.2% I-131 Te-132 4.5% Pd-109 59% 20 h Ce-134 4.3% 1-133 4.8% 3.5% 218 \$9% Ru-103 Xe-133 13.2 d Mb-99 4.2% Zr-97 3.1% Ce-141 4.6% Nd-147 3,4% 30 y I-135 42% 1-131 3.1% Nd-147 3.3% Ce-144 1.4% 12.8 d 40.2 6 Te-132 2.8% Ba-140 2.9% Zr-95 2.2% 1.4% Ri-106 31.5 d 2.4% Sr-91 Xe-135 1.9% Rh-105 1.2% Te-129m 0.39% 1:4 d Ru-105 1.5% Pd-109 1.4% Sb-127 0.76% Te-132 0.32% Xe-133 0.69% En-156 0.29% 1.4% Ru-103 1.4% Ce-143 noble gasi 1.3% 0.23% Xe-131m 0.13 Ce-141 Xe-133m 9.1 h Xe-133m 0.42% Xe-131m 0.05% 2.19 d

Xe-131m 0.0058%

Figure 2.3. Radionuclides of interest in CTBT compliance monitoring.

Detections of radioxenon that results from the irradiation of uranium for medical isotope production share most characteristics with those from uranium-based nuclear explosions, given that in both situations, there is some delay between the time noble gasses are produced and when they are released. This poses a problem for environmental monitoring operations in the context of nuclear nonproliferation and CTBT verification, since industrial isotope production facilities (IPFs) contribute xenon isotopes to the environment, which are subsequently detected.

11.9 d

2.2 Relative radioxenon productions

Radioxenon isotopes are produced by nuclear weapons and nuclear reactors as mentioned above. Although nuclear power reactors produce *much* greater quantities of radioxenon than IPFs, most of the xenon is retained in the reactor's fuel cladding, containment vessels, and noble gas delay systems. To *process* irradiated materials for isotope production in an IPF, however, fuel cladding is dissolved only a short time after irradiation, and xenon effluents are released and may subsequently enter the atmosphere. Figure 2.4 illustrates the relative quantities of xenon emissions in the environment from different sources.



Figure 2.4. The relative releases of radioxenon isotopes from the various sources involved.

As Figure 2.4 indicates, the *average* daily release of radioxenon from an IPF is considerably greater than that of a nuclear power plant, but is less than a "not-so-well contained" nuclear explosion. Generally, air samples analyzed to be high in ¹³⁵Xe and/or ^{133m}Xe are indicative of a nuclear explosion, while measurements high in ^{131m}Xe are more indicative of civil applications.

2.3 Confusion of detection

The high rate of radioxenon release from IPFs poses a problem for environmental monitoring associated with nonproliferation. The CTBT noble gas monitoring station in Germany regularly detects emissions from the IPF of National Institute for Radioelements (IRE) in Fleurus, Belgium (Figure 2.5); and the station at Charlottesville, Virginia, USA, can detect emissions from the Chalk River IPF in Canada (Figure 2.6).



Figure 2.5. Detections of the Fleurus IPF by the xenon monitoring station in at Schauinsland, Germany.



Figure 2.6. Detection of Chalk River emissions at Charlottesville, USA.

The regular detection of industrial xenon emissions by networks set up primarily for nuclear explosion monitoring requires robust means of source differentiation. A key to this differentiation problem rests with the changes in isotopic composition of a radioxenon release over time, and with the relative magnitudes of isotopic ratios and ratios of ratios.

2.4 Radioxenon ratios

The quantity of xenon isotopes in an irradiated sample grows over the course of nuclear reactor operation, and then decreases due to radioactive decay once fission ceases, as illustrated Figure 2.7.



Figure 2.7. Trends in Xe isotope activities during production by fission in a nuclear reactor and after cessation of irradiation (indicated by blue line after 9 days). Dashed lines indicate decoupling of isotopes from their parent nuclides due to dissolution of target material.

Reactor production of xenon can be compared with activity trends from radioxenon following a nuclear explosion, as shown in Figure 2.8. In a nuclear explosion, if instant noble gas release is assumed, the shaded area indicates a difference in ^{133m}Xe production in detonations of U- and Pu-fuelled devices. Since ¹³³Xe activity varies little based on the fuel of an explosive device, this constant can be used to calculate ratios that help discriminate between device types, provided that uncertainties are low enough.



Figure 2.8. Activity of radioxenon isotopes from a 1 kt nuclear explosion.

Activity ratios vary with time elapsed from the start of reactor production, as shown in Figure 2.9 (the same operating conditions as those in Figure 2.7 are assumed). Figures 2.9 and 2.10 also show that plotting ratios of xenon shows trends over time.



Figure 2.9. Changes in Xe ratios over time during and after irradiation.



Figure 2.10. Ratio 133m Xe/ 133 Xe vs 135 Xe/ 133 Xe (red line is explained in Figure 2.12).



Figure 2.11. Ratio 133m Xe/ 131 mXe vs 135 Xe/ 133 Xe.

Comparisons of ratio plots provide a tentative means of distinguishing between civilian and military sources of radioxenon, as shown in Figure 2.12.



Figure 2.12. Differentiation between reactor and weapon emissions on the basis of Xe isotope ratios. The green area signifies reactor production and the blue weapons. The dashed red line separates the two as a possible "discrimination line."

Although the possibility of source discrimination seems clear in Figure 2.12, the reality is different. Unfortunately, when modeled ratios are compared with actual measurements, the differentiation is not so clear-cut. Figures 2.13, 2.14 and 2.15 show plots of isotope ratios for detections related to emissions from three IPFs: the National Institute for Radioelements (IRE), Belgium; NTP, South Africa; and Australian Nuclear Science and Technology Organisation (ANSTO), Australia, respectively. These plots show a scatter of ratios across the theoretical "discrimination line," into both reactor and weapon sectors of the ratio plot.



Figure 2.13. Comparison of ratios at IRE, Belgium.



Figure 2.14. Comparison of ratios at NTP, South Africa.



Figure 2.15. Comparison of ratios at ANSTO, Australia.

It is clear (figures 2.13 - 2.15) that characteristics of IPF emissions can overlap into the weapon explosion region of the isotopic ratio plot, allowing for detections of civilian emissions to be confused with those from military sources. So, while ratio comparison shows promise as a means of differentiation, it cannot be relied upon to always facilitate *unambiguous* source attribution. This is the essence of the problem confronting the environmental monitoring and radiopharmaceutical communities, balancing the need for accurate source attribution with the need for continued and increasing medical isotope production. Thus, the reduction and abatement of radioxenon emissions at the source (i.e. IPFs) may be the only way to definitively solve the problem for nuclear explosion monitoring, as addressed elsewhere in this report.

3.0 Global ⁹⁹Mo production and vulnerability of supply

Radiopharmaceuticals are used primarily in diagnosis and treatment planning. Radionuclides suitable for use as radiopharmaceuticals must satisfy a number of practical criteria to be appropriate for use. They must have half-lives long enough to support medical supply logistics, but short enough to decay soon after imaging or treatment is complete. They must have decay energy suitable for the given application, should be relatively easy to produce, and preferably can be available via a "generator" used on site.

Technetium-99m, produced through the decay of ⁹⁹Mo, is by far the most heavily used isotope, applied in about 30 million procedures per year worldwide. To supply ^{99m}Tc, ⁹⁹Mo generators ("cows") are provided to a medical imaging facility, where technicians can "milk" the decay product ^{99m}Tc as needed. The supply of the parent ⁹⁹Mo is therefore the main topic of this discussion. Because of its short half-life (65.9h), it is not practical to stockpile ⁹⁹Mo, so supply is sensitive to facility shutdowns and transport disruptions.

The normal production routes for ⁹⁹Mo involve ²³⁵U fission, ⁹⁸Mo activation, ¹⁰⁰Mo photolysis, or ²³⁸U fission, as depicted in Figure 3.1.



Figure 3.1. The possible production routes for ⁹⁹Mo.

The ⁹⁹Mo supply chain is dominated by a small number of major producers as shown in figures 3.2 and 3.3. Conditions for a supply crisis are right: aging facilities require frequent or unexpected closures, and demand is growing. Historically, five reactors have provided the global supply of ⁹⁹Mo, yet all are now

over 45 years old. At present, nine reactors are involved, but all except one are over 38 years old. This aging reactor fleet faces prolonged and even permanent shutdowns. In the period 2001 - 2010, various facilities were shut down for periods of six days to 14 months. They are all fission based using highly enriched uranium (HEU) or low-enriched uranium (LEU), though pressure for facilities to switch to LEU has been increasing due to concerns related to nuclear security. Global demand is anticipated to increase about 30% by 2030, and the current reactor-based supply will not be sufficient to meet this.



Figure 3.2. The major producers of ⁹⁹Mo.



Figure 3.3. Major ⁹⁹Mo producers, reactors and associated facilities.

Unless new lines of ⁹⁹Mo production are established, shortages will inevitably arise if ⁹⁹Mo demand projections hold true. Under these conditions, as indicated in Figure 3.4, a shortfall will occur in 2016-2017 or 2021.



Figure 3.4. Projections of ⁹⁹Mo shortages.

The supply chain for the United States is particularly vulnerable since it current relies on sources outside the U.S., as depicted in Figure 3.5.



Figure 3.5. The supply chain for ⁹⁹Mo-based pharmaceuticals approved for use in the U.S.

4.0 Environmental monitoring technology

The International Monitoring System (IMS) is the environmental monitoring network that is a key component of CTBT verification. It aims to detect all nuclear tests in all environments (underground, atmospheric, and underwater) through three waveform technologies (seismic, infrasound, and hydroacoustic monitoring) and through radionuclide monitoring, which supports detection of tests in all three environments. An International Data Centre provides analysis of IMS data as the backbone of the CTBT verification regime, which also includes on-site inspections (OSI), consultation and clarification, confidence-building measures, and national technical means. The IMS network is depicted in Figure 4.1.



Figure 4.1. The IMS network (as of June 2012).

Underpinning the future of verification are technology development frameworks and processes, engagement with the science and technology communities, engagement with the user community, and engagement with medical isotope producers in the matter of xenon source attribution, as discussed here.

Waveform technologies' strength lies in the detection, location, and timing of events. But, the only way to establish unequivocally the *nuclear* nature of the event is the detection of fission products. Radionuclide technology provides such evidence as the "smoking gun" of a nuclear test explosion.

The IMS radionuclide network has an approximate 90% probability of detecting a 1 kt nuclear explosion anywhere on earth within 14 days. The IMS is still under construction but will eventually comprise 80 particulate stations and 40 noble gas stations, supported by 16 international radionuclide laboratories.

Following atmospheric detonations, most fission products condense and/or attach to aerosols in the atmosphere, forming particulate radionuclides. Noble gas fission products are chemically inert and remain as gases. They are therefore less likely to be washed out in rain, and more likely to escape from underground explosion chambers. Among the noble gases, monitoring focuses on the radioxenon isotopes because of their high fission yields and their half-lives suitable for atmospheric transport to and detection at monitoring stations.

The radionuclide monitoring process involves the following components:

- Data acquisition (remote sampling of aerosol and noble gases);
- Data analysis (analysis of spectra and characterization of results); and
- Atmospheric transport modeling application for source location.

Particulate monitoring is depicted in Figure 4.2.



Figure 4.2. Components of the IMS particulate radionuclide monitoring system.

Radioxenon measurement involves extraction of stable Xe from the atmosphere and measurement of xenon isotope activities. Stable xenon volume is also determined as a measure of the sampled volume. The system is outlined in Figure 4.3.



Figure 4.3. The basic Xe sampling scheme.

This design concept is also utilized in some national xenon monitoring programs, such as that of the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) in Italy. There, the system has three components as illustrated in Figure 4.4: collection (sampling and adsorption), processing (gas extraction and purification), and gamma spectroscopic analysis.



Figure 4.4. Design concept for the ENEA xenon system.

During sampling, air is passed through a filter and water trap (silica gel), and then through a cryogenic trap of activated charcoal cooled by liquid nitrogen. About 100 m^3 of air is sampled in one week. The collection and xenon desorption components of the ENEA system are illustrated in figures 4.5 and 4.6.

After sampling, the xenon is desorbed after the adsorbent cartridge is heated to 533K and helium carrier gas is used to remove CO_2 and H_2O , using the system illustrated in Figure 4.6.

Finally, the gas container is analyzed directed by high-resolution gamma spectroscopy.





Figure 4.5. Air sampling system for the ENEA xenon monitor.

Figure 4.6. ENEA xenon desorption system.

5.0 Impact of ⁹⁹Mo production on monitoring operations

5.1 History of radioxenon detection in Germany

The sensitivity of environmental monitoring operations to civilian xenon releases is illustrated by the history of monitoring in Germany, where releases from anthropogenic sources in Europe dominate the atmospheric background. The German Federal Office of Radiation Protection has monitored atmospheric xenon since the mid-1970s. Time-series data from the Freiburg monitoring station are summarized in Figure 5.1. Activity distributions for various monitoring sites in Germany are summarized in Figure 5.2.



Figure 5.1. Long-term ¹³³Xe activity concentration measured at Freiburg, Germany.

Since the year 2000, an IMS SPALAX system has been operating at the Schauinsland station (DEX33), time-series data for which are shown in Figure 5.3. The effects of the IPF at Fleurus and the Fukushima incident are clear.

The civilian nature of xenon sources detected at station DEX33 is demonstrated by the isotope ratio plot, Figure 5.4, where discrimination is clear.



Figure 5.2. ¹³³Xe activity distributions in Germany, 1990 – 2010.



Figure 5.3. Radioxenon time series at Freiburg, 2000 – 2012, as measured by the IMS SPALAX system.



Figure 5.4. Radioxenon discrimination at DEX33, 2004 – 2012 for events in which all four xenon isotopes are detected.

It is expected that Germany's phasing out of nuclear power up to 2022 will reflect the influence of nuclear power plants (NPPs) on radioxenon levels, which may possibly increase monitoring sensitivity to emissions from IPFs.

5.2 Southern Hemisphere detections and modeling

Known xenon sources in the Southern Hemisphere are mapped in Figure 5.5.



Figure 5.5. Xenon sources in the Southern Hemisphere.

The ANSTO IPF has an influence on IMS stations AUX04 (Melbourne, Australia), AUX09 (Darwin, Australia) and NZX46 (Chatham Island, New Zealand). Figure 5.6 graphs the percentage of days when ¹³³Xe concentrations, modeled to be released from ANSTO, exceed the minimum detectable concentration of IMS noble gas instrumentation (estimated at 0.3 mBq/m³) at these locations.





Figure 5.7 illustrates the modeled impact of all southern-hemisphere xenon sources on the 13 IMS stations in the southern hemisphere.



Figure 5.7. The modeled influence of southern IPFs on Southern Hemisphere IMS stations.

Models of the frequency with which IPF emissions impact IMS stations raise the question of whether the IMS mission is significantly (and scientifically defensibly) impacted by the current levels of radioxenon emissions from IPFs. Further, questions arise about whether reducing xenon emissions is practicable in an operational environment, or whether IPFs are the only category of facilities that should reduce xenon emissions, considering the large number of other civilian sources like NPPs (Figure 5.8).

With these questions in mind, all existing and anticipated future IPFs (Figure 5.9) were included in a model that predicts the frequency of detection, by IMS station, based on release volume (Figure 5.10). When emissions fall below 5×10^9 Bq/d, average ¹³³Xe concentrations appear to be localized to the emission source, thus limiting these detections' impact on the IMS mission. NPP emissions are below this average (about 10^9 Bq/d), and some large IPFs have already achieved averages $< 5 \times 10^9$ Bq/d. This (voluntary) emissions limit strikes a balance between what IPFs can attain and what affects the IMS.







Figure 5.9. Exiting and possible future IPFs.



Figure 5.10. ¹³³Xe detections in the IMS network from current and future IPF and NPP plants using an MDC of 0.1 mBq/m^3 .

5.3 The 2011 European ¹³¹I incident

In 2011, national monitoring systems throughout Europe widely reported detection of 131 I. Figure 5.11 illustrates the areas affected. The incident proved to be a good test of international cooperation within the environmental monitoring community and of the applicability of atmospheric tracking systems.



Figure 5.11. Detections of ¹³¹I in Europe during November 2011.

Atmospheric backtracking, coupled with measurements from various locations, indicated that the most likely source of ¹³¹I was the Institute of Isotopes, Hungarian Academy of Sciences (KFKI) in Budapest. The total release between January and October was initially estimated at about 700 GBq.

Possible sources of ¹³¹I at the KFKI Budapest campus included the radioisotope laboratory and research reactor (although the latter would be accidental only). A common stack receives ventilation gas from both these facilities after ventilation from hot-cells is routed through particulate and activated charcoal filters.

During 2011, production of 131 I was confirmed to be 21,700 GBq, and the atmospheric release was defined at 660 GBq. This high release was due to a deficiency in the filtration system. Individual releases are plotted along a timescale in Figure 5.12.



Figure 5.12. Release of ¹³¹I from the Budapest facility during 2011.

To prevent a recurrence, an emission control system has been improved, including the installation of multi-step filtration systems and additional monitoring units.

6.0 Xenon emission reduction and stack monitoring

6.1 Emission studies at Nordion

The Nordion IPF, Canada, processes liquid samples containing ⁹⁹Mo received from Chalk River Laboratories. Emissions from the ⁹⁹Mo processing chain at Nordion have been studied in detail, using an in-line proportional counter and sodium iodide (NaI) (Tl) scintillation counter coupled to a Multichannel analyzer, as shown in Figure 6.1.



Figure 6.1. The processing line in the Nordion plant showing the placement of radiation detectors.

The activities of radioactive xenon released during ⁹⁹Mo processing and their rates of release are illustrated in figures 6.2 and 6.3.



Figure 6.2. Activity released during ⁹⁹Mo processing, versus process time, at the Nordion IPF.



Figure 6.3. Xenon release rate during ⁹⁹Mo processing at Nordion.

A high xenon release rate at the beginning of the process, as visible in Figure 6.3, is associated with opening bottles with liquid samples upon receipt from Chalk River Laboratories.

Table 6.1 compares annual radioactive xenon releases with their associated isotope production process during the period 2008 - 2011.

GBq's	Xe-133 Processing	Mo-99 Processing			
Year	Xe-133	Xe-133	Xe-135	Xe-135m	
2008	17 975	48 862	48 205	60 236	
2009	11 986	14 401	14 439	20 441	
2010	3 191	5 875	6 407	9 366	
2011	19 144	15 822	17 239	27 688	

Table 6.1. Trends in xenon isotope releases from the Nordion IPF, 2008 – 2011.

6.2 Activated charcoal vs zeolites as adsorbers for ¹³¹I or Xe at IRE

Xenon and iodine emissions are most commonly controlled with cryogenically cooled activated charcoal as an adsorber, although comparisons have been made with the effectiveness of zeolites. At the IRE IPF in Fleurus, zeolites were found to have relatively little trapping ability for ¹³¹I, as determined by alternating zeolite and charcoal beds, with ¹³¹I found in charcoal beds on either side of zeolite, but no ¹³¹I in zeolite. IRE found total trapping when four charcoal beds were in series and replaced daily. It appears that charcoal cartridges act more like "retarders" than filters, and releases can be underestimated if account is not taken of this.

Silver-exchanged zeolites (SZ) are known to be highly selective for xenon and might be possible to use in place of activated charcoal in normal off-gas treatment systems. IRE assessed the relative effectiveness of SZ and charcoal by monitoring the time it took for xenon to break through the trap, such that the final xenon concentration equaled the initial level, as illustrated in Figure 6.4. Results indicate that SZ is more effective than charcoal at room temperature, but charcoal is considerably better at 200K. Work is ongoing at IRE concerning whether SZ can be used as a viable alternative for charcoal.



Figure 6.4. Comparison of effectiveness of activated charcoal and SZ in controlling xenon emissions at IRE.

6.3 Improvements at ANSTO

Charcoal and molecular sieve columns are also used at the ANSTO IPF in Australia; Figure 6.5 shows the process used at ANSTO.





To minimize gas escapes from the stages indicated in Figure 6.5, sealed column holders are used to prevent escape from decaying ion exchange columns, plastic tubing has been replaced with stainless steel, instant shut-off valves are used, and all process transfers are achieved under vacuum. Vacuum is maintained by a buffer tank, re-evacuated for every run, and more buffer tanks have been added to avoid

need to pump tanks down during the process. When buffer tanks are evacuated an activated charcoal column and a molecular sieve column are used on the pump outlet. These steps seem to be effective in limiting emissions.

6.4 Stack monitoring at ANSTO

ANSTO monitors the release of six isotopes of regulatory concern from its facility: ¹³³Xe, ¹³⁵Xe, ¹³⁵Mze, ^{85m}Kr, ⁸⁷Kr, and ⁸⁸Kr. In 2011, a high-purity intrinsic germanium system was installed for high-resolution stack monitoring, particularly for ^{131m}Xe and ^{133m}Xe. The earlier low-resolution NaI system uses a 100 mL flow-through sampling chamber viewed by a 2" \times 2" NaI crystal encased in lead shielding. The new high-resolution system uses a 35% high-purity intrinsic germanium with a 4.5L Marinelli flow cell, with automated analyses involving 15-minute counting times. Greater efficiency of NaI results in greater sensitivity, but the high-resolution system enables detection of ^{131m}Xe and ^{133m}Xe, which assists in source attribution.

7.0 New Technologies

The continuation of supply of ⁹⁹Mo depends on the emergence of new technologies and the upgrading of existing reactor systems, assisted by government policies. The U.S. government, for example, has set two goals concerning medical isotope production: to minimize HEU usage while establishing reliable supplies, and to minimizing the impacts of effluents from IPFs. To that end, it provides assistance in converting facilities from HEU to LEU, while also aiding development of non-HEU facilities for ⁹⁹Mo production in the U.S. to achieve a reliable domestic supply. Its objective is that domestic sources can meet 100% U.S. demand without HEU. The U.S. government has encouraged development of the following technologies:

- Neutron capture technology;
- LEU solution reactor technology; and
- Accelerator technology.

Considering the difficulties in discriminating sources of radioxenon detections, reducing the sources of emissions is considered to be a far more effective solution than improving signature discrimination. In addition to abatement methods, new technologies provide another means of doing this.

New non-HEU technologies are being developed as indicated in Figure 7.1, including homogeneous and heterogeneous reactor systems and accelerators. Many countries are conducting research and development for new facilities, including Argentina, Australia, Belgium, Brazil, China, Egypt, France, Germany, India, Indonesia, Republic of Korea, Netherlands, Poland, Romania, Russia, South Africa and the United States. The International Atomic Energy Agency is encouraging diversification and development of local supplies through various Coordinated Research Projects.



Figure 7.1. New technologies becoming available for ⁹⁹Mo production.

The Organisation for Economic Cooperation and Development has adopted the following principles for ⁹⁹Mo production:

- Full-cost recovery should be implemented;
- Reserve capacity should be paid for by the supply chain;
- Governments should enable safe and efficient market operation; and
- Governments should support HEU-LEU conversion.

Argonne National Laboratory supports the development of three technologies:

- Electron- Linear particle accelerator (LINAC)-accelerator production from the γ/n reaction of ¹⁰⁰Mo (NorthStar Medical Technologies, LLC.; and Los Alamos National Laboratory);
- Production of fission-product ⁹⁹Mo from an aqueous homogeneous reactor (Babcock & Wilcox);
- D/T-accelerator-driven production of fission-product ⁹⁹Mo in an aqueous LEU target solution (Morgridge Institute for Research, SHINE Medical Technologies, Los Alamos National Laboratory).

7.1 Cost of HEU – LEU conversion

Various costs are associated with converting from HEU to LEU systems. One way to reduce or eliminate process efficiency costs is through the use of silicide, rather than aluminate targets. With the reduction of

U enrichment from 91% to 19.75%, the density of the targets must be increased to offset reduced U content, if production economy is to stay the same. UAl_x density is 8.1g/mL, while that of U_3Si_2 is 4.8g/mL. An additional oxidizing step is required in silicide processing, but it leads to a 100% compensation for the loss of HEU. LEU- U_3Si_2 thus results in a monetary return similar to that of HEU, whereas LEU- UAl_x returns about 30% less. The chemical processes associated with the two alternatives are slightly different, as illustrated in Figure 7.2, but this is not considered to be a significant issue.



Figure 7.2. Comparison of treatment processes for UAl_x and U_3Si_2 targets.

7.2 The SHINE system

The SHINE system offers several advantages: it involves mature accelerator technology, it is inherently safe (needs to be driven to operate), uses LEU (19.75%), it reduces nuclear waste (no reactor, recyclable solution), and the aqueous target simplifies chemical extraction. Development work is underway on the following points:

- Mo-recovery system using TiO₂ sorbent;
- Use of a LEU-modified Cintichem process for Mo purification;
- Preparation of uranyl sulphate target solution;
- Periodic cleanup of irradiated target solution;
- Measurement of rates and composition of radiolytic gas generation;

- Measurement of changes in solution composition; and
- Facility design and waste treatment.

Data collected from experiments associated with this development work will aid the final design of the off-gas treatment and Mo-recovery systems. An industrial-scale prototype is now under development, with an anticipated production of >500 6-day Ci/week.

Overall, the SHINE system aims to produce high-specific-activity medical isotopes without a nuclear reactor. The technology has two key aspects:

- Primary neutrons are created using a high-output D-T source;
- Neutrons enter a LEU solution where they multiply subcritically and create isotopes.

In neutron production, D (²H) gas flows into an ion source and is ionized by microwaves; an accelerator pushes ions towards a target chamber (300keV); the accelerated deuterons strike T (³H) gas in the target, creating neutrons at $2x10^9$ n/s/W. The neutron production system is illustrated in Figure 7.3, while the ⁹⁹Mo extraction and processing procedures are illustrated in Figure 7.4.



Figure 7.3. The SHINE neutron production system.



Figure 7.4. ⁹⁹Mo extraction and processing in the SHINE system.

The aqueous fission creates challenges of radiolysis of water (to H_2 and O_2) and volatile fission products (iodine, noble gases). Off-gas treatment elements include cryogenic traps to condense and freeze Xe, from which the Xe will be routed to a storage system pending release, together with the following:

- Recirculating nitrogen atmosphere;
- Closed system at negative pressure;
- Heat exchanger to lower water content;
- Entrainment filter to allow only gas in to system;
- Silver-coated zeolite traps for iodine capture;
- Catalytic recombiner for H₂ and O₂; and
- Heat exchanger to remove heat of recombination and condense water vapor.

The treatment system is illustrated in Figure 7.5.



Figure 7.5. Off-gas treatment system for the SHINE system.

7.3 Accelerator systems

Another system under development by Northstar aims, in the short term, to use the Missouri University Research Reactor to eventually produce ⁹⁹Mo for 50% of U.S. demand. In the longer term, LINAC methodology will be used for ⁹⁹Mo production. The LINAC facility will house up to 16 LINAC machines capable of producing >3000 6-day Ci/week. Both the n, γ and γ ,n ⁹⁹Mo production processes use stable Mo isotopes as the target material, and no U, Pu or fission products are in the waste. These processes are compared Figure 7.6.



Figure 7.6. Comparison of electron accelerator systems.

7.4 Neutron activation

Neutron activation of ⁹⁸Mo is another alternative to fission systems, as compared in Figure 7.7.

С	Comparison of Current Production						
	Methods						
	²³⁵ U(n,f) ⁹⁹ Mo	⁹⁸ Mo(n,γ) ⁹⁹ Mo					
	Produces high specific activity 99Mo	Produces low specific activity Mo-99					
	Requires enriched ²³⁵ U target	Requires highly enriched Mo- 98 target					
	Complex chemical processing	Simple chemical processing					
	Requires dedicated processing facility	Requires high flux neutron source					
	Generates high-level radioactive waste	Generates minimal waste					

Figure 7.7. Comparison of fission and activation techniques for ⁹⁹Mo production.

8.0 Other isotope production facilities

WOSMIP III included representation from IPFs which had not participated in previous meetings.

8.1 Uzbekistan

The Institute of Nuclear Physics, Uzbekistan Academy of Sciences, Tashkent, has the following facilities: a research reactor (10 MW), two cyclotrons (22 MeV and 20 MeV), an electron accelerator (408 MeV), a ⁶⁰Co gamma irradiation facility, and a neutron generator. Figure 8.1 notes which isotopes the Institute produces, and at which facilities.



Figure 8.1. Isotope production in Uzbekistan.

Some of the isotopes produced there, and the reactions employed are as follows:

$${}^{32}P[{}^{32}S(n,p){}^{32}P]; {}^{33}P[{}^{33}S(n,\gamma){}^{33}P]; {}^{125}I[{}^{124}Xe(n,\gamma){}^{125}I]; {}^{55}Fe[{}^{54}Fe(n,\gamma){}^{55}Fe];$$

 ${}^{35}S[{}^{35}Cl(n,p){}^{35}S]; {}^{131}I[{}^{130}Te(n,\gamma){}^{131}Te]; and {}^{99}Mo[{}^{98}Mo(n,\gamma){}^{99}Mo].$

Figure 8.2 summarizes the nuclear security and nonproliferation activities undertaken by Uzbekistan.



Figure 8.2. Nuclear security and non-proliferation activities undertaken by Uzbekistan. O. Aviv: Overview of recent activities at Soreq NRC, Israel

8.2 Israel

The nuclear research center at Soreq, Israel, includes a CTBT radionuclide laboratory (ILL09), for which certification was currently in progress at the time of WOSMIP III, but which has since been certified. Located at the center is SARAF (Soreq Applied Research Accelerator Facility), a 4 MeV d/p beam used to produce various radionuclides. It has a proton/deuteron beam of 40 MeV, 4 mA, used to produce ⁶He (for science applications), ⁸Li (for nuclear astrophysics), and ¹⁰³Pd (for prostate cancer therapy).

9.0 Conclusion

WOSMIP-III succeeded in continuing to increase mutual understanding and knowledge of the challenges that face medical isotope producers and the environmental monitoring community. Most importantly, the workshop helped to deepen knowledge about how radioxenon emissions link the important mission of the IMS to the imperatives and constraints of radioisotope production for healthcare purposes.

While some limited discrimination between radioxenon sources through analytic methods (e.g. activity ratio comparison) is possible, the long-term solution probably depends on emission abatement and cleaner isotope-production technologies.

Efforts by the major contributors to the global supply of ⁹⁹Mo represented at the Workshop were thus instrumental in facilitating the knowledge exchanges that comprised the success of WOSMIP-III, particularly in the following areas:

- Undertaking and demonstrating the viability of efforts to reduce or delay the emission of radioxenon into the atmosphere;
- Sharing results of experiments to develop new methods of reducing emissions;
- Monitoring facility releases of radioxenon and sharing data; and
- Generating and sharing knowledge about facility practices in isotope processing.

Progress on new ⁹⁹Mo production technologies shared at WOSMIP-III is also highly encouraging. While these technologies not only succeed in reducing or eliminating the possibility of large xenon emissions, increasingly, their contribution is also significant in that they present viable models of new sources of ⁹⁹Mo, which may help resolve concerns about the sustainability of the current global market.

The participation of new representatives from Uzbekistan and Israel highlights the importance of the matters at hand, as well as reaffirms the interest and good faith of all participants in finding practical solutions. Continued engagement and dedicated efforts by these and all participants will attract the interest of new partners whose participation is equally important to making further progress.

Presentations at WOSMIP-III added to the documentation of the impact of radioxenon emissions to CTBT verification efforts; the modes by which xenon is produced, contained, and detected; and the state of the global ⁹⁹Mo supply chain and market. As a result, the first three meetings of WOSMIP have solidified the research base about the effects of ⁹⁹Mo production on the mission of the IMS, potential

solutions that can be implemented by monitors and at IPFs, and the constraints in which the participants operate.

Given this solid foundation, *the success of Workshops in the future depends on the efforts by WOSMIP participants to continue evaluating and implementing technically and economically feasible solutions* to mitigate the impact of radioxenon emissions on environmental monitoring. While adding to foundational research will support this goal, only practical evaluation and implementation of proposed solutions will highlight the opportunities and challenges that they present, and thereby further discussions about how WOSMIP participants can bring about a sustainable future for their important contributions to international peace and security, and human health.

Toward this goal, and short of implementing known methods capable of reducing radioxenon emissions to levels that will no longer interfere with the IMS mission, WOSMIP participants should collaborate to:

- Increase confidence that 5×10⁹ Bq/d is a goal limit that is both necessary (i.e. uncertainty associated with discrimination analysis cannot be reduced further) and sufficient (i.e. producers will not be asked to reduce emissions in the foreseeable future again);
- Consider how governments, international organizations, and other entities can support the common goals of WOSMIP;
- Continue to develop more efficient and cost-effective methods of reducing emissions;
- Design facility-specific approaches to limiting radioxenon emissions to 5×10^9 Bq/d;
- Identify challenges of implementing these approaches and methods of overcoming them; and
- Evaluate the economic costs and benefits of reaching the voluntary target limit.



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