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WOSMIP IV—Workshop on Signatures of Medical and Industrial Isotope Production

Vienna International Center, Vienna, Austria
November 11–13, 2013

July 2014

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Executive Summary

The fourth Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP IV) was held November 11-13, 2013, at the Vienna International Center in Vienna, Austria. The workshop brought together 82 experts from 25 countries from the isotope production and radionuclide nuclear explosion monitoring communities to continue discussions on the challenges for nuclear explosion monitoring presented by effluents from medical isotope production. This workshop continued to promote coordination and collaboration between these two distinct scientific communities in an effort to discover ways to mitigate the effects of isotope production on monitoring, while continuing to support efficient, reliable production of isotopes. This fourth workshop included a brief overview from each community, followed by detailed discussions on specific technical challenges and opportunities for future collaboration. The major outcomes and observations of WOSMIP IV were discussed with all participants at the conclusion of the workshop and are summarized below.

- ***Radioxenon Emissions Pledge.*** Four more producers signed the pledge with the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) to help mitigate the effects of radioxenon emissions by, for example, reducing emissions, sharing stack monitoring data, and continuing to collaborate with the WOSMIP community. These include two current producers—the Australian Nuclear Science and Technology Organisation (ANSTO) and PT Batan Teknologi Company of Indonesia— and two prospective future producers—the Korean Atomic Energy Research Institute (KAERI), and Coquí RadioPharmaceuticals Corporation of the United States. Counting the signing of the pledge by the Belgian-based Institute for Radioelements (IRE) in June 2013, there are now five signatories to the pledge. Following the signing ceremony, several more producers expressed interest in learning more about the pledge.
- ***A Coalition of Willing Collaborators.*** WOSMIP has always encouraged collaboration between the isotope production and monitoring communities. This year's workshop specifically encouraged willing experts to work towards openly available technical solutions to issues such as emission mitigation and stack monitoring. Several workshop participants expressed interest in meeting between sessions to discuss open-source solutions to the emissions problem that could be shared, with the ultimate goal of designing a system with very low emissions.
- ***Sharing of Stack Monitoring Data.*** Participants agreed that the sharing of stack monitoring data is very important for the CTBTO verification mission. It was noted that some producers already share stack data with the CTBTO on an ad hoc basis, though more producers willing to do so are needed. During roundtable discussions, several producers requested that the CTBTO provide additional clarity on the modalities for sharing information, including confidentiality practices, data storing, etc. In response to concerns expressed by producers, the CTBTO stated that stack monitoring data are used only for treaty verification purposes by the CTBTO and State National Data Centers (NDCs), and the data are not made available to the public. The Provisional Technical Secretariat (PTS) agreed to provide a written explanation of the procedures for ensuring that stack data are only viewed by authorized users for treaty verification purposes. This will hopefully mark the beginning of the formal discussions between the CTBTO and the producers on the sharing of stack data. It was also noted that, while the CTBTO is the primary beneficiary of data-sharing arrangements, once in receipt of stack monitoring data, the CTBTO and the International Atomic Energy Association (IAEA) could, if asked, potentially assist medical isotope producers (MIPs) in resolving public concerns over releases from facilities.

- ***Accepting the 5×10^9 Bq/d as a Goal.*** WOSMIP participants acknowledged the emission limit/recommendation of 5×10^9 Bq/d that was proposed at last year's workshop. While it is understood that not all current facilities may be able to meet the 5×10^9 Bq/d threshold, participants have accepted this number as a goal for the community. Additionally, participants discussed the merits of making 5×10^9 Bq/d an internationally accepted value. Further discussions at the workshop focused on good practices in the use of abatement systems that can be used to reach this emission limit. Results from experiments and modeling that focused on reducing emissions during dissolution and separation processes at MIP facilities were presented. Discussions also focused on different detector system technologies that can be used to monitor facility stack emissions, as well as software that can be used to streamline stack data management and visualization. There were also several discussions focused on alternate production technologies and products that have essentially zero emissions to minimize dependence on fission-produced ^{99}Mo.
- ***Validation of Models.*** Participants from both communities recognized the need for continued improvements in validation of models for isotope production and dispersion. Overviews of atmospheric transport modeling methods were discussed, as well as examples of different analysis scenarios highlighting the impact of MIP on nuclear explosion monitoring. As with all models, it was stressed during the discussions that validation with stack monitoring data is very important. During stack data-sharing discussions it was determined that a two-step process for sharing data may be the most logical path forward. A first step would involve sharing historical stack data to improve models and the second step would involve sharing stack data in as near real-time as practicable.

As this process moves forward, it is increasingly important that the two communities stay in frequent contact. Participants generally agreed that annual meetings with work between sessions are important to ensure continued progress.

While experts from the CTBTO, IAEA, many current and future producers, and other interested parties were in attendance, participants acknowledged the importance of continued expansion to those not yet part of the WOSMIP community. Currently, most of the radioxenon background originates from a few facilities across the world, and it is important to continue to collaborate with these current major producers. Equally important is the need for WOSMIP to continue to engage with potential new producers in order to ensure that the monitoring situation remains manageable.

Acknowledgments

Lori Metz, Ted Bowyer, Justin McIntyre and Judah Friese would like to thank their coauthors and Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP) presenters for their contributions to this report.

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Acronyms and Abbreviations

AEA	Atomic Energy Authority, Egypt
AECL	Atomic Energy of Canada Limited
ANM	ANSTO Nuclear Medicine
ANSTO	Australian Nuclear Science and Technology Organisation
ATM	Atmospheric Transport Model
AUD	Australian dollar
CNEA	National Atomic Energy Commission
CRL	Chalk River Laboratories
CTBT	Comprehensive Nuclear-Test-Ban Treaty
CTBTO	Comprehensive Nuclear-Test-Ban Treaty Organization
DPRK	Democratic People's Republic of Korea
EMS	Environmental monitoring system
EU	European Union
GSG	Gamma-Service Group International GmbH
HEU	Highly enriched uranium
HPGe	High purity germanium
HVAC	Heating, ventilation, and air conditioning system
IAEA	International Atomic Energy Agency
IDC	International Data Centre
IMS	International Monitoring System
INR	Institute for Nuclear Research (Romania)
INR	Institute for Nuclear Research, Romania
INVAP	Investigaciòn Aplicada (Argentinean high-technology systems development company)
IRE	National Institute for Radioelements, Belgium
KAERI	Korean Atomic Energy Research Institute
LEU	Low-enriched uranium
LINAC	Linear particle accelerator
MDC	Minimum detectable concentrtion
MIP	Medical isotope production
NDC	National Data Center
NRC	U.S. Nuclear Regulatory Commission
NTP	NTP Radioisotopes Ltd, South Africa
PET	Positron emission tomography
PNNL	Pacific Northwest National Laboratory
PTS	Provisional Technical Secretariat

RPF	Radioisotope Production Facility of the AEA
SAUNA	Swedish Automated Unit for Noble Gas Analysis
SCK•CEN	Belgian Nuclear Research Center
SHINE	Subcritical Hybrid Intense Neutron Emitter; also a medical technology company
SPECT	Single-photon emission computed tomography
TSV	Target solution vessel
TXL	Transportable xenon laboratory
WOSMIP	Workshop on Signatures of Medical and Industrial Isotope Production

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1.0 Introduction

Medical and industrial isotopes are highly valuable tools used in science, medicine, and industry. Their principal use is for medical diagnostics, with approximately 30 million procedures per year; and medical therapy, with approximately 3 million treatments per year. Technetium-99m, daughter of ^{99}Mo, is by far the most heavily used. Broad applications of ^{99m}Tc include examining the function of the heart, liver, thyroid, and blood flow, as well as detection of tumors in the prostate, breast, and bone. The main method used to produce ^{99}Mo is reactor-based and includes fission of ^{235}U. In this process, fission gases including xenon and krypton are released into the atmosphere. Unlike reactors, gaseous signatures released during the production of medical isotopes are similar to a nuclear explosion because the process involves irradiation of uranium followed by dissolution as soon as possible. Medical isotope production can thereby inadvertently create a radioxenon background that impedes nuclear explosion monitoring. In particular, this background causes challenges for verifying compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

The Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP) brings together the isotope production and nuclear explosion monitoring communities to discuss ways to mitigate the effects of isotope production on the monitoring community without disrupting the supply of isotopes, and to better understand the isotopic and chemical signatures created through isotope production mechanisms. The four meetings to date (in 2009, 2011, 2012, and 2013) and other efforts intersession have raised awareness of issues of interest and concerns among isotope production facility operators and those responsible for environmental monitoring associated with nuclear security. Great progress has been made in bridging gaps and encouraging cooperation.

WOSMIP I in July 2009, defined and outlined the problem and introduced the monitoring and production communities to each other. During WOSMIP II in July 2011, the problem was more distinctly refined and scientific and political boundaries, as well as identification of ways for the communities to work together to move forward, were explored. The monitoring community continued to communicate and show that isotope production is easily observed from International Monitoring System (IMS) measurements at distance, promoting the realization that something can and should be done. Examples of continued progress in developing technology for emissions control and real examples of emissions being lowered were shared. Questions about new producers remained: Who were they? Where were they located? At what levels do they produce?

In July 2012, WOSMIP III produced concrete steps that were explored and exploited. A number of important observations were made at this meeting, including the fact that medical isotope production at current levels can be seen by IMS of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) every day. Stack monitoring data were considered a crucial portion of the required solution as well as reduction at the source.

Six major topics identified from past WOSMIPs are listed below. The topics are followed by explanatory text. The grade that follows the topic was given by one participant and explained in parentheses.

1. *Improve signature discrimination tools*, understanding of emission rates, isotopes emitted, and locations for isotope production. Grade: **B.** (Some atmospheric transport model [ATM] tools are improving.)

2. *Make stack data available to the International Data Centre (IDC),* either with an experiment or routinely. Grade: **B+**. (At least one producer has agreed to give data to the IDC.)

3. *Establish a code of conduct (producer's pledge) for medical isotope production*; to include best practices for emission mitigation, stack monitoring data, and the willingness to share information on emissions at levels important to the nonproliferation community, etc. Grade: **A-** (The producers pledge was established and one producer, National Institute for Radioelements (IRE), signed in between WOSMIP 3 and 4.)

4. *Establish multi-tiered levels of adherence to emission reduction*. This may appear similar to the Leadership in Energy & Environmental Design standard for energy-efficient buildings and may provide an incentive for pursuing best practices. Grade: **C.** (Though some thought has gone into this.)

5. *Identify all producers around the world, existing and new.* Grade: **C.** (Several gaps still exist and it is not clear why.)

6. *Increase the visibility of the problem with relevant national and international bodies*. The International Atomic Energy Agency (IAEA) and CTBTO were well represented at the senior staff level at WOSMIP, and both organizations seem to realize the importance of the issue; Grade: **B+.** (See Figure 1.1.)

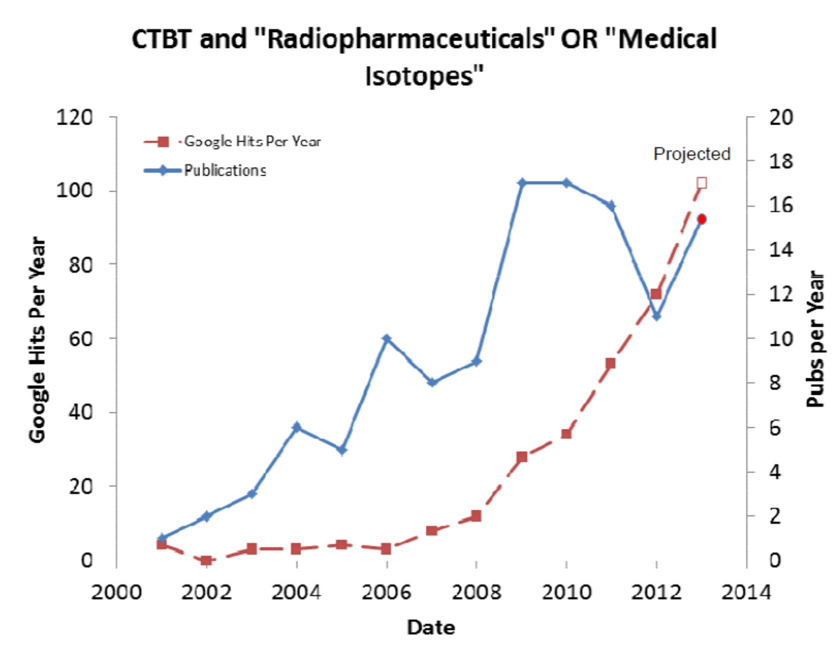

Figure 1.1. The impact of WOSMIP and other related collaborations

The fourth workshop continued this trend and enhanced capabilities on both sides of the subject. The goals of WOSMIP IV were to find ways to break down barriers, identify gaps that scientific investigations will fill, and to take advantage of new political decisions and new strategic relationships.

WOSMIP IV was hosted by the CTBTO and held at the Vienna International Center in November, 2013. The workshop began with a message from Mr. Randy Bell, Director of the IDC of the CTBTO, who stressed the importance of the workshop. A message was also delivered on behalf of Ms. Vorian Maryssael, Director of the IMS of the CTBTO, expressing appreciation for the participation by the medical isotope producers and acknowledging that past WOSMIPs have been very productive.

This report provides a summary of highlights from the 2.5-day workshop. Unavoidably, some presentations and events 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.

2.0 Introduction to the Problem

The first session of the conference provided overviews of both the nuclear explosion monitoring community and the ^{99}Mo production communities. While this session was a review to many repeat participants of WOSMIP, it was very important background to enable newer participants to fully understand issues faced by each community. These overviews were provided by the IAEA and by the Provisional Technical Secretariat (PTS) of the Preparatory Commission for the CTBTO. Immediately following the session, interested workshop attendees toured the IDC/Noble Gas Monitoring Station.

2.1 Overview of the Problem

The workshop started with an overview of some of the nuclear physics involved with both the production of the medical isotope ^{99m}Tc and nuclear explosions. This was an important review of material presented in previous WOSMIPs to convey that both processes release similar isotopic ratios of radioxenon into the air (Table 2.1). The global total emission from the four largest medical isotope production facilities is approximately four times higher than the emissions of all nuclear power plants worldwide; however, these releases are well below health and safety regulation thresholds.

Table 2.1. Sources of Radioxenon in the Atmosphere, with the Maximum Amount Produced for a Nuclear Explosion Shown in the Last Row. The actual amount released into the atmosphere from a nuclear detonation may be much less (>10^3) than this value.

Source	Order of Magnitude of Xenon Release
Hospitals	~10^6 Bq/day
Nuclear power plants	~10^9 Bq/day
Radiopharmaceutical plants	~10^9 - 10^{13} Bq/day
1 kTon nuclear explosion	~10^{16} Bq/day

The different processes release the same radioxenon isotopes but at slightly different radionuclide abundances. During fission of ^{235}U in a nuclear reactor, thermal (slow) neutrons are used, whereas during a nuclear explosion the fission is induced by fast neutrons. There is little time for complex activation build-up in a nuclear explosion (microseconds). There is sufficient time for production of many activation products during fission-based isotope production (days) or in a nuclear power reactor (several months), but the emissions from the production of medical isotopes can be very similar to a nuclear explosion because the dissolution happens very soon after irradiation of uranium. Because subtly distinct but similar radionuclide abundances are produced by the different processes, isotopic ratios of these fission products could potentially be used for source identification. Figure 2.1 shows the four-isotope plot that can be used to differentiate between nuclear explosions and reactor emissions if sufficient data are available. At the workshop, examples were discussed comparing fission simulations in irradiated uranium targets for radioxenon to real environmental and some stack measurements from medical isotope production facilities by plotting results on the four-isotope graphic shown in Figure 2.1. The theoretical data fit well for most facilities and only partially with the measurements from one of the facilities.

Additional ^{131m}Xe as a decay product of ^{131}I from other sources could be one possible explanation, as well as different operations within the facility.

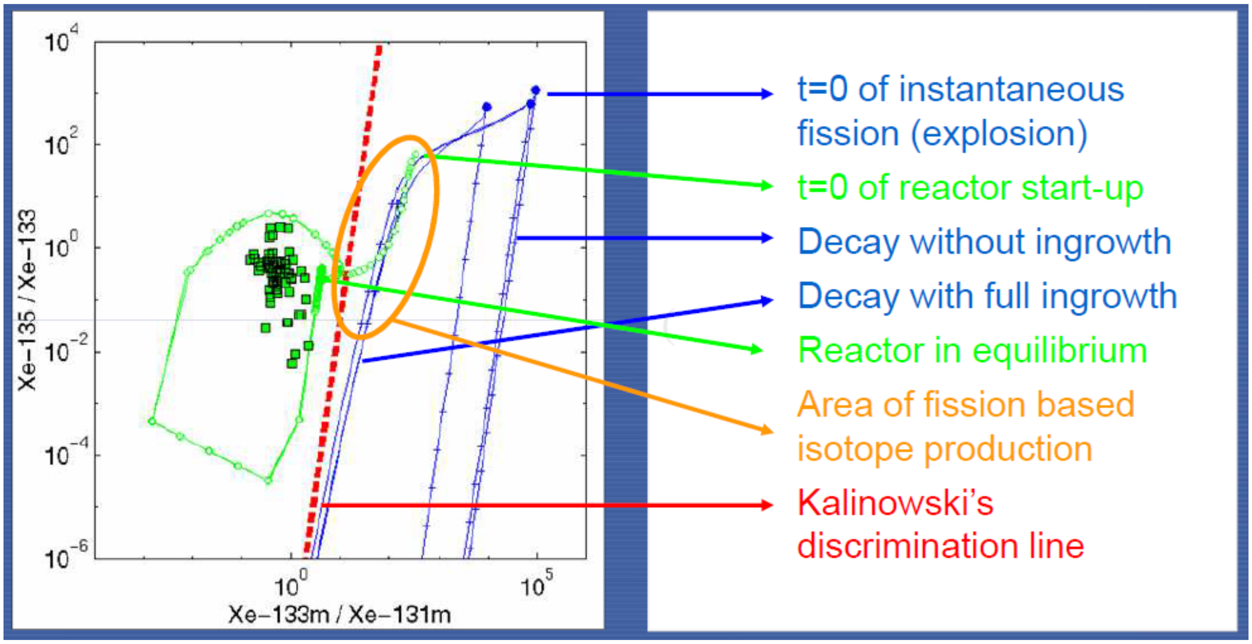

Figure 2.1. Plotting emission to differentiate between nuclear explosions and reactor emissions

Even though these isotopic ratios could potentially differentiate between medical isotope production and a nuclear explosion, the data from the two processes can be very similar and difficult to discriminate. High emissions from medical isotope production (MIP) facilities create a global radioxenon background that can create ambiguity for the highly sensitive IMS for nuclear explosion monitoring.

2.2 Overview of CTBTO Monitoring

An overview of the CTBTO monitoring processes explained the importance and difficulties associated with obtaining sensitive and accurate measurement of radioxenon isotopes in the environment for the detection of nuclear tests. Current processes involve measurement of radionuclide gases in all types of nuclear tests (atmospheric, underground, and underwater) as a verification tool to monitor CTBT. The same radioxenon isotopes that are released from medical isotope processes are used for CTBTO monitoring. The 80 particulate stations and 40 noble gas systems of the IMS are very sensitive and are able to see other natural and man-made variations such as radiopharmaceutical production, nuclear accidents and natural isotopes in the air. The CTBTO has shown that medical isotope production across the world is observed essentially every day. The background of xenon in the world from these civilian nuclear activities is shown in Figure 2.2.

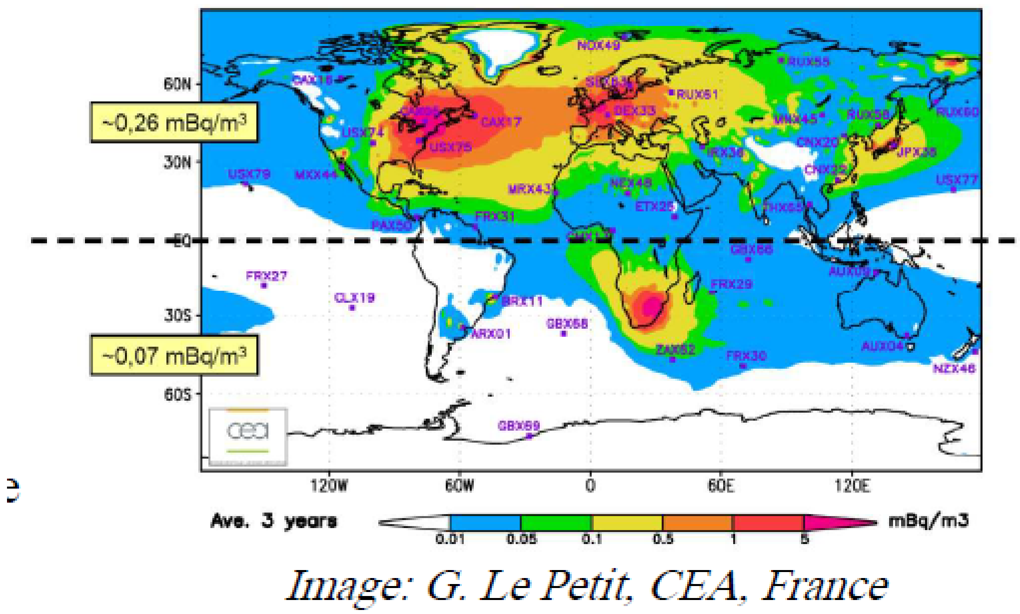

Figure 2.2. Noble gas background of xenon in the world due to civilian nuclear activities

Examples were presented to demonstrate the difficulties that this can pose for CTBT verification. Measurements from the Democratic People's Republic of Korea (DPRK) -announced nuclear test in February 2013 were presented and discussed. Figure 2.3 shows actual seismic and xenon measurements right after the nuclear test in February. The CTBTO needs to understand what is typical for the station and, if possible, be able to calculate the effect of known sources on detections.

No immediate abnormal detections were observed for the test, but 55 days later, on April 7 something unusual was seen. Table 2.2 shows the two reliable, simultaneous measurements. Additionally, small signals were seen on April 12–14, 2013 at detector station RUX58. Figure 2.4 shows the $^{131m}Xe/^{133}Xe$ station history, including all detections above minimum detectable concentration (MDC) after the Fukushima accident. A ratio around 2 percent is not typical for this station; rather this might be expected from processing of radiopharmaceuticals in a hospital.

There are two ways to handle the situation: reduce emissions and/or collect accurate information on xenon emission to calculate their contribution to the CTBT samples. Emission reduction is voluntary but recommended highly by the xenon monitoring community. In some cases emission reduction can be rather expensive if not considered in the facility design phase. WOSMIP has defined a target value for daily xenon emissions in the range of 5×10^9 Bq/d on each facility. There are different methods that can be used to reduce emissions (e.g., retention tanks, charcoal or silver-zeolite trapping, iodine trapping, etc.) and these were discussed in detail in the emissions abatement session of the workshop (see Section 5). The second half of the solution is for medical isotope producers to share emissions data with the CTBTO. To be able to calculate how large a contribution known noble gas sources pose to CTBT xenon detection, it is essential to know emissions from known facilities (i.e., stack monitoring data), source-receptor-sensitivity (computed daily in the PTS), and detections from the CTBT monitoring network. Emissions data based on an annual average have proved to be inadequate. The possibility of flagging the samples using a “xenon flagger,” in which the flag would represent expected contribution to the measurement from the known sources, was discussed. The CTBTO needs more accurate emission data to better understand these contributions from known MIP facility sources.

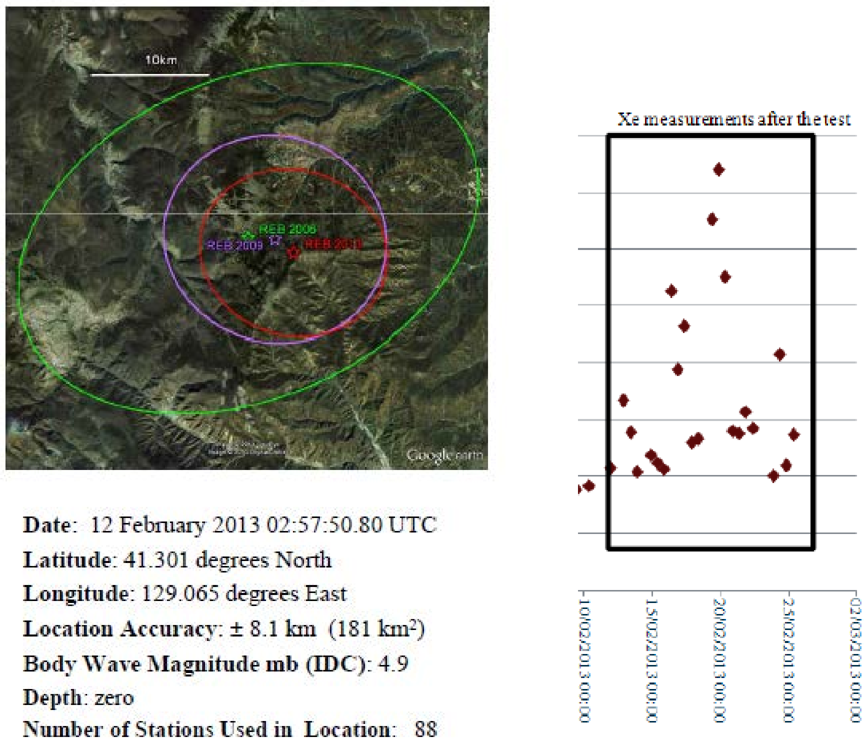

Figure 2.3. Actual seismic (left) and xenon (right) measurements after the DPRK announce nuclear test in February 2013 (Takasaki, Japan)

Table 2.2. Measurements on April 7 Following the Announced DPRK Nuclear Test

DETECTOR CODE	COLLECTION STOP	Xe-131m mBq/m³	Xe-133 mBq/m³	RATIO Xe-131m / Xe-133
JPX38_002	08-APR-2013 06:54	0.27	2.08	0.13
JPX38_001	08-APR-2013 18:54	0.73	3.05	0.24
JPX38_002	09-APR-2013 06:54	0.35	1.87	0.19
JPX38_001	09-APR-2013 18:54	0	0.72	0

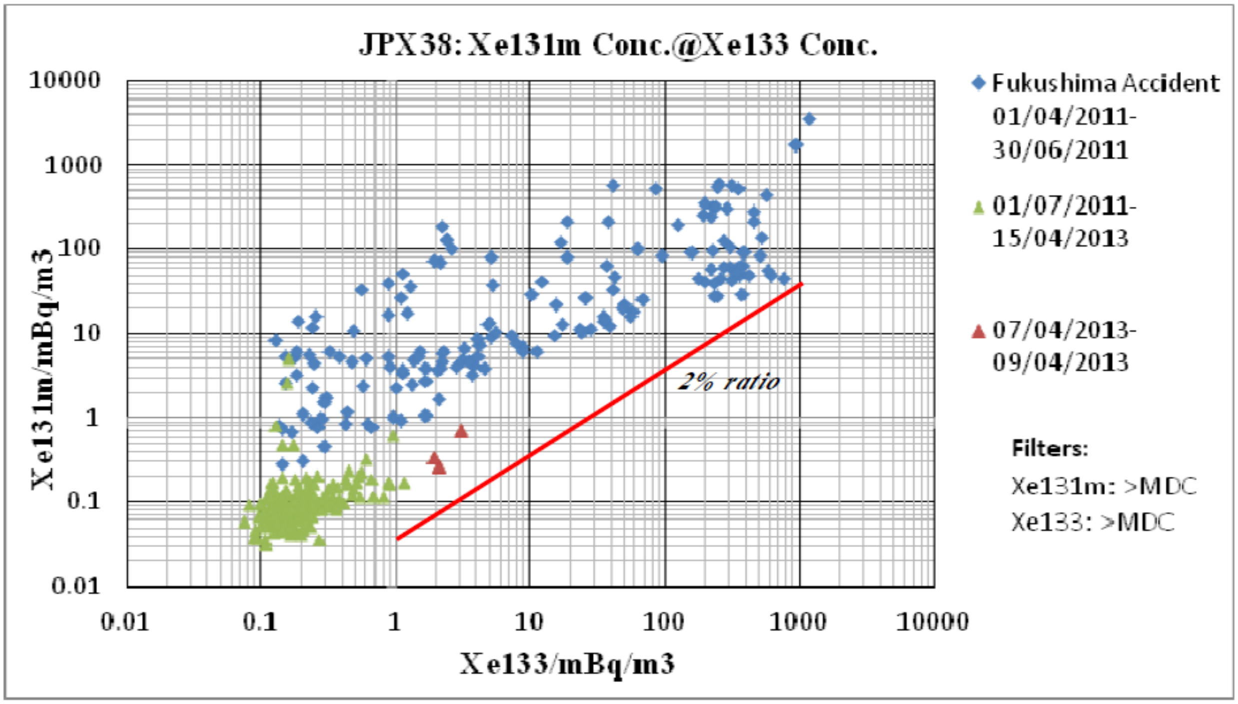

Figure 2.4. Xenon-131m/^{133}Xe station history, all detections above MDC after the Fukushima accident

2.3 Overview of Fission Production of Molybdenum-99

The overview session concluded with an update on the fission product production of ^{99}Mo at research reactors and accelerators worldwide. The importance of ^{99}Mo/^{99m}Tc production was stressed as they are versatile isotopes with broad medical applications and very high global demand.

A brief history of the more than 60 years of ^{99}Mo production was presented. The ^{99}Mo/^{99m}Tc market and supply chain development (including related policy decisions) created legacy issues that recently resulted in two worldwide supply crises. Measures taken from 2008 through 2010 mitigated the immediate supply threat but the risk of shortage remains. Recent changes in production were discussed, including Australian Nuclear Science and Technology Organisation's (ANSTO's) capacity increase, diversification in Europe, Russia commencing production, and SAFARI-1's and BR-2's increased production (in periods of shortage). Figure 2.5 shows the major current production routes, more than 90 percent of which are fission-based.

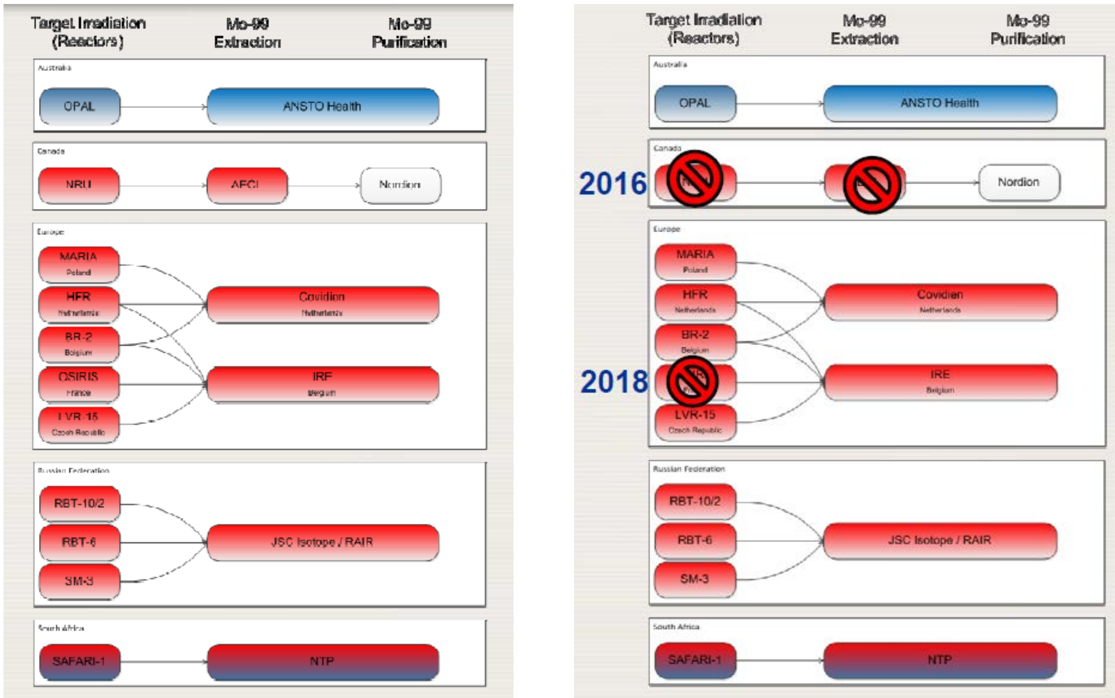

Figure 2.5. Current major production of ^{99}Mo and ongoing challenges. A majority of the world's producers still use HEU (red); ANSTO has switched fully to LEU (blue); and SAFARI-1/NTP is converting to LEU.

Molybdenum-99 demand is expected to continue to increase a few percent per year from both mature and emerging markets, and the current-reactor based supply is not sufficient to meet future demand. Supply shortages are expected as early as 2016 with the current fleet of producers. Actions to further reduce the supply risk include market and policy reforms, new production facilities and increased capacity, increased efficiency and better organization of clinical procedures, novel production technologies (some of which do not involve the fission process), and use of alternative radioisotopes with equivalent or comparable characteristics. Government support to address externalities remains indispensable. The IAEA is providing support to ensure ^{99}Mo supply security to all Member States while transitioning production away from highly enriched uranium (HEU). Potential future production for fission-based and non-fission-based methods as well as new research reactor projects is captured in Table 2.3 and in Figure 2.6. Future possible cooperation between the IAEA and WOSMIP/CTBTO could involve provision of current information on the production of ^{99}Mo via fission, and identification and support for targeted participants with possible co-financing of participation.

Table 2.3. Potential Future New Fission-based Production, Non-Fission-based Production, and New Research Reactor Projects

Country	Production Source
Potential future new fission-based production	
Argentina	New RA-10 RR + new processing capacity
Australia	OPAL RR + new processing capacity
Belgium	New MYRRHA (ADS-RR) (~replace for BR-2)
Brazil	New RR + new processing capacity
China	CARR reactor + new processing capacity (RR not yet full power)
Egypt	ETRR-2 + new processing capacity (hot tests done)
France	New JHR (~replace for OSIRIS)
Germany	FRM-II (existing European processing capacity?)
India	BARC RR + new processing capacity
Indonesia	GA SIWABESSY RR + new processing capacity (?) (foil /mod. Cintichem)
Korea	new KJRR + new processing capacity
Netherlands	New PALLAS reactor (~replace for HFR)
Poland	MARIA + new processing capacity
Romania	INR + new processing capacity (?)
Russia	RIAR RRs + new processing capacity
South Africa	New SAFARI-II (~replacement for SAFARI-I)
USA	B&W New solution RR + new processing capacity (?)
USA	Morgridge / Shine + new processing capacity
Potential non-fission-based future production	
Canada – domestic supply projects	• Advanced Cyclotron Systems Inc. • Canadian Light Source Inc. • TRIUMF • The Prairie Isotope Production Enterprise
USA	MURR + new processing/generator capacity (NorthStar, (n,γ))
USA	Accelerator (NorthStar)
USA	Accelerator / heavy water (AMIC)
New research reactor projects (capture and/or fission-based)	
Jordan	5 MW (+10) under construction + new processing capacity
Nigeria	6 MW planned + new processing capacity

Saudi Arabia	20 MW planned + new processing capacity
Vietnam	~10 MW (+20) planned + new processing capacity

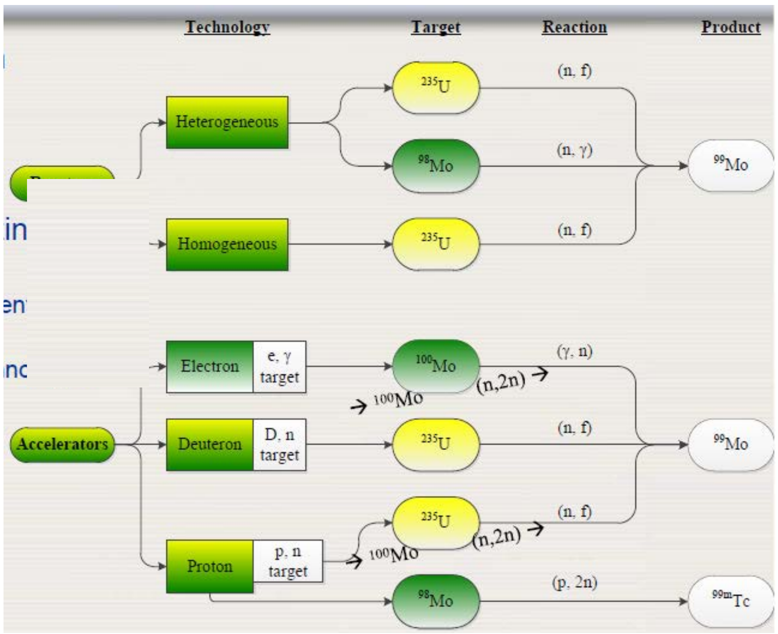

Figure 2.6. Potential future (non-HEU) production technologies

3.0 Current and Planned Molybdenum-99 Production Methods

The second session of the workshop included several presentations from current and future ^{99}Mo medical isotope producers. During this session many producers provided updates on the status of their current production methods, as well as on methods planned for future use. Different considerations related to potential xenon release from these various production methods and some plans for xenon mitigation were also discussed.

3.1 Production at NorthStar Medical Technologies, United States

Two different future production methods were discussed for both near-term and long-term production of ^{99}Mo at NorthStar Medical Technologies in the United States. The near-term method (expected to begin mid-2014) will involve a neutron capture production method in collaboration with the Missouri University Research Reactor (MURR). The long-term production plan (expected to start in late 2016) will involve the use of a photo-nuclear reaction, ^{100}Mo (γ,n) ^{99}Mo, with NorthStar's linear accelerator (LINAC) methodology. A new facility location is planned in Beloit, Wisconsin. A LINAC production method can potentially be used to respond to daily production needs as opposed to the weekly response reactor-based methods can provide. Both production methods use high-purity or enriched molybdenum target material, not uranium. Also, since both methods are non-fission based, no xenon production impact on monitoring is predicted.

3.2 Overview of the ANSTO Nuclear Medicine Project, Australia

The main objectives, design concepts, and timelines of the ANSTO Nuclear Medicine fission-based ^{99}Mo project known as ANSTO Nuclear Medicine (ANM) were presented. The ANM project is focused on continuing to supply Australia's ^{99}Mo needs beyond 2017 (which marks the end of the initial design life of the current facility) as well as on capitalizing on the commercial opportunity to export up to 3000 6-day curies of ^{99}Mo per week to the global community. ANM is also focused on providing a Synroc solution for the treatment of intermediate-level liquid waste from legacy waste and the new ANM ^{99}Mo production plant. It is also developing a capability to engineer customized nuclear waste treatment solutions based on Synroc technology. To date, the preliminary engineering designs for the ANM ^{99}Mo program have been completed and ANSTO is evaluating tender bids for the detailed design and construction. The initial design concept included a central hydrogen cell and two dissolver cells, each with its own bank of processing hot cells, and dedicated carbon columns (individually shielded carbon columns for safety and ease of maintenance). The planned completion date for the ANM ^{99}Mo program is mid- to late 2016.

The planned xenon emission control for the ANM ^{99}Mo project was also discussed. Some design considerations exist, along with future emission targets, cost and performance concerns, operational complexity and maintainability. The primary criterion is to keep emissions at or below current levels despite a three-fold increase in ^{99}Mo production. The emission control system is designed to have all air exhausted via carbon traps, whether producing ^{99}Mo or not. Gases from dissolution will be stored for 7 weeks in decay tanks. The gases in the hot cell atmosphere that exit via carbon column will have more than a 2-week delay. Design involves always having 1 carbon column online, which creates a 9-week delay before dissolution gases reach the discharge stack (60 columns, 12 hours service for each column).

The airflow and temperature will be controlled to approximately 10–15° C and columns will be monitored for performance/breakthrough. This emission control system design provides guaranteed delay/decay at all times and should control inadvertent or spurious emissions. The dominant design criteria for the ANM off-gas treatment system is the requirement for limiting the ^{133}Xe annual emissions to below the current notification level of 280,000 GBq. This translates to an average minimum of 622 GBq per run, based on 450 production runs per annum. An estimate of the predicted ^{133}Xe emission from ANM is provided in Figure 3.1.

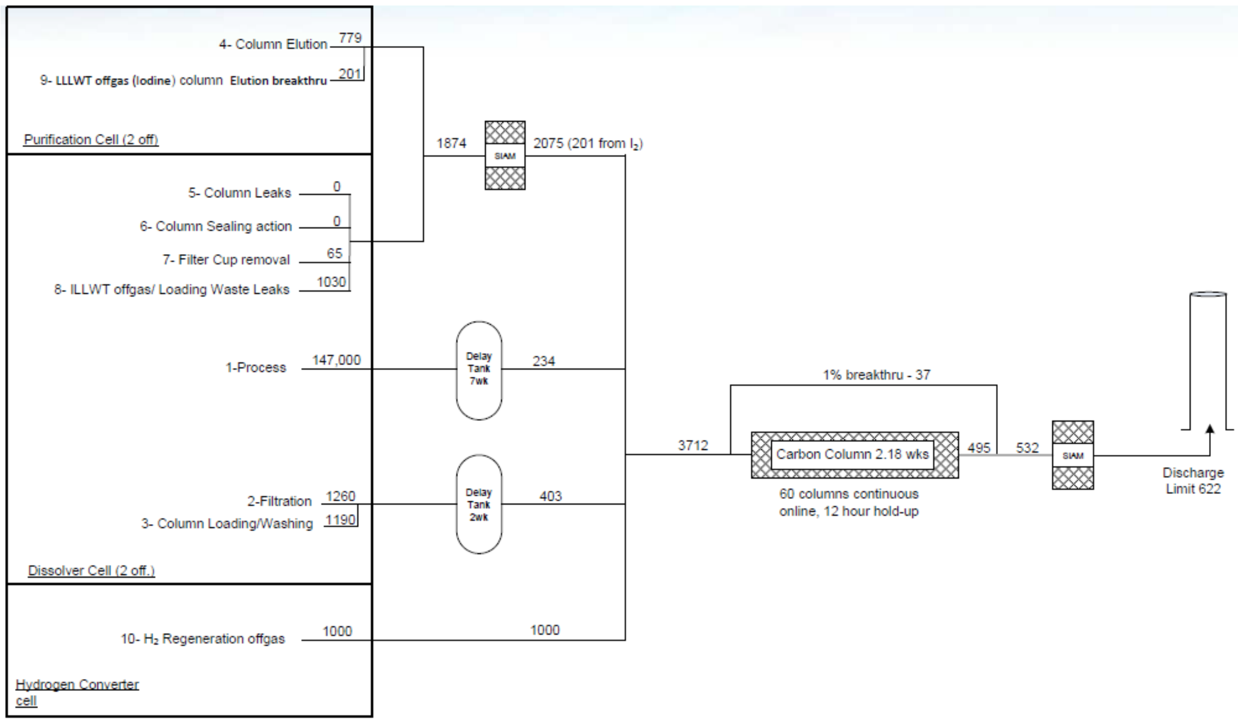

Figure 3.1. Estimate of ^{133}Xe emission for ANM (GBq)

Current estimates for a carbon column delay system that enables ANM to meet ANSTO's current annual emission limits of 280 TBq or 770 GBq per day run to $3.1 million AUD. Scaling up to reduce emissions to 5 GBq/day is estimated to add $7.1 million AUD to that estimate for the additional mechanical plant; if the additional building floor space and associated infrastructure requirements are included, the total estimate is $10.2 million AUD. ANSTO described the gap between their current emissions limits and the WOSMIP lower emissions goal as a funding shortfall.

3.3 Development Status at the Korea Atomic Energy Research Institute, South Korea

The Kijang New Research Reactor project with a fission molybdenum production facility has been operating at the Korea Atomic Energy Research Institute (KAERI) since 2012. This project is intended to address the major issue of the insecurity of the ^{99}Mo supply in Korea. The project is in the conceptual and basic design phase and KAERI is aiming to complete their ^{99}Mo process development by 2017. KAERI is targeting a fission-based ^{99}Mo production capacity of 2,000 Ci/week. Notable progress has been made in both low-enriched uranium (LEU) target and process development. Molybdenum separation has been demonstrated through cold experiments and now in a tracer experiment phase. Figure 3.2 provides a diagram of the overall fission molybdenum production process.

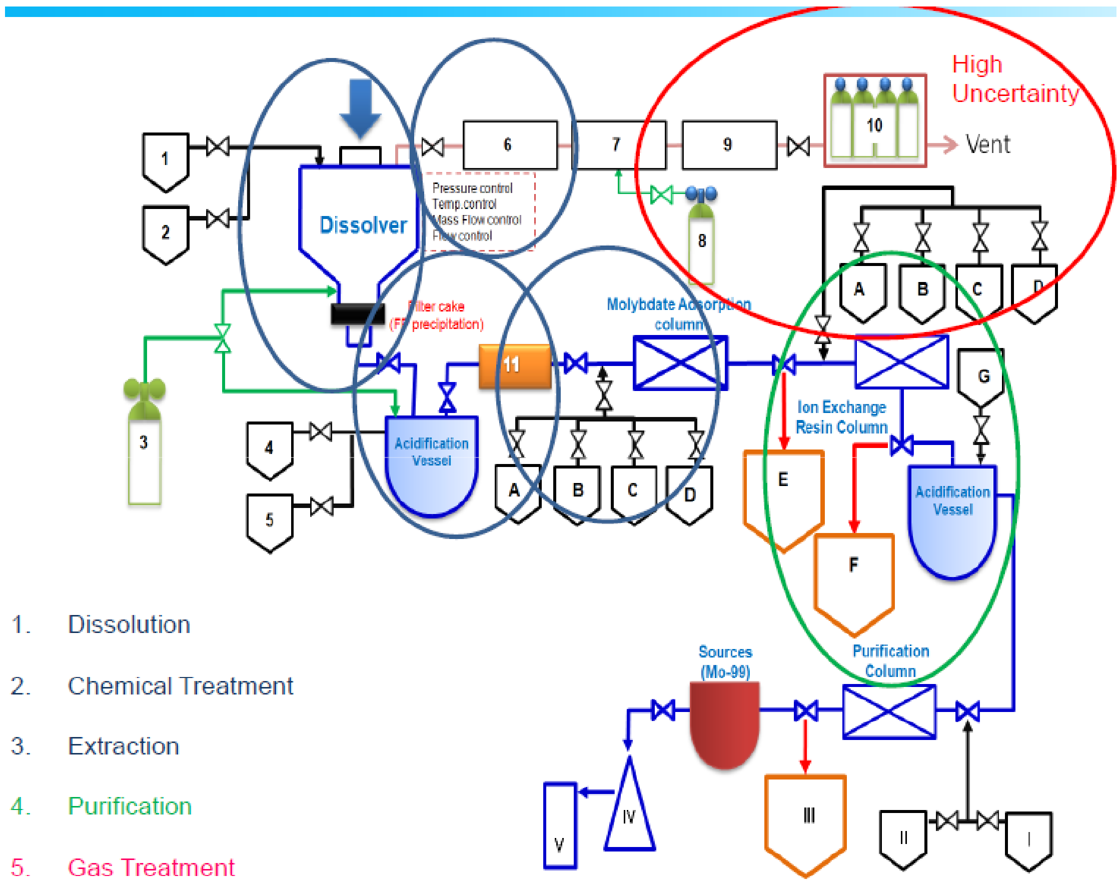

Figure 3.2. Overall fission ^{99}Mo process planned at KAERI

Current design concerns are related to the release of radioactive iodine and xenon. Figure 3.3 is a diagram of the conceptual gaseous waste flow. International technical supports and consulting on the facility design and waste management are necessary to successfully meet project goals and timelines.

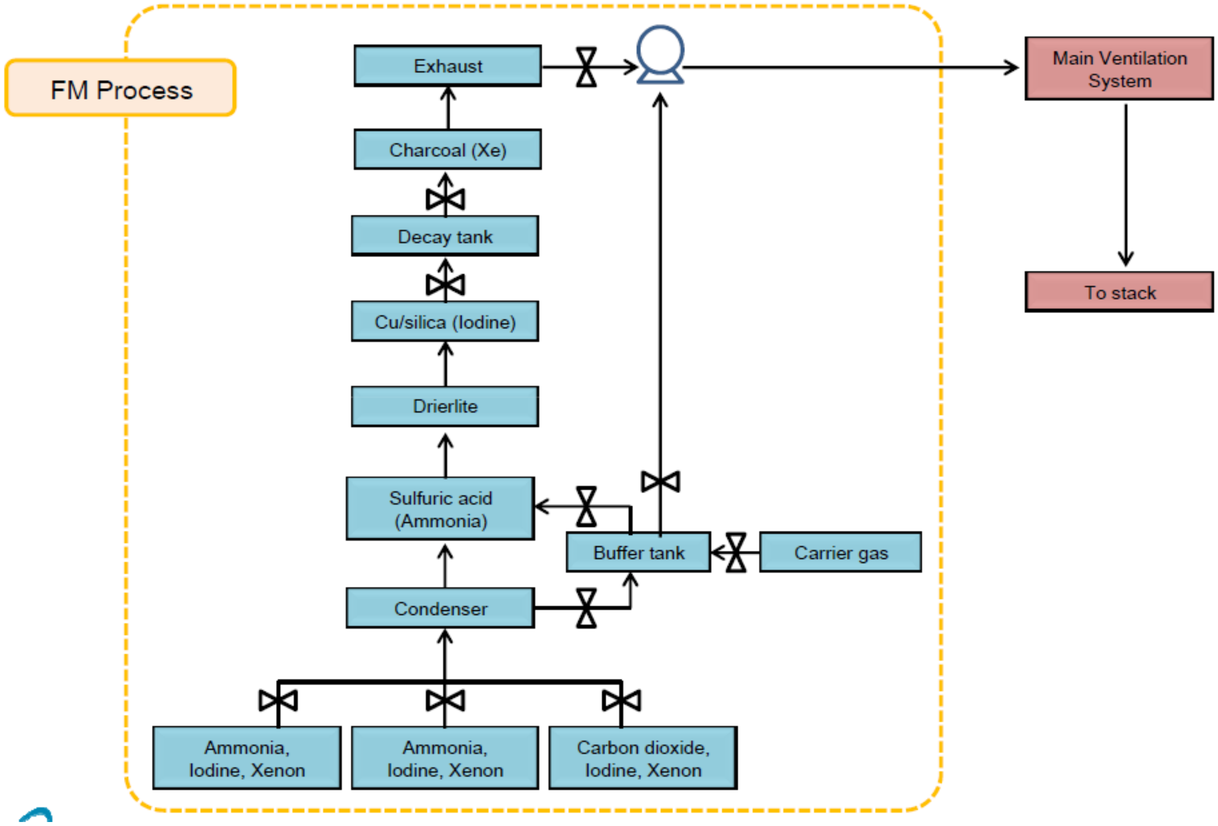

Figure 3.3. Conceptual gaseous waste flow for the fission ^{99}Mo process at KAERI

3.4 Production in the Radioisotope Production Facility - Atomic Energy Authority, Egypt

Atomic Energy Authority (AEA), Egypt presented the production stages of the ^{99}Mo fission-based production cycle at their Radioisotope Production Facility (RPF). The facility's current aim is to produce 500 Ci of ^{99}Mo per batch (activity concentration >10 Ci/ml, specific activity >27 KCi/g Mo). The process uses low-enriched uranium aluminide target plates (19.75% enrichment). A diagram of the ^{99}Mo fission production cycle using UAl_x target plates is shown in Figure 3.4. Details about the target material used, irradiation conditions and cooling time, and the chemistry and logistics associated with each production stage were discussed.

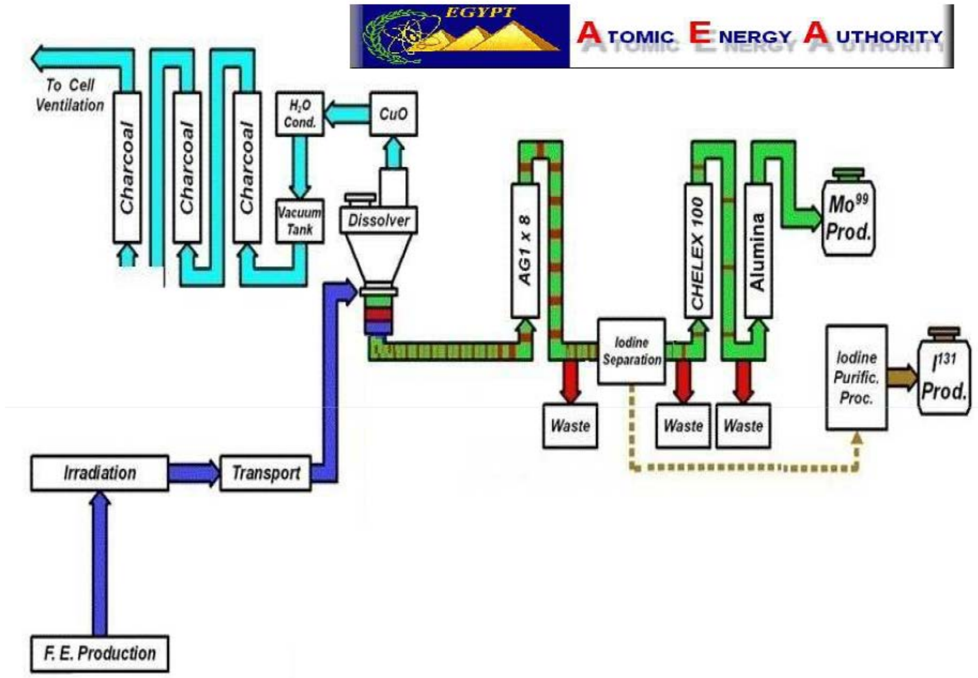

Figure 3.4. Molybdenum-99 fission production cycle using UAlx Target Plates at AEA

3.5 Plans at Coquí RadioPharmaceuticals, Corp., United States

An overview of the fission-based ^{99}Mo production plans at Coquí RadioPharmaceuticals Corporation of the U.S. was presented. The production plan includes twin production reactors, a radioisotope processing plant, a waste management unit, and support services and administration offices. Coquí will work with Investigaciòn Aplicada (INVAP) of Argentina, a company dedicated to the development of high-technology systems, to develop a production plan based on established technologies. The planned target weekly production is 7000 six-day Ci of ^{99}Mo. Coquí will collaborate with the University of Florida College of Engineering. The proposed location for the future facility is close to the University of Florida Reactor in Progress Corporate Park in Alachua, Florida. The current timeline includes finalizing financing for licensing stage in December 2013, submitting an environmental report by May 2014, submitting an application to the U.S. Nuclear Regulatory Commission (NRC) by August 2014, obtaining a construction license from the NRC by August 2015, and beginning commercial operations by December 2017.

3.6 SHINE Medical Technologies™ Planned Production System, United States

Plans for the SHINE Medical Technologies™ ^{99}Mo production system and off-gas control technologies were discussed. SHINE is developing a system that can potentially produce high specific-activity medical isotopes without a nuclear reactor. The new technology involves two key aspects: primary neutrons created by high-output deuterium-tritium source (full-power demonstration was achieved early 2013) and neutrons entering an LEU solution where they multiply sub-critically and create medical isotopes. The aqueous target simplifies subsequent chemical extraction processes and can be recycled for reuse. The plan for the initial construction is to produce nationally relevant quantities of ^{99}Mo and other medical isotopes at about 3000 six-day Ci/wk. An update on their recent progress included that their preliminary design has been completed, the environmental report and preliminary safety analysis report (PSAR) application have been submitted to the NRC (the environmental report has been docketed, and the first round of requests for additional information are complete), and a prototype target solution vessel (TSV) and off-gas system have been built. Future plans include beginning construction in 2015 and operations in 2017.

Aqueous fission can create many challenges, such as the radiolysis of water, which creates hydrogen, oxygen, and volatile fission products (iodine, noble gases). The planned facility radioactive gaseous emissions controls include a TSV off-gas system, a noble gas removal system, and the facility heating, ventilation, and air conditioning (HVAC) system. The TSV off-gas system is the primary system coupled to the production device. Figure 3.5 shows the different characteristics of this system, which include recirculating atmosphere, closed system at slightly negative pressure, Heat Exchanger 1 to lower the water content of gas, entrainment filter to allow only gas into system, silver-coated zeolite traps for iodine capture, catalytic recombiner for H_2 and O_2, and Heat Exchanger 2 to remove the heat of recombination and condensed water vapor. The noble gas removal system collects and processes TSV off-gas system purge gases after each irradiation cycle. The gases are compressed and stored for decay and then discharged to stack once the gas meets release criteria. The maximum hypothetical accident consists of a release of the contents of this system. The facility HVAC is a closed/open loop system with high-efficiency particulate absorption (HEPA) filtration and four cascading differential pressure zones to limit spread of airborne materials.

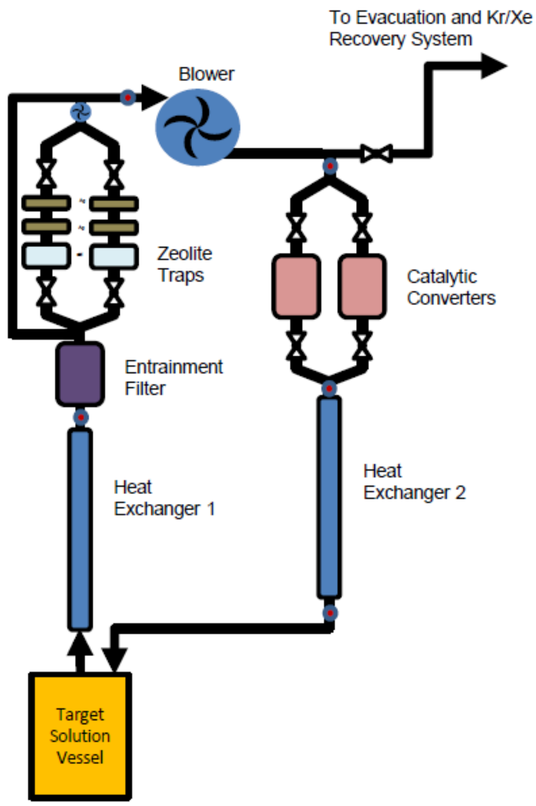

Figure 3.5. Target solution vessel off-gas system for SHINE Medical Technologies

Discussion ensued regarding gaseous release limits based on NRC requirements compared to the voluntary release limit of 5×10^9 GBq/d accepted as a goal by the WOSMIP community. SHINE showed calculations to explain that the hold time would potentially have to double from the 40 days required to meet NRC limits to 80 days to meet the WOSMIP-agreed goal. SHINE explained that the WOSMIP goal would not be achievable without financial assistance (similar to the discussion from ANSTO above). Implementing methods to reduce releases during facility startup can also present challenges because adding the scope and costs associated with increased abatement can be difficult for a company that is not well-established.

In addition to the update provided by SHINE Medical Technologies™, Argonne National Laboratory provided an update on the Mini-Subcritical Hybrid Intense Neutron Emitter (Mini-SHINE) experiments. These experiments are integral for the development of this new ^{99}Mo production technology using an accelerator-driven aqueous target solution. Data collected from the mini-SHINE experiments will be important to the final design of the off-gas-treatment and molybdenum recovery systems for SHINE. Micro-SHINE experiments are underway and uranyl mini-SHINE experiments were predicted to begin in December. Argonne National Laboratory is also assisting the development of the production methods in cooperation with NorthStar Medical Technologies (discussed in Section 3.1).

3.7 Status of Production Methods at BATAN, Indonesia

BATAN discussed their current radioisotope production capability and progress on their efforts to minimize xenon release from the fission product separation of ^{99}Mo. Recent activities at the facility include conversion from HEU to LEU targets with a modified Cintichem process for the separation of ^{99}Mo (Figure 3.6), production of various radioisotopes such as ^{99}Mo, ^{131}I, and ^{192}Ir, and preparation of ^{99m}Tc-, Na-I Oral and Na-I Hip radiopharmaceuticals. A typical ^{99}Mo production run is designed to produce 50 six-day Ci with 25% being exported internationally. In their efforts to reduce and monitor radioxenon releases, BATAN is working to optimize cryogenic conditions and facilitate the collection and analysis of monitoring data. Training was requested to teach staff how to analyze the high-resolution detector system recently installed by Pacific Northwest National Laboratory (PNNL) and the CTBTO in the facility stack. In collaboration with PNNL and CTBTO, BATAN also performed studies with a locally positioned transportable xenon laboratory (TXL) Swedish Automated Unit for Noble gas Analysis (SAUNA) system.

Planned activities include upgrading production quantities and building a new reactor. A business plan that involved isotope production reactor development with an aqueous homogeneous reactor technology was discussed. The current roadmap shows potential production with the new reactor technology by the end of 2017.

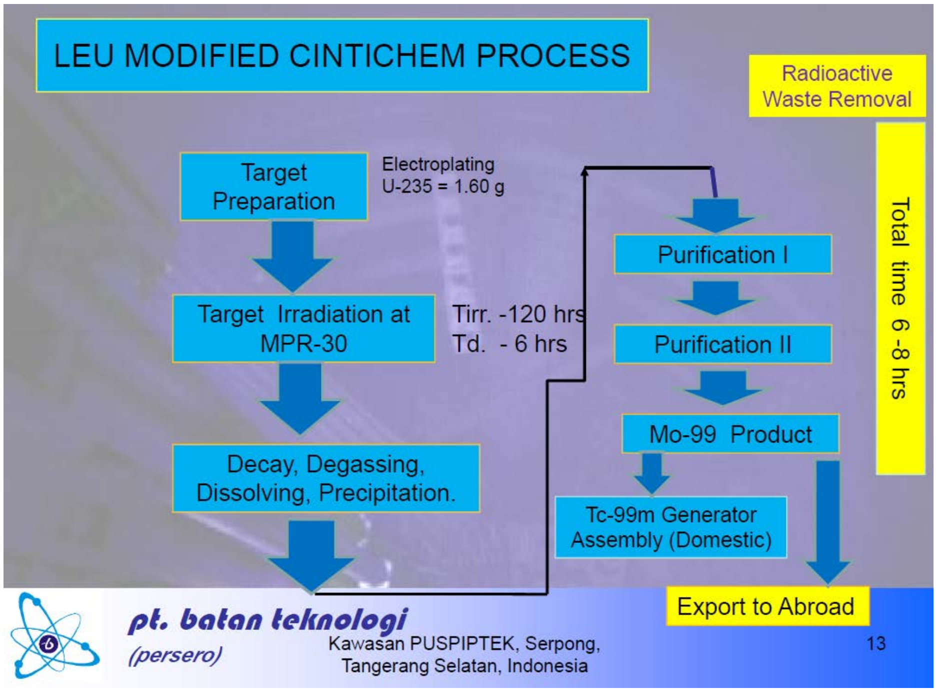

Figure 3.6. Process steps of the LEU-modified Cintichem method at BATAN

3.8 Additional Developments for Production

The Institute for Nuclear Research (INR) in Romania presented its infrastructure for potential production of radioisotopes and its progress in ^{99}Mo production method development. The INR has the ability to manufacture targets using LEU metallic foils, irradiate them in a TRIGA 14-MW reactor, ensure handling, disassembling, and chemical processing of the target in the existing hot cell facility, and handle, treat, and condition radioactive waste. Several fume hood experiments were conducted to demonstrate the Cintichem process: all gases resulting from target dissolution were retained in a cold finger and iodine trap. The quality of the final sodium molybdate solution is satisfactory; the yield of the process still needs improvement. More experiments are planned to improve yields and conduct work inside the hot cells. Additionally, procedures will be developed for high-level radioactive liquid waste treatment.

Developments in high-density MoO_3 pellets were discussed for potential ^{99}Mo domestic production in Iraq. To enhance supply to the country's current medical isotope demand within a national program, an electron accelerator technique was suggested. The idea of this project is to produce ^{99}Mo using a high current linear electron accelerator. Technetium-99m can be obtained in the course of photonuclear processes with ^{100}Mo used as a target. Various experiments were conducted to test different high-density MoO_3 pellet production methods, including various sintering techniques such as natural convection, microwave, and radiofrequency induction furnaces. Various measurements were performed on the pellets for characterization and results were presented. Future irradiation tests with these high-density pellets require collaboration with another facility.

4.0 Monitoring Technology and Data Sharing

4.1 Atmospheric Transport Modeling

4.1.1 Overview of ATM use for Monitoring

The U.S. NDC presented an overview from a monitoring user's perspective explaining how ATM is used for nuclear explosion monitoring. Radioisotope data alone may not identify the source (e.g., may not contain timing information or unique source information) so a combination of various technologies is used to aid monitoring identification efforts. Meteorology, specifically ATM, and waveform technologies, which may identify a specific event time and location, are used. Challenges arise when no waveform event correlates to radionuclide detection and when there is a highly variable radionuclide background with multiple possible sources.

The methodologies of meteorology and ATM were briefly described. Forecast models transform atmospheric measurements into predicted wind fields. Transport and dispersion models use the wind fields as input to estimate and calculate the movement of material from one point to another. The resulting transport models can then be used in forward and reverse modes. The models are then used to simulate conditions in the IDC's field of regard, which provides the general source region of air sampled at a site during a given collection; forecasts which sites may detect a potential debris cloud; and applies diagnostics, which involves the application of ATM to determine the most likely source of detected radionuclides.

Examples were provided for using ATM for both forecasting a plume and diagnosing specific cases. Figures 4.1 and 4.2 show its diagnostic use. It was stressed that ATM is an essential component of the radionuclide technique for nuclear explosion monitoring. Multiple possible sources may exist in a region, which can make identifying a specific source difficult, so the more that is known about these possible sources, the better these tools can be used to identify sources of detected radioisotopes. This is especially true for MIP; the more information and data are gathered about production methods and releases into the environment, the better these tools can be used.

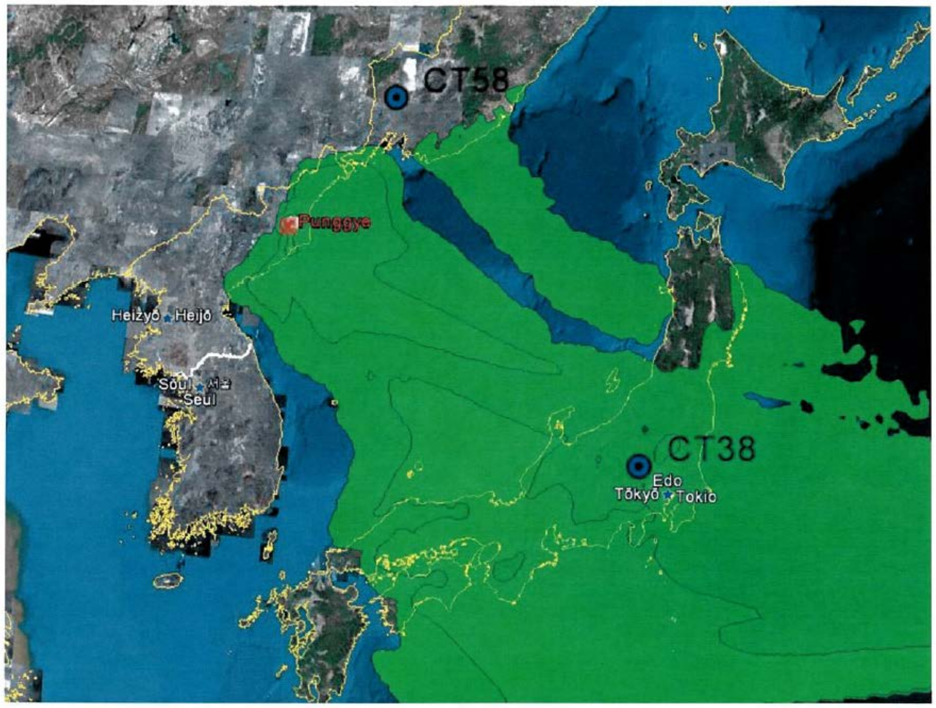

Figure 4.1. April 2013 forward plume projection following the announced DPRK nuclear test

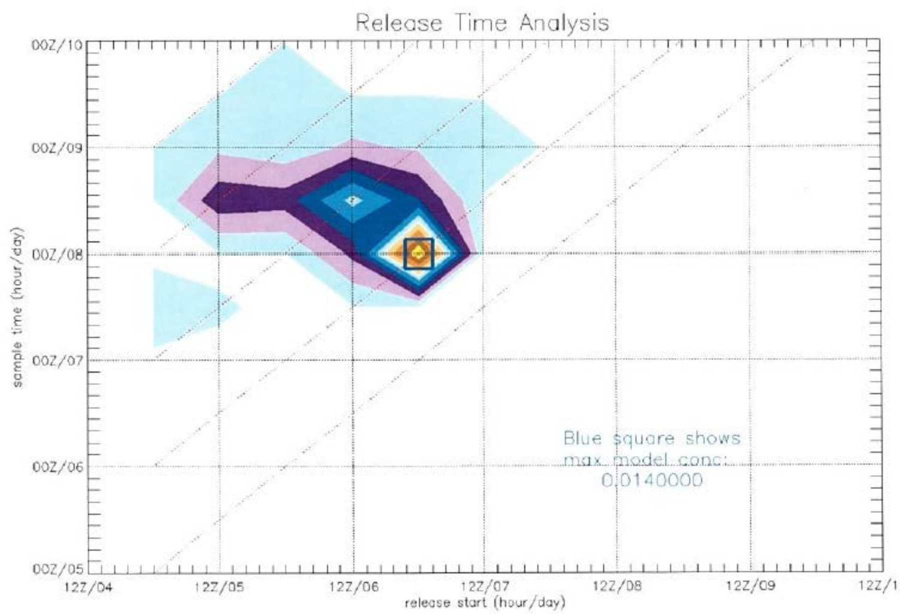

Figure 4.2. Release time analysis for Takasaki xenon collections

An interesting idea that was presented later in the workshop but that is related to the current topic was the concept that emissions from ^{99}Mo production facilities could be used to develop verification techniques for ATM. A presenter from Northwest Institute of Nuclear Technology stated that, although mitigation methods are very important, in the meantime it may be possible to use the released noble gases as trace gases for studying field observations of ATM. The stack monitor can provide the source term of the xenon isotopes being released from a specific medical isotope facility, and this source term could be monitored by several mobile stations within approximately 1000 km. Long-term stack and field observation data can be obtained and used to improve ATM tools, which help improve CTBT verification.

4.1.2 Predicting Effects of MIP

ATM can be used to predict the potential effects of medical isotope production on nuclear explosion monitoring. Modeling was used to determine that for most locations, medical isotope emissions in the range of ~5×10^9 Bq/d are an acceptable goal for reducing the impact on nuclear explosion monitoring (Matthews and Bowyer 2013). At levels higher than 5×10^9 Bq/d, a constant presence of xenon can cause a background that can be described as a xenon "fog." This xenon background may be subtracted, but this "fog" is the same isotope nuclear explosion monitors are looking for and therefore it affects the statistical precision to which they can subtract it. To further the analogy, stack monitoring can shed some light into the "fog," but does not solve the problem completely.

A specific example was discussed to show how production on the Korean peninsula may affect future measurements made by the IMS. As discussed in previous presentations and earlier in this report, approximately 55 days after the seismic event that followed the DRPK-3 announced nuclear test, two xenon isotopes were detected at the JPX (Takasaki) and RUX58 (Ussuriysk) stations. The results were consistent with a nuclear test; MIP could be ruled out. However, if nearby MIP facilities had been operating, there could have been real and surmised problems in discriminating this event from an admixture that included medical isotope emissions. Had such an admixture contained a factor of two of the amount of ^{133}Xe, the ability to age-date this event could have been obscured and an erroneous conclusion might have been reached. Serious doubt as to the nature of the event might have entered into people's minds if medical isotopes had been in production close by (as demonstrated in simulations and shown in Figure 4.3 and Figure 4.4). This is exacerbated by that it is not always possible to use the more accurate two- or three-isotope plots if multiple isotopes are not found. For example, following DPRK-1 at the Yellowknife station, a single isotope was measured. Pertinent to this discussion, as mentioned in Section 3.3 of this report, production of medical isotopes in South Korea is planned.

Calculations have been performed at various levels to predict the effect of emissions, and have confirmed that the previously estimated maximum releases of 5×10^9 Bq/day would be low enough to have minimal effect on international monitoring.

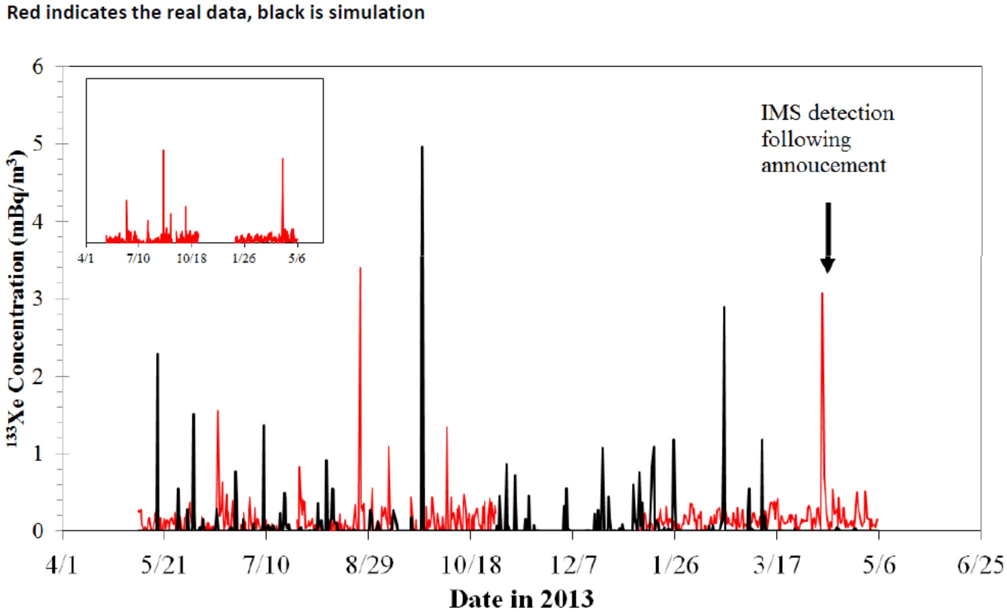

Figure 4.3. Medical isotope simulations for Takasaki

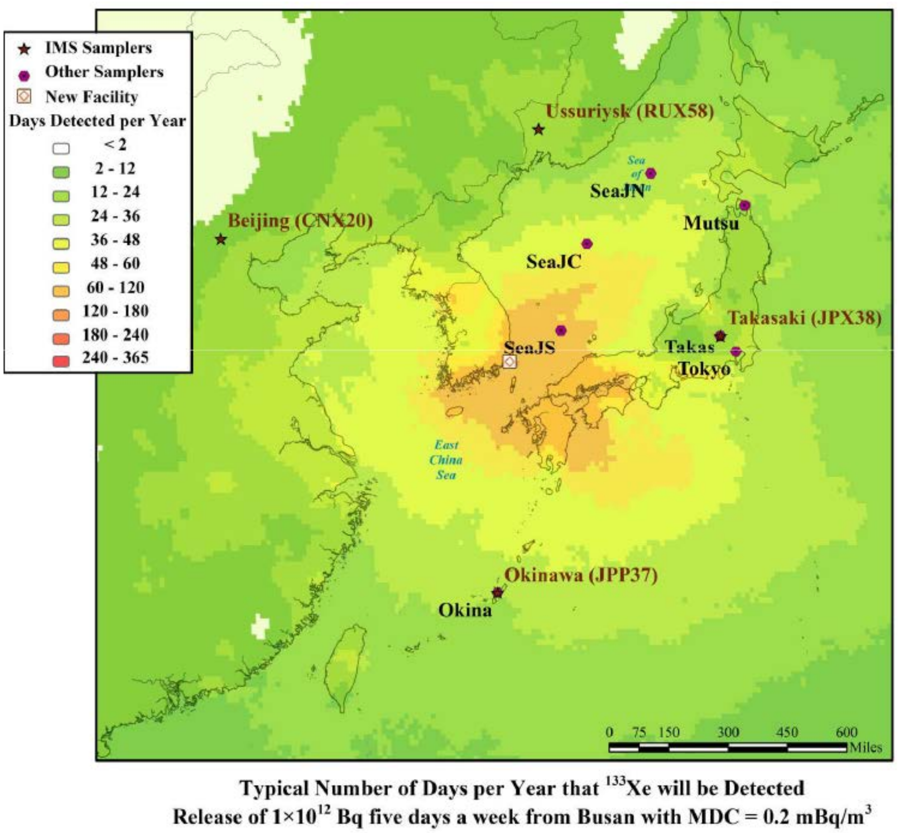

Figure 4.4. Annual detections expected at 10^{12} Bq/day

4.2 Detector System Technologies

As stated previously in the discussion of production method updates at BATAN (Section 3.7), a high-resolution detector system was recently installed by PNNL and the CTBTO in the BATAN/BaTeK medical isotope facility stack. More details about the design requirements for the stack monitor, final system schematic and specifications, installation, and data collection and analysis were presented. The system is portable and modular and consists of a LaBr detector, a sample beaker (1.2 liter) with inlet and outlet valves, lead shielding, a charcoal filter unit, and an air flow monitor. Figure 4.5 shows an example spectrum from the stack monitor system. It performs with good resolution and data are saved every 10 minutes. Software tools with the system allow high-speed data viewing and analysis and processing months of data in a few minutes. Also, results from the studies performed with a locally positioned TXL SAUNA system were discussed. The stack monitor data (~1-2×10^{12} Bq ^{133}Xe) were compared to the releases calculated using ATM and TXL data (~3×10^{12} Bq ^{133}Xe).

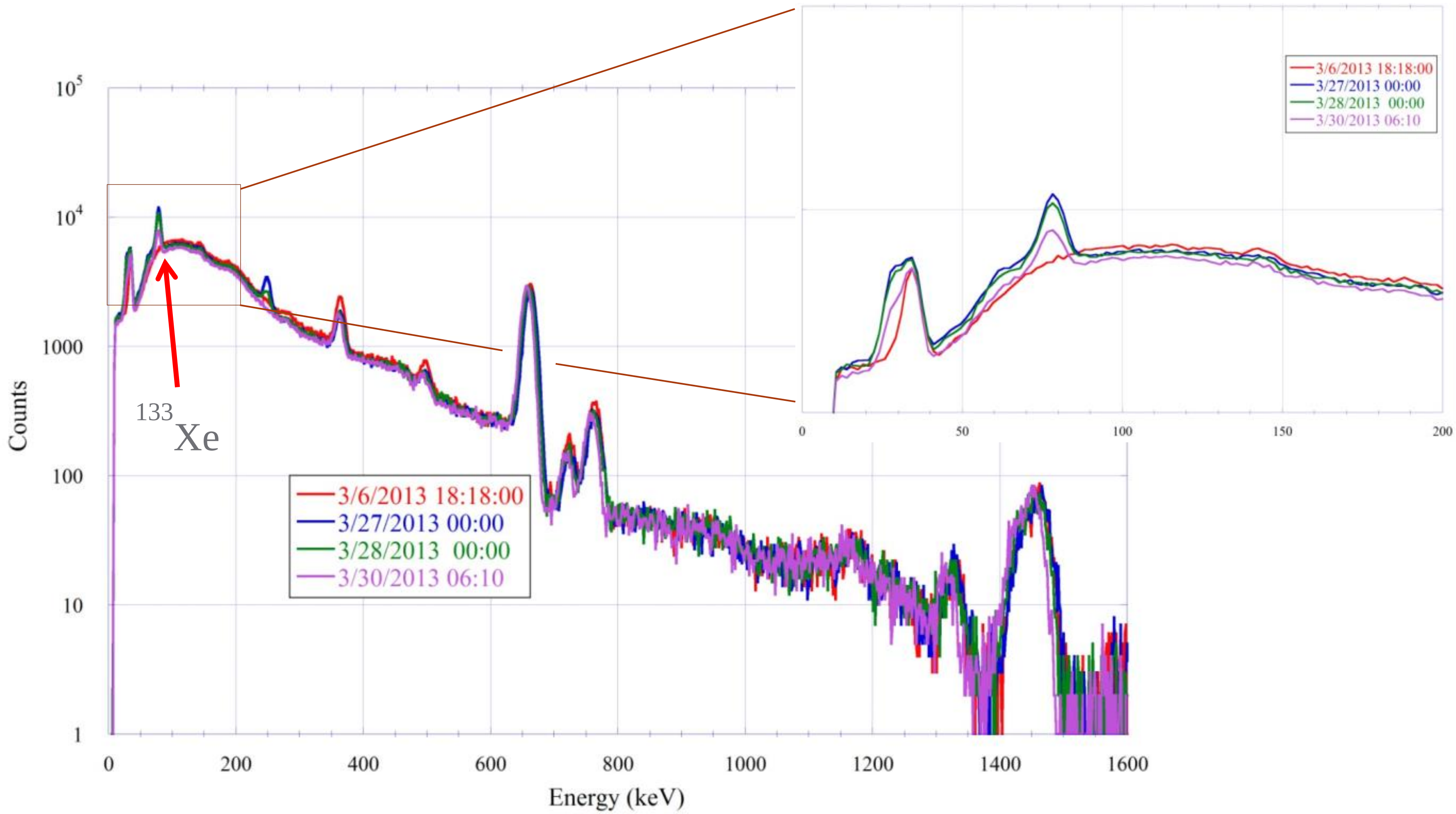

Figure 4.5. Example spectrum from the high-resolution stack monitor system recently installed at BATAN/BaTeK

INVAP of Argentina presented the general design features, start-up measurements, and new designs for a stack monitor for Argentinian isotope production facilities. The current gaseous effluent monitor (AEMi) generates real-time data regarding some specific isotopes and quantity release in the stack emission. Its general design features include an aerosols, iodine, and noble-gas air effluent monitor; stand-alone online noble-gas gamma detection channel; and minimization of iodine plate-out and aerosols deposition in the sampling lines. Xenon emission was determined from gamma spectra taken online by the AEMi NaI detector. Gross counts of the 81 keV (^{133}Xe) and 249 keV (^{135}Xe) peaks counted from a single channel analyzer were used to calculate emission. During start-up measurements at the radioisotope production facility, samples taken in a 100-mL container at the exit of the AEMi were measured with only a 5-min. delay in a high-purity germanium (HPGe) detector for normalization. Figure 4.6 shows the measurements collected over time. The current system keeps track of noble gas emissions online, with subsequent iodine measurements off-line. After analyzing the emissions data of the start-up runs, a correlation between emission peaks and operations inside the hot cells was performed. The highest emissions were recorded when the H converter regeneration started and during conditioning of hoses for iodine purification. Dissolver gases coming from washing solution charge and those coming from AG elution are collected in the buffer tank. These gases are released to the hot cell when there is need to use the tank. Gases produced during the decay of the iodine solution in ^{99}Mo cell are stored in the purge tank. These gases are released before the next production process. They should be stored in a tank allowing, them to decay before being released to the cell. As a result of this analysis, and with the purpose of minimizing emissions, more storage tanks are foreseen in future designs. The lesson learned from this study is that even if the goal is start-up, actions must be performed to keep emissions reduction in mind. A design for a new detector that would potentially include two detectors for noble gas measurement online with aerosols and iodine measured continuously “almost online” (beta for aerosols, gamma for iodine) and off-line with better resolution (with an HPGe detector) was discussed at the end of the presentation.

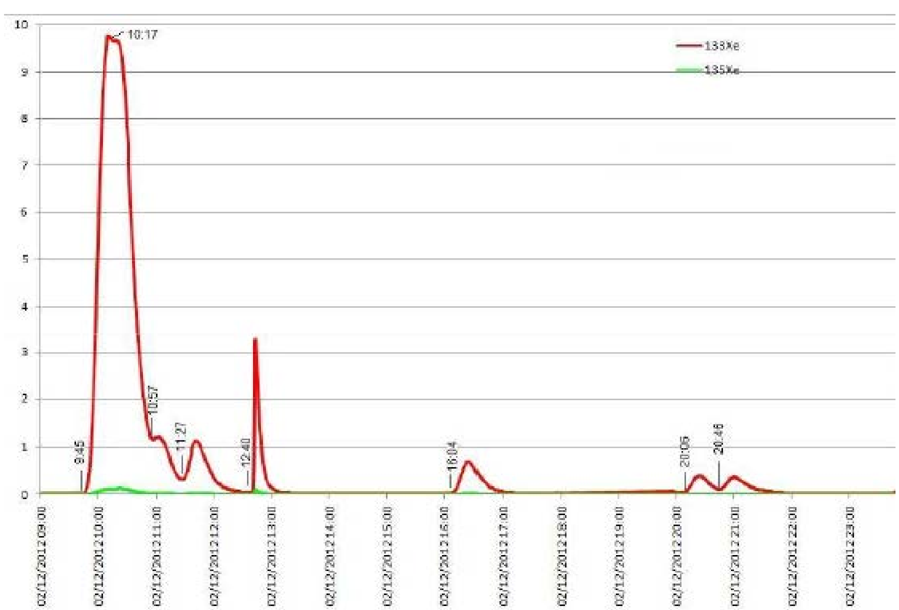

Figure 4.6. Emission of ^{133}Xe and ^{135}Xe during a ^{99}Mo run during start-up measurements with AEMi stack monitor at a radioisotope production facility

4.3 Stack Data Management and Visualization

A representative from Health Canada presented progress on stack monitoring with Atomic Energy of Canada Limited (AECL) and other areas of CTBT cooperation. Current project goals are focused on improved understanding of Chalk River Laboratories (CRL) emissions and their impact, automated processing of data collected from NaI stack monitoring equipment, and historical archiving of emissions data with an impending MIP shutdown in 2016. AECL has provided information to Health Canada and CTBTO community on CRL emissions but it has been a time-consuming manual process to extract data, and reports are in units inappropriate for regulatory requirements. A more flexible and automatic solution was desired to assist both AECL and Health Canada. Health Canada and AECL collaborated to develop a convenient emissions analysis platform and the details about the system, data export, Excel® data display, charting capability, etc. (Figure 4.7). This convenient emissions platform is better for reporting data for regulatory purposes, CTBT monitoring, and other analyses by Health Canada. It also provides more complete emissions records. AECL has expressed a concrete willingness to continue their support to the CTBT community and are considering they can best enhance their support.

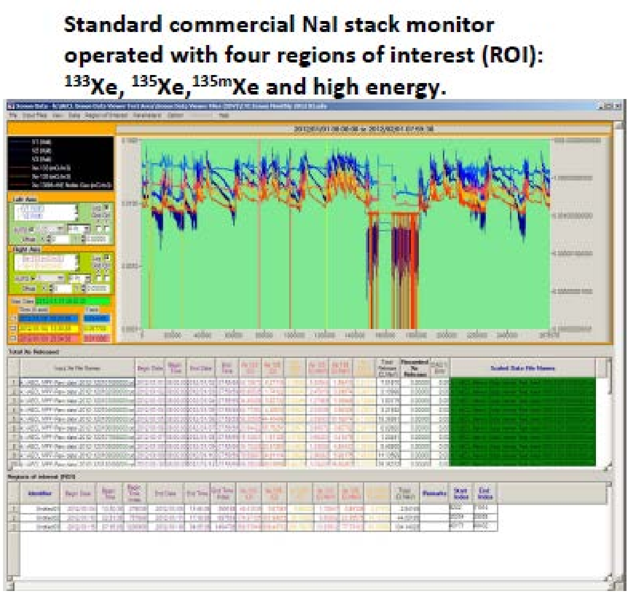

Figure 4.7. Health Canada and AECL developed convenient emissions analysis platform

ANSTO also discussed using web-based data visualization software to monitor emission trends at ^{99}Mo production facilities. ANSTO's real-time environmental data, project to upgrade the supporting software applications, and new data visualization tool were discussed. ANSTO had a need for a new tool because their old tools involved old programming languages that their IT department no longer supports; they needed to maintain multiple programs and databases; they were not able to retrieve some data; and they had limited data analysis and trending. ANSTO designed their own web-based application known as the environmental monitoring system (EMS) to suit their specific needs with a uniform platform on centralized servers (Figure 4.8). The new tool provides cost savings on software and license fees, can maintain and refine system over time, provides calibration and change traceability, and offers long-term storage of historical data. It also allows internal and external access to various types of real-time data—

weather, radiation stations, and emissions. Various features of the EMS tool were demonstrated and discussed. Data from recent ^{99}Mo stack monitor measurements were shared. The potential to share this application with WOSMIP colleagues was also discussed.

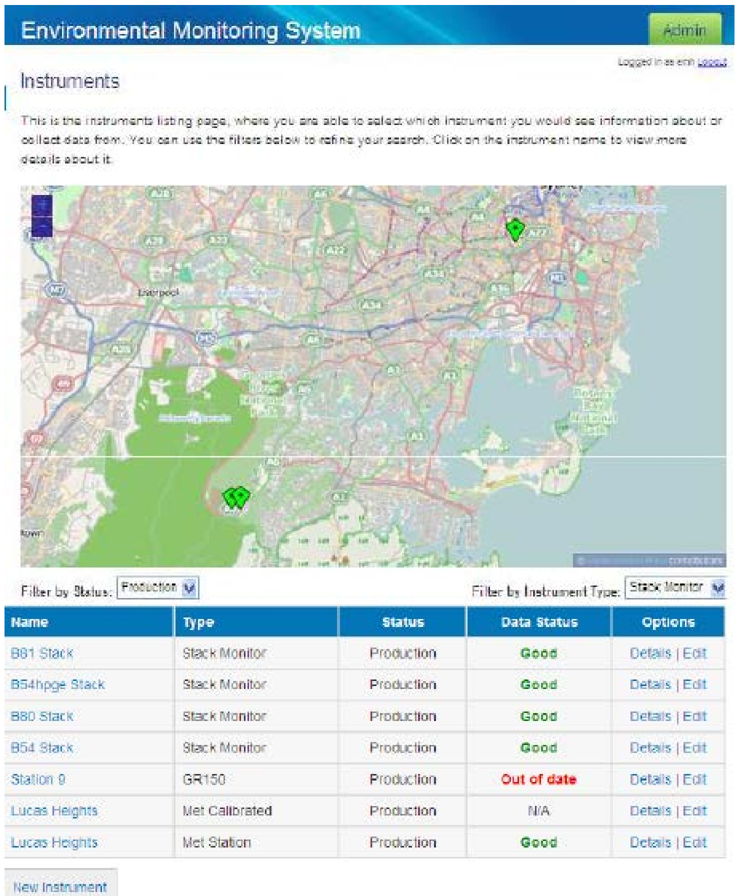

Figure 4.8. ANSTO environmental monitoring system tool

4.4 Supply and Use of Stack Monitoring Data

The importance of sharing stack monitoring data has been thoroughly discussed during all WOSMIPs, and both the monitoring and medical isotope communities understand and acknowledge its value. The next step is for both communities to agree upon a plan to share and use the stack data. A roundtable discussion was held to foster communication between both communities and to reveal the potential barriers and challenges associated with sharing data. Most of the discussion focused on the need for the CTBTO to share a standard operating procedure for protecting the data with the medical isotope producers. It was discussed that stack monitoring data will only be used by state parties (upon request) for treaty verification purposes only. Producers requested documentation from the CTBTO explaining that stack data are not openly available, defining who owns the data, and defining the partnerships, modalities, and confidentiality provided by CTBTO. Additional points and questions discussed at the roundtable are listed below.

- Through data sharing and other collaboration, the CTBTO can show how MIP is a peaceful and safe use of nuclear technology and help to rectify false public health risk claims
- Even with emission reduction, the CTBTO still needs data to verify a detection is not MIP
- Who will pay for the additional technology needed for data sharing, and who sets it up?
- What are the data requirements (i.e., confidence levels)?
- Data sharing could be a two-step process: start with historical data to improve models and then move to on-line sharing

5.0 Emission Abatement

5.1 Good Practices

A scientist from PNNL discussed the main principles to be followed for achieving low emissions. The three principles include operational sharpness, chemistry, and abatement systems/engineered systems (Figure 5.1). Operational sharpness involves highly trained and experienced staff, routine operations, and optimal facility operation at all times. Chemistry can be optimized for low emissions by focusing designs on capturing and isolating iodine (which decays to xenon) as a primary goal, using chemical steps that are automatable in sealed/closed systems and developing waste management procedures that can be performed in closed/isolated systems. Abatement systems/engineered barriers involve capturing all gases from the process and retaining them until the decay of ^{133}Xe released is less than 5×10^9 Bq/d. About 8 weeks of retention in tanks or on charcoal could be required to meet this limit, although this is variable depending on production amounts. Xenon and iodine retention on charcoal is optimized by lowering air flow, dehydrating air, and cooling air (and cooling the charcoal beds).

As discussed in previous presentations from the medical isotope producers, these abatement systems are significantly more restrictive than required for regulatory controls, and thus more costly. Stack monitoring requirements were also presented as part of this discussion. For stack monitoring data to be efficiently used by the IDC and NDCs, isotopic information (^{131m}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe), large dynamic range, time resolution in 3-hour intervals, and spectral data are needed.

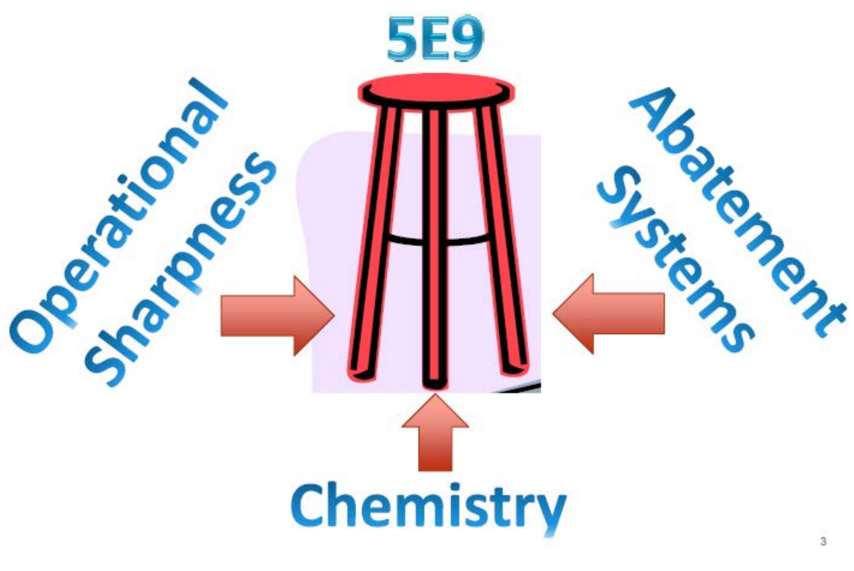

Figure 5.1. Three areas of good practices that make up the basis for reduced emissions

An expert/consultant on production cycles of irradiated nuclear fuels for medical isotopes discussed proven designs for effective abatement. He showed that the distribution of ^{133}Xe within the production process is mostly released during the target digestion/dissolution step, with a smaller amount coming from the generation by decay of the ^{133}I remaining in the system (Figure 5.2). The chemistry of the digestion and hydrogen conversion were discussed. Several schematics were shown and discussed as designs for effective abatement for different parts of the medical isotope production process. Figure 5.3 shows how the dissolver offgas can be handled to reduce emissions. Different options were discussed for a batch-wise long-time storage approach of fission gases in vacuum tanks followed by release into the cell atmosphere. Designs were shown for handling and storage of fission noble gases, xenon enrichment from the dissolver off gas, minimization of ^{133}Xe release during chromatographic purification, and cell and equipment ventilation.

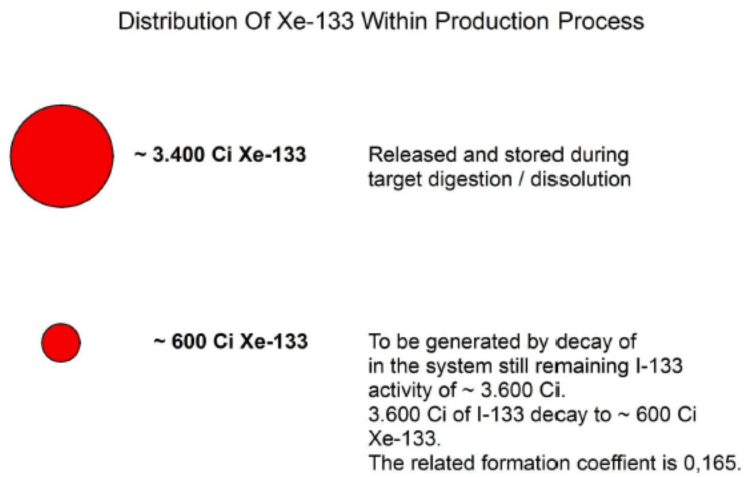

Figure 5.2. Distribution of ^{133}Xe within production process

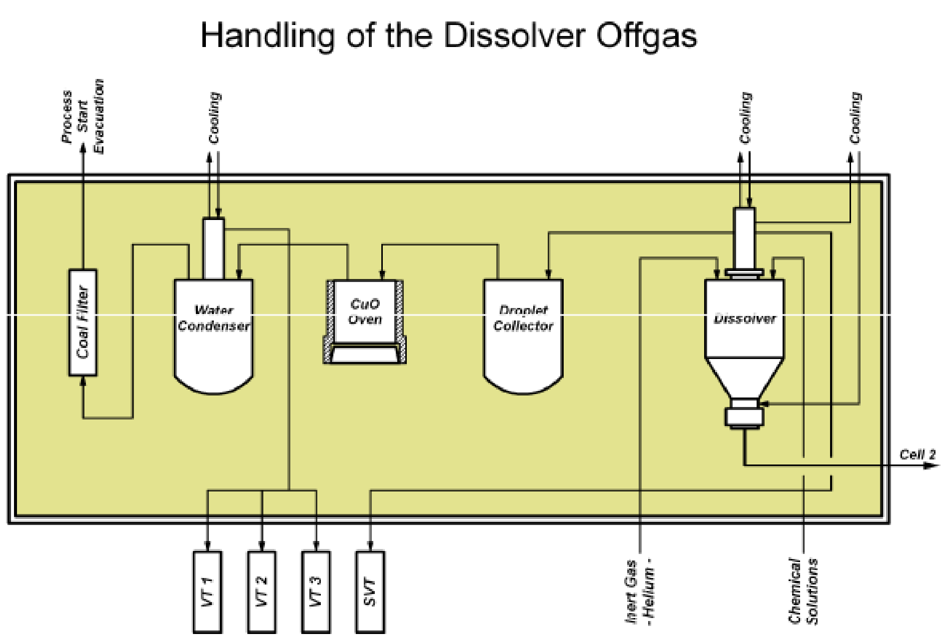

Figure 5.3. Handling the dissolver offgas

Participants also discussed the impact of conversion on processing output and the challenges associated with the chemistry of LEU targets in comparison to HEU. The processing of irradiated UAl_x, applying the highest practically available uranium density, will require no change in the dissolution process, but offers a maximum achievable output of 67% of the HEU operation. The processing of irradiated U_3Si_2 targets with the standard uranium density will require additional selective dissolution steps with HF under oxidizing conditions that will permit the compensation of the HEU operation to 100%. The challenges associated with this chemistry were discussed.

5.2 Experiments and Modeling

EU-Joint Action 5 funded a pilot study to reduce xenon emissions in the existing radiopharmaceutical facility at IRE in Belgium. Information gained from this pilot study could influence emission reduction in other existing facilities and future facilities. An overview and first results of this xenon mitigation project were provided with the main objective of developing and testing a mobile pilot system for the reduction of xenon emission (based on physical adsorption). These studies involve selecting an optimized adsorbent material, designing and optimizing trapping under different physical conditions (flow rate, temperature, adsorbent material), and installing and testing at different steps of the IRE MIP process. Figure 5.4 shows the experimental setup used at the Belgian Nuclear Research Center (SCK•CEN) to test various adsorbent materials (silver zeolites, activated carbons, and molecular sieves) under various conditions (temperatures, flow rates, and column geometries). Global treatment is not possible, so mitigation is tested for use at different steps in the process. Results are shown from many different absorption experiments. Silver zeolites are yielding promising results.

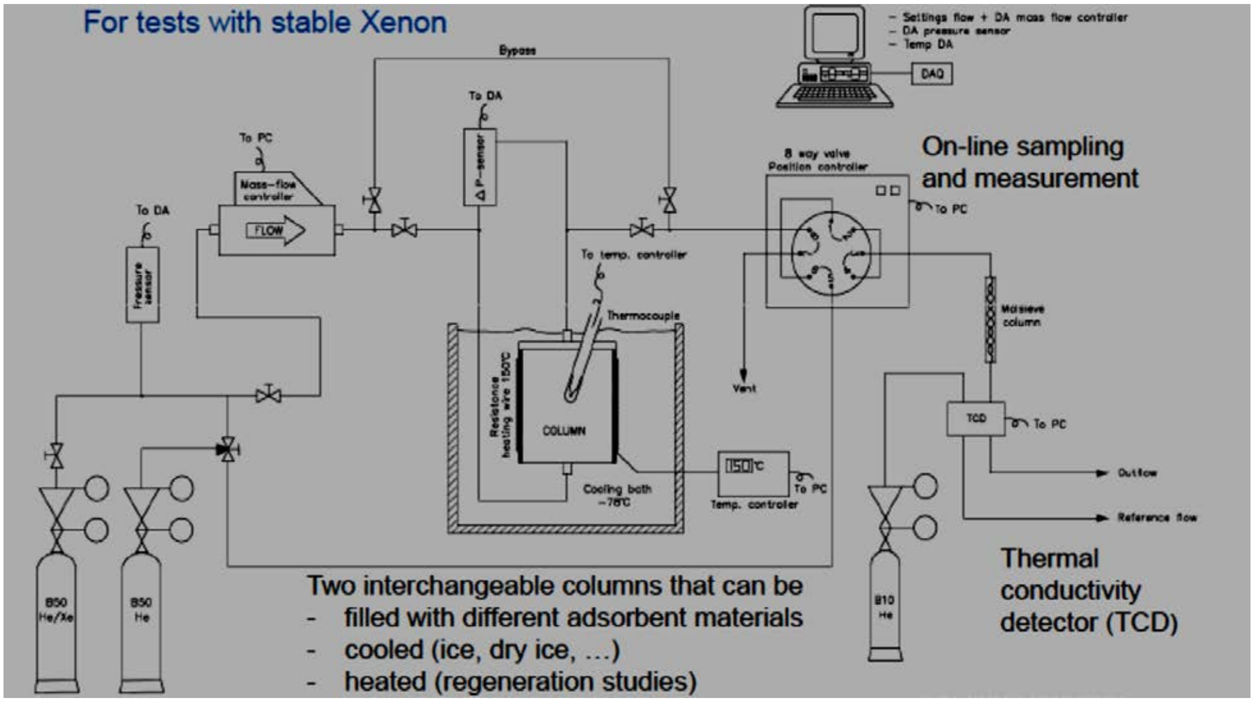

Figure 5.4. Experimental setup for tests installed at SCK•CEN

Considerations for scaling-up the experimental system to an operational system at IRE included total xenon released from each production step, number of production days, decontamination factor required, the effect of pulsed verses continuous release, xenon concentrations, and flow rates. Modeling the adsorption process was discussed in a subsequent presentation as a useful step. Simulation of the adsorption process is important to optimize the column geometry, study the adsorption for a pulsed/continuous release, specify dimensions of the column according to the decontamination factor desired and the operation conditions. The calculations used to simulate the physical processes were discussed. The accuracy of the modeled adsorption process is verified by fitting to experimental data. Figure 5.5 shows examples of modeled adsorbed concentrations as a function of time in the column. The development of an adsorption model is needed to simulate the adsorption of zeolites. Models can be expanded to include desorption and decay predictions.

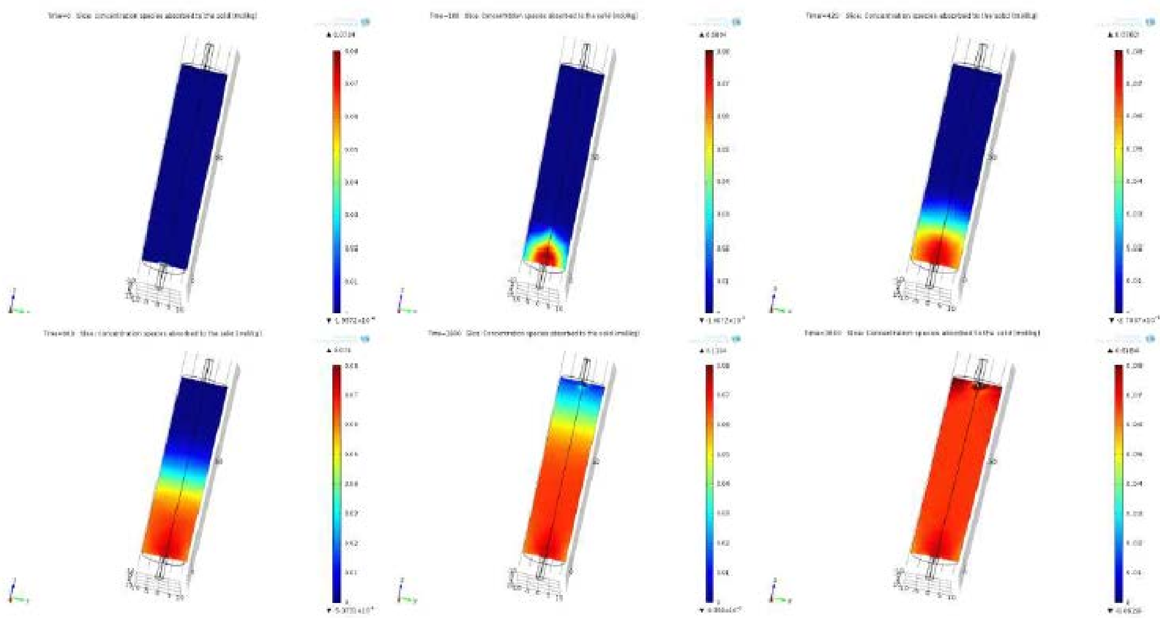

Figure 5.5. Modeled adsorbed concentration as function of time in column

IRE is working on the implementation of a LEU process in its medical isotope facility. This provides an opportune time to review the methodology of managing radioactive gas releases from the facility. IRE is working on various studies to reduce emissions, including the study discussed in the previous paragraphs/presentations with the SCK•CEN to compare the performances of various adsorbents. As mentioned previously, zeolite Ag-ETs10 appears to be a very good candidate for retention of xenon from the initial stable xenon studies at SCK•CEN. IRE has started preliminary tests with radioxenon (^{133}Xe) on Ag-ETS10. The results from the first adsorption experiments involving ^{133}Xe activity scale-up to ~200 Ci was presented. Two different scenarios that could lead to a reduction of xenon residence times inside decay tanks by using Ag-EST10 were discussed (Figure 5.6). Example results and some promising initial conclusions from this recent set of experiments are presented in Figure 5.7. Next steps will continue work on the IRE/SCK•CEN/CTBTO collaboration to define/identify critical parameters to achieve good reliability and to potentially develop a portable system for xenon retention. With the new LEU manufacturing line, it is possible to test the device in both online and post-treatment configurations.

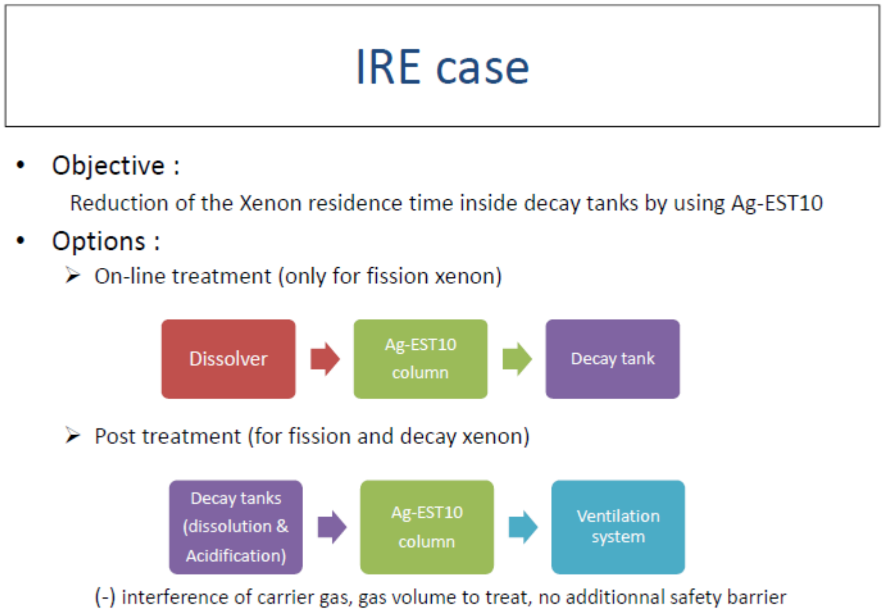

Figure 5.6. Two options for reduction of xenon residence times inside decay tanks by using Ag-EST10

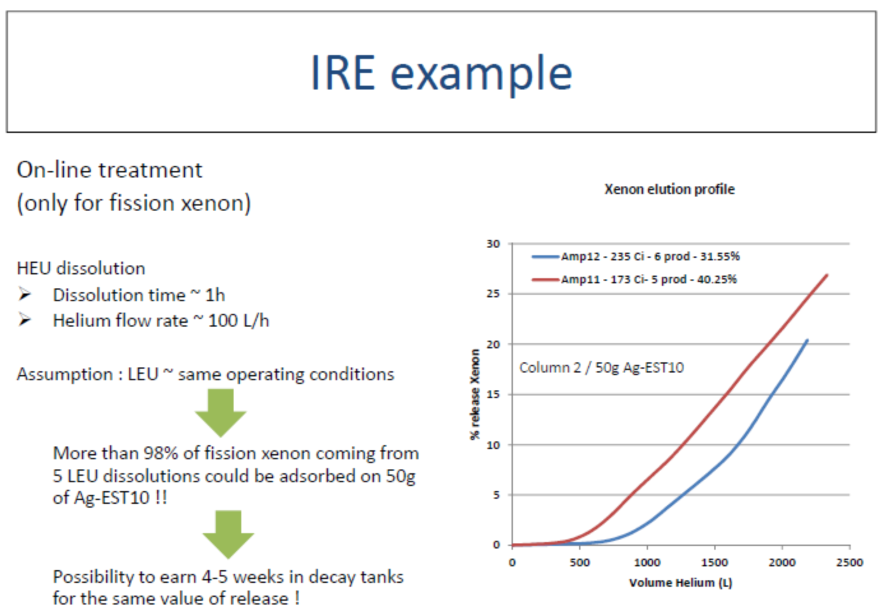

Figure 5.7. Example results from the recent set of experiments testing 133Xe retention in Ag-EST10 columns

Methods of retention and separation of hydrogen and noble gases generated in the dissolution of aluminum-uranium targets at the CNEA processing facility in Argentina were discussed. The presentation described targets of aluminum-uranium characteristics, the dissolution process in sodium hydroxide, and the generation of hydrogen. The activities of noble gases and iodine are quantified in the production process. Different methods for managing the hydrogen generated—evacuated tanks, hydrogen converter, membrane Pd–Cu, and hydrides-forming materials—were presented. The advantages and disadvantages of each were mentioned. The records of emission of ^{133}Xe and ^{135}Xe in the recent years were also presented and shown in Figure 5.8. The releases average 1.21E^{11} Bq and 5.37E^{10} Bq per week for ^{133}Xe and ^{135}Xe, respectively. The increase in releases corresponds to an increase in ^{99}Mo production at CNEA.

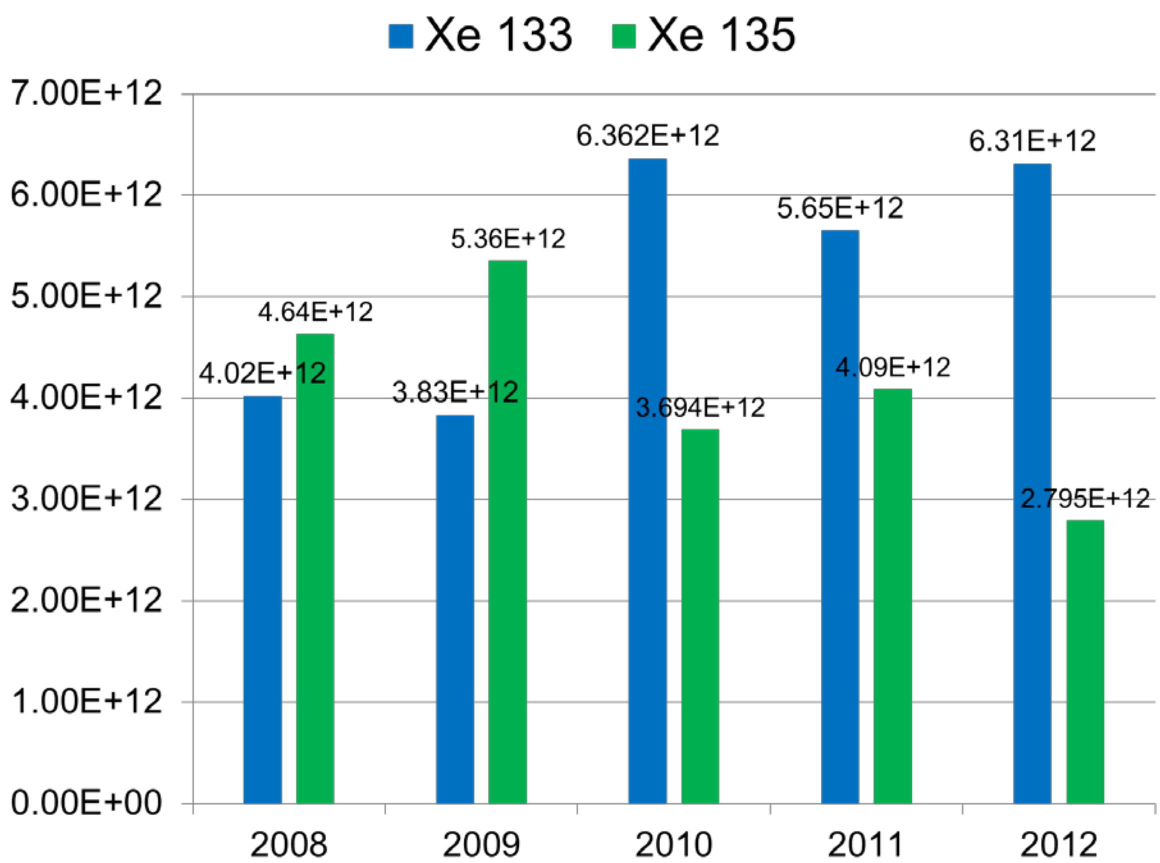

Figure 5.8. Annual releases from CNEA

5.3 Large-scale Technology with Emissions below 5 GBq/d

The scientific director from Gamma-Service Group International GmbH (GSG) shared some highlights of their various ^{99}Mo production technologies. The discussion started with highlights in the history of German fission ^{99}Mo production technologies, GSG's portfolio of proven fission ^{99}Mo technologies (both large- and small-scale), and turnkey customized fission ^{99}Mo facilities installed and commissioned by GSG. GSG provided examples of the technologies they designed and installed in the Pakistan Institute of Nuclear Science and Technology (2006–2009) and at the Research Institute of Atomic Reactors (2010–2012). Figure 5.9 summarizes the advantages of each large-scale technology. The only proven minimal environmental impact technology is Karlsruhe-Sameh Technology with an average release/day at 2.5×10^9 Bq/d (Matthews et. al. 2010).

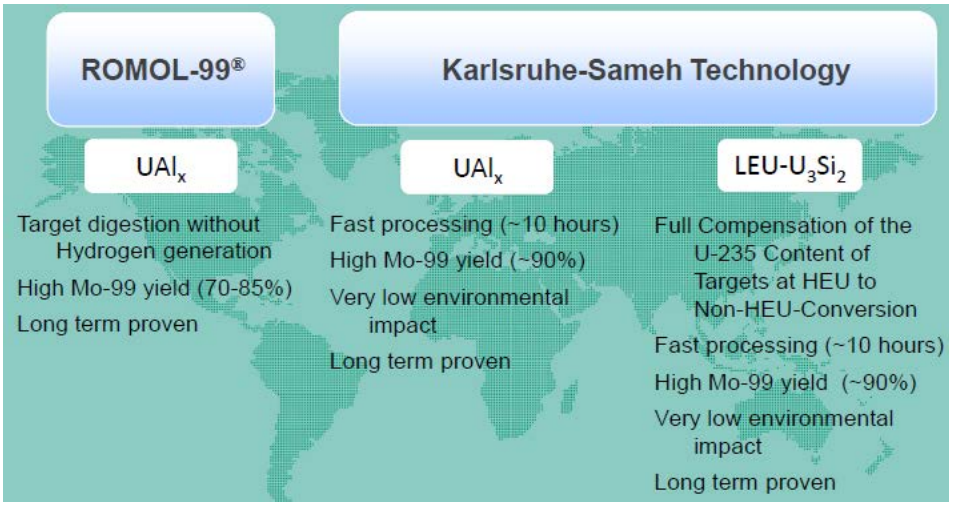

Figure 5.9. Highlights of GSG's proven large-scale processes

6.0 Alternate Approaches to Mitigation

A representative from the IAEA discussed ideas for fostering alternate technologies and products to minimize dependence on fission-based ^{99}Mo. Two approaches were discussed for identifying alternate non-fission technologies: the use of low-specific activity ^{99}Mo through the (n,γ) route at research reactors and ^{99m}Tc or ^{99}Mo production through other routes (e.g., accelerator). Amenable generator technologies were listed along with the locations where they are being used. Other potential routes of producing ^{99}Mo and ^{99m}Tc were listed:

- Direct production of ^{99m}Tc
  - >99.5% enriched ^{100}Mo (p,2n) ^{99m}Tc ; 16-20 MeV p accelerators; proven; clinical trials
- Photo nuclear reaction
  - >97% enriched ^{100}Mo (γ,n) ^{99}Mo ; 15-20 MeV electron LINAC; demonstrated
- Enriched ^{100}Mo(n_{fast},2n) ^{99}Mo ; demonstrated
- Enriched ^{96}Zr(α,n) ^{99}Mo
- ^{98}Mo(n,γ) or ^{100}Mo (γ,n) ^{99}Mo followed by (off-line) mass separation of ^{99}Mo

Different approaches were discussed for the reduced use of ^{99m}Tc and the adoption of alternate products, including the use of other radiopharmaceuticals that can give the same information as the ^{99m}Tc products and make more efficient use of generators (sharing, extended use, "central radiopharmacy" supply). Diagnostic radionuclides alternate to ^{99m}Tc that could provide similar information were listed:

- **Fluorine-18** ($T_{1/2}$=110 min) : well established worldwide; ^{18}F-FDG, a glucose-like molecule, the most widely used radiopharmaceutical of ^{18}F; positron emission tomography (PET) images
- **Gallium-68** : availed from ^{68}Ge- ^{68}Ga generator; increasingly used/popular; new molecules emerging; PET images
- **Thallium-201** : very well established, accelerator produced isotope; single-photon emission computed tomography (SPECT) images
- **Iodine-123** : old; well established; high quality SPECT images; compatible with biological molecules – warrant a revisit – 13-h half-life is not an issue when produced in a cyclotron near the end user

Fostering supplies from central radiopharmacy and enhanced use of generator capacity were also topics of discussion. A paradigm shift is needed such that fission-based ^{99}Mo is not the only or the major route to ^{99m}Tc and so concerted development efforts are investigated to support non-fission-based methods and use of alternate radionuclides. The vital role of nuclear medicine and radiopharmaceuticals need not be affected while implementing policies for eliminating civilian use of HEU and global monitoring for nuclear events.

A representative from the American Association for the Advancement of Science also discussed ideas for potential radioxenon mitigation through non-fission medical isotope production. Nuclear reactors have historically produced most medical isotopes, but he discussed how accelerator technology is an available alternative, offering potential cost competitiveness in addition to reduced waste streams, reduced proliferation risk, and the effective elimination of radioxenon effluent. Accelerators are suitable for any country, but are particularly advantageous for new and small-scale producers. It was suggested that a 100 million-person country could meet U.S.-level demand for all isotopes with 8 photo-nuclear targets for ^{99}Mo, 17 neutron beam lines (1014 n/sec) (15 would be devoted to ^{192}Ir), and 5 cyclotrons (30 MeV, 0.5 mA).

The benefits of voluntary self-regulation of radioxenon emissions by industry were a final topic for the oral presentation sessions. Self-regulation was defined as a voluntary approach to mitigating externalities of commerce that industries (e.g., MIPs) pursue as a supplement to state regulation. Self-regulation works because market conditions and other incentives push firms to adopt practices that leave them more benefits than without self-regulation. MIPs are likely to adopt further voluntary radioxenon mitigation to the extent that it offers opportunity to enhance sustainable business. Stakeholders' identification of technical and business requirements would structure continued work on best practices, implementation, and formation of accountable monitoring mechanisms.

7.0 Radioxenon Emissions Pledge Signing Ceremony

The Radioxenon Emission Pledge is a voluntary, non-binding agreement by medical isotope producers to reduce radioxenon emissions and to share information on emissions levels with the CTBTO. A standard copy of the pledge is provided in Appendix A. The first pledge was signed in June 2013, during the CTBT Science and Technology 2013 Conference organized by the Institute for Radio Elements (IRE), Belgium. Lassina Zerbo, CTBTO Executive Secretary and Jean-Michel Vanderhofstadt, CEO-General Manager, IRE signed the pledge in a signing ceremony at the conference. A second pledge-signing ceremony was held on the third day of the WOSMIP IV workshop. Lassina Zerbo signed the pledge together with Yudiutomo Imardjoko Bernadib, President Director, CEO of PT Batan Teknologi Company, Indonesia; In Cheol Lim, Vice President of Korea Atomic Energy Research Institute, Republic of Korea; Carmen Irene Bigles, CEO of Coquí RadioPharmaceuticals Corp., USA; and Hefin Griffiths, Chief Nuclear Officer of Australian Nuclear Science and Technology Organization, Australia (see Figure 7.1). The representatives of the permanent missions of Belgium, Indonesia, Korea, and the United States attended the ceremony.

Figure 7.1. The Five Pledge Signatories with the CTBTO Executive Secretary (from left: In-Cheol Lim, Carmen Irene Bigles, Lassina Zerbo, Yudiutomo Imardjoko Bernadib, Emmy Hoffmann, Benoit Deconninck)

8.0 Conclusions

At the conclusion of the workshop, participants discussed the next steps for WOSMIP and agreed on five major outcomes, listed below.

- The workshop promoted a coalition of willing experts to work together and produce openly available solutions to emissions mitigation. All participants generally agreed that annual meetings with intersessional work would be best for continued progress, as well as encouraging the participation of additional attendees from each community.
- Participants agreed that the sharing of stack monitoring data is crucial for the CTBTO verification mission; the PTS will produce some clarity on the modalities for sharing information, including its confidentiality practices.
- Participants acknowledged the emission limit/recommendation of $5x10^9$ Bq/d discussed in WOSMIP III and recognized it as a goal for the industry.
- Participants from both communities recognized the need for continued improvements in validation of models.
- Four more producers signed the pledge to reduce radioxenon emissions and share data with the CTBTO, bringing the total number of signatories to five. These include two current producers—ANSTO and PT Batan Teknologi Company of Indonesia—and two prospective future producers—KAERI and Coquí RadioPharmaceuticals Corporation of the United States. The first signing was by the IRE in June 2013. More signatories are sought.

Everyone agreed that tremendous progress has been made over the four WOSMIPs. The monitoring community has gained a better understanding of the challenges and pressures facing the medical isotope supply chain, and the medical isotope producers have an enhanced understanding of the challenges associated with nuclear explosion monitoring. Participants stressed the need for at least one meeting per year to maintain and grow this successful relationship between the two communities. They also expressed the need for intercessional technical exchange meetings to facilitate continued detailed discussions on specific emissions reduction or data sharing solutions. It is important to engage with additional medical isotope producers in future WOSMIPs. Further collaboration and coordination between the CTBTO and the IAEA may promote participation from other medical isotope producers and other related companies or agencies.

9.0 References

Matthews M, and TW Bowyer. 2013. *WOSMIP III - Workshop on Signatures of Medical and Industrial Isotope Production*. Report No. PNNL-22721, Pacific Northwest National Laboratory, Richland, WA.

Matthews M, PR Saey, TW Bowyer, GF Vandergrift, N Ramamoorthy, C Cutler, B Ponsard, R Mikolajczak, Y Tsipenyuk, L Solin, DR Fisher, GM Dolinar, R Higgy, I Schraick, EC Carranza, SR Biegalski, B Deconninck, A Ringbom, AA Sameh, D Amaya, EL Hoffman, LA Barbosa, J Camps, EB Duran, M Zaehringer, AP Rao, J Turinetti, D Mercer, M Auer, P Achim, VY Popov, G Steinhauser, S Hebel, A Becker, and S Solomon. 2010. *Workshop on Signatures of Medical and Industrial Isotope Production - A Review* . Report No. PNNL-19294, Pacific Northwest National Laboratory, Richland, WA

Appendix A

Radioxenon Emission Pledge

CTBTO PREPARATORY COMMISSION | preparatory commission for the comprehensive nuclear-test-ban treaty organization

Radioxenon Emissions Pledge

The signatories to this pledge have agreed to work voluntarily to minimize, mitigate, and resolve, where possible, interference of medical isotope radioxenon emissions with monitoring for nuclear explosions.

Recognizing the efforts of the International Community supporting the establishment of an effective verification system for the Comprehensive Nuclear-Test-Ban Treaty (CTBT);

- Recognizing the role of the Provisional Technical Secretariat of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO/PTS) to supervise, coordinate and ensure the operations of the International Monitoring System (IMS), a worldwide network of stations supported by an International Data Centre (IDC) in Vienna to monitor for signatures of nuclear explosions, using inter alia a radioactive noble gas component to detect the presence of radioxenon isotopes;
- Recognizing the importance of radioisotopes to medical treatment of people world-wide and the urgent need for continued medical isotope production;
- Recognizing the IMS can detect even trace amounts of radioactive xenon; and that radioactive xenon emissions from fission-based medical and industrial isotope production are detected and reported by the IMS on a daily basis;
- Recognizing that efforts to detect nuclear explosions are hindered by other emissions of radioxenon;

The producer of isotopes for use in medicine and industry recognize and voluntarily agree to:

- Minimize isotopic releases so that the monitoring efforts of the international CTBT community are minimally impacted. Whenever feasible, they will pursue radioxenon emissions at or below the levels recommended by the CTBTO/PTS;
- Explore means to share xenon monitoring (i.e., "stack" or facility monitoring) data with the CTBTO/PTS for use in screening out xenon detections from the IMS that result from isotope production, the modalities of this communication will address the timing, processing of the data and confidentiality issues; and
- Further support the CTBTO/PTS upon request with information regarding radioxenon emissions in order to improve the interpretation or clarification of IMS radioxenon data.

The Provisional Technical Secretariat of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization agrees to:

- Assist producers in clarifying any concerns due to elevated IMS readings of radioxenon isotopes,
- Recognize and cooperate with producers who are taking steps to mitigate the effects of emissions on the IMS.
- This collaboration offered by the CTBTO will aim at direct support in expertise, technological development and possible financial support.

Date:	Date:
Lassina Zerbo Executive Secretary, CTBTO	N.N.

Appendix B

WOSMIP 2013 Agenda

Appendix B

WOSMIP) 2013 Agenda

Workshop on the Signatures of Medical and Industrial Isotope Production (WOSMIP) 2013

11-13 November 2013

Vienna International Centre

Room C3 (7th floor of the C-Building)

Monday 11-Nov-13

08:00	- 09:00	Badging/Registration
09:00	- 09:30	Welcome Mr. Randy Bell, Director, International Data Centre (IDC), CTBTO Ms. Vorian Maryssael, Director, International Monitoring System (IMS), CTBTO Mr. Ted Bowyer, Pacific Northwest National Laboratory (PNNL)
		Session 1 – Introduction to the Problem **Chair: Ted Bowyer, Mika Nikkinen**
09:30	- 09:45	Overview of the Problem Saey, P.
09:45	- 10:00	Past WOSMIPs, and This One! Bowyer, T.
10:00	- 10:30	Overview of CTBTO Monitoring Nikkinen, M.
10:30	- 11:00	Update on Fission Production of Mo-99 at Research Reactors and Accelerators Ridikas, D.
11:00	- 11:30	Fostering Alternate Technologies and Products to Minimise Dependence on Fission-Produced Mo-99 Ridikas, D.

11:30	-	12:30	Tour of the International Data Centre/Noble Gas Monitoring Station
12:30	-	13:45	**Lunch (VIC Cafeteria)**
			Session 2 –Production Methods **Chairs: Luis Barbosa, Judah Friese**
13:45	-	14:10	Overview of Production at NorthStar Medical Technologies Harvey, J.
14:10	-	14:35	Overview of the ANSTO Nuclear Medicine Project and Planned Emission Abatement System Hoffman, E.
14:35	-	15:00	Development Status of Fission Mo Production Process in Korea Lee, J.
15:00	-	15:25	Mo-99 Production in RPF- EAEA (Egypt) Aydia, M.
15:25	-	15:50	Plans at Coquí Bigles, C.
15:50	-	16:30	**Coffee Break** Featured Posters: Production of Iodine-131 from Nuclear LEU U-235 Fission Products (RPF-AEA-Egypt) Faseih, T. Real-time Tracking of Radioxenon Plumes at ANSTO using NaI(Tl) Detector with Rapid Peak Identification Software Hoffman, E. Bayesian Method for Xenon Source Reconstructions Using Atmospheric Transport Methods Ungar, K.
16:30	-	16:55	The SHINE Medical Mo-99 Production System and Planned Off-Gas Control Technologies Pitas, K.

16:55 - 17:20	Update Mini-SHINE Experiments for Development of Mo-99 Production Using an Accelerator-Driven Aqueous Target Solution Vandegrift, G.
17:20 - 17:45	Infrastructure for Production of Radioisotopes and Progress in Mo99 Production from LEU Targets Ivan, A.
18:00	**Reception, hosted by CTBTO** Function Room III in the VIC Restaurant area, F Building

Tuesday 12-Nov-13

Session 2 –Production Methods (continued)

Chairs: Luis Barbosa, Judah Friese

08:30 - 08:55	Production of Mo-99 from Fission Process with Xenon Release Below the Permitted Limit Ibrahim, H.
08:55 - 09:20	Developments in high-density MoO3 Pellets for (y, n) 99 Mo domestic production Sarkis, Z.

Session 3: Monitoring Technology

Chairs: Matthias Auer, John Lucas

09:20 - 09:45	Atmospheric Transport Modelling from a User's Perspective Lucas, J.
09:45 - 10:10	Tracking Medical Isotope Plumes with Stack Monitoring at the BATAN Teknologi (BaTeK) Facility McIntyre, J.
10:10 - 10:35	Effects of the Backgrounds of Radioxenon from Potential Future Medical Isotope Production on the Korean Peninsula Bowyer, T.

10:35	-	11:15	**Coffee Break** Featured Posters: Radionuclide Monitoring Program for the Local Production of Mo-99/Tc-99m Generators in the Philippines Bulos, A. Workshop on the Signatures of Medical and Industrial Isotope Production (WOSMIP IV) Soe, N.
11:15	-	11:40	Progress on Stack Monitoring with AECL and Other Areas of CTBT Cooperation Ungar, K.
11:40		12:05	Stack Monitor for IPF's Emissions Di Tada, M.
12:05	-	12:30	Monitoring Radioxenon Emission Trends at the ANSTO Medical Isotope Production Facility Using Web-Based Visualisation Software Hoffman, E.
12:30	-	13:45	**Lunch (VIC Cafeteria)**
			Session 4: Emissions Abatement **Chairs: Benoit Deconninck, Emmy Hoffman, Lori Metz**
13:45	-	14:10	Good Practices for Abatement Systems Friese, J.
14:10	-	14:35	Overview and First Results of the Xenon Mitigation Project Gueibe, C.
14:35	-	15:00	Modeling Xenon Adsorption Camps, J.
15:00	-	15:25	Xe-133 Adsorption Experiments for Reducing the IRE Radioxenon Emission After the HEU-LEU Conversion Moyaux, D.

15:25	-	16:05	**Coffee Break** Featured Posters: Stack Monitoring Testbed at PNNL's Radiochemical Processing Laboratory Metz, L. Canberra Stack Monitoring Technology De Baerdemaeker, L.
16:05	-	16:30	Methods of Retention and Separation of Hydrogen and Noble Gases Generated in the Dissolution of Aluminium-Uranium Targets Carranza, E.
16:30	-	16:55	Large Scale fission Mo-99 Production Facilities with Xe-133 Emissions Below 5 GBq/d Dittrich, S.
16:55	-	17:20	Utilization of Noble Gas released from Nuclear Facilities Liu, L.
19:00			**Dinner, Hosted by Pacific Northwest National Laboratory & Scienta SAUNA Systems** Hajszan Neumann Grinzingerstrasse 86, A-1190 Wien

Wednesday 13-Nov-12

08:30	-	10:00	Roundtable Discussion: Supply and Use of Stack Monitoring Data
10:00	-	10:30	**Radioxenon Emissions Pledge Signing Ceremony** Dr. Lassina Zerbo, Executive Secretary, CTBTO
10:30	-	11:00	**Group Photo (M Building)**
			Session 5: Alternate Approaches to Mitigation **Chairs: Ian Cameron, Paul Saey**
11:00	-	11:25	Conversion of Fission Mo-99 Production Targets as a Challenge for

			Processing Innovations Sameh, A.A.
11:25	-	11:50	Radioxenon Reduction Through Non-Fission Based Medical Isotope Production Updegraff, D.
11:50	-	12:15	Voluntary Radioxenon Regulation Mahoney, C.
12:15	-	13:45	**Lunch (VIC Cafeteria)**
			Session 6: Next Steps for WOSMIP **Chairs: WOSMIP Scientific Committee**
13:45	-	15:00	Roundtable Discussion: Next Steps for WOSMIP
15:00	-	15:30	**Coffee Break** Awarding of the Wozzie for Demonstrated Commitment Towards Mitigating the Effects of Emissions from Medical and Industrial Isotope Production
15:30	-	16:30	Workshop Conclusions

Posters:

Bulos, A., "Radionuclide Monitoring Program for the Local Production of Mo-99/Tc99m Generators in the Philippines"

De Baerdemaeker, L., "Canberra Stack Monitoring Technology"

Faseih, T., "Production of Iodine-131 from Nuclear LEU U-235 Fission Products (RPF-AEA-Egypt)"

Hoffman, E., "Real-time Tracking of Radioxenon Plumes at ANSTO using NaI(Tl) Detector with Rapid Peak Identification Software"

Metz, L., "Stack Monitoring Test Bed at PNNL's Radiochemical Processing Laboratory"

Soe, N., "Workshop on the Signatures of Medical and Industrial Isotope Production (WOSMIP IV)"

Ungar, K., "Bayesian Method for Xenon Source Reconstructions Using Atmospheric Transport Methods"

Please note that all posters will be displayed for the whole duration of the workshop; however, only the presenters listed for each coffee break are required to be present at their poster to answer questions at the times of the respective coffee breaks.

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