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# **WOSMIP VI**

# Workshop on Signatures of Man-Made Isotope Production

San Carlos de Bariloche, Argentina November 28 to December 2, 2016

Hosted by the Provisional Technical Secretariat (PTS) of the Preparatory Committee Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom), Investigación Aplicada (INVAP), and Pacific Northwest National Laboratory (PNNL)

## January 2018



Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830



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This report was compiled by Pacific Northwest National Laboratory from presentations, materials, and discussions at the sixth Workshop on Signatures of Man-Made Isotope Production.



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## WOSMIP VI - Workshop on Signatures of Man-Made Isotope Production

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This report was compiled by INVAP and PNNL from presentations, materials and discussions at the sixth Workshop on Signatures of Man-Made Isotope Production (WOSMIP)

October 2017

Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830 Pacific Northwest National Laboratory Richland, Washington 99352

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### **Executive Summary**

The sixth Workshop on Signatures of Man-Made Isotope Production (WOSMIP) was held in San Carlos de Bariloche, Argentina from 28 November through 2 December, 2016. The focus of the workshop was to bring together representatives from the isotope production and nuclear explosion monitoring communities in order to discuss effects on the verification efforts for the Comprehensive Nuclear-Test-Ban Treaty (CTBT) resulting from nuclear facility radioxenon emissions. In attendance there were 81 participants from 27 countries. Discussions at WOSMIP VI focused on alternative xenon sources (nuclear power plants, research reactors, production, handling, and use of medical isotopes, industrial isotopes, etc.), updates from current and prospective medical isotope producers (MIP) on production processes and facilities, R&D efforts toward radioxenon emission reduction, the use of nuclear facility stack emission data for verification purposes, and atmospheric transport modeling (ATM). The major outcomes of the workshop are presented in this summary.

The workshop began with a welcome and workshop overview session. As part of this session, an overview of the issues unique to the medical isotope producer perspective was given—during this presentation, it was stated that most producers are happy to share release data and believe that lowering their radioxenon releases is a good thing, although clear design objectives are necessary. In addition, an update was given on the International Atomic Energy Agency (IAEA) coordinated research project (CRP) on the sharing and developing of protocols to further minimize radioactive gaseous emissions to the environment in MIP, and real world examples of the use of International Monitoring System (IMS) measurements related to announced Democratic People's Republic of Korea (DPRK) events were shared.

A session on alternative xenon source terms explored the broader range of potential sources of xenon isotope emission other than <sup>99</sup>Mo production. Alternative sources of radioxenon that were discussed included next-generation nuclear reactors with potential for increased radioxenon releases, production of isotopes other than <sup>99</sup>Mo at accelerator and research reactor facilities, nuclear power plants, radioxenon that is used in medical and industrial settings, and the use of an aqueous homogeneous reactor for production of <sup>99</sup>Mo in Tajikistan.

Sessions overviewing current and future <sup>99</sup>Mo production featured recent and planned activities at isotope production facilities with updates from eight current or prospective medical isotope producers. Several producers are working toward lowering their impact on the IMS by reducing emissions and volunteering to share stack release data with the Preparatory Commission for the Comprehensive Nuclear Test-Ban-Treaty Organization (CTBTO PrepCom). Detailed information shared by producers will help the community better understand the effects of MIP on monitoring.

A session focused on R&D efforts on emission reduction highlighted recent advances in decreasing radioxenon emissions. In this session, discussions regarding considerations and challenges in radioxenon abatement system design were held, projects by the European Union (EU) and IAEA to advance radioxenon mitigation technology were overviewed, and the plan for an upcoming feasibility study to deploy and test the prototype xenon mitigation system developed by SCK•CEN at IRE's MIP facility was presented.

The session devoted to the use of stack emission data for the understanding of the global radioxenon backgrounds from industry introduced a new concept, the Source Term Analysis of Xenon (STAX) project. This project would develop methods, tools, and technology for the sharing of stack emissions release data to be used along with ATM to quantify the effect of industrial releases on CTBT verification. Topics relating to the STAX network concept and stack release data sharing were also discussed, including data confidentiality, software requirements, and progress on the stack data upload experiment. Additionally, stack effluent detection technologies were discussed.

The Atmospheric Transport session focused on characterization of the impact of MIP emissions on the global radioxenon background. This session gave ATM experts the opportunity to share challenges related to accurate modeling of radioxenon background concentrations, such as the impact of industrial radioxenon sources; seasonal variations in xenon background; and uncertainty in source data. In addition, results from the 2<sup>nd</sup> ATM challenge to ascertain the agreement between modeled radioxenon concentrations and backgrounds measured by the IMS were shared. The results of this challenge showed a strong correlation between the modeled and actual background, although the magnitude of the concentrations was more difficult to accurately calculate.

A concluding discussion was held to summarize thoughts and ideas from the WOSMIP community. A number of positive outcomes from WOSMIP VI were noted, including expansion of the workshop to include emission sources other than MIP; willingness of the producers to share stack release data with the CTBTO PrepCom; sessions in which abatement and stack release data sharing were discussed; discussing how the uncertainty in ATM calculations can be quantified; and that it was the first WOSMIP to be held outside of Europe. It was commented that it may be beneficial to increase communication and build awareness of issues relating to radioxenon emissions with organizations outside of the current WOSMIP community and that building trust between organizations is important to the success of WOSMIP. Looking forward, it was agreed that the next WOSMIP should be held in approximately 18 months. Lastly, there was an announcement that a WOSMIP session will be added to the CTBT Science and Technology 2017 (SnT17) meeting that will be held at the Hofburg Palace in Vienna, Austria from June 26-30, 2017, if there is enough interest as measured by the number of abstracts submitted.

## Acknowledgments

The workshop hosts, the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom), Investigaciones Aplicados (INVAP) and Pacific Northwest National Laboratory (PNNL) would like to thank WOSMIP participants for their contributions to the workshop and this report. The agenda contains more information on individual roles in relation to the workshop; see Appendix A.

Participant	Country	Role	Affiliation
A. Axelsson	Sweden	Presenter	Swedish Defence Research Agency
J. Baré	CTBTO PrepCom	Presenter	Preparatory Commission for the Comprehensive Nuclear- Test-Ban-Treaty Organization
R. Berg	Canada	Presenter	Radiation Systems and Field Specialist; Health Canada
T. Bowyer	United States	Co- organizer, Session Chair and presenter	Laboratory Fellow; Pacific Northwest National Laboratory
T. Boytsova	Russia	Presenter	Khlopin Radium Institute
R. Brown	United States	Presenter	Senior Director; Mallinckrodt Pharmaceuticals
E. Carranza	Argentina	Co-organizer	Head of Fission Radioisotope Production Plant; Head of Radiological Protection
I. Cameron	United States	Co-organizer	Specialist – Arms Control and Nonproliferation; Pacific Northwest National Laboratory
P. Cristini	Argentina	Presenter	Manager of Radioisotope Production; Argentinian National Atomic Energy Commission
L. De Baerdmaeker	Belgium	Presenter	Mirion Technologies
R. DeCaire	Canada	Presenter	Health Physicist; Nordion
B. Deconninck	Belgium	Session Chair and Presenter	Environmental and Metrology Dept. Manager; National Institute for Radioelements ELiT
M. Di Tada	Argentina	Co- organizer, Session Chair and Presenter	Radiation Monitoring Systems Design; Investigaciones Aplicados
J. Dix	Austria	Presenter	International Atomic Energy Agency
C. Doll	United States	Co-organizer and presenter	Research Scientist; Pacific Northwest National Laboratory
J. Domínguez	Argentina	Presenter	Argentinian National Atomic Energy Commission
D. Ferraro	Argentina	Presenter	Investigaciones Aplicados

J. Friese	United	Co-	Lab Fellow: Pacific Northwest National Laboratory
	States	organizer, Session Chair and presenter	
S. Generoso	Argentina	Presenter	Commissariat à l'Énergie Atomique et aux Énergies Alternatives
L. Glascoe	United States	Presenter	Lawrence Livermore National Laboratory
I. Goldman	United States	Presenter	Lantheus Medical Imaging
F. Gubernator	Germany	Presenter	University of Hamburg
C. Gueibe	Belgium	Session Chair and Presenter	Scientific Collaborator; Belgian Nuclear Research Centre
D. Haas	United States	Presenter	University of Texas
E. Hoffman	Australia	Presenter	Manager; Nuclear Assurance Services; Australian Nuclear Science and Technology Organisation
Y. Imardjoko	Indonesia	Presenter	President Director; PT INUKI (Persero)
M. Kalinowski	CTBTO PrepCom	Co- organizer, Session Chair and presenter	Head of the Scientific Methods Unit in the IDC Division; Preparatory Commission for the Comprehensive Nuclear- Test-Ban Treaty Organization
M. Khalid	Pakistan	Presenter	Pakistan Institute of Nuclear Science and Technology
K. Khrustalev	Austria	Presenter	Instrumental Software Technologies Inc.
Y. Kijima	Japan	Presenter	Japan Atomic Energy Agency
J. Kusmierczyk- Michulec	CTBTO PrepCom	Presenter	Preparatory Commission for the Comprehensive Nuclear- Test-Ban-Treaty Organization
A. Malkawi	Jordan	Presenter	Section Head; Jordan Atomic Energy Commission
C. Maurer	Austria	Presenter	Central Institute for Meteorology and Geodynamics
S. Mekhaimr	Egypt	Presenter	National Data Center National Research Institute of Astronomy and Geophysics
L. Metz	United States	Co-organizer	Research Scientist; Pacific Northwest National Laboratory
D. Moyaux	Belgium	Presenter	Researcher; R&D Department; Belgian Nuclear Research Centre
K. Muminov	Tajikistan	Presenter	Academy of Sciences of the Republic of Tajikistan
J. Perrotta	Brazil	Presenter	Brazilian Nuclear Energy Commission
R. Plenteda	CTBTO PrepCom	Presenter	Preparatory Commission for the Comprehensive Nuclear- Test-Ban-Treaty Organization

P. Saey	IAEA	Co- organizer, Session Chair and Presenter	Nuclear Safeguards Inspector; International Atomic Energy Agency

## Acronyms and Abbreviations

ANSTO	Australian Nuclear Science and Technology Organisation
ANM	ANSTO Nuclear Medicine (facility)
ARIX	Russian Analyzer of Xenon Radioisotopes
ARN	Argentine Nuclear Regulatory Authority
ATM	Atmospheric Transport Model/Modeling
CNEA	Argentinian National Atomic Energy Commission
CNEN	Brazilian Nuclear Energy Commission
CNL	Canadian Nuclear Laboratories
CPU	Central Processing Unit
CRP	Coordinated Research Project
CTBT	Comprehensive Nuclear-Test-Ban Treaty
CTBTO PrepCom	Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization
DPRK	Democratic People's Republic of Korea
ECMWF	European Centre for Medium-Range Weather Forecasts
EU	European Union
FLEXPART	FLEXible PARTicle dispersion model
GFR	Gas-Cooled Fast Reactor
HC	Health Canada
HEU	Highly enriched uranium
IAEA	International Atomic Energy Agency
IDC	International Data Centre
IMS	International Monitoring System
INUKI	PT Industri Nuklir Indonesia (formally PT Batan)
INVAP	Investigación Aplicada
IRE	National Institute for Radioelements, Belgium
JRTR	Jordan Research and Training Reactor
LEU	Low-enriched uranium
LFR	Lead-Cooled Fast Reactor
MDC	Minimal Detectable Concentration
MIP	Medical Isotope Producer/Medical Isotope Production
MIPF	Medical isotope production facility
MSR	Molten Salt Reactor
MURR	Missouri University Research Reactor Center
NCEP	National Centers of Environmental Prediction
NDC	National Data Center

NPP	Nuclear Power Plant
NRU	National Research Universal
NTP	NTP Radioisotopes Ltd, South Africa
PARR 1/PARR 2	Pakistan Research Reactor
PINSTECH	Pakistan Institute of Nuclear Science and Technology
PNNL	Pacific Northwest National Laboratory
PTS	Provisional Technical Secretariat
RIPF	Radioisotope Production Facility
RMB	Brazilian Multipurpose Research Reactor
SAUNA	Swedish Automated Unit for Noble Gas Analysis
SCK•CEN	Belgian Nuclear Research Centre
SGE	Selective Gaseous Extraction
SPALAX	French Automated Unit for Noble Gas Analysis
SRS	Source-Receptor Sensitivity
STAX	Source Term Analysis of Xenon
vDEC	virtual Data Exploitation Centre
VHTR	Very-High-Temperature Reactor
VQ	Ventilation/Perfusion Nuclear Study
WOSMIP	Workshop on Signatures of Man-made Isotope Production

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### 1.0 Introduction

The Provisional Technical Secretariat (PTS) of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom) operates and maintains an extensive, worldwide network of sensors to monitor for signatures of nuclear explosions. This network, the International Monitoring System (IMS), has the ability to detect seismic, hydroacoustic, infrasonic, and radionuclide signatures of nuclear explosions. Of the radionuclide signatures expected from a nuclear explosion, the noble gas radioxenon is perhaps the most important, as it is one of the only signatures expected from an underground nuclear explosion that can confirm the nuclear nature of the event.

Radioxenon is also a common waste product from the fission-based production of <sup>99</sup>Mo, the daughter of which, <sup>99m</sup>Tc, accounts for 80% of the nuclear medicine procedures conducted worldwide with the vast majority being produced via fission resulting in global backgrounds of radioxenon. Radioxenon is also produced during the operation of nuclear power plants (NPPs) and other reactors, but because the production process involves the dissolution of a uranium target shortly after irradiation and the near immediate release of radioactive noble gases, the gaseous signature from medical isotope production (MIP) can appear more similar to that of a nuclear explosion than those from NPPs and other reactors, where short-lived radionuclides are trapped in the fuel and allowed to decay prior to release. The routine release of these fission gases during MIP—at levels compliant with regulatory limits and far below levels that would impact human health—is detected in the IMS on average every day, which can challenge the interpretation of IMS data by the PTS.

The Workshop on Signatures of Man-Made Isotope Production (WOSMIP) enlists the help of two groups of experts—those from the nuclear explosion monitoring and isotope production communities—to explore solutions to the challenges presented by the radioxenon emissions, while respecting the need for a stable supply of these life-saving isotopes. WOSMIP has evolved since its inception in 2009. Where the first workshop was focused on introducing the isotope production and explosion monitoring communities to the issue and encouraging further scientific collaboration, subsequent workshops began to more distinctly refine the problem; establish scientific and political boundaries; demonstrate observations of radioxenon emissions from MIP detected by the IMS; and share examples of emission mitigation technology being developed and used in real-world facilities to lower radioxenon releases. At recent workshops, attendees have recognized the importance of sharing stack release data, reducing emissions at the source, and identifying and investigating knowledge gaps in both explosion monitoring and emissions monitoring

WOSMIP VI expanded the scope from previous workshops to better understand all man-made sources of radioisotopes in the environment (NPPs, research reactors, production, handling and use of medical isotopes, industrial isotopes, etc.) that have the potential to be observed by the CTBT verification regime. A main objective of the workshop was to facilitate the characterization of radioxenon detections in the IMS through improved understanding of the global radioxenon background created by all known sources and environmental variations. WOSMIP VI was held in San Carlos de Bariloche, Argentina, 2016, from 28 November to 2 December and was hosted by Investigación Aplicada (INVAP), the Provisional Technical Secretariat (PTS) of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom) and Pacific Northwest National Laboratory (PNNL), see Figure 1.



Figure 1. Group photo at WOSMIP VI (bottom). SCK•CEN received the WOZZIE award for its contribution to the design of abatement technologies (right center).

This report provides a summary of highlights from the five-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 Welcome and Workshop Overviews: Introductory Contributions

The introductory session presentations focused on each organization's involvement in understanding and reducing the effect of radioactive gaseous releases from MIP on nuclear explosion monitoring. PNNL, IMS, the International Data Centre (IDC), the Medical Isotope Producers (MIP) community and the International Atomic Energy Agency's (IAEA) all provided overviews.

#### 2.1 Workshop Overview – WOSMIP VI

PNNL presented an overview of the workshop, focusing on the evolution of WOSMIP from its origin in 2009 to the present. WOSMIP was first established to better understand radionuclides that are detected daily by the IMS in the environment and to bring together the monitoring and medical isotope communities to find solutions to the effect of the global radioxenon background resulting from isotope production emissions on nuclear explosion monitoring.

The previous workshop, WOSMIP V, was held in Brussels and co-hosted by the Belgian Nuclear Research Centre (SCK•CEN); see Figure 2. Highlights from this workshop included agreement from the community that stack release data in conjunction with ATM have potential to be used as a tool for determining the effect of radioisotope emissions on distant monitoring to within  $\pm 10-15\%$ , under ideal circumstances. Several studies were underway that focused on emission control at the source. These studies, activities, and experiments to lessen and correct for the effect of emissions are key to discovering a solution to the problem.



Figure 2. WOSMIP V was held in Brussels, Belgium May 2015

A number of accomplishments that are related to WOSMIP activities include significant advancements in ATM-related tools; and continued work on emission control by several groups, including basic research by SCK•CEN to develop an engineered emission control system and stack monitoring systems designed by INVAP. Other related issues are the IAEA coordinated research project (CRP) on the sharing and developing of protocols to further minimize radioactive gaseous emissions to the environment in the

manufacture of medical radioisotopes and the U.S. National Nuclear Security Administration's efforts to establish reliable supplies of <sup>99</sup>Mo produced without highly enriched uranium.

These accomplishments point to a hopeful future in which there will be an improved understanding of radioxenon emission points and conditions; incorporation of stack measurement technology—combined with ATM; development of improved tools to discriminate between industrial and nuclear explosion activities; and, eventually, emission reductions at the source.

Keep in mind that the WOSMIP community is not advocating for reduction of <sup>99</sup>Mo production, purposefully or otherwise. Any action should be voluntary to benefit nuclear non-proliferation efforts. We do believe that we can make a difference in global monitoring—and already have—using these principles.

WOSMIP VI continued this tradition with a 2<sup>nd</sup> ATM challenge, reports on continued studies of collecting and storing fission gases at the production location, discussions of stack data collection and methods for data confidentiality, and reporting on emissions from the other sources (e.g., reactors).

#### 2.2 International Monitoring System Overview and Noble Gas Detection Capability

The CTBTO PrepCom presented an overview of the IMS focused on noble gas detection. The CTBT was opened for signature in 1996 and currently has 183 signatory states of which 166 have ratified the treaty. The treaty will enter into force after ratification by the eight remaining Annex 2 states. Article I of the CTBT states that a verification regime to monitor compliance with the treaty must be in place at the time of entry into force:

Each State Party undertakes **not to carry out any nuclear weapon test explosion or any other nuclear explosion**, and to prohibit and prevent any such nuclear explosion at any place under its jurisdiction or control. For verification of the CTBT, a comprehensive global system is required to which can monitor atmospheric, underground, under water, outer space for nuclear explosions.[1]

To establish and maintain a verification capability, the CTBTO PrepCom has more than 260 staff members from more than 70 countries and an annual budget of approximately \$125,000,000 USD. The verification regime for the CTBT has several components, including the IMS network for global and continuous surveillance of compliance; the IDC, which oversees data processing, data analysis, and provision of data to the member states; and OSI—conducted by a team comprising inspectors from state parties.

The IMS consists of monitoring stations situated globally that incorporate four technologies for the detection of signatures resulting from nuclear explosions. The waveform technologies include 50 primary and 120 auxiliary seismic stations, 11 hydro-acoustic stations, and 60 infrasound stations. Radionuclide technologies comprise 80 radionuclide stations, which measure aerosol borne radionuclides; 40 of these stations will also be equipped with sensors to measure radioxenon isotopes. Currently, >80% of the network is installed and certified (283 out of 331 monitoring stations and radionuclide laboratories).

Xenon isotopes are of particular interest for monitoring underground nuclear explosions and are a focus of the IMS because underground and underwater detonations are expected to have low noble gas releases to the atmosphere; see Figure 3. Xenon was chosen for nuclear explosion monitoring because noble gases are chemically inert, remain gaseous, and are likely to escape from underground nuclear explosions. In addition, noble gases are not washed from the atmosphere by precipitation. This situation is in contrast

with most fission products, which condense and attach to aerosols in the atmosphere or, in the case of an underground nuclear explosion, are retained in the ground. Xenon has several unique properties among the noble gases, including

- It has the highest fission yield
- The half-lives that are appropriate for atmospheric transport from the source to the detector (the half-lives for <sup>131m</sup>Xe, <sup>133m</sup>Xe and <sup>135</sup>Xe are between 9.1 hours to 11.8 days)
- The half-lives are short enough to keep atmospheric "memory" low (e.g., ensure low atmospheric background concentrations (unlike <sup>85</sup>Kr)



Figure 3. Signatures of nuclear explosions

Three systems are deployed for IMS noble gas monitoring: SPALAX (France), SAUNA (US), and ARIX (Russia). These automated systems were specifically developed for the IMS and use similar principles to detect  $\leq 1 \text{ mBq/m}^{3 \text{ 133}}$ Xe in the atmosphere by extracting and concentrating xenon before placing the sample in front of the detector (this is not the case for facility stack detectors). The challenge to the IMS is to be able to distinguish background emission from emission from nuclear explosions; see Figure 4.

Source	Emissions
1 kT nuclear explosion	Atmospheric test: 10 <sup>16</sup> Bq Underground test: 10 <sup>14</sup> Bq Global impact / all isotopes
Nuclear reactors	Global release: 10 <sup>15</sup> Bq / a Regional impact / mainly 133Xe
Hospitals	Local impact / <sup>131m</sup> Xe
<sup>99m</sup> Tc production	Per facility 10 <sup>15</sup> Bq/a Global impact / all isotopes

Figure 4. Radioxenon emissions from different sources

There are two approaches to estimate the impact of radioxenon background on the ability of IMS to detect a nuclear explosion. First, with the knowledge of the background source term (i.e., MIP stack monitoring), ATM can be used to estimate the impact of the background on IMS stations. The second approach is to characterize the background using mobile systems to measure radioxenon backgrounds in different areas of the planet. Several radioxenon background measurement campaigns have been conducted with the objective of discriminating between CTBT-relevant events vs. radioxenon background near major contributors such as MIP facilities, NPPs, and hospitals. These campaigns in large part have been funded by the European Union (EU) through Council Decisions II, III, V, and VI (ongoing). These campaigns have been conducted since 2008 in Belgium, Kuwait, South Africa, Thailand, Indonesia, and Japan. High backgrounds measured near Jakarta are potentially from the INUKI MIP facility. These high backgrounds decrease the sensitivity against a real event by increasing stations' minimum detectible concentration (MDC) for real events. In addition, they can lead to false positive categorization of an event due to the continuous presence of radioxenon and treaty relevant radioxenon ratios.

Radioxenon monitoring is a key component in verification of the CTBT; among all IMS monitoring technologies, it can provide the strongest evidence for underground nuclear explosions. MIP has a major effect on the global radioxenon background; therefore, distinction of this background from nuclear explosion emission is crucial for signal interpretation. This leads to the following question: How can the impact of radioxenon background on the data analysis be lessened to a manageable level?

#### 2.3 Radionuclide Processing at the IDC

The CTBTO PrepCom gave an overview on IDC processing of radionuclide data received from the IMS. The IDC is a central element of the CTBT monitoring and verification mechanism that collects, processes, and analyzes data originated in the 337 IMS stations and radionuclide laboratories. Results from the data received from these stations are presented as lists of events, bulletins, and reports to Member States. Based on this information, States are enabled to make judgments about the nature of a suspect event, whether such an event has taken place, and whether such an event may indeed have been a nuclear explosion. The IDC also archives all data and data bulletins in its computer center.

The IMS radionuclide network is designed to have at least a 90% capability to detect a 1 kt nuclear explosion within approximately 14 days by identifying radionuclides in the atmosphere originating from an underground or underwater detonation. Data collected from radionuclide stations—80 particulate, 40 equipped with noble gas when the IMS is fully built—is analyzed by the IDC processing pipeline for radionuclides; see Figure 5. This pipeline consists of automatic processing, interactive review, categorization, and reviewed products. The types of radionuclides typically seen by the IMS include naturally occurring isotopes are the following:

- Radon decay products: <sup>212</sup>Pb, <sup>212</sup>Bi, <sup>208</sup>Tl, <sup>214</sup>Bi, <sup>214</sup>Bi, etc.
- Terrestrial radionuclides: <sup>226</sup>Ra, <sup>228</sup>Ac, <sup>235</sup>U, <sup>234</sup>Th, <sup>40</sup>K, etc.
- Cosmic-ray induced radionuclides: <sup>7</sup>Be, <sup>22</sup>Na, Ge isotopes, etc.

The IMS also typically sees the following anthropogenic isotopes:

- Non-relevant radionuclides that are not associated with nuclear weapons, including some radiopharmaceuticals and accelerator products: <sup>241</sup>Am, <sup>109</sup>Cd, etc.
- CTBT-relevant radionuclides: possibly from nuclear explosions (fission and activation products)

Radionuclides of interest are those that would be expected to be released from a nuclear explosion. Two radionuclides, <sup>135</sup>Xe and <sup>133</sup>Xe, are abundant in a 1 kt nuclear explosion, with activities increasing after a couple of days due to product formation from beta-decay chain from precursors. These radionuclides are considered relevant as nuclear explosion indicators because their presence or their mutual ratios can be used to discriminate other possible sources of radioactivity.



Figure 5. IDC processing pipeline for radionuclides

There have been recent enhancements to IDC processing of particulate samples. Within 2015 IDC development efforts, several areas were identified for further improvement to enhance the quality of automatic processing results of particulate samples and to reduce the workload on analysts in interactive

mode. The priorities of these efforts are to reduce the 11,000 observed false peaks and 33,500 observed false positives and to better understand CTBT relevant nuclides in background. Several solutions were defined to accomplish these goals:

- Automatic commenting-out of false peaks
- Optimization of the key lines of 7 nuclides
- Automating the discrimination tool between <sup>99m</sup>Tc and <sup>75m</sup>Ge
- Implementing a background subtraction module in automatic mode
- Updating <sup>212</sup>Pb and <sup>214</sup>Pb nuclear data in the library

Several of the defined goals have been addressed. First, software to perform automatic commenting-out of false peaks has reduced the work required by analysts to comment-out by 40-50%. This automatic labeling is visible to analysts via the review tools (Saint2 @IDC, OpenSpectra@NDC) and analysts can act on pre-review results as appropriate, using the same interactive functionalities as for their own review actions. If not changed during the interactive review, automatic labeling of peaks will be reflected in the reviewed radionuclide report after the sample is released. Second, a tool for discrimination of the CTBT-relevant nuclide <sup>99m</sup>Tc (140.51keV) from <sup>75m</sup>Ge (139.68 keV)—which is produced by interaction of neutrons from cosmic radiation with the germanium crystal of the detector itself—was developed and implemented. This tool uses energy discrimination between <sup>75m</sup>Ge and <sup>99m</sup>Tc (only applied if the peaks are large enough to allow centroid determination with low uncertainty). Peak ratio between <sup>75m</sup>Ge to <sup>71m</sup>Ge is used to determine whether <sup>99m</sup>Tc is present. Finally, improvements in <sup>212</sup>Pb family, <sup>214</sup>Pb, and <sup>109</sup>Cd nuclear data in the library have been implemented. The overall impact on the quality of automatic results is an improvement in the consistency rate of automatic categorization of particulate samples vs. implemented software solutions; see Figure 6.



#### Improvements in the consistency rate of automatic categorization of particulate samples VS. implemented software solutions

Figure 6. Overall impact on the quality of automatic results for radionuclide data

#### 2.4 Issues in the MIP World

Nordion gave a presentation on issues that are a concern in the MIP world. Prior to the first WOSMIP, the MIP community did not realize there was a problem related to emissions released from MIP facilities until issues were discussed and it became understood that radioxenon emissions were being detected by the IMS. These releases from MIP were well below health or safety limits; Nordion's design goal of release mitigation was limiting dose to public <1 mSv/yr. Release limits for a facility are calculated based on the site limits and the pathway of exposure. Historically, producers have released approximately 0.01 mSv/yr (approximately 1% of the limit). These releases are small compared to natural doses, which range from 1.3 -3.5 mSv/yr in major Canadian cities—1.8 mSv/yr is the Canadian average dose from natural sources.

It is not necessarily easy for a producer with existing processes to lower emissions. Before a real discussion related to reduction of releases can begin, a design objective is required. It is easier to design a new process facility to achieve lower emissions than to modify an existing facility.

When reducing releases from a facility, there are Action Level concerns with regulators. Regulators expect licensees to have control of their processes. Control is demonstrated by the consistency in facility emissions. Regulators may require Action Levels even if emissions are below regulatory release limits if a loss of control is observed; see Figure 7. Therefore, if a new mitigation system does not function perfectly, lack of reliability may cause a requirement to report to regulators—even when there is no effect on safety.





Other issues that the MIP community is dealing with include conversion from HEU to LEU; establishment of full cost recovery with many <sup>99</sup>Mo production subsidized by national governments; institutional inertia—it takes time to influence and get institutions on board; an aging nuclear infrastructure, which makes changes to existing processes difficult; and quality assurance concerns—the community wants consistency because changes affect quality assurance.

To help alleviate the problem created by detection of MIP emissions by the IMS, the producers are being asked to incorporate processes that have potential reliability problems, increase costs, add complexity to the process, increase waste management needs (e.g., more waste containers), and share data that a competitor may use against a producer's business. Meanwhile, demands for medical isotopes are increasing, in this business where time of processing is critical and reliability is crucial in the marketplace. However, there are reasons for optimism because most producers are happy to share release data (as long as it is kept confidential) and think lowering releases is a good thing; new processes and

LEU conversion gives producers an opportunity to implement new design release objectives; and WOSMIP has provided a format for discussing a new design objective and sharing emission reduction technology.

#### 2.5 Progress on the IAEA CRP on Sharing and Developing Protocols to Further Minimize Radioactive Gaseous Releases to the Environment in the Manufacture of Medical Radioisotopes, as Good Manufacturing Practice

The IAEA has been involved in several activities related to <sup>99</sup>Mo production. These activities include conversion from HEU to LEU targets, indigenous production using non-HEU targets, small-scale production of <sup>99</sup>Mo or <sup>99m</sup>Tc for local use and associated regulatory aspects, new alternatives to <sup>99m</sup>Tc radiopharmaceuticals, and coordination and participation in the Organization for Economic Co-operation and Development's Nuclear Energy Agency's High-level Group on the Security of Supply of Medical Radioisotopes. The main focus of the presentation was an update on the CRP on Sharing and Developing Protocols to Further Minimize Radioactive Gaseous Releases to the Environment in the Manufacture of Medical Radioisotopes, as Good Manufacturing Practice. This CRP began when a request was received from Australia, Belgium, the Netherlands, Republic of Korea, and the United States in May 2014. This request proposed that the IAEA initiate a CRP on the topic of technologies to reduce emissions from MIP facilities. The CRP will identify important technical issues and is open to all Member States.

The overall CRP objective is to share and develop protocols to further minimize radioactive gaseous releases to the environment in the manufacture of medical radioisotopes, as good manufacturing practice. To achieve this goal, steps will be identified in the medical radioisotope production process that need proper gaseous emission monitoring and trapping; research focused on efficient mitigation methods of treatment and processing of radioactive gaseous emissions will be conducted; stack measurement systems for radioactive gases will be evaluated; and data exchange methodologies and formats will be explored.

The CRP is expected to produce a document containing guidelines on how to minimize and mitigate the radioactive gaseous releases to the environment resulting from the production of medical radioisotopes via the irradiation and processing of uranium targets, according to the best available technologies and Good Manufacturing Practice (GMP) and Good Laboratory Practice (GLP) regulations.

The first Research & Coordination Meeting was held 17–21 August 2015 in Vienna, Austria. During this meeting, the group presented on their activities supporting the CRP's objectives; formulated work plans for each participant; established specific areas of cooperation between participants; prepared conclusions from the meeting and provided a set of recommendations; and drafted country reports. A second Research & Coordination Meeting is scheduled for 6-10 March 2017 in Vienna, Austria.

#### 2.6 IMS Radionuclide Monitoring after the Democratic People's Republic of Korea 2016 Announced Tests

A discussion of IMS data related to the Democratic People's Republic of Korea (DPRK) announced nuclear explosions was presented by the CTBTO PrepCom. While seismic data may be sufficient to identify an explosion, seismic data alone cannot identify an explosion as nuclear. To identify whether the explosion is nuclear, the IMS radionuclide network would be expected to detect radionuclides in the atmosphere. Hence, radionuclides must be released to the atmosphere in an underground nuclear explosion for detection by an IMS radionuclide station. Whether radionuclides reach the atmosphere from

an underground nuclear explosion is dependent on many technical factors, including depth, geological conditions, explosive yield, prevailing atmospheric pressure conditions, etc.

On October 9, 2006, DPRK announced a test that was detected by IMS seismic stations; see Figure 8. Using Provisional Technical Secretariat (PTS) Atmospheric Transport Modeling, radioactive noble gas observations at the Yellowknife, Canada station were shown to be consistent with a hypothetical release of radioactive xenon at the time and location of the DPRK event. Other known sources of man-made radionuclides could be excluded.



Figure 8. Seismic measurements after the DPRK announced nuclear test on October 9, 2006

Another DPRK nuclear test was announced on 25 May 2009. The relevant radioactive xenon signals were not detected by the IMS or reported by other agencies. In this instance, no conclusion can be drawn on the nature of the event due to the lack of an observed radioxenon signal.

A third nuclear test was announced by DPRK in February 2013. Although initially there were no relevant radioactive xenon signals detected by the IMS, there were three consecutive noble gas detections at the JPX38 station that were among the four highest detections at that station during the past 15 months. In addition, these three detections differed in their <sup>131m</sup>Xe and <sup>133</sup>Xe ratios from previous detections. If the detections were the result of fission, the isotopic ratio indicates that the fission occurred approximately 51 days earlier (+5/-3 days); this information corresponds to the announced DPRK test that was conducted 55 days before the detections.

DPRK announced a fourth test on January 6, 2016. Again there was no immediate release from the DPRK event, but 42 days after the seismic event, a Level C episode, an observation above the radioxenon threshold for that station, was assigned due to detection of <sup>133</sup>Xe at station JPX38. Occasional "abnormal" values like this episode occur about 10 times per year whereas normal concentrations are regularly observed at this station, and about 500 detections occur at the JPX38 station annually with two samples

taken per day. Therefore, it is important to continue efforts to understand the background for State Signatories to interpret measurements.

Another DPRK event was conducted on September 9, 2016. ATM forward modelling of the event, assuming a release at the DPRK test site, shows the plume broadly spreading and possibly reaching the radionuclide stations at Takasaki, Japan; Ussuriysk, Russian Federation; Guangzhou, China; Beijing, China and Ulaanbaatar, Mongolia. Following this event several samples categorized at level C were detected at the Chinese station CNX22 between September 22 and 27 of 2016, although there was no significant correlation between actual detections of <sup>133</sup>Xe and <sup>131m</sup>Xe at CNX22 and ATM simulation. No other level C categorized samples or observations were detected at nearby stations in the relevant time-frame of the September 9 DPRK test.

There was no relevant radionuclide observation determined to be associated with either of the 2016 seismic DPRK events. For these events, the estimated depth of burial would make the probability of rapid venting low although operational releases could come at any time. Typical radionuclide background in the atmosphere is expected and can account for some observed measurements above normal; therefore, it is important to continue efforts to understand the background.

Based on past experience (explosions in 2006, 2009 and 2013) the behavior of detected radionuclides can vary.

- In 2006: observations were consistent with a release from the announced test
- In 2009: there were no radionuclide observations
- In 2013: the observations of a late release were clearly related to the event

In 2016, a late release from the DPRK site could have been observed, and therefore any significant detection was followed by careful analysis for its possible association with the seismic event of January 6 and September 9, 2016.

It should be noted that no conclusion can be drawn on the nature of the event from non-observation of related radionuclides. After entry into force of the CTBT, detecting of the seismic event could be sufficient for an on-site inspection to find the evidence for Member States to make a final judgement on whether the event was a nuclear explosion.

### 3.0 Alternative Xenon Source Terms: The Broader Range of Isotope Sources

This session dealing with alternative xenon sources emphasized their impact on global radioxenon backgrounds, particularly on the next-generation NPPs, the diagnosis and treatment with nuclear medicine, and efforts set in the conversion from HEU to LEU.

#### 3.1 Potential Sources of Isotope Emissions from Next-generation Nuclear Reactors

A discussion by the University of Texas detailed relevant features of prominent next-generation nuclear power reactor designs, their potential impact on isotope release levels, and potential steps for mitigation.

These new generator designs, Generation III+ and IV, hold the potential to be safer and more efficient than current light-water reactor technology, but new approaches may result in increased isotope releases at levels relevant to nuclear explosion monitoring. Current nuclear reactors mainly consist of boiling water reactors, which contain xenon within the fuel and therefore have minimal releases. A number of reactor designs—Gen IV Reactors—are being investigated for development, including Supercritical-Water-Cooled Reactors (SCWR), Very-High-Temperature Reactors (VHTR), Gas-Cooled Fast Reactors (GFR), Lead-Cooled Fast Reactors (LFR), Sodium-Cooled Fast Reactors (SFR), and Molten Salt Reactors (MSR).

It was concluded that SCWR technology would not have major differences in the radioxenon releases compared to current reactors. The VHTR and GFR use a different pellet fuel. These pellets have been found to fail over time and may release radioxenon gas. More research is needed to understand gas transport in this fuel matrix. If isotopes are likely to migrate out of the matrix and into the coolant gas, then the WOSMIP community should engage with GFR and VHTR designers. Although the MSR is presented as being safer and more efficient than current reactors based on light water technology, they would potentially increase isotope releases, particularly radioxenon and radioargon. The levels of radioxenon released from an MSR would likely be relevant for nuclear explosions monitoring. If the current worldwide fleet was replaced with MSR reactors producing 1000 GW<sub>th</sub>, then they would have the potential of releasing  $10^{20}$  Bq/d <sup>133</sup>Xe (no filtration). Xenon filtration of 99.9999999% would be required to meet a  $10^9$  Bq/day <sup>133</sup>Xe release level (29 half-lives – 152 days). Therefore, it was suggested that the WOSMIP community should engage with MSR developers.

#### 3.2 Radioxenon Potential Emission Analysis from Acceleratorproduced Alpha-particle Emitting Radionuclides Facilities

INVAP presented a conceptual analysis of the potential production and release of radioxenon from accelerator-produced alpha-particle emitting radionuclides facilities using <sup>232</sup>Th and <sup>226</sup>Ra targets to verify the potential impact to the global CTBT verification, together with the comparison with main production parameters from other typical MIP facilities.

Radioisotope production by accelerators is a well-developed technology. Since 2014, the Argentinian National Atomic Energy Commission (CNEA) has shown an increasing interest in developing technology to produce <sup>225</sup>Ac through cyclotron proton irradiation. The work presented was an analysis of <sup>133</sup>Xe production for a facility to producing <sup>225</sup>Ac through two different channels:

- a)  $^{232}$ Th (p,4n)  $^{229}$ Pa where  $^{299}$ Pa  $\rightarrow \alpha + ^{225}$ Ac (0.48%)
- b)  $^{226}$ Ra (p,2n)  $^{225}$ Ac.

The expected fission products from <sup>232</sup>Th and <sup>226</sup>Ra targets were modeled to understand the potential amount of radioxenon produced; see Figure 9. The calculated results indicate that <sup>133</sup>Xe produced from the accelerator produced reactions is orders of magnitude below that produced by a typical <sup>99</sup>Mo plant. In addition, <sup>133</sup>Xe and <sup>133m</sup>Xe activities are higher in the <sup>232</sup>Th reaction (~1-2e9 Bq/run <sup>133</sup>Xe produced) than for the <sup>226</sup>Ra reaction. Based on these results, it was concluded that complex emission abatement technologies are not foreseen as necessary for those facilities. This finding is important as radioisotope production using accelerators is expected to become more important in the future.



Figure 9. Example of calculated fission yields

#### 3.3 Problems of Radon Isotopes' Fixation and Detection during the Processing of Irradiated <sup>226</sup>Ra

The Kholpin Radium Institute presented their results on radon isotope fixation and detection during processing of irradiated <sup>226</sup>Ra to produce  $\alpha$  emitters for medical treatment. An increasing number of radioisotopes are used in the field of diagnosis and treatment of different diseases. In that regard, a lot of scientific research is related to use of alfa emitting nuclides, such as <sup>213</sup>Bi, <sup>212</sup>Pb, and <sup>209</sup>Bi, in radiotherapy of cancerous diseases. The most effective way to obtain these radionuclides is to irradiate <sup>226</sup>Ra with the subsequent processing of irradiated material; however, this processing is accompanied with generation of gaseous radon isotopes. Due to their low chemical activity and components that hinder radon isotopes are alpha emitters and their intake can cause serious harm. Therefore, methods for the detection, capture, and fixation of radon are being studied.

To effectively capture <sup>222</sup>Rn, a sealed dissolution unit is required that incorporates a method to capture the radon. In order to design an apparatus to capture <sup>222</sup>Rn, methods to detect and determine <sup>222</sup>Rn in the apparatus are being investigated. Possible methods for detection include radiometric methods, measurement of radioactive decay products, and concentration by chemical coupling or adsorption. An initial study of activated carbon zeolite and fullerene adsorbents found that the activated carbon had the largest adsorption coefficient for radon. Other potential methods for the capture of radon are formation of nonvolatile salt complexes or cryogenic trapping.

#### 3.4 RA3 Research Reactor, Base of the Reactor Radioisotope Production in Argentina

CNEA gave a presentation on the RA3 reactor. The RA3 reactor is a 10 MW open pool type reactor that was converted from HEU to LEU in 1989. In addition to radioisotope production, the reactor is used for neutron activation analysis, nuclear instrumentation qualification, research in irradiation damage, and nuclear fuel qualification. The most important radioisotopes produced at RA3 are <sup>99</sup>Mo and <sup>131</sup>I, which are both produced by fission of LEU targets. Twelve LEU targets are typically irradiated weekly for <sup>99</sup>Mo production, each containing 1.4g of <sup>235</sup>U.

Annual noble gas releases from the RA3 reactor were reported for 2005-2015; see Table 1. These releases are almost exclusively <sup>41</sup>Ar, which varies between 22 TBq to 36 TBq. The contribution of <sup>133</sup>Xe and <sup>135</sup>Xe are negligible, and <sup>131</sup>I release varies between 0.92 MBq to 1.48 MBq.

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
<b>Xe-133</b> [TBq]	1,56E-02	1,75E-02	1,58E-02	8,67E-03	6,86E-04	9,13E-04	6,47E-04	5,60E-04	4,89E-04	3,72E-04	3,72E-04
<b>Xe-135</b> [TBq]	5,47E-02	1,19E-01	1,09E-01	1,30E-02	2,16E-03	1,58E-03	1,68E-03	1,51E-03	1,21E-03	1,31E-03	7,53E-04
<b>Ar-41</b> [TBq]	2,94E+01	2,77E+01	2,69E+01	2,53E+01	2,93E+01	3,34E+01	3,56E+01	3,12E+01	2,72E+01	2,63E+01	2,26E+01
E1 [TBq]	2,94E+01	2,78E+01	2,70E+01	2,53E+01	2,93E+01	3,34E+01	3,56E+01	3,12E+01	2,72E+01	2,63E+01	2,26E+01

Table 1. Annual noble gas releases from CNEA

#### 3.5 Radioxenon Emissions from Research Reactors

A study of radioxenon emissions from TRIGA Mark II research reactor was presented by the Vienna University of Technology and the Swedish Defense Research Agency (FOI). The 250 kW reactor—250 MW in pulse mode—has a mixed core loading uranium zirconium hydride (UZrH) fuel (LEU and HEU) and has been in operation since 7 March 1962 (10,000 days or 50,000 hours of operation). This study measured xenon released from the reactor using three different methods.

A Swedish Automatic Unit for Noble Gas Acquisition (SAUNA II) designed to monitor the CTBT relevant xenon isotopes—<sup>131m</sup>Xe, <sup>133m</sup>Xe, <sup>135</sup>Xe—was used to monitor the background near the reactor. Sampling was conducted during different operation periods: Background sampling occurred on weekends when the facility was not in operation; normal operating sampling was conducted during normal operation with the cooling on; and sampling was conducted during operation without cooling (enhanced convection and diffusion).

A second measurement at the reactor was conducted to detect fuel element failure. Reactor failure typically occurs in the region of contact of graphite-fuel cylinders and can be observed via activity-increase in primary circuit resulting from a major leakage. The wet sipping method was used to identify leakage. This method draws water through a loop from above the reactor core around a germanium detector and back to the water tank; see Figure 10. Leaking elements can be determined by comparing the measured value for <sup>89</sup>Kr and <sup>139</sup>Xe at different locations and comparing these values to the expected values for a non-leaking system.



Figure 10. The wet sipping method draws water from above the reactor core through a loop to be measured by a germanium detector

A third measurement of air collected from within the reactor was analyzed. Radioxenon detected in air collected at multiple locations inside the reactor could originate from several possible sources, including pinhole leaks in the fuel cladding, activation of air, and uranium contamination on fuel surface.

The measured and theoretical counts for each of the three measurements were compared (see Figure 11), and the measured values matched well with simulated plots. It should be noted that some of the theoretical counts and observed measurements fall in the nuclear test domain of the plots (to the right of the red line), see Figure 12.



Figure 11. Comparison of measured and theoretical plots

In February 2010, a leaking fuel element was identified from the sudden release of gas bubbles that contained moderate activities of different xenon isotopes and <sup>85</sup>Kr (total activity in the range of kBq). The leaking fuel element was removed from the core, and in December 2012, the last HEU fuel was exchanged with LEU. In light of these changes, a new measurement campaign would be of interest.

# 3.6 Brazilian Multipurpose Reactor: The New Brazilian Multipurpose Research Reactor

The Brazilian Nuclear Energy Commission (CNEN) presented an update of technical information and the development status of the 30 MW LEU Brazilian multipurpose reactor (RMB) project. The Brazilian reactor is planned be built about 100 km from Sao Paulo and include radioisotope production capacity that will reduce Brazil's dependence on external sourcing of radioisotopes used in nuclear medicine. Other capabilities of this reactor include fuel and material irradiation testing, a neutron beam laboratory, a neutron activation analysis laboratory, and education and training; see Figure 12.

The RMB is a key factor for supplying radiopharmaceuticals for nuclear medicine applications in Brazil. One thousand 6 day Ci/wk <sup>99</sup>Mo will be produced from LEU target irradiation and processing. In addition, radioisotopes for injectable radiopharmaceuticals will be produced including: <sup>99</sup>Mo/<sup>99m</sup>Tc ,<sup>131</sup>I, <sup>51</sup>Cr, <sup>153</sup>Sm, <sup>177</sup>Lu, <sup>166</sup>Ho, <sup>90</sup>Y, <sup>188</sup>W, and <sup>32</sup>P.

Conceptual and basic engineering designs have been completed for the RMB project and the development stage for fuel assembly development and fabrication is underway. In addition applications for nuclear and environmental licensing are ongoing.



Figure 12. Planned RMB nuclear research and production facility

#### 3.7 Contribution of Normal Operational Releases from Nuclear Power Plants to the Global Radioxenon Emission Inventory

The CTBTO PrepCom presented information on NPP contributions to the global radioxenon emission inventory. The presentation provided updates for the release period of the calendar year 2014. In addition, main sources of information for generation of new updates were introduced.


Figure 13. Map of nuclear power plants (Red Dots) and IMS Stations (Blue Dots). Nuclear power plants are concentrated in certain regions.

Radioxenon releases from NPPs were estimated for the year of 2014 using methodology outlined in Kalinowski & Tuma, 2009[2]. These estimations were based on generic annual releases per reactor with no distinction between continuous and pulsed releases. In addition, reactors not existing in the Kalinowski & Tuma, 2009 study were considered, and reactors locations are mapped in Figure 13. The expected radioxenon released from these NPPs is plotted on a 4 xenon isotope plot in Figure 14. Three of the NPPs were found to have expected releases to the right line of the discrimination line in this plot. Therefore, NPPs should be considered as radioxenon sources.



Figure 14. Expected radioxenon released from NPPs. Three of the results have similar radioxenon ratios to nuclear explosions.

An estimation of radioxenon activity concentration at IMS locations was also studied. These ATM calculations used FLEXPART incorporating operational Source-Receptor Sensitivity (SRS) fields from the IDC and historical European Centre for Medium-Range Weather Forecasts (ECMWF) numerical weather data. Observations at IMS stations were compared with the calculated data to determine the average observed vs estimated activity concentration. In average over all samples, each IMS noble gas system shows an underestimation of the real observations. The IMS stations BRX11, DEX33 and SEX63 had the best agreement with the majority of samples being estimated within a factor of 10 of the IMS detections. All other stations had a larger under-estimation. This underestimation could be from other sources such as an additional MIP, continuous assumption instead of pulsed release, or an underestimation of releases.

#### 3.8 Xe-133 Medical Use Diagnosis of Pulmonary Embolism

Lantheus Medical Imaging presented information on products used to diagnose coronary artery disease, congestive heart failure, stroke and other diseases focusing on the use of <sup>133</sup>Xe gas. Xenon-133 gas is the typical medical imaging agent for pulmonary embolism diagnosis in the United States but is being replaced by Technegas (<sup>99m</sup>Tc) procedures in Europe and Canada. In addition to medical applications, <sup>133</sup>Xe is used industrially as a tracer for oil and gas well logging and as an NPP tracer.

There are approximately 600,000 pulmonary embolism cases per year and it is the third leading cause of U.S. hospital death. The projected U.S. pulmonary embolism examinations are 1.4M/year. Currently, there are two imaging methods to assess pulmonary embolism: Computed Tomography Pulmonary Angiography which is the preferred modality per medical guidelines due to accuracy, high specificity, availability, radiation exposure concerns for some patient populations, and poor patient tolerance of contrast media; and Ventilation/Perfusion Nuclear Study which uses a diethylenetriamine-pentaacetic acid (or DTPA)-<sup>99m</sup>Tc aerosol or <sup>133</sup>Xe gas. In both of these methods, a mismatch comparison between ventilation and perfusion images provides the diagnosis of pulmonary embolism; see Figure 15.



A) After inhalation of 20.1 mCi of Xenon-133 gas, scintigraphic images were obtained in the posterior projection, showing uniform ventilation to lungs



B) After intravenous injection of 4.1 mCi of Technetium99m- labeled macroaggregated albumin, scintigraphic images were obtained, shown here in the posterior projection.

Figure 15. Example of a mismatch comparison in a lung ventilation and perfusion (VQ) study

To reduce radioxenon emissions, Lantheus uses a trapping system to capture radioxenon. This system passes exhaled air through a cold trap followed by a desiccant column and charcoal trap before being released. Environmental stack monitoring of <sup>133</sup>Xe releases is performed using an in-stack Geiger-Mueller detector connected to scaler with background subtraction. Release data is shown in Figure 16. Regulatory concentration limits for exhaust stack air emissions at Lantheus are  $5x10^{-7} \mu$ Ci/mL annual concentration limit.



Figure 16. Xenon-133 released from the Lantheus facility from 2004 to 2015

IRE currently provides <sup>133</sup>Xe to Lantheus and on January 21, 2015 Lantheus announced a new strategic agreement with IRE for future supply of <sup>133</sup>Xe gas. IRE provides the unprocessed radiochemical <sup>133</sup>Xe to Lantheus for processing and finishing. Additional diversification options and LEU-based <sup>133</sup>Xe are also being investigated.

#### 3.9 On the State Program Tajikistan on Restoration of the "Argus" Liquid Nuclear Reactor and Plans for the Production of Radiopharmaceuticals

Tajikistan introduced plans for the solution nuclear research reactor "Argus-FTI" which is installed at the Physical-Technical Institute; see Figure 17. The 50kW reactor uses a 25L aqueous  $UO_2SO_4$  fuel solution (90% <sup>235</sup>U). Some possible applications of the reactor include: neutron activation analysis, neutronography, industrial exposure, isotope production, filter manufacturing, training, and neutron modification of materials.



Figure 17. Model of the ARGUS FTI reactor

The ability to place the Argus FTI reactor in the city without compromising the public and the environment (sanitary protection zone of 50 m) opens up the potential for use in medicine, molecular biology, and other fields (in particular, production of <sup>99</sup>Mo (<sup>99m</sup>Tc); <sup>89</sup>Sr; etc.). After modernization for manufacture of isotopes, the small capacity reactor (~30-50 kW) will be able to possess the guaranteed safety based on properties of self-regulation on low enriched fuel; be reliable and simple in industrial operation; have small volume of active zone (~25 L); have low cost of modernization (compared with building a new reactor complex); and produce insignificant quantity of radioactive wastes. In addition to housing the Argus reactor, the nuclear-technological complex on manufacture of medical radioisotopes will be equipped for extraction of isotopes and preparation of generators.

The reactor will also provide activation analysis of most elements with a capacity of 300 thousand elements per year and the sensitivity of  $10^{-6}$ - $10^{-9}$ g/g for use in various fields of economy and physical studies on the beams of neutron radiation.

The modernization project will be conducted in two stages. Stage I of the project (2017-2019) will include determining general maintenance and safety rules for the reactor and nuclear-technological complexes; conducting professional training, design, and exploration studies; determining design documentation on reactor reconstruction; developing documentation on technological reactor equipment; and developing documentation on building a nuclear-technological complex for manufacture of radioisotopes. Stage II (2020-2022) will include reconstruction of the buildings' reactor complex, Argus FTI; building nuclear-technological complex on manufacture of radiopharmacy isotopes; purchasing, installing and placing into operation reactor and nuclear-technological complex equipment; maintaining Physical-Technical Institute equipment for carrying out of research work on reactor and nuclear-technological complexes; documenting problems of environmental protection and working with the public; and carrying out of annual international conferences on the applications of nuclear-physical methods in a national economy to prospects of nuclear medicine.

#### 4.0 Current Mo-99 Production Overview: Recent and Planned Activities at Isotope Production Facilities

The overview session on current <sup>99</sup>Mo production was initiated with the IAEA's global perspective on the topic and it was followed by presentations from specific countries and industries. Detailed information shared by producers will help the community better understand the effects of MIP on monitoring.

#### 4.1 Mo-99 Production Overview

The IAEA presented in their current work related to <sup>99</sup>Mo production beginning with a review on medical isotope supply being conducted by the Organization for Economic Co-operation and Development. The review highlighted the many factors affecting the current <sup>99</sup>Mo supply situation, including the shutdown of OSIRIS reactor in December 2015; the return to service of the BR2 reactor on July 2016; cessation of MIP at National Research Universal (NRU) on Oct. 31, 2016; conversion to LEU targets; and new production planned to enter the market. The review found that <sup>99</sup>Mo global demand for 2016 has been maintained at a level of around 9,000 6-day Ci per week. The current irradiator and processor supply chain capacity should be sufficient throughout 2021 if well-maintained, planned and scheduled— assuming no unplanned outages; see Figure 18. However, there is still a need to add processing capacity by 2017, which could be accomplished through the on-time introduction of substantial conventional processing capacity in Australia and the introduction of alternative irradiation and processing technologies. In addition, the extension of the NRU operating period could be a useful stop-gap in 2017 and early 2018, with the provision of substantial contingency capacity. The supply situation will continue to require careful and well-considered planning to minimize supply risks. While supply is secure, economic sustainability remains a challenge.



**Figure 18**. Current irradiator and processor supply chain capacity should be sufficient throughout 2021. Current demand of 9000 6-Day Ci <sup>99</sup>Mo/Week (red line); current demand +35% ORC vs. total irradiation capacity and total processing capacity (green line); total processing capacity (orange line); and total irradiation capacity (blue line). Note: predictions assume 50% of assessed new projects will succeed one year later than scheduled and there are no unscheduled outages of existing facilities.

A report on <sup>99</sup>Mo for medical imaging was released in 2016 by the National Academies of Sciences, Engineering, and Medicine. This report includes a list of facilities that produce <sup>99</sup>Mo for medical use and whether these facilities use HEU; a summary of the international production of <sup>99</sup>Mo over the previous five years, including new production; unexpected halted production; decommissioned or otherwise permanently removed from service; progress on establishing domestic (U.S.) production; the adequacy of <sup>99</sup>Mo supplies to meet future domestic medical needs, particularly in 2016 and beyond; and progress made to eliminate worldwide use of HEU in targets and MIP facilities.

Currently, several major <sup>99</sup>Mo producers have converted to LEU targets. Australia (ANSTO) uses only LEU targets, and South Africa (NTP) was the first producer to convert to LEU production in June 2010 and has been steadily increasing LEU production (2014: 38%, 2015: 47%, 2016: 77%). Cold commissioning with LEU targets is ongoing in Belgium (IRE) and conversion to LEU targets is expected in 2017. Mallinckrodt, in the Netherlands, has completed cold runs, R&D runs, and yield test runs, and complete conversion is expected by the end of 2017. Russia (RIAR) has made no formal commitment but currently has mock-up LEU targets and has stated plans to convert to LEU targets in 2017/2018.

#### 4.2 Radioisotope Production at National Atomic Energy Commission of Argentina

CNEA presented a review of radioisotope production in Argentina. The presentation referred to their evolution through 50 years of production, the development of production technology with LEU and technology transferences of the method. CNEA has been producing <sup>99</sup>Mo from fission commercially since August 1985, and in 2002, the reactor was converted from HEU to LEU. In addition to <sup>99</sup>Mo several other

isotopes are produced by irradiation, including the reactor products <sup>131</sup>I, <sup>153</sup>Sm, <sup>32</sup>P, <sup>51</sup>Cr, <sup>181</sup>Hf, <sup>177</sup>Lu, <sup>192</sup>Ir, and <sup>133</sup>Xe and <sup>60</sup>Co.

The current commercial production of fission <sup>99</sup>Mo and <sup>131</sup>I at CNEA fulfills the national demand in Argentina and one-third of the Brazilian market (weekly exportation). Additionally, 15–20% of total production volume is being exported to other Latin American countries through two national private companies, in the form of <sup>99m</sup>Tc generators and <sup>131</sup>I doses and capsules.

The RA-10 multipurpose reactor project began on June 2010 with the purpose to consolidate and enhance radioisotope production, supply facilities for irradiation of fuel elements and materials, and offer new applications and services in the fields of science and technology. The installation of the RA-10 reactor will be accompanied by a new fission radioisotope production plant. This new plant will have nine main hot cells and seven auxiliary cells, a capacity of 2500 Ci <sup>99</sup>Mo/wk (6 day) and 400 Ci <sup>131</sup>I/wk. They plan to start construction of the new building at the end of 2017 and start production in 2020.

#### 4.3 Update on Mo-99 Production and Xenon Abatement at ANSTO

ANSTO gave an update on MIP and emission abatement systems at their current and new MIP facilities. The current <sup>99</sup>Mo operations use a base digestion process. The original capacity was 1100 six-day Ci <sup>99</sup>Mo per week—normally run below full capacity. In 2015, ANSTO was granted regulatory permission for periods of increased capacity up to 1300 Ci per week to mitigate world <sup>99</sup>Mo shortage. This capacity limit is determined by emission levels, which are mitigated using trap and decay strategy for noble gas emissions and carbon adsorption for iodine emissions.

The new <sup>99</sup>Mo facility will increase capacity from four to five runs per week using twelve plate irradiations each run. This improved capacity will result in an increase from 1100 to 2250 six-day Ci <sup>99</sup>Mo per week; see Figure 19. Currently, the project has received regulatory approval for changes to process; the existing <sup>99</sup>Mo facility upgrade is complete; and the transition from the existing facility to the new ANSTO Nuclear Medicine (ANM) facility has begun. Full transition to ANM is expected by the second half of 2017. In addition, the Synroc Waste Plant is expected to be fully operational in 2019.

### **ANSTO/ANM** Capacity

#### 3500 3000 2250 2250 2000 1250 1250 Q1 Q1 Q2 Q3 Q4 Q2 Q3 2016 2017

Projection of 6 day Ci Production advised to OECD

Figure 19. Expected increase in <sup>99</sup>Mo capacity during transition to the new ANM facility

The xenon and iodine abatement systems for the new ANM facility were designed to be below the 2012 limits and ~ 5 times lower than current release levels for xenon and iodine. To achieve this design criterion, the system incorporates a low-input airflow of 60L/hour, 82 delay tanks, and 60 carbon columns that incorporate a double baffle design. An expected 2-week residence time on carbon columns and a total system delay of 7 weeks and fewer fugitive emissions should smooth out the xenon emission profile, which is expected to be mainly <sup>133</sup>Xe with little or no <sup>135</sup>Xe (9.2 hr T1/2), <sup>135m</sup>Xe (15 min.), or <sup>133m</sup>Xe (2.2 d)—some <sup>131m</sup>Xe is possible (11.9 d).

ANSTO has been involved in several WOSMIP activities, including participation in the EU Radioxenon Mitigation Project's trial of silver zeolite xenon abatement system on the existing MIP facility (Christophe Gueibe, SCK•CEN -Damien Moyeaux, IRE); testing of a compact stack monitoring system being developed by German Radiation Protection Agency and Hamburg University; and participation in the stack data upload experiment.

#### 4.4 IRE Update

IRE gave a presentation that informed on the ongoing conversion of their <sup>99</sup>Mo chemistry from HEU to LEU, which began in 2012. They are currently proceeding with qualification of the new LEU target in European reactors, testing a new transport container, and conducting processing modifications; see Figure 20.



Figure 20. Process modifications for the conversion to LEU

In conjunction with the conversion to LEU, a new xenon management system is being implemented. This new system incorporates xenon trapping improvements at each critical step, a new method of storage using 56 decay tanks, and circuit redundancy.

Cold commissioning of the new LEU processing is ongoing with a progressive increase in batch size from 3-15 targets/run. This processing will be used to develop new Standard Operating Procedures and perform operator training. Cold commissioning issues have delayed the project by about 9 months but more than 200 LEU targets have been processed in the LEU production line (more than 20 runs) and approximately 100 tests have been performed (R&D and Production). These tests conform to expectations for <sup>99</sup>Mo and <sup>131</sup>I yield, process time, product quality (metallic impurities), vacuum consumption of the decay tanks, etc.

Processing authorization has begun, and hot commissioning is planned to commence in early 2017. Once the hot commissioning is complete, validation and approvals are planned for 2017 followed by HEU phase-out in 2018 for a progressive transition to full LEU capacity.

#### 4.5 Mo-99 Production and Effective Control of Xe-133 Emissions

Mallinckrodt began their presentation by announcing that they have entered into an agreement to sell the nuclear medicine business to IBA Molecular. This sale was still subject to a number of closing conditions, including approval from the U.S. Nuclear Regulatory Commission and the U.S. Committee on Foreign Investment. The sale will include transfer of Mallinckrodt's radiopharmaceutical plants in Maryland Heights, Missouri and Petten, Netherlands. This transfer of the business to IBA Molecular is expected to close in the first half of 2017.

Mallinckrodt provides bulk <sup>99</sup>Mo to key strategic markets of which 80% is produced at their facility in the Netherlands. This facility has been producing <sup>99</sup>Mo since the 1990s and was engineered with a system using vacuum storage tanks and banks of charcoal filters for decay to effectively minimize <sup>133</sup>Xe releases.

Mallinckrodt currently produces about 182,000 Ci <sup>99</sup>Mo per year (6-day) and will have the ability to produce as many as 260,000 Ci per year in 2017. Production has increased from four to five days per week and will go to six in 2017. Although the number of <sup>99</sup>Mo production runs has increased, there has

not been a corresponding increase in <sup>133</sup>Xe emissions from the site. Emission data is tracked daily and is well below any regulatory criteria. Although levels of <sup>99</sup>Mo production vary by day and week, <sup>133</sup>Xe emission rate is fairly uniform; see Figure 21.

# April 1, 2015 - March 1, 2015 - April 1, 2015 - April 1, 2015 - April 1, 2015 - August 1, 2015 - July 1, 2015 - July 1, 2015 - July 1, 2015 - August 1, 2016 - August 1, 2016 - June 1, 20

Figure 21. Xenon-133 daily releases from the Mallinckrodt facility are consistent

Mallinckrodt supports the principles of the CTBTO PrepCom and the tracking of <sup>133</sup>Xe emissions and will continue to closely control and monitor <sup>133</sup>Xe emissions from <sup>99</sup>Mo production operation. Mallinckrodt has provided specific <sup>133</sup>Xe release data to WOSMIP when requested and will continue to respond to individual CTBTO PrepCom requests for specific release data related to time windows of interest.

#### 4.6 Nordion Update

Nordion is owned by Streigenics but functions as a standalone company consisting of two business units: Gamma Technologies using <sup>60</sup>Co in sterilization, radiotherapy, and food irradiation/disinfestation; and MIP of a variety of radionuclides from cyclotrons, reactor-based fission, and n-gamma.

Nordian has historically received raw <sup>99</sup>Mo (from HEU) as an acidic solution from the NRU facility. Nordion's portion of radioiodine and radioxenon releases are being studied to pinpoint release points in a mitigation and initial results indicate that Nordian only releases 1% of the amount released by NRU who irradiates and dissolves the targets. Nordion freely shares release data with CTBTO PrepCom community upon request, and annual releases are posted in the Nordion website; see Figure 22.

Year	Xe-133 (GBq/yr)	Xe-135 (GBq/yr)	Xe-135m (GBq/yr)		
2011	34,967	17,239	27,688		
2012	36,153	23,943	39,498		
2013	30,735	28,193	43,383		
2014	15,018	13,075	18,170		
2015	11,916	8,237	10,758		

Figure 22. Release data posted on Nordion website as part of public information program

Routine production at NRU will stop November 2016 and the facility will be on standby until March 2018. To replace the supply of <sup>99</sup>Mo provided by NRU, a project with General Atomics, the University of Missouri Research Reactor (MURR), and Nordion was announced in February 2015 to develop a new Selective Gaseous Extraction (SGE) Process for <sup>99</sup>Mo production. This LEU process uses raw <sup>99</sup>Mo from irradiation at MURR and will undergo final processing at Nordion. This new process is expected to remove 95% –99.9% of radioiodines during the SGE process at MURR. If raw <sup>99</sup>Mo arrives at Nordion with 5% residual radioiodine, then mitigation will be required to maintain <sup>133</sup>Xe releases below 5 GBq/day. In addition, Nordion believes that the slow flow rates and small volumes of SGE will make xenon capture easier than traditional production. Nordion expressed interest in collaboration with members of the WOSMIP community for a potential mitigation project.

#### 4.7 Production of Radioisotopes in Pakistan

The Pakistan Institute of Nuclear Science and Technology (PINSTECH) presented information on its radioisotope production activities and their application to medical treatments. They use the Pakistan Research Reactor-1 (PARR-1), a 10,000 kW pool reactor, for research, isotope production (<sup>99</sup>Mo, <sup>131</sup>I, <sup>32</sup>P, <sup>177</sup>Lu, <sup>82</sup>Br, etc.), neutron scattering, neutron radiography, nuclear chemistry, and training. A second 30,000 kW Miniature Neutron Source Reactor, PARR-2, is also used for Neutron Activation Analyses (NAA) and radioisotope research and production of short-lived isotopes.

The main goal of the Isotope Production Division is to maintain an uninterrupted supply of high-quality radioisotopes/radiopharmaceuticals and freeze-dried kits for <sup>99m</sup>Tc-radiopharmaceuticals to their users. Products of the isotope production division include <sup>99</sup>Mo, <sup>99m</sup>Tc generators, <sup>131</sup>I, <sup>32</sup>P, <sup>177</sup>Lu, <sup>198</sup>Au, and <sup>153</sup>Sm, which are supplied to various private and government hospitals.

For <sup>99</sup>Mo production, HEU targets are irradiated at PARR-1 and transferred to the <sup>99</sup>Mo facility where alkaline dissolution, filtering, iodine removal, and acidification occur followed by separation and purification of the <sup>99</sup>Mo product; see Figure 23. Eight cylinders are used to trap and decay xenon captured from dissolution that is released through the facility stack. After purification, the product is transported to the <sup>99</sup>mTc Generator Production Facility where approximatly1500 generators are prepared each year.



Figure 23. Separation technique for <sup>99</sup>Mo at PINSTECH

#### 5.0 R&D Efforts on Emission Reduction: Recent Advances for the Reduction of Radioxenon Emissions

This session addressed recent advances achieved in xenon abatement technologies.

#### 5.1 Radioxenon Abatement Technology Overview

INVAP presented information on the use of chromatographic adsorption on activated carbon and in combination with decay tanks for the design of noble gas abatement systems used in fission MIP plants. For this study, a reference production facility producing 3,500 six-day Ci <sup>99</sup>Mo per week was modeled to understand the carbon adsorption process and to perform a cost analysis.

Xenon and iodine originate from irradiation and decay products and can be released as off-gas from several different processing steps including dissolution and filtration. The off-gas has the potential to leak from the closed processing into the hot cell during controlled venting and connection-disconnection operations. Methods commonly used for xenon emission reduction include adsorption on activated carbon, zeolites, and molecular sieves; containment in holding or buffer tanks; and cryotrapping. The method of chromatographic adsorption by activated carbon was chosen for this study due to the reasonable cost and effectiveness. Many factors affect the adsorption capacity of activated carbon, including the type of carbon; the flow rate of gas carrier (air, N<sub>2</sub> and He); the presence of other impurities such as H<sub>2</sub>O, CO<sub>2</sub>, NO<sub>x</sub>, N<sub>2</sub>, I<sub>2</sub>, etc.; humidity; temperature; and system operating total pressure (Langmuir isotherm).

The radioxenon inventory for the modeled facility was calculated to be 250 GBq/day/1 six-day Ci <sup>133</sup>Xe and 210 GBq/day/1 six-day Ci <sup>135</sup>Xe. An additional 20 GBq/day/1 six-day Ci <sup>133</sup>Xe would be expected from iodine decay. Leaks of <sup>133</sup>Xe into the hot cells were estimated to be 1 GBq/day/1 six-day Ci. While this amount is small, it is diluted into the large flow rate of the hot cell ventilation off-gas, making it difficult to capture for abatement.

Two systems were considered for this study: a passive and non-passive system. The requirement of low temperature and low humidity demands the use of a non-passive system (cooling equipment, compressor, pump, etc.). Typically, high airflow rate is a key design parameter for noble gas delay in carbon columns, which mandates large carbon volumes; this study introduced the concept of recirculation of hot cell off-gas to allow for a reduction in the airflow rate through carbon beds and to achieve the same performance as with large carbon volumes and higher flow; see Figure 24.



Figure 24. Hot cell ventilation off-gas recirculating concept

A cost analysis for an abatement system that would meet the design objective of releasing less than 5 GBq/day <sup>133</sup>Xe was conducted to compare the cost of systems designed to capture different amounts of xenon and using different temperatures for carbon columns; see Figure 25. The system modeled for the cost analysis uses a combination of holding tanks and activated carbon columns. For this modeled facility, these tanks are used in conjunction with the process off-gas (recirculating) system and not the hot cell ventilation system (due to the high volume and flow rate). This model incorporated three holding tanks, which would be used one per day (one in use, one venting and one in standby) and be vented through a carbon column. The hot cell ventilation off-gas for the modeled facility uses a very high-volume carbon column abatement system which has three lines with 50% capacity each (one in standby).

Leaks to hot cell ventilation off-gas [GBq/day]	Carbon Temperature [°C]	Carbon Volume [m³]	Relative Cost [arbitrary units]	
4000 (1% leaks)	20	123	100	
2000 (0.5% leaks)	20	113	94	
400 (0.1% leaks)	20	89	76	
4000 (1% leaks)	0	72	60	
2000 (0.5% leaks)	0	66	53	
400 (0.1% leaks)	0	52	40	

Figure 25. Cost analysis of abatement system options

The main cost driver for this system was found to be the activated carbon columns. This cost is dependent on the hot cell ventilation off-gas airflow rate (high sensitivity); chromatographic efficiency of carbon column (high sensitivity); radioxenon and radioiodine activity inventory leaks to hot cell; and ventilation off-gas (medium sensitivity). General considerations for system design are that over 50% of the cost is related to carbon columns; therefore, reduction of the hot cell ventilation off-gas flow rate and cooling to 0° Celsius—which reduces the quantity of carbon required to 42% compared to ambient temperature has dramatic impact on cost. It was noted that while reduction of the carbon columns' working temperature has a dramatic impact on cost, it adds complications related to the non-passive cooling system.

#### 5.2 Past, Present, and Future Progress in Radioxenon Mitigation

SCK•CEN presented their design for a mobile system for the reduction of radioxenon emissions from radiopharmaceutical facilities. Reduction of noble gas emissions from large civilian radioxenon sources is a key issue in increasing the sensitivity of the IMS and would significantly enhance CTBT verification capability. In this framework, SCK•CEN was contracted under the EU Council Decision V to design a mobile system for the reduction of radioxenon emissions from radiopharmaceutical production facilities.

The EU Council Decision V, which ended December 2015, investigated material and design study for optimized mitigation of radioxenon at radiopharmaceutical facilities. The project was subdivided in three phases: i) investigation and selection of xenon adsorption materials, ii) study of operational conditions and trap design, and iii) construction and testing of a mobile trap. The outcome of this project was the construction of a compact mobile prototype system, which uses silver zeolite, for the reduction of xenon emissions for testing at IRE; see Figure 26. The EU Council Decision VI (June 2016–December 2017) builds on the previous project by 1) analyzing the scale-up and the long-term behavior of the prototype at IRE, 2) performing design studies at up to three additional facilities, and 3) investigating further the stack releases and stack monitoring at IRE. In addition to this project, SCK•CEN has been participating in the IAEA CRP, which will provide the community with a roadmap to guide the international community of

medical radioisotope producers (established and newcomers) on ways to address and reduce the emission of radioactive gases. SCK•CEN will investigate new adsorbent materials and participate in sampling campaigns near IRE and study regional atmospheric dispersion and transport using concentration levels of xenon measured in the air at the German noble gas network.



Figure 26. Prototype of mobile compact radioxenon trapping system

It was noted that a comprehensive radioxenon mitigation system requires consideration of all major radioxenon release pathways and would need to work on different gaseous effluents. A global design approach is required to consider the entire radioxenon release of a facility. Existing and planned facilities have different requirements—installation of a system in a planned facility is straightforward, while retroengineering an existing facility is more difficult. A global design approach should be applicable to a large variety of facilities; it should thus consider differences in production processes, waste management, and constraints of each facility.

#### 5.3 Feasibility Study of the Xenon Retention with the Ag-ETS10 Zeolite Prototype System Developed by SCK•CEN

A presentation by IRE discussed integration of the prototype compact radioxenon trap designed by SCK•CEN in the new LEU production line of the IRE MIP facility (dissolution circuit). This work was conducted as part of the EU Council Decision VI Xenon Mitigation project discussed in the previous presentation by SCK•CEN. Task I of the project will be to scale up the prototype system and evaluate long-term behavior of the system. To accomplish this, the system will first be integrated into the IRE production setup and preliminary validation will be performed. The next step will be a full prototype validation of critical parameters such as column reliability and reproducibility. Finally, the prototype system will be assessed under real operating conditions in the LEU process. Current results of the prototype integration and preliminary validation that has been completed includes a direct purge of <sup>133</sup>Xe vial for establishment of a breakthrough curve and connection of system to the dissolution circuit.

To determine the breakthrough curve for the system, a known amount of radioxenon was introduced to the system and the xenon container was purged at a constant flow rate. The same column was used for each test and is regenerated by thermal desorption operation at the end of each adsorption test. For Tests 1 and 2, a low activity of <sup>133</sup>Xe was introduced followed by 4 weeks of purge. No signal of <sup>133</sup>Xe release was detected. For Tests 3 and 4, the activity was increased and breakthrough curves were established; see Figure 27.



Figure 27. Example of elution profile for breakthrough Test 4

The measured breakthrough curves were compared with a theoretical adsorption model. The adsorption capacity for Test 3 was higher than expected. To correct this, the input parameters were optimized with higher adsorption capacity and diffusion coefficient values for Test 4. With the new optimized input parameters, good agreement was achieved with the theoretical values and the Test 4 results. The breakthrough investigation is continuing and will be further investigated.

Further experiments will be conducted for full validation of the system, including the effect of different parameters (e.g., xenon concentration and activity with associated dose rate and column reuse) on the xenon elution profile; impact of other gases on the breakthrough curve; follow-up of the zeolite behavior during the scaling up of the LEU process; and tests with irradiated targets of increasing activity.

#### 6.0 Future Mo-99 Production: Progress on Development of New Isotope Production Facilities

The future of <sup>99</sup>Mo production session contained updates from companies in the process of developing new isotope production facilities.

## 6.1 Radioisotope Production in Jordan Research and Training Reactor (JRTR)

The Jordan Research and Training Reactor (JRTR) gave a presentation on their planned radioisotope production activities. With the demand for radioisotopes in Jordan steadily increasing, stable supply is very important, especially for the medical uses of short half-life radioisotopes. Therefore, production of many medical isotopes is planned, including <sup>131</sup>I, <sup>192</sup>Ir, and <sup>99</sup>Mo.

JRTR is Jordan's first nuclear reactor and the focal point for the Jordan National Nuclear Center; see Figure 28. The JRTR project includes a reactor, radioisotope production facility, education and training building, radioactive waste facility, and cold neutron facility. JRTR is a 5 MW open pool reactor that is upgradable to 10 MW with a maximum Flux of  $1.45 \times 10^{14}$ .



Figure 28. The Jordan Research and Training Reactor (JRTR)

The Radioisotope Production Facility (RIPF) contains three banks and ten hot cells and has capacity for the <sup>131</sup>I capsules and solutions, <sup>192</sup>Ir source assembly and <sup>99</sup>Mo in addition to other isotopes, including <sup>197</sup>Hg, <sup>32</sup>P, <sup>198</sup>Au, <sup>60</sup>Co, and <sup>24</sup>Na. The hot cell laboratories are installed at Floor 0 of the RIPF, and irradiated target capsules can be transferred from the reactor building to RIPF by using manually driven carts.

The option of producing <sup>99</sup>Mo through fission of <sup>235</sup>U was rejected at JRTR due to IAEA safeguard expected determinations in the future and the availability of <sup>235</sup>U targets. Instead, <sup>99</sup>Mo will be produced by neutron activation of MoO<sub>3</sub> target material (<sup>98</sup>Mo(n,<sup>y</sup>) <sup>99</sup>Mo). The MoO<sub>3</sub> targets will be >99.5% purity and 90–120g per batch. The <sup>99</sup>Mo produced will be dispensed in 10–15 mL vials as a sodium molybdate solution.

The RIPF is designed to handle 4,000 Ci <sup>192</sup>Ir/batch (one batch per two weeks), 40 Ci <sup>131</sup>I/batch (one batch per week), and 20 Ci <sup>99</sup>Mo/<sup>99m</sup>Tc/batch (one batch per week). This production will help meet Jordan's annual consumption of radioisotopes.

#### 6.2 PT INUKI Update/ New Indonesian Medical Isotope Production Plant

It was announced that PT INUKI will be under new management but plans to continue working with the WOSMIP community. INUKI has completed revitalizing their facility and will start production 6 January 2017. The LEU target is irradiated and cooled for six hours. Following irradiation, the <sup>99</sup>Mo processing at INUKI will begin by submerging the dissolution target capsule in liquid nitrogen. The dissolved capsule is heated in a rotator, and iodine is collected via a cold trap. Next, the gas phase is removed from the target assembly. The cocktail solution is drained and the precipitate is collected for further purification of the <sup>99</sup>Mo product.

A new Indonesian company is planning to build a MIP plant based on a homogeneous reactor built specifically for isotope production and fueled using uranium and thorium; see Figure 29. This new technology will extract isotopes from the fuel solution. Advantages of this homogenous reactor include a simple design, high-production capacity with small reactor power and the low cost of construction. This new facility is planned to produce 3000 Ci/wk <sup>99</sup>Mo and is expected to be operational in 2018 and will create a world class facility.



Figure 29. Schematic of medical isotope production plant planned for Indonesia

#### 7.0 Stack Emission Data: Understanding the Global Radioxenon Backgrounds from Industry

This session discussed the use of MIP facility stack emission data to better understand global radioxenon backgrounds from industry. The session introduced new ideas, issues, and solutions related to implementation of stack data sharing and analysis.

#### 7.1 Stack Release Data Confidentiality

The CTBTO PrepCom discussed issues related to maintaining data confidentiality. State signatories require open and transparent sharing of data from the CTBTO PrepCom, and the data providers want to maintain nuclear facility confidentially. When data is received from the IMS network, it is stored at the IDC and the stored data may be retrieved using several access procedures by different users for treaty verification (authorized users), civil applications, and scientific use. There are currently 1769 authorized users and other users can request data under contract.

The virtual Data Exploitation Centre (vDEC) is a technical support platform supported by voluntary contributions from the EU. The vDEC provides scientists access to IMS data to conduct research and publish new findings. Access to vDEC can be established through a cost-free contract that does not contain any monetary remuneration between the parties for services or property but does contain legal requirements. The online registration form can be found at <u>http://www.ctbto.org/specials/vdec/</u>. The main provisions of a vDEC contract, as defined by the CTBT/PC-13/1/Annex II policy, are as follows:

- Each contractor will access only those IMS data and IDC products necessary for fulfilling its task.
- The IMS data and IDC products will be used only for research associated with the development of the IMS and IDC or for the purposes stated in the contracts.
- Access provided to an organization for the purposes of fulfilling a contract will terminate when the contract is completed (usually after 2 years).
- Data access is provided only to a maximum of three individuals per contract.
- The vDEC access credentials and the retrieved data may not be shared with others.
- The restrictions placed on all users will not exclude the presentation of data or products (or information derived therefrom) for peer review at scientific meetings or in scientific journals and other scientific publications. The inclusion of IMS data and IDC products in scientific journals and other scientific publications will be limited to those required to reflect the scientific achievements. This is subject to approval by CTBTO PrepCom.

In addition to the cost-free contract, each contractor has to sign a separate confidentiality agreement. The vDEC contract provides a provision for "Other Data," which includes data that is neither IMS data nor IDC products but which have been acquired by the Commission from other sources. This provision was used for the 2016 ATM Challenge in which participants used the vDEC platform to access data for the challenge.

The CTBT lists some general provisions related to confidentiality in Article II. These provisions are listed below:

- Article II, Paragraph 6: "[The Organization] shall take every precaution to protect the confidentiality of information on civil and military activities and facilities coming to its knowledge in the implementation of this Treaty..."
- Article II, Paragraph 7: "Each State Party shall treat as confidential and afford special handling to information and data that it receives in confidence from the Organization in connection with the implementation of this Treaty. It shall treat such information and data exclusively in connection with its rights and obligations under this Treaty."

#### And in Article IV:

• Article IV, Paragraph 8: "...all necessary measures shall be taken to protect the confidentiality of any information related to civil and military activities and facilities obtained during verification activities."

Transparency is a basic requirement for data access. State Signatories must have access to all data for reconstruction of IDC event analyses; however, that data must only be used for Treaty-relevant purposes. However, there is precedence for non-IMS data being shared; data has been used to investigate mining explosions and World Meteorological Organization Regional Specialized Meteorological Centres results on atmospheric transport modeling.

It was stated that any stack data sharing would be voluntary and information of interest would include the location of a MIP facility, the facility operational status (on/off), absolute release data (time resolution, relevant isotopes, uncertainties, MDAs, and other relevant information on measurement technology and analysis), analysis reports on spectral data, and raw stack detector data. The level of data disclosure would be determined by the data provider; see Figure 30. While qualitative data is helpful, detailed information would be ideal and may include raw data (spectral pulse height data), release values (isotopic activities or concentrations), resulting concentration predictions at IMS stations broken down by source, and resulting overall predicted background at IMS stations.

Draft access restriction matrix						
	Security through access restrictions for classes of users (1-5) and levels of information disclosure (a-d)	Raw data (spectral pulse height data)	Release values (isotopic activities)	Resulting concentration predictions at IMS stations broken down by source	Resulting overall predicted background at IMS stations	
		a)	q	<b>;</b>	ਰ	
1.	Relevant CTBTO staff					
2.	Briefing to delegations of State Signatories	n/a				
3.	CTBTO contractors through vDEC with confidentiality clause in their contract		AS ; b	requi	red	
4.	Authorized users as designated by the State Signatories by email (through SWP once operational) under confidentiality rules like for IMS data	L	Providers			
5.	Scientists (including PNNL) for approved purpose through vDEC under the confidentiality rules of the zero-cost contract					
6.	Publicly open (no restrictions)					

Figure 30. Data to be shared with the CTBTO PrepCom is determined by the provider

Security for ATM Challenge occurred before the dedicated vDEC platform was available and data sharing security was assured in an ad hoc basis. Moving forward, a virtual machine will be installed for a second vDEC platform dedicated to the stack release data experiments. Its operating system foreseen security features are a Linux account with username/password security; access over Secure Shell with IP-based whitelisting; file storage segregation based on Linux user and group rights; dedicated vDEC database security measures; and access restriction on a per-data-provider basis without additional licensing cost.

#### 7.2 Data Upload IDC Software Requirements

The CTBTO PrepCom outlined an overview of projects to better understand the global radioxenon background and IDC software requirements related to data collection. The CTBTO PrepCom conducts monitoring of air to detect any traces of radioxenon isotopes from any potential nuclear tests. This monitoring detects radioxenon emitted from clandestine underground nuclear explosions and in some cases civil nuclear facilities, such as NPPs, MIP, etc. Emissions from civil nuclear facilities are creating a radioxenon background at the IMS detection systems that affect the capability of the noble gas network to detect nuclear explosions—these emissions make discrimination of CTBT related events vs. background very difficult. Two approaches to estimate and reduce the impact of the radioxenon background at IMS stations include xenon background measurements and support for voluntary xenon mitigation.

In 2012, an investigation of radioxenon background in Jakarta, Indonesia found that all four treatyrelevant radioxenon isotopes were detected. These detections were postulated to occur due to the proximity of the sampling system to the local MIP facility (INUKI), which was ~20.3 km away. The consequences of this proximity are an increase in the minimum detectable concentration for radioxenon at stations located near MIP facilities, false categorization of events, and difficulty in discrimination between background and CTBT-related events. This particular case shows the importance of reducing the radioxenon releases from civil nuclear facilities.

A LaBr detector system is installed at the Vienna International Centre. The system consists of a .5'x1.5' LaBr detector assembly fixed on rails inside a Snow White air sampler hood. The system provides continuous measurement of the collection filter, saving data every 10 minutes. About two thousand files have been collected and processed.

A project collaboration among BATAN, PNNL, and the CTBTO PrepCom installed an online LaBr<sub>3</sub> scintillation detector for detection of radioxenon releases from the INUKI facility (formally BATAN). This work was funded by the EU and Contributions in Kind (CiK) from the United States. This system collected approximately 55,000 raw data files between March 2013 and March 2014. The data collected from the INUKI facility has been used to test methods to send, process, store, and analyze data; see Figure 31. The files converter step was successfully completed and can read several input formats; distinguish between raw and processed data; standardize data to UTC time/units of measurement values; take into account non-operational times; and automatically send data in the dedicated pipeline. Data analysis of the converted data was performed using Saint2/iNSPIRE to integrate an automatic recalibration process for each spectrum (e.g., based on internal lines/ 40K /annihilation peaks).



Figure 31. Testing phase for handling of stack data received from the INUKI facility (sending, testing, processing, storage, and analysis)

Shared stack data will be used by the IDC for scientific studies to help understand the impact of stack data on the IMS noble gas network, improve the IMS noble gas network performance, and improve discrimination between background and CTBT-relevant events. The data format for shared data is undetermined but may include raw, processed, or other types of data. Collected data may be shared through the vDEC platform or other platforms that are to be determined.

It was concluded that the IDC has the capacity and resources to collect, process, and analyze shared data and experience with data storage in a secured environment and from LaBr detector operations. Developments to optimize their standard database and software are on track.

#### 7.3 Stack Data Upload Experiment

Sharing radioxenon stack release data with the CTBTO PrepCom has been discussed as a tool to be used to discriminate between radioxenon released from MIP and nuclear explosions. At WOSMIP V in 2015, a small working group was formed to conduct an experiment to develop and demonstrate methods for confidential transfer of stack release data from MIP facilities to a central data repository. The objective of this experiment was to create a data repository, transfer example data, and produce software and database compatible with IMS architecture; see Figure 32.



Figure 32. Outline of data upload experiment

The experimental working group comprises three teams: the producer team—ANSTO, IRE, INUKI—who agreed to share example data; the data team—CTBTO PrepCom and PNNL; and the software team, which includes members from the producer and data teams.

Progress on the experiment in 2016 began with a draft document from CTBTO PrepCom outlining the stack release data software requirements specification described earlier. PNNL established a data repository based on cloud computing, Cloudberry Explorer, which is a scalable platform (easily integrated with Amazon Web Services computing, database, and other cloud services as the demand grows). Example data was uploaded to the repository by INUKI, ANSTO, and IRE and includes raw spectra, processed data, and concentration and air volume data.

Moving forward, the received data will be used to determine data format specifications based on available data from the producers, develop methods for the conversion of diverse data to a standard format, and determine the most efficient means to transfer data between organizations.

#### 7.4 STAX – New Concept for Using Stack Data for the CTBT

The concept for a Source Term Analysis of Xenon (STAX) project was introduced by PNNL to better understand emissions from MIP facilities and their impact on the IMS. The goal of this project would be to collect radioxenon stack release data from MIP facilities (shared voluntarily), transfer the data to a database that is accessible by the IDC for use in determination of whether detections at IMS stations are consistent with releases from MIP or other sources, and convert the data into a usable format. This project would develop methods, tools, and technology for the use of MIP radioxenon stack release data for CTBT verification. Technical solutions required for the project include data collection technology, processes for data transfer from the facility to the IDC, data security, and methods for stack data use by the IDC and NDCs. Implementation of this project would create a system linking the IDC (and NDCs upon request) to shared stack release data from participating producers; see Figure 33.



Figure 33. Stack release data sharing as envisioned for the STAX project

Data requirements for the STAX project would be determined through discussions with IDC and various NDCs and will include specifications for types of data needed (e.g., gamma spectra, airflow rates, detector State of Health, and quality assurance/quality control calibration data). Some general suggested requirements are that the four treaty-relevant radioxenon isotopes are able to be detected with a 10% uncertainty in the presence of inferences; the detector should have a large dynamic range to accommodate fluctuations in radioxenon emissions from the facility (typically in the range of 1GBq-10 TBq); and detection of non-xenon CTBT-relevant radionuclides may be of interest. It was noted that there will need to be options in how and what data is collected because a "one size fits all" solution may not work for all facilities.

Detector technology will need to meet the data requirements that will be established by the project. Current detectors at INUKI, ANSTO, and INVAP will be compared along with other commercially available systems to determine which meet the project needs. When the STAX project is implemented, an agreed framework for stack data with standardized technology for data collection and processing will be needed. In addition, implementation of data confidentially methods will be a priority.

# 7.5 Ideas on the Implementation of the Stack Monitoring Network and Data Analysis

Instrumental Software Technologies Inc. (ISTI) gave a presentation introducing ideas on software that would support stack data sharing with the IDC. ISTI provides expertise in numerous technologies for monitoring nuclear tests. A comparison was made that highlighted similarities between the IMS and a stack detector network with 18 or more detectors located at MIP facilities (compared to 80 IMS radioxenon stations). Both networks would require a unified standard for reporting and analyzing data. These similarities make adaption of the existing Radionuclide Station Software Interface (RSSI) an ideal platform for a stack monitoring network.

The RSSI platform is an open source cross-platform that can control acquisition of up to two spectrometers; allows for auxiliary data collection (power, VPN status, room temperature, and humidity); can store and back up data; authenticate data; has VPN communication; provides web-interface for remote connection, configuration, monitoring, and control; has a dashboard overview of the system status, State-of-Health, alerts, and data visualization; supports typical station operations (detector background, detector calibration, problem reporting); and has automatic recovery after power failure. Some advantages of RSSI over other platforms are that the RSSI platform is independent and is compatible with Windows and Linux; it is easily maintainable; it is compliant with PTS coding standards; it supports both Canberra and Ortec spectrometers; it supports the same date authentication infrastructure as PTS/IMS network; and it is fully developed.

To adapt RSSI for stack data, several modifications to the software will be required, including adapting WebUI; adapting the components to stack monitoring specifics; introducing new components for stack-monitoring as needed; performing configuration management; integrating with analysis tools; and introducing Message Queuing communications (more reliable than email).

Stack data analysis requirements are both qualitative and quantitative in time and magnitude. For this data to be useful, realistic ATM uncertainties, minimal uncertainties in stack emissions, high timing resolution, low concentrations uncertainties, and reliable isotopic ratios are required.

## 7.6 A Compact Set-up for Measuring Radioxenon Concentrations in the Stack Releases of Nuclear Facilities

The Carl Friedrich von Weizsäcker Center for Science and Peace Research (ZNF) conducted a project to study technologies to monitor emissions of isotope production facilities in the close-up to long-distance range. The project was conducted under the IAEA CRP-F23031 to study possible implications to verification systems of nonproliferation treaties. The proposed research objective was to monitor releases from MIP facilities at or near a facility using room temperature detectors and to evaluate data taken at Radionuclide Station 33 located at Schauinsland, Germany, for the detection of possible releases from nuclear facilities, and to identify the impact of released activities on the detection capability of monitoring stations.

A cadmium zinc telluride detector (CZT) semiconductor detector was chosen for this project. Advantageous properties of this detector are the ability to operate at room temperature, a low-temperature susceptibility, medium energy resolution, high density, and high effective atomic number. However, the crystal size and sensitivity are limited. Other aspects that make this detector ideal for this application are the detectors integrated electronics, very compact size, and low energy usage.

The first campaign in support of the project was conducted at IRE to monitor gaseous emissions released from the facility stack. A compact system incorporating a cadmium zinc telluride detector and Raspberry Pi was developed to monitor the stack effluent at a point collocated with the facility high-purity germanium (HPGe) regulatory monitoring system; see Figure 34. Measurements were collected in 10-min intervals for about 3 weeks. The experimental CZT system was found to be simple, mobile, and inexpensive. While capable of detecting <sup>133</sup>Xe and <sup>135</sup>Xe, detection of <sup>131m</sup>Xe and <sup>133m</sup>Xe need further development to be useful in source characterization.



Figure 34. A compact system for measuring radioxenon concentrations in the stack release of nuclear facilities

#### 7.7 Stack Air Effluent Monitor: Present and Future

INVAP presented the design status of its Air Effluent Monitor (AEM) equipment for nuclear facility stacks. This system has been developed for radioisotope plants that the company is designing and constructing for various clients. INVAP demonstrated experiments and simulations with NaI and CdTe detectors and introduced the next-generation of AEM.

The AEM for nuclear facilities has three different measuring channels: iodine, aerosol, and noble gas. The noble gas channel is the most relevant from the point of view of CTBTO PrepCom due to its contribution

to accurate determination of radioxenon emissions at the source. The current system measures aerosols and iodine through continuous sampling followed by measurements conducted in the laboratory while noble gases are measured in real time using a CdTe detector, NaI detector, and on-line spectrometry. The system comprises two units—a control and processing unit and the sampling and measurement unit; see Figure 35. The sampling unit has noble gases chambers optimized for the activity range with modular shielding for each detector (versatile equipment to be used in MIP or research reactors). Two detectors, a NaI scintillator and a CdTe, are used for detection of <sup>133</sup>Xe and <sup>135</sup>Xe in the gaseous effluent. This dual detector arrangement allows the system to take advantage of the low geometrical efficiency of the CdTe for high concentrations and the high efficiency of the NaI for low concentrations. In addition, aerosol filter and iodine cartridge holders for this unit have two parallel sampling lines for continuous sample collection—the removable cartridges are glass fiber for aerosol and charcoal for iodine. After removal from the system, high-resolution gamma spectroscopy is used to analyze the cartridges in the laboratory.



#### Stack Air Effluent Monitor: present status

Figure 35. The air effluent monitor consists of a control and processing unit and a sampling and measuring unit

The current software for the AEM system has evolved since 2003. The most recent version stores historical spectra for future analysis in a relational database. Eluted peaks are analyzed in time with the number of counts for each peak calculated with background subtraction. The system communicates with the plant's control and monitoring system and provides real-time data on an operator's screen at the front of the hot cell.

The aim of this new design is to obtain better energy resolution for improved accuracy in the facility activity reports. In addition, this system will serve as an introductory design for possible requirements of on-line measuring of iodine and noble gases. For these applications, the use of HPGe and LaBr<sub>3</sub> detectors are also being analyzed.

## 7.8 Evolution of HPGe Detectors: When Performance and Reliability Count

Canberra Industries presented information on their technology related to HPGe detectors including analysis of performance and reliability of their current cooling systems and the Cryo-Pulse 5 detector; see Figure 36.



Figure 36. Canberra HPGe detectors and cooling systems

The Cryo-CycleTM cryostat is a cooling system that combines the reliability of  $LN_2$  with the convenience of electricity. This cryostat is compatible with existing  $LN_2$  detectors and has advantages, including low maintenance, low power consumption, a long lifespan, quiet operation, and no compromise on performance specifications.

The Cryo-Pulse 5 is an HPGe detector that features  $LN_2$  free operation by using a sealed compressor-cold head (linear pulse tube technology). This cooling technology uses a high-frequency gas pressure wave principle for cooling that has no moving parts and therefore no mechanical wear. Canberra states that the system has many applications due to the low power consumption, a long life with no maintenance, and is quiet with virtually no vibration.

Some real-world reliability data for the Cryo-Pulse 5 was presented for the Pulse-Tube detectors that have been in service since 2006. More than 1100 Cryo-Pulse 5-PLUS units (Pulse Tube cooler) and 800 Cryo-Cycle II units (Stirling cooler) have been installed since 2006. These units have been operating 24/7 at +/-50% of maximum cooling power. During this time, there have only been two Cryo-Pulse 5-PLUS cooler related failures compared to fifteen Cryo-Cycle II C(-II) cooler related failures. From this data it was concluded that the lifetime for the Cryo-Pulse 5-PLUS is greater than 10 years with 99.9% probability for the Cryo-Pulse technology compared to approximately 9 years with 85% probability with the Stirling cooler technology. Both technologies show enormous improvement over older, low-cost cooler types.

Two new items were introduced including a new water-cooled system, the Cryo-Pulse 5 PLUS/W, which allows operation of the detector and cryostat up to  $+50^{\circ}$ C ( $120^{\circ}$ F) and an intelligent preamplifier for germanium detectors.

Networking capabilities of the HPGe system were also discussed. Currently the systems can be networked through Ethernet converters, but in the future these capabilities will be expanded to allow a Lynx MCA to be used as an interface to monitor and control the system and allow remote access.

#### 7.9 Evaluation of the Ag-ETS10 Zeolite in the Stack Monitoring System of a MIP

IRE presented preliminary results and analysis of work to evaluate the behavior of zeolite in their stack monitoring system instead of charcoal. This work was conducted in collaboration with SCK•CEN under Task III of the xenon mitigation project. The analysis of the zeolite studied the impact of flow rate on zeolite retention capacities, the impact of other isotopes on retention capacities, and a comparison of charcoal and zeolites for monitoring applications.

To use the industrial monitoring system of IRE to perform this investigation, several factors had to be considered. Because the safety equipment of IRE is not intended to be used for research and development, authorization to make changes was required based on the design of specific testing procedures. In addition, the zeolite cartridges would have to be sized to match the current monitoring system cartridge holder and new efficiency curves for the HPGe detector would have to be calculated.

IRE stack monitoring of xenon is accomplished by differential measurement, while iodine, which is captured on the charcoal cartridges, is determined by integration measurement. Replacing charcoal with zeolite should allow measurement of xenon using integration measurements, but how will the zeolites behave in a high-flux mix of air, moisture, and chemical species?

The current experimental progress was reported in which IRE has completed design of the zeolite cartridges, efficiency calculations for the zeolite cartridge, and some initial testing of the zeolite. Conditions at IRE are ideal for this testing and include a continuous background release, spectra with all xenon isotopes present (<sup>133</sup>Xe, <sup>135</sup>Xe, <sup>133m</sup>Xe, <sup>135m</sup>Xe, etc.), spectra with only <sup>133</sup>Xe during flushing operations and weak <sup>133</sup>Xe releases from the secondary stack. However, there is interference due to moisture and other chemical species released into the stack air. Saturation of the zeolite with water was expected, but how fast this would occur was unknown.

Two stack monitoring systems are installed at IRE, allowing a direct comparison between charcoal and zeolite. Both systems have an I-CAM detector and a self-cooled HPGe detector. The first measurements with two zeolite cartridges were performed at approximately 90% humidity. During this measurement, the cartridge became saturated with water in approximately fifteen minutes; the cartridge weight increased by approximately 7%; and the xenon had been cleared from the cartridge; see Figure 37. A second measurement used a silica gel cartridge inline before the zeolite cartridge in similar conditions—again, the cartridge was quickly saturated.

When used for monitoring conditions, moisture saturates the zeolite very quickly (more than expected). To evaluate the performance of the zeolite under these conditions, a drying system will be required upstream. Further tests will be performed with a high-capacity column charged with standard zeolite or silica gel. If good results are obtained, Ag-ETS10 zeolite may have applications, including accurate daily compatabilisation (reference measurement), weak or high dilution releases (increase of sensitivity), and environmental monitoring of xenon.

## Task achievements



Figure 37. Results from the first measurement incorporating Ag-ETS10 zeolite into the IRE stack monitoring system. The zeolite cartridge became saturated quickly and the retained xenon was cleared.

#### 8.0 Atmospheric Transport: Characterization of the Atmospheric Isotope Background

The session on ATM focused on numerical simulation studies, which used hypothetical values of MIP emissions to analyze the impact on the global radioxenon background and the implications this has for the CTBTO PrepCom's IMS.

#### 8.1 2<sup>nd</sup> ATM Challenge 2016

The Central Institute for Meteorology and Geodynamics (ZAMG) presented an overview of results from the 2<sup>nd</sup> ATM Challenge. One purpose of the 2<sup>nd</sup> challenge was to ascertain the level of agreement one can achieve between real IMS measurements and those simulated using only the stack release data and ATM. In support of this objective, the IDC started collecting stack release data following WOSMIP V. Another purpose was to compare results of the current challenge in terms of model performances with those from the 1<sup>st</sup> ATM Challenge.

For the  $2^{nd}$  ATM Challenge, stack release data from the ANSTO MIP facility was chosen along with several IMS stations. Stations were chosen that are known to be influenced by radioxenon releases from ANSTO (AUX04, AUX09, FRX27 and NZX46), have no influence from ANSTO releases (BRX11), and one (GBX68) that is located 11900 km from ANSTO and has slight influence from ANSTO releases. This distance between the source and the selected IMS station is much larger than for the previous challenge in which the largest distance was 380 km. In addition, the locations of the two challenges (~50°N versus ~34°S) are nearly on opposite sides of the globe and, consequently, are expected to have very different atmospheric circulation patterns.

The challenge included seventeen participants from nine different countries, twelve of which had already participated in the 1<sup>st</sup> ATM Challenge. Each organization contributed up to six models using HYSPLIT (9), FLEXPART (14), MLPD (2), IdX (1), and NAME (1), and metrological data from various sources was incorporated into these models.

The overall statistics for the stations were compiled and it was found that the models underestimated the measured values; the normalized average absolute deviation, which excludes time steps for which simulations and measurements add up to zero, was calculated to be around 200%; the Kolmogorov-Smirnov parameter has limited significance for these models due to the many zero or near-zero values.; the highest ranking run has the best correlation and best Factor of Exceedance; and two runs with moderate rank have the best root mean square error and normalized average absolute deviation.

The results were arranged according to common characteristics and compared with the 1<sup>st</sup> ATM Challenge in 2015. It was found that forward modeling was better than backward (technical reasons may contribute); there is no advantage to using a meteorological field resolution of less than 1.0°; and that daily resolved emission values did not result in any disadvantage compared to higher resolved values. An example time series for AUX04 (Melbourne) with daily source resolution compares measured and simulated data; see Figure 38.



Figure 38. Time series for AUX04 (Melbourne) with daily source resolution

This challenge found that the performance of individual models at individual stations is quite diverse and that no single model-meteorology or model combination (ensemble) performs best for all stations (and daily source time resolution). The overall best run for the current challenge was found using National Centers for Environmental Prediction Global Forecast System (NCEP-GFS) meteorological input and HYSPLIT, a result that is contrary to findings from the 1<sup>st</sup> ATM Challenge, indicating that FLEXPART was superior to HYSPLIT and ECMWF was superior to NCEP. However, results indicating that a coarse resolution of meteorology (1°) and a coarse resolution of the source (1 day) were not detrimental to ATM are supported by this challenge; in addition, the results indicated that forward simulations were superior to backward simulations.

Although the overall statistics are worse than for the 1<sup>st</sup> ATM Challenge, comparable ranks (average and maximum) are reproducible for station FRX27. The station statistics did not depend on the distance between the source and the individual stations—some remote stations had better statistics than close ones (e.g., FRX27 vs. AUX04). Poorer statistics at certain stations could be related to high-pressure systems and thus impaired representation of mixing. The average deviation for simulated values with measurements or simulations above MDC adds up to 200%, including phase shifts of simulations with regard to measurements. Ensemble modeling seems to have had little or no benefit with regard to rank statistics as was the case for the 2015 Challenge. The best individual models score higher in rank.
## 8.2 Analysis Results of 2<sup>nd</sup> ATM Challenge 2016 by JAEA/NDC

The Japan Atomic Energy Agency (JAEA) gave an overview of the 1<sup>st</sup> ATM Challenge and reported on their results from the 2<sup>nd</sup> ATM Challenge.

The task of the 1<sup>st</sup> ATM Challenge was to use stack monitoring data from the IRE radiopharmaceutical plant in Belgium along with ATM to predict the time-history of <sup>133</sup>Xe activity concentrations at the DEX33 IMS station. Participants from seven nations (Austria, Canada, France, Germany, Japan, United Kingdom, and United States) and CTBTO PrepCom took part in the challenge. Results of this challenge showed a strong correlation between the calculated <sup>133</sup>Xe concentrations when the concentrations measured at the DEX33 station were relatively high but a weaker correlation when lower concentrations were measured at DEX33; see Figure 39. It was also determined that the best ensembles for ATM simulations with 2, 3, and 4 members have lower Mean Square Error than the single best analysis result when using the ensemble mean of the individual analysis results. For this study, it was found that FLEXPART simulations were better than HYSPLIT, and ECMWF meteorological data was better than NCEP Global Data Assimilation System from the perspective of correlation strength between calculated and measured concentration on average.



**Figure 39**. Results of the 1<sup>st</sup> ATM Challenge showing a correlation between the calculated <sup>133</sup>Xe concentrations and measured concentrations at the DEX33 IMS station

It was concluded that IRE was a dominant emission source of <sup>133</sup>Xe measured at DEX33 during the period from 10 November to 9 December, 2013; however, a study on other emission <sup>133</sup>Xe sources is needed because it is highly possible that some of the <sup>133</sup>Xe measured at DEX33 originated from sources other than IRE. A combination of multiple models may provide more accurate predictions than use of a single model, although future research is needed to identify optimal methods for selecting ensemble members.

The task of the 2<sup>nd</sup> ATM Challenge was to predict the time-history of <sup>133</sup>Xe activity concentrations at six IMS stations located in the Southern Hemisphere using ATM and stack monitoring data from the ANSTO radiopharmaceutical facility in Australia. Distances between the source (ANSTO) and each IMS station in this challenge ranged from 700 km (AUX04) to 12,000 km (GBX68). There are two portions to this challenge: 1) the "blind phase (phase 1)" in which the amount of each emission (S*i*) is 1 Bq. and 2) the

"open phase (phase 2)" where a comparison review between calculated and measured concentration for the six selected IMS stations will be conducted using stack monitoring data from the ANSTO facility.

JAEA used HYSPLIT code along with NCEP meteorological data to calculate "daily" and "half-daily" simulations for the "blind phase (phase 1)" of the challenge. An atmospheric dispersion simulation for three1-day hypothetical releases were determined (see Figure 40), and the dilution factor at each station was calculated based on the 31-day Hypothetical Release. These results indicate that the <sup>133</sup>Xe plume would arrive at the NZX46 station first, the FRX27 station has one sample per day, the other five stations have two samples per day, and BRX11 is expected to have the lowest average dilution factor among the six stations. The "open phase (phase 2)" of the 2<sup>nd</sup> ATM Challenge will be completed after the stack monitoring data from ANSTO is made available.



Figure 40. Atmospheric dispersion simulation for 31-day hypothetical release

### 8.3 2<sup>nd</sup> ATM Challenge from CTBTO PrepCom Perspective: An Approach to Quantify Uncertainties in Forward and Backward Modeling

The CTBTO PrepCom presented an approach to quantify uncertainties in forward and backward modeling to better understand the quality of data from ATM simulations.

Challenges faced in validation of ATM-simulated results include uncertainties related to meteorological data, emission data, the model, and observations. For the ATM Challenge, the same emission data and measurements were used for all simulations but there are still many possible sources of discrepancies to be considered; some examples include the use of either ECMWF or NCEP meteorological data that require different assimilation algorithms; spatial resolution (e.g., 1 degree vs 0.5 degree); stack data release options (24 hours, 12 hours, 3 hours, etc.); and the number of virtual particles released during simulations.

It should also be kept in mind that radioxenon measured at a given station may be the result of several different sources. Thus, underestimation of simulated results may result from the comparison of one source versus multiple sources. The overestimation of simulated results may also have different reasons, such as the release being higher than assumed, modeling errors, or other sources. In addition, seasonal changes have been shown to affect the <sup>133</sup>Xe source contributions observed at IMS stations.

In this study, the activity concentration values for <sup>133</sup>Xe were estimated for four stations (FRX27, NZX46, AUX09, and AUX04) using FLEXPART in the backward mode with both ECMWF and NCEP meteorological data. These results were compared with measured values to determine uncertainties and mean values for 24-, 12-, and 3-hour release values, as shown in Figure 41, and to calculate MDC median values for each time (dif\_24h= 0.29 MDC, dif\_12h= 0.27 MDC, dif\_3h= 0.20 MDC, and dif\_back= -1.17 MDC).



Figure 41. Difference between simulated and measured values at 24, 12 and 3 hours and for background measurements

### 8.4 Impact of Non-treaty-related Radioxenon Sources on CTBT Verification—Nuclear Power Plants in SEX63 Data

FOI shared information related to the potential effect of radioxenon from nuclear power plant (NPP) treaty verification. Power reactors are typically "weak" sources of radioxenon, but there are many reactors that contain and could emit (and occasionally have emitted) sizeable amounts of radioxenon. Typically, operational xenon signatures are quite different from signatures from a "fresh" nuclear explosion, but signals from nuclear explosions cannot always be expected to be "fresh"—the "full signature" of four xenon isotopes is rarely detected and there are a surprisingly large number of exceptions to "schematic" NPP operations. Therefore, a better understanding radioxenon emission from reactors is needed.

There are three NPPs located in Sweden that surround the SEX63 IMS radionuclide station; see Figure 42. To investigate the impact of these NPPs on the SEX63 station, the 6608 air samples that were collected at the station between 2006 and 2016 were analyzed using quality control (QC) drift correction. Of the original samples, 5732 samples were retained after excluding those that did not meet the QC requirements or were influenced by Fukushima. Radioxenon detections found in the retained samples were further analyzed for xenon signatures of NPPs using 3- and 4-isotope plots. The reactor detection rates observed in 3- and 4-isotope plots are comparable to or lower than false-positive and random coincidence rates, making analysis challenging but not impossible. Where measured, xenon ratios are available for observed radioxenon detections; they could often be attributed to known NPP operations or operational patterns. While detections of one or two isotopes are more common, they offer less

independent information. Whether the source of radioxenon detection is an NPP needs to be confirmed by ATM and/or information received from the NPP.



Figure 42. NPPs in Sweden near the SEX63 IMS station

The study concluded that when NPPs are present near an IMS station, radioxenon signatures from the NPPs can be detected fairly frequently and occasionally more distant (regional) NPPs can be detected. While NPP-related detections can sometimes be identified by their radioxenon ratios, it is rare because radioxenon isotopic ratios emitted from NPPs often stay within expected standard operations cycle trajectories, but there are exceptions that do not require anything like an emergency to happen (e.g.,  $^{135}$ Xe/ $^{133}$ Xe >> 1). A better understanding of operations at NPPs will help better identify NPP emissions, and the more isotopes detected increases the chance of identification/discrimination.

### 8.5 Characterization of Global Xe-133 Atmospheric Background

The Commissariat à l'Énergie Atomique et aux Énergies Alternatives (CEA) conducted a study to simulate the <sup>133</sup>Xe atmospheric background considering the main identified industrial sources worldwide for a two-year period (2013-2014) using ATM. The goal of this study was to characterize the <sup>133</sup>Xe atmospheric background across the globe and at particular points of interest (IMS stations). A comparison of the simulation vs measurements from the 29 operational IMS stations was presented.

ATM simulations were based on radioxenon releases from fifteen source groups, nine MIP facilities, and six groups of NPPs—the sources were combined based on similarities and consisted of nine MIP facilities and 383 total reactors. The daily release of <sup>133</sup>Xe from these sources was 48 TBq for MIP and 3TBq for the NPPs for a combined daily release of 51 TBq <sup>133</sup>Xe per day. The dispersion of each release was calculated over 30 days with 12-hour output resolution using FLEXPART.

Simulated <sup>133</sup>Xe annual average activity concentrations indicated that there was a factor of four difference between the Northern and Southern Hemispheres with an average <sup>133</sup>Xe concentration of 0.4 mBq/m<sup>3</sup> in the Northern Hemisphere and 0.1 mBq/m<sup>3</sup> in the Southern Hemisphere. This difference is likely influenced by differences in the wind circulation of the two hemispheres because the daily <sup>133</sup>Xe emissions for the Southern Hemisphere are approximately two-thirds of the Northern Hemisphere

emissions (19 TBq in SH vs 32 TBq in NH). The simulations also indicate that the average  $^{133}$ Xe concentration near MIP facilities can exceed 5 mBq/m<sup>3</sup>, while the contribution from NPPs to the background remains mostly regional and limited to the eastern half of North America, Europe, and Southeast Asia.

The contributions of different release sources at individual IMS radionuclide stations was investigated using the simulation data and, with the exception of five stations located near the Inter Tropical Convergence Zone (ITCZ), releases from sources in the Northern and Southern Hemispheres only contribute to the atmospheric background at IMS stations located in the Northern and Southern Hemispheres respectively; see Figure 43. The results also indicated the contribution of multiple xenon sources to the total average background at each IMS station.



**Figure 43**. Contribution of identified sources to the annual average of <sup>133</sup>Xe simulated at the 39 IMS radioxenon stations. Yellow arrows indicate stations located at the tropical convergence zone.

Seasonal variations were investigated to forecast the <sup>133</sup>Xe atmospheric background changes, and it was predicted that the occurrence of <sup>133</sup>Xe detections at IMS stations will increase during winter periods for both hemispheres.

The simulation data for annual <sup>133</sup>Xe average activity concentrations at IMS stations was compared to IMS measurements. There was good agreement between the measured and simulated data for the 6000 measurement points considered for average activity concentration and variability of observed levels between stations. Of the 39 IMS stations only five have an average concentration greater than 1mBq/m<sup>3</sup>. It should be noted that the daily activity concentrations have a large variability that cannot be seen in annual averages. While there is still more to understand, this simulation gave a good comprehension of the "global picture."

# 8.6 Canadian Nuclear Laboratories Emissions Study in the Ottawa Valley

Health Canada (HC) presented data collected as part of an ongoing study to collect emission data from locations near the Canadian Nuclear Laboratories (CNL) medical isotope facility and the Ottawa Valley.

Since 2001, HC has used these emissions as the basis of a real-world measurement and "modeling laboratory" employing its radiation monitoring equipment throughout the Ottawa valley and its access to data from more sensitive equipment operated in the global international monitoring system of the CTBTO PrepCom. HC, in partnership with CNL and other partners, has undertaken an extensive measurement campaign to gather as much emission information as possible in and around the facility before its closing. The outcome of the project will be a shareable time-series database with detailed emission, corrected time-series data, meteorological data on- and off-site, isotropic ratios during different plant processes, regional sensory measurements, and detailed atmospheric transport modeling. Information on the types of data collected and current data analysis were shared.

Stack monitoring data collected from 2010 until present will be used to provide a correlation between medical isotope processing and noble gas releases. The radioactive noble gas source terms for this study were the stacks of the MIP facility and the NRU reactor stack. The goals of this study are to provide an accurate source term for modeling, understand the release from different plant processes, and understand the temporal relationships between regional and global monitors. In addition, this information along with detailed information on the chemistry being performed will help researchers understand the factors that influence the various xenon ratios and the distribution of the ratios MPF stack monitor data.

A high-resolution stack monitor data measurement is also being conducted at the MIP facility stack by placing a Canberra Falcon detector in-line before and after the iodine cartridge to determine if the iodine cartridge creates a holdup of noble gasses. This data will be used to characterize emissions from the facility, relate them to facility processing, and compare real measurements to theoretical values.

The NRU reactor emits approximately 1 TBq/hr noble gas release (including argon). In addition to stack monitoring at NRU, regional measurements have been collected by HC's network of seven NaI detectors and CNL's Geiger-Mueller tube measurements around and on the Chalk River site. This data is being analyzed to determine local noble gas concentrations downwind to understand MIP on global concentration.

Air sampling in the region has been conducted collecting 500 L and 800 L bag sample for xenon analysis. Data collected from these samples will be used to determine local noble gas concentrations for comparison with data from the NaI detectors and to determine local concentrations for ATM modeling verification.

Aerosol samplers for <sup>135</sup>Cs, the product of <sup>135</sup>Xe, will be collocated with SPALAX near CNL and in Ottawa. These filters will be analyzed by accelerator mass spectrometry to determine local concentrations of <sup>135</sup>Cs and compare these concentrations with measured <sup>135</sup>Xe concentrations at the same location.

Meteorological data for the periods of the study were obtained from local weather stations and the Canadian Centre for Meteorological and Environmental Prediction's Atmospheric Dispersion Suite.

The collected data is currently being used to calculate local noble gas concentrations by ATM, as shown in Figure 44, which is compared with measured concentrations from collected air samples. Upon completion, the study will provide shareable data sets, increased understanding of the influence of MIP processing on emissions, improved ATM for accident response and global transport, and a better understanding of the impacts of MIP on global measurements.



Figure 44. Surface concentration plume for a 6-hour emission period from the MIP facility stack starting October 18 2013 UTC

# 8.7 Subsurface Gas Measurements near Chalk River, Canada

The University of Texas at Austin presented results from their second sampling campaign to study imprinting of radioxenon near Chalk River, Canada. Imprinting is a mechanism whereby radioxenon in the atmosphere becomes pressed into subsurface pores and fractures in the ground. The original sampling campaign was carried out in 2014 where subsurface radioxenon was quantified. The elevated subsurface radioxenon concentrations were shown to have a source term from radiopharmaceutical production. The most recent sampling effort in 2016 aimed to increase the understanding of the atmosphere/soil gas interface as well as environmental sampling and associated technologies. Sampling depth was also investigated as a variable within the sampling methodology.

Samples were collected by auguring holes into the ground and drawing air samples from the holes at 1.4 L/min through a subsurface gas sampler. Once an entire sample was collected, it was compressed into SCUBA bottles and shipped to PNNL for beta-gamma analysis of xenon. The experiment successfully collected 26 total samples (13 subsurface and 13 atmospheric). Subsurface samples were collected at 103 cm (8) and 189 cm (5).

The first-ever measurements of radioxenon imprinting into subsurface gas was conducted by the University of Texas in 2014. From this sampling campaign, it was concluded that radioxenon levels on the order of 10% of the average atmospheric radioxenon concentration could be detected by subsurface sampling. The second sampling campaign in 2016 found that <sup>133</sup>Xe concentrations began increasing after first plume passage and a maximum subsurface concentration of 48 Bq/m<sup>3</sup> corresponded to 2 days of highly elevated atmospheric concentrations.

Subsurface transport modeling performed using the Subsurface Transport over Multiple Phases (STOMP) code developed by PNNL was used to analyze the data. The site was modeled as a single, homogenous, porous media with the general properties of sand and a simulated depth of 15 m. Modeled results were averaged over sampling periods and compared with measured results.

For the 2014 experiment, the model predicted a higher xenon concentration than was measured, although the model did predict the same trend. This result may be because the model does not account for xenon interactions with geology, water, adsorption or absorption, or sampling effects. Modeled results for the most recent samples (2016), which were collected at 1 m depth, also predicted higher concentrations than for the measured concentrations. Again, the model predicted radioxenon imprinting and had a similar trend for both the measured and modeled values. The best results were achieved for samples collected at 1.9 m depth; see Figure 45. The model predicts the trend fairly well and a xenon concentration factor of 0.8 lower than measured. This combination of higher predictions at 1 m and lower predictions at 1.9 m may indicate an inaccurate representation of the actual permeability by the model corresponding to geology change, as noted during hole emplacement, which could lead to an underestimation for the upper 1 m of soil.



Figure 45. Comparison of modeled (Blue) and measured (Red) values for imprinted radioxenon concentration

Results of both sampling campaigns show that imprinting of radioxenon into the subsurface does occur and was observed during periods of increasing and decreasing pressure—models confirm that when sampling, barometric pumping is not the primary driver in porous media. Simple descriptions of the site geology were sufficient to predict the expected subsurface radioxenon concentrations within order of magnitude (at least in porous media). Based on these results, it is predicted that a radioxenon plume containing at least 1 Bq/m<sup>3</sup> in areas with consistently high atmospheric xenon concentrations could be seen days after passing over the area if samples were taken at a 10 m depth. Future work related to imprinting will include studies with different geology (rock instead of sand), a simultaneous depth profile, and examination of mitigation techniques.

## 8.8 Web-Grape Continuously Emitting Sources Functionality—a Handy Tool Connecting Nuclear Facilities and IMS Stations

The CTBTO PrepCom discussed recently added functionalities of Web-Grape software, which enables visualization and post-processing of ATM outputs used to trace the travel paths of radionuclides. The IDC calculates SRS fields for each radionuclide sample taken at the IMS radionuclide stations. The ATM operational system used for these calculations is based on a Lagrangian Particle Dispersion Model, FLEXPART (ver. 9.02). Daily calculation of SRS fields uses global wind field analysis data provided by ECMWF and NCEP. To enable visualization of the ATM outputs, the IDC has designed and developed the web-connected graphics engine (Web-Grape) software.

Web-Grape (v. 1.8.1) was designed by the IDC as a convenient viewer and integrated assessment tool that requires IDL virtual machine runtime environment software. The Web-Grape platform is an independent application available for almost any platform (e.g., Mac OS X) and those supported by the PTS (Solaris 10, Linux and Windows for any CPU architecture).

A Continuously Emitting Source (CES) functionality was recently added to Web-Grape which allows the user to analyze the effect of continuously emitting sources such as nuclear facilities on the measured concentrations at monitoring stations. The resulting time-series of predicted measurements per station can be displayed as line plots and are also stored in CSV format. One possible application of this new functionality is distinguishing between contributions from different individual facilities or from groups like MIP facilities and NPPs to the measured concentrations at monitoring stations. Examples were shown that used data from the 2<sup>nd</sup> ATM Challenge for IMS stations AUX09, NZX46, and FRX27 and daily values of <sup>133</sup>Xe emission from ANSTO, Batek, Ezeiza, and Pelindaba, as shown in Figure 46, and use of the software to compare seasonal changes in wind field pattern at IMS stations.







**Figure 46**. Example data generated using Web-Grape showing the contribution of <sup>133</sup>Xe from different sources

## 8.9 Source Uncertainty Quantification by Using LAM Ensemble Dispersion Modeling

The Egypt National Data Center, National Research Institute for Astronomy and Geophysics gave a presentation introducing their investigations on quantification of source uncertainty. Source determination of man-made radioactive isotopes, which may be observed by CTBTO PrepCom's IMS radionuclide stations, is a critical matter due to the existence of many uncertainties in the inverse modeling (meteorological data, radionuclide concentrations, and inherent atmospheric models uncertainties). The ability of an ensemble technique to quantify source uncertainty was investigated using an idealized numerical experiment, where the dispersion model (FLEXPART-WRF) using an ensemble of seventeen different atmospheric simulations for each <sup>133</sup>Xe source was run in the forward mode to estimate the concentration of <sup>133</sup>Xe at IMS stations. Using the concentrations calculated for the IMS station, the dispersion model was run in the backward mode to estimate the SRS fields, which were then used to estimate the source location by the Bayesian approach; see Figure 47.



Figure 47. Experimental outline

This study concluded that the ensemble technique can reduce the uncertainty in source determination and that a combination of the ensemble and the Bayesian techniques gives the best results. Atmospheric uncertainties were found to be associated mainly with regions of baroclinic systems (low pressure and trough regions). It was also determined that uncertainties in the middle latitude region propagate from west to east and that uncertainties increase with time for short-period simulations.

## 8.10 The Impact of Interfering Plumes on Nuclear Explosion Monitoring and a Possible Mitigation Using Numerical and Bayesian Analysis

Lawrence Livermore National Laboratory illustrated the impact of plumes originating from industrial facilities by simulating a hypothetical release from an underground nuclear explosive test in the presence of temporally and spatially varying background radiological air concentrations. Separation of background noise through analysis of both the interference radioxenon plume and the radioxenon plume of monitoring interest requires source characterization. When the source of the interfering plume cannot be well characterized, Bayesian methods under development may help estimate both the interference noise and the signatures of interest.

Bayesian and genetic algorithm methods may help discriminate a treaty violation from other releases but require careful source characterization because localized meteorological data can affect longer-range transport/dispersion. New methodologies incorporating test data and simulated subsurface gas transport are better for characterizing dynamic sources for atmospheric transport. These capabilities that

incorporate CFD-LES are helping to resolve surface meteorological complexities near the source. Some examples using these methods were discussed. A study that incorporated the Markov chain Monte Carlo (MCMC) approach to reconstruct the 1998 Algeciras release through a forward simulation was able to identify the likely source locations to within 100 km of the true source. In another analysis, a regional-scale Bayesian inversion was used to characterize an NPP source with uncertain release and meteorological conditions. This analysis identified the source inputs as 80% of variance and meteorological conditions as 20% for this historical study. Lastly, a study using genetic algorithms to optimize a greenhouse gas observation network in California was discussed in which single objective performance genetic algorithm (SOGA) was used to identify sensor locations identified using a multi-objective performance genetic algorithm (MOGA) to access maximize accuracy and minimize deployment costs. By considering performance and cost together, a flexible sensor deployment plan was developed; see Figure 48.



Figure 48. Comparison of sensor locations determined using Single Objective Performance Genetic Algorithm (SOGA) and Multi-Objective Performance Genetic Algorithm (MOGA) methods

# 9.0 Roundtable Discussions

Three roundtable discussion sessions were held during the workshop to promote the exchange of ideas and conversation on abatement technology, the use of stack data, and path forward/action items. To help stimulate conversation, questionnaires were answered in small groups during working lunch breaks, and the answers were discussed during the roundtable sessions.

# 9.1 Abatement Technology

The first question for discussion was what would you like your monitoring/MIP counterpart to understand about your industry? The producers stressed that they want to do their best to reduce emissions, but there are costs, added complexity, and reliability concerns related to abatement technology and that clear goals would be helpful. The monitoring community would like the producers to understand the following:

- Small releases of treaty-relevant nuclides (4 radioxenon and 83 particulate) can be seen at long distances
- The importance of understanding MIP release patterns
- Data shared with the monitoring community is used for science monitoring only
- Producers do not want to reduce <sup>99</sup>Mo production

When asked what the biggest barriers to facilities adopting additional abatement methods are, the producers stated that licensing and regulatory changes would be required; introduction of new systems would increase operation cost and complexity; there are physical space limitations in existing facilities; and the risk of failure. If systems were to be provided to the producers, then all costs associated with installation and operation must be covered, not just the technology. These costs would include costs related to facility shutdown for upgrades, lobbying the regulator, legal advice, and quality assurance involvement, ongoing maintenance costs, etc.

Abatement technologies considered likely to succeed in lowering emissions are charcoal and delay tanks, which have been proven but require considerable facility space; other technologies such as silver zeolite and cryo-based techniques would need long-term testing to ensure reliability; and passive systems are thought to have a higher likelihood for success. Incorporation of technology for real-time feedback at the hot cell was thought to be a good idea. Cost will be the driver as to what type of system can be employed, and an approach combining technologies is likely the best solution.

Possible experiments that could be conducted at a facility were discussed and included bypassing the abatement system altogether and monitoring the effect on the IMS system; performing long term testing of new abatement technologies under real conditions; identifying and repairing leaks in the current system; monitoring facility releases; performing ATM modeling; sharing operator knowledge on lowering emissions (tricks of the trade). Issues related to performing experiments at an operational facility include liability if the test produces a release and limited funding.

# 9.2 Use of Stack Data

The session began by discussing the largest barriers to overcome for facilities to begin stack data sharing. Security and confidentiality of shared data was the largest concern. Having the necessary resources for system upkeep and quality assurance/quality control by the producers was also a large concern. Solutions to these barriers were discussed, including the following:

- Incentives for stack monitoring equipment and upkeep.
- Automated data analysis with human review only in special instances to reduce costs.
- Incorporation of a delay time before data is shared—it was thought that a delay time of one week may be too long but 1-3 days would be acceptable. Receiving data in near real time would be best for increasing the quality of IDC products.

The discussions moved to whether stack data or other data would be useful to the MIP. Several benefits of stack data were identified by the producers, including the following:

- A better overview of the processing and when releases occur (identifying trends in emissions)
- Possible backup to the regulatory system
- Use for research and development studies
- Use by operational staff—real-time stack release information would help improve their processes

Types of data that could be shared were discussed. It was stated that sharing of raw spectra data may be difficult for some MIP facilities; that processed time-series data may be easier; and there could be issues if these values were significantly different from those determined by the facility's regulatory system. The monitoring community responded that while time-series data for treaty-relevant isotopes would be fine for routine collection, raw data would be helpful for confirmation of unusual events.

The roundtable session determined that the decision of whether to share stack release data was dependent on the facility and could be the Chief Executive Officer, other company personnel, or a government official. Each facility is unique, but to share data, each facility would need to know how the data was being used and that it was secure, at minimum.

During an off-normal event (accidental release), a delay time would be important for facility personnel to deal with the issue. Although facility personnel would be busy, communicating with the monitoring community would be important if treaty-relevant isotope ratio releases occur.

### 9.3 Path Forward/Action Items

During the final roundtable discussion, the community was in agreement that WOSMIP is making a difference. The workshop has resulted in upgrades to existing facilities, implementation of solutions, and application of ATM to better understand the problem. Some of the most useful aspects of this WOSMIP were thought to be the producer updates, discussion on regulatory pressures and need to increase communication, the exchange of ideas, abatement discussions, discussion of challenges related to discrimination of sources and ATM, working lunches, the ATM challenge, understanding releases from MIP, and progress of technologies.

Suggestions to improve the WOSMIP technical program included involvement of more students, having a dedicated poster session, expansion of the Other Sources session, more focus on abatement technology, inclusion of other data (seismic, hydroacoustic, argon isotopes, etc.), and making more time for ATM (potentially a side meeting).

Although this was one of the largest WOSMIPs to date, there are others that would benefit the workshop, including a representative from the NPP community, representatives of the hospital and end users, and producers not present at this workshop (e.g., Russia, South Africa, India, China, United States).

Other fora available to discuss similar topics such as the CTBT Science and Technology conference and the Mo-99 Topical Meeting were discussed as well as the possibility of holding a joint meeting with other conferences.

A number of experiments that would benefit the WOSMIP community were suggested, including a stack monitoring challenge, abatement experiments, sequestration experiments, ATM experiments, experiments to understand NPP emissions, comparison of new abatement systems installed at ASNTO and IRE, and experiments to understand emissions from aqueous targets (ARGUS, ENSTERNA, SHINE).

Some final comments from the participants included that it was good to have the workshop outside of Europe; the workshop has expanded to include other sources (not just MIP) of radioxenon emissions; the producers that participated are willing to share data; increased communication would be beneficial; and it is important to continue to build trust between the MIP and CTBT communities.

# **10.0 Workshop Conclusions**

A concluding discussion was held to summarize thoughts and ideas from the WOSMIP community. A number of positive outcomes from WOSMIP VI were noted: expansion of the workshop to include emission sources other than MIP; willingness of the producers to share stack release data with the CTBT; sessions in which abatement and stack release data sharing were discussed; discussion of how the uncertainty in ATM calculations can be quantified; and it was the first WOSMIP to be held outside of Europe. It was commented that it may be beneficial to increase communication and build awareness of issues relating to radioxenon emissions with organizations outside of the current WOSMIP community and that building trust between organizations is important to the success of WOSMIP. It was agreed that the next WOSMIP should be held in approximately 18 months. Lastly, there was an announcement that a WOSMIP-related session will be added to the CTBT Science and Technology 2017 (SnT17), Vienna, Austria from June 26-30, 2017, if there is enough interest as measured by the number of abstracts submitted.

The workshop was extremely successful. Many producers presented their updates and showed not only their interest but also the methods put into practice to reduce radioxenon emissions from MIPs. There were significant advancements in R&D. SCK•CEN received the WOZZIE award for its contribution to the design of abatement technologies. CTBTO PrepCom put emphasis on stack data confidentiality in its experiment.

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# Appendix A: Workshop Agenda

#### Workshop on the Signatures of Man-Made Isotope Production (WOSMIP) 2016

Hosted by the Preparatory Committee for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom), Investigación Aplicada (INVAP) and Pacific Northwest National Laboratory (PNNL)

#### 28 November–2 December 2016

San Carlos de Bariloche, Argentina

#### **Final Agenda**

#### Monday, 28 November 16

8:00 - 9:00 BADGING AND REGISTRATION

#### Session 1: Welcome and Workshop Overviews Theme: Introductory Contributions Chair: Kalinowski/CTBTO PrepCom

- 9:10 9:20 Welcome Pedro Pessatti/Vice-Governor of the Province of Rio Negro
- 9:20 9:30 Welcome Pedro Villagra Delgado, Chief of Cabinet, Foreign Affairs Ministry
- 9:30 9:40 Welcome M. Kalinowski/CTBTO PrepCom
- 9:45 10:15 Workshop Overview Ted Bowyer/PNNL
- 10:15 10:35 International Monitoring System Overview Romano Plenteda/CTBTO PrepCom
- 10:35 10:55 Radionuclide Processing at the IDC M. Kalinowski/CTBTO PrepCom
- 10:55 11:20 **BREAK**
- 11:20 11:50 Issues in the MIP World Richard DeCaire/Nordion
- 11:50 12:15 Progress on the IAEA CRP on Sharing and Developing Protocols to Further Minimize Radioactive Gaseous Releases to the Environment in the Manufacture of Medical Radioisotopes Joanie Dix/IAEA

- 12:15 12:45 IMS radionuclide monitoring after the DPRK 2016 announced tests Martin Kalinowski/CTBTO PrepCom
- 12:45 14:20 WORKING LUNCH (Group Photo)

#### Session 2: Alternative Xenon Source Terms Theme: The Broader Range of Isotope Sources Chair: di Tada/INVAP

- 14:20 14:40 Potential Sources of Isotope Emissions from Next-Generation Nuclear Reactors Derek Haass/University of Texas at Austin
- 14:40 15:00 Radioxenon potential emission analysis from accelerator-produced Alpha-particle Emitting radionuclides facilities Diego Ferraro/INVAP
- 15:00 15:20 Problems of Radon isotopes' fixation and detection during the processing of irradiated <sup>226</sup>Ra Tatiana Boytsova/Khlopin Radium Institute
- 15:20 15:50 BREAK
- 15:50 16:10 RA3 Research Reactor, base of the reactor radioisotope production in Argentina Jorge Alberto Quintana Domínguez/CNEA
- 16:10 16:30 Radioxenon Emissions from Research Reactors Paul Saey
- 16:30 16:50 Brazilian Multipurpose Reactor (RMB) Project and Status Jose Perrotta/CNEN (Brazilian National Nuclear Energy Commission)
- 16:50 17:15 Contribution of Normal Operational Releases from Nuclear Power Plants to the Global Radioxenon Emission Inventory Martin Kalinowski/CTBTO PrepCom
- 17:15 17:40 Xe-133 Medical Use Diagnosis of Pulmonary Embolism Ira Goldman/Lantheus

- 17:40–18:00 On the State Program Tajikistan on Restoration of the "Argus" Liquid Nuclear Reactor and Plans for the Production of Radiopharmaceuticals Khikmat Muminov/Academy of Sciences of the Republic of Tajikistan
- 17:40 End for the day
- 18:30 PNNL SOCIAL HOUR

#### Tuesday, 29 November 16 Session 3: Current Mo-99 Production overview Theme: Recent and planned activities at isotope production facilities Chair: Saey/IAEA

- 9:00 9:10 Daily Welcome Chair
- 9:10 9:35 Mo-99 Production overview Joanie Dix/IAEA
- 9:35 9:55 Radioisotope Production at National Atomic Energy Commission of Argentina Pablo Roman Cristini/CNEA
- 9:55 10:15 Update on MIP and emission abatement systems at ANSTO Emma Hoffmann/ANSTO
- 10:15 10:35 IRE Update Dominique Moyaux/IRE
- 10:35 10:55 **BREAK**
- 10:55 11:15 Mo-99 Production and Effective Control of Xe-133 Emissions Brown/Mallinckrodt
- 11:15 11:35 Nordion Update Richard DeCaire/Nordion
- 11:35 11:55 Production of Radioisotopes in Pakistan Khalid/PINSTECH
- 12:15 13:45 Working Lunch (MIP Issues, Abatement tech and experiments)

#### Session 4: R&D efforts on emission reduction Theme: Recent advances for the reduction of radioxenon emissions Chair: Gueibe/SCK•CEN

- 13:45 14:05 Radioxenon Abatement Technology Overview Juan José Merino/INVAP
- 14:05 14:25 Past, present and future progress in radioxenon mitigation Christophe Gueibe/SCK•CEN
- 14:25 15:00 Feasibility study of the xenon retention with the Ag-ETS10 Zeolite prototype system developed by SCK-CEN Moyaux/IRE
- 15:00–15:30 **BREAK**
- 15:30 16:55 Round Table on Abatement Technology ALL

#### Session 5: Future Mo-99 Production Theme: Progress on development of new isotope production facilities Chair: Benoit Deconninck/IRE

- 16:55–17:15 Radioisotope Production in Jordan Research & Training Reactor (JRTR) Ahmad Malkawi/JAEC
- 17:15 End for the day
- 19:00 Pick up at Edelweiss Hotel and drive to Villa Huinid Resort
- 19:30 Showcase Concert INVAP
- 20:15 Conference Dinner CTBTO PrepCom

#### Wednesday, 30 November 16 Session 6: Future Mo-99 Production Theme: Progress on development of new isotope production facilities Chair: Benoit Deconninck /IRE

- 9:00 9:10 Daily Welcome Chair
- 9:10 9:30 PT INUKI Update/ New Indonesian Medical Isotope Production Plant Yudiutomo Imardjoko Bernadib/PT INUKI

#### Session 7: Stack Emission Data Theme: Understanding the global radioxenon backgrounds from industry Chair: Friese/PNNL

- 9:30 9:55 Stack Release Data Confidentiality Martin Kalinowski/CTBTO PrepCom
- 9:55 10:25 Data upload IDC software requirements Jonathan Baré/CTBTO PrepCom
- 10:25 11:00 **BREAK**
- 11:00 11:25 Stack Data Upload Experiment Doll/PNNL
- 11:25 12:00 STAX New Concept for Using Stack Data for the CTBT Judah Friese/PNNL
- 12:00 12:25 Ideas on the implementation of the stack monitoring data sharing and analysis network Kirill Khrustalev/ISTI
- 12:25 14:00 WORKING LUNCH (Stack sharing issues/solutions)
- 14:00 14:25 A compact Set-Up for Measuring Radioxenon Concentrations in the Stack Releases of Nuclear Facilities Florian Gubernator/ZNF Germany
- 14:25 14:50 Stack Air Effluent Monitor: present and future Mariana Di Tada/INVAP
- 14:50 15:15 Evolution in HPGe detectors, when performance & reliability counts Luc De Baerdmaeker/Canberra

15:15 – 15:45	Break
15:45 - 16:10	Evaluation of the AgETS10 Zeolite in the stack monitoring system of a RPF Benoit Deconninck/IRE
16:10 - 17:50	Round Table on use of stack data ALL

17:50 End for the day.

#### Thursday, 1 December 16 Session 8: Atmospheric Transport Theme: Characterization of the atmospheric isotope background Chair: Bowyer/PNNL

- 9:00 Daily Welcome Chair
- 9:00 9:30 2<sup>nd</sup> ATM Challenge 2016 Christian Maurer/ZAMG
- 9:30 9:50 Analysis Results of 2<sup>nd</sup> ATM Challenge 2016 by JAEA/NDC Yuichi Kijima/JAEA
- 9:50 10:10 2nd ATM Challenge from CTBTO PrepCom Perspective: An Approach to Quantify Uncertainties in Forward and Backward Modeling Jolanta Kusmierczyk-Michulec/CTBTO PrepCom
- 10:10 10:30 Impact of Non-treaty-related Radioxenon Sources on CTBT Verification—Nuclear Power Plants in SEX63 Data Anders Axelsson/FOI
- 10:30 10:50 Characterization of Global Xe-133 Atmospheric Background Sylvia Generoso/CEA
- 10:50 11:20 BREAK (WOZZIE award presentation)
- 11:20 11:40 Canadian Nuclear Laboratories Emissions study in the Ottawa Valley Rodney Berg/Health Canada
- 11:40 12:00 Subsurface Noble Gas Measurements near Chalk River, Canada Christine Johnson/University of Texas
- 12:00 12:20 WEB-GRAPE Continuously Emitting Sources functionality-a handy tool connecting nuclear facilities and IMS stations Kusmierczyk-Michulec/CTBTO PrepCom
- 12:20 12:40 Source Uncertainty Quantification by using ensemble Dispersion modeling Sayed Mekhaimr/Egypt NDC

12:40 – 13:00 The impact of interfering plumes on nuclear explosion monitoring and a Possible mitigation using numerical and Bayesian analysis Lee Glascoe/LLNL

#### 13:00 – 14:30 WORKING LUNCH (Path Forward/Action Items)

14:30 – 15:30 Round Table discussion (Path Forward/Action Items)

#### Session 9: Workshop Conclusions

15:30 - 15:45 Review of WOSMIP

Lori Metz/PNNL; Charles Doll/PNNL, with support of INVAP Rapporteurs Elsa Iturbe - Alejandra Roncallo /INVAP

- 15:45 16:15 WOSMIP Path Forward and Action Items ALL
- 16:15 16:30 CTBTO PrepCom Concluding Remarks Martin Kalinowski/CTBTO PrepCom
- 16:30 16:40 Greetings & Farewell words Eduardo Nassif/INVAP
- 16:40 End for the day

#### Friday, 2 December 16 Session 10: Technical Tours Chair: INVAP

- 8:45 12:30 Daily Welcome-Travel to Technical Tour Locations (Part I) INVAP
- 12:30–13:15 Travel to Technical Tour Location (Part II) INVAP
- 13:15 14:20 WORKING LUNCH (Location TBD)
- 14:30 19:00 Technical Tour (Part II) INVAP
- 19:00 End for the day

#### **Posters**

Note: All posters will be on display throughout the workshop. Authors will be present to discuss their poster during the assigned sessions (below).

#### Poster Session 1: Monday, 28 November 16, 15:10–15:40

Radioisotope Application and production in Myanmar

M. Min Htwe, Ministry of education - Myanmar

CNEA Argentina Update

E. Carranza - Comisión Nacional de Energía Atómica de Argentina (CNEA)

Comparison and modeling of different design of steel shielding for radioisotopes production hot cells

Romero Marengo, N. Brunori, E. Cuello, R. Rey, F. Albornoz, R. Cocco - INVAP S.E

Future Medical Isotope Production in Argentina: a Regulatory Overview

Eduardo Quintana – ARN

#### Poster Session 2: Tuesday 29-November-16, 15:25-15:50

Medical Radioisotopes Production and Supply in Bangladesh

M. Waheed

Recent Developments in Addressing Radioisotope Requirements for Medical and Industrial Applications in the Philippines

A. Bulos, M. Borras, G. Ciocson, I. Nunez - Philippine Nuclear Research Institute

Global Spatial Database of Anthropogenic Isotopes

S. Shah – National Disaster Management Authority (NDMA)

ORIGEN-S evaluation of radioxenon isotopes signatures from Molybdenum production facilities for long irradiation times Donato Maurizio Castelluccio, Federico Rocchi - ENEA

#### Poster Session 3: Wednesday 30-November-16, 11:00-11:30

Development of CTBT- relevant Xe isotopes in the atmosphere between 2010 and 2015

M. Elbahrawy<sup>1</sup>, S. Mekhaimr<sup>2</sup>–<sup>1</sup> Atomic Energy Authority P.O. Box 13759 Cairo-Egypt, National Data Center, Cairo, EGYPT; <sup>2</sup> National Data Center, National, National Data Center, Cairo, EGYPT Research Institute for Astronomy and Geophysics, Cairo, EGYPT

Medical Isotopes Production Facilities: Atmospheric Transport Modeling and Dose Calculation

M. Alessi<sup>1</sup>, C. Field<sup>1</sup>, S. Eguía<sup>2</sup>, M. Brizuela<sup>1</sup> - <sup>1</sup> INVAP; <sup>2</sup> INVAP and UNRN

Medical isotope production and monitoring in Thailand

P. Krisanangkura – Office of Atoms for Peace

#### Poster Session 4: Thursday 01-December-16, 10:50-11:20

The Impact of Power Reactor Noble Gas Retention systems on Discriminating Xenon Releases of Nuclear Tests and Nuclear Reactors

A. Heise, G. Kirchner – University of Hamburg

 $\beta$ (PS)- $\gamma$ (HPGe) Coincidence System for Radioxenon Isotopes Measurement in CNL06

Q. Li, S. Wang, H. Jia, Y. Fan, Y. Zhao, X. Zhang – Beijing Radionuclide Laboratory

On the usability of event zero time determinations using xenon isotopic activity ratios given the real atmospheric background observations

K. Yamba<sup>1</sup>, M. B. Kalinowski<sup>2</sup>, O. Sanogo<sup>1</sup>, J. Koulidiati<sup>3</sup> – <sup>1</sup>CNRST - NDC/ Burkina Faso; <sup>2</sup>IDC/CTBTO PrepCom; <sup>3</sup>LPCE/University of Ouagadougou

FLEXPART-WRF Solutions of the ATM Challenge

J. Kusmierczyk-Michulec,, M. Schoeppner/CTBTO PrepCom

- 1. PrepCom, C., Comprehensive Nuclear-Test-Ban Treaty. 1996.
- 2. Kalinowski, M.B. and M.P. Tuma, *Global radioxenon emission inventory based on nuclear power reactor reports*. Journal of Environmental Radioactivity, 2009. **100**(1): p. 58-70.





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