

PNNL-28870

# The 7th Workshop on Signatures of Man-Made Isotope Production

## 3-7 December 2018 in Sydney, Australia

J L Burnett, T W Bowyer, J I Friese, L A Metz, T M Weber



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3-7 December 2018 in Sydney, Australia

April 2019

J L Burnett, T W Bowyer, J I Friese, L A Metz, T M Weber

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

Pacific Northwest National Laboratory Richland, Washington 99354

### **Executive Summary**

On 3-7 December 2018, the 7th Workshop on Signatures of Man-Made Isotope Production (WOSMIP VII) was held in Sydney, Australia. The workshop was hosted by the Australian Nuclear Science and Technology Organization (ANSTO) and supported by the Pacific Northwest National Laboratory (PNNL). Its aim was to support radioxenon monitoring by the International Monitoring System (IMS) operated by the Provisional Technical Secretariat (PTS) of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), and unite experts from the radioisotope production and nuclear explosion monitoring communities for a 4-day scientific meeting. A total of 91 experts from 19 countries attended and participated in technical discussions relating to the impact of radioisotopes released from civil and industrial sources on nuclear explosion monitoring in support of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). During WOSMIP VII, sessions were held on background sources of man-made isotopes, research to reduce the radioxenon impact on the IMS of the CTBT, current and planned <sup>99</sup>Mo production activities, and radioxenon stack measurements.

WOSMIP VII commenced by ANSTO reaffirming its commitment to mitigating the impact of <sup>99</sup>Mo production emissions on the verification regime of the CTBT. This included the recent installation of a radioxenon stack monitor at the ANSTO Nuclear Medicine (ANM) <sup>99</sup>Mo facility in support of the Source Term Analysis of Xenon (STAX) project, officially opened by ANSTO and PNNL on the day prior to the start of the workshop. Subsequent discussions of background sources identified this as one of the most important topics pertinent to the nuclear explosion monitoring community. While it was recognized that WOSMIP had facilitated a significant improvement in the understanding of background sources, the need for more research and development was identified. It was estimated that for only approximately 50% of IMS detections was the radioxenon source adequately understood.

The significance of research reactor emissions was discussed, and while not generally considered to be a significant background source compared to isotope production, significant gaps in understanding their releases were identified, including spurious releases and potential air activation signals. These may become more important as radioxenon monitoring systems become more sensitive and the WOSMIP community's understanding of the background improves. It was noted that, the contribution of nuclear power reactors is more significant than research reactors and may be important at certain locations or at certain times of the year, since the source terms can vary due to startup and shutdown operations and subsequent meteorological conditions for atmospheric transport. The use of radioxenon in tracer operations by the oil and gas industry was also identified as a potentially important and enigmatic source. Anomalous particulate IMS detections were also considered, including detections of ruthenium isotopes (<sup>103</sup>Ru, <sup>106</sup>Ru) in Europe during late 2017. As with radioxenon measurements, a need to more fully understand these background fluctuations and source terms was recognized.

New WOSMIP focus areas were identified, including understanding the impacts of new medical isotope producers and advancements in emissions mitigation and stack monitoring technology. Recent developments in xenon abatement technologies demonstrated a need to focus on smaller, passive, cost effective, robust, and selective methods. Key to this success are collaborations with additional partners from the medical isotope production and monitoring communities. This was also apparent from the successful partnerships by both ANSTO and the Belgian Institute for Radioelements (IRE) with PNNL for the installation of STAX stack monitors for radioxenon emissions.

The workshop concluded with the presentation of the WOZZIE award to Dr. Anders Ringbom from the Swedish Defense Research Agency (FOI) in recognition of his significant contributions to radioxenon detection. It was followed by a tour of the ANM <sup>99</sup>Mo facility and Open Pool Australian Lightwater (OPAL) reactor provided by ANSTO at their main facility at Lucas Heights, situated on the outskirts of Sydney.

### **Acknowledgments**

The Pacific Northwest National Laboratory (PNNL) would like to thank the Australian Nuclear Science and Technology Organisation (ANSTO) for hosting WOSMIP VII. PNNL would also like to thank all WOSMIP participants for their contributions to the workshop and this report.

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### 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		
AWE	Atomic Weapons Establishment		
CNEA	Argentinian National Atomic Energy Commission		
CNEN	Brazilian Nuclear Energy Commission		
CNL	Canadian Nuclear Laboratories		
CPU	Central Processing Unit		
CRP	Coordinated Research Project		
СТВТ	Comprehensive Nuclear-Test-Ban Treaty Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty		
CTBTO PrepCom	Organization		
CRP	Co-ordinated Research Plan		
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		
HPC	High Performance Computing		
HPGe	High Purity Germanium		
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		
MCNP	Monte Carlo N-Particle		
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		

NG	Noble Gas
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
RR	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
TXL	Transportable Xenon Laboratory
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**

#### 1.1 Overview

The 7th Workshop on Signatures of Man-Made Isotope Production (WOSMIP VII) was held in Sydney, Australia, 3-7 December 2018. The workshop was hosted by the Australian Nuclear Science and Technology Organization (ANSTO) and supported by the Pacific Northwest National Laboratory (PNNL). WOSMIP VII provided a unique forum for technical discussions and collaborations to discuss the impact of radioisotopes released from civil and industrial sources on nuclear explosion monitoring in support of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). It aimed to unite experts from the radioisotope and nuclear explosion monitoring communities for a 4-day friendly scientific meeting with a technical tour provided at ANSTO on the final day of the workshop. Recognizing the immense importance of both radioisotope production and nuclear treaty monitoring, WOSMIP VII sought to bring together both communities to better understand the challenges created by radioisotope effluents released from the peaceful uses of nuclear technology and to propose methods to mitigate these emissions.

#### 1.2 Background

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), can detect seismic, hydroacoustic, infrasonic, and radionuclide signatures of nuclear explosions. Of the radionuclide signatures expected from a nuclear explosion, radioactive noble gas xenon isotopes (radioxenon) are 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 fission 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 from xenon releases during production. Radioxenon is also produced during the operation of nuclear power plants (NPPs) and research reactors, but in fission-based <sup>99</sup>Mo production, the process involves the dissolution of a uranium target shortly after irradiation and the near immediate release of radioactive noble gases. Therefore, 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.

WOSMIP enlists the help of the radioisotope and nuclear explosion monitoring communities to explore solutions to the challenges presented by the radioxenon emissions, while respecting the need for a stable supply of radioisotopes. The first WOSMIP was held in 2009, and focused on introducing both communities to one another, and facilitating future scientific collaborations. Since then subsequent workshops have become increasingly engaged in addressing the scientific problems facing the International Monitoring System (IMS). This includes establishing scientific boundaries; demonstrating observations of radioxenon emissions from MIP detected by the IMS; and sharing examples of emission mitigation technology being developed and used in real-world facilities to lower radioxenon releases. WOSMIP attendees have recognized the importance of

sharing stack release data, reducing emissions at the source, and identifying and investigating knowledge gaps in both nuclear explosion monitoring and emissions monitoring and abatement. At WOSMIP VI the workshop scope was extended to better understand all man-made sources of radioisotopes in the environment (NPPs, RRs, production, handling and use of medical isotopes, industrial isotopes, etc.) that have the potential to be observed by the CTBT verification regime. WOSMIP VII continues this important scientific work with an emphasis on background sources of man-made isotopes, research to reduce the radioxenon impact on the IMS of the CTBT, current and planned <sup>99</sup>Mo production activities, and radioxenon stack measurements.

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.

#### 1.3 Location

The venue for WOSMIP VII was at the Pullman Sydney Hyde Park Hotel in Sydney, Australia (Figure 1). Located opposite the iconic Hyde Park in the epicenter of the arts, retail and business precincts, the hotel provided an accessible and attractive workshop location. Additionally, this location provided convenient access to the ANSTO Nuclear Medicine (ANM) <sup>99</sup>Mo and Open Pool Australian Lightwater (OPAL) reactor for a tour on the final day of the workshop.





#### 1.4 WOZZIE award

At WOSMIP VII, Dr Anders Ringbom from the Swedish Defence Research Agency (FOI) received the prestigious 2018 WOZZIE Award (Figure 2). This was in recognition of his outstanding contributions to providing a better understanding of global radioxenon isotopic sources, in addition to his development of techniques and incredibly sensitive instrumentation for radioxenon detection.

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Figure 2. Dr Anders Ringbom (center) receiving the WOZZIE Award from Dr Jonathan Burnett (left) and Dr Emmy Hoffmann (right).

### 2.0 Session 1: Welcome and workshop overviews

#### 2.1 Welcome to WOSMIP VII

The workshop was opened by Dr Adrian Paterson, CEO of ANSTO, who expressed the need to understand the challenges facing the verification regime of the CTBT, by both producers and the monitoring community working together. ANSTO has been proactive within this area by collaborating with PNNL and installing a radioxenon stack monitoring system on its ANM facility. In doing so, ANSTO stated it hopes that it will encourage other producers to also become more involved. Once fully operational, the ANM facility could produce up to 30% of the current demand for <sup>99</sup>Mo. Global demand is expected to increase, and to meet this demand, collaborations are essential to ensure that nuclear explosion monitoring efforts are not negatively impacted. Utilizing the latest advances in technology is important to mitigate the impact of the plants, and this has been successfully implemented at the ANM facility. This has a lower emission footprint than the older ANSTO facility, despite a higher production capacity, due to more effective emission controls. Key to continuing this success is ensuring that the intellectual agenda develops, encouraging other producers to join ANSTO's work.

#### 2.2 WOSMIP VII kick-off

Emmy Hoffman (ANSTO) further welcomed participants to the workshop and provided an invitation to visit ANSTO and observe the radioxenon stack monitoring system, the OPAL reactor and ANM facilities.

#### 2.3 The history of WOSMIP

Paul Saey (IAEA) provided a welcome and overview of the history of WOSMIP. Medical and industrial isotopes are fundamental tools used in science, medicine and industry. The principal use is for medical diagnosis with approximately 30 million procedures per year and therapy with approximately 3 million procedures per year. The radioisotope <sup>99m</sup>Tc, the daughter of <sup>99</sup>Mo, is by far the most heavily utilized for a broad range of applications including assessing the function of the heart, liver, thyroid, blood flow, prostate, breast and for bone tumor detection. To produce <sup>99m</sup>Tc, it is necessary to produce <sup>99</sup>Mo, and the main production is reactor-based through the fission of <sup>235</sup>U. It is this process that releases fission gases such as radioactive xenon into the atmosphere (Figure 3). The basis for the interest in a better understanding of source terms associated with fission-based isotope production is that during <sup>235</sup>U fission in a nuclear reactor, thermal (slow) neutrons are used, whereas during a nuclear explosion the fission is induced by fast neutrons. There is little time for complex fission product build-up in a nuclear explosion (microseconds) – but there is sufficient time for production of many fission products during fission-based isotope production days) or in a nuclear power reactor (several months). These differences produce subtlety different radionuclide abundances.



Figure 3. A common technique for <sup>99</sup>Mo production.

WOSMIP aims to bring together two communities (isotope production and the nuclear explosion monitoring communities) to better understand each other's concerns. It aims to find solutions to the concerns by discussing the scientific issues, creating confidence building measures and developing solutions. It also aims to discuss ways to mitigate the effects of isotope production on the monitoring community without disrupting the supply of isotopes. Ultimately this venue aims to better understand the isotopic and chemical signatures created through isotope production mechanisms.

The first WOSMIP (WOSMIP I) was held on 1-3 July 2009 in Strassoldo, Italy and was attended by more than 70 professionals from the medical isotope production and the international monitoring communities from 16 countries. The workshop was hosted and organized by PNNL, and the impacts of medical isotope production on the IMS were discussed. The need to understand the emissions from the large amounts of medical isotopes produced was identified. WOSMIP I provided the first opportunity for the communities to come together to discuss the impacts their missions have on each other. It also provided a forum to foster communication and build a stronger collaboration with information sharing between scientists. The need to facilitate informal discussions was especially important. This resulted in a better understanding of the isotopic and chemical signatures created through isotope production mechanisms and the trace quantities that are detected in the environment. The findings of the workshop were published in the Journal of Environmental Radioactivity (Matthews, Bowyer et al. 2012).

The success of the first workshop continued with WOSMIP II, which was held on June 2011 in Strassoldo, Italy. Utilizing an increasing volume of monitoring data from around the world, specific measurement campaigns took place in close co-operation with several medical isotope producers including Belgium, South Africa and Indonesia. Based upon these observations, there was no

doubt on the influence of the emissions on the IMS and the seriousness of the issue was very clear. However, the Fukushima Daichi accident on 11 March 2011 confounded the issue of medical isotope production by introducing another set of source-term variables. WOSMIP III on June 2012 at Strassoldo, Italy continued the co-operation between the medical isotope production and monitoring community. The level of this interaction increased, with information exchange of some producers with the monitoring community. Some of the first substantial experiments related to emissions from isotope production were conducted by the Belgian Nuclear Research Centre (SCK-CEN) at the Institute for Radioelements (IRE). These collaborations continued at WOSMIP IV held on November 2013 in Vienna, Austria. This workshop helped facilitate the issues of medical isotope production to the Science and Technology Conference of the CTBT, where SCK-CEN received the European Star Award for their work on emission reduction. Five medical isotope producers also pledged to work towards reducing their emissions impact with the CTBTO.

In May 2015, WOSMIP V was held in Brussels, Belgium. The workshop was attended by 14 current or prospective medical isotope producers who shared detailed information on current and future medical isotope production. The discussions at this workshop focused on new developments in the IMS noble gas network and sensors, and the science of radioxenon emissions and atmospheric transport. Updates were also provided by medical isotope producers on production processes and facilities, and the technologies used to measure radioxenon stack releases. At request by the isotope production community, it was shown that a 5 x 10<sup>9</sup> Bq day<sup>-1</sup> release level from an MIP would not significantly interfere with monitoring when investigated using atmospheric transport modelling (ATM), and further that this level was near to the same value already being released by nuclear power reactors. Methods were also discussed for data sharing between the communities.

WOSMIP VI was held on November 2016 at San Carlos de Bariloche, Argentina. The workshop was attended by 81 participants from 27 countries. The discussions focused on alternative xenon sources, such as Nuclear Power Plants (NPPs), Research Reactors (RRs), isotope production and handling, and the use of medical and industrial isotopes. It included updates from current and prospective medical isotope producers on the use of nuclear facility stack emission data for verification purposes, and ATM. Production processes and facilities were also discussed, and research and development efforts toward radioxenon emission reduction.

#### 2.4 WOSMIP VII Overview

Ted Bowyer (PNNL) provided an overview of WOSMIP and its objectives. The workshop is designed to allow for open discussions of issues among the participants on factors affecting the airborne concentration of isotopes in the environment. It exists to encourage engagement between two distinct communities whose work overlaps and discuss creative solutions to mitigate the effect of emissions. It also provides benefits to participants, such as time to talk with peers on the issues of importance to them (e.g. during facility tours of INVAP and ANSTO). The CTBTO PrepCom continues to be interested in the emissions of isotope production, and policy discussions in Vienna have been useful to understand several views on this subject. However, the IMS detects the effects of emissions from isotope production essentially every day and this impact continues to be of concern. Focus areas for WOSMIP VII were also identified (Figure 4). This included new area of interest to the community, such as anomalous background detections (e.g. <sup>131</sup>I and <sup>106</sup>Ru detections), new producers on the horizon, emissions mitigation technology and developments in stack monitoring technology.



Figure 4. WOSMIP VII focus areas.

# 2.5 Updated worldwide background of CTBT relevant xenon isotopes based on IMS data and mobile systems

Tammy Taylor (CTBTO) summarized the analysis results of the noble gas (NG) data based on 6 years of International Data Centre (IDC) operations which included 25 certified IMS NG systems. Notable achievements were discussed, including the implementation of a NG categorization scheme into IDC operations in August 2012, and the inclusion of <sup>131m</sup>Xe for independently triggering Level C categorizations in January 2014. An important highlight was the release of NDC-in-a-Box 3.0 in May 2018 which has integrated NG categorization and product generation. Ongoing IDC activities include the development of Integrated Software Platform for Interactive Radionuclide Review (Inspire) with promotion to IDC operations in 2019. The acceptance testing of Next Generation NG systems is also continuing, including the Sauna III in Stockholm, Sweden and the Spalax NG in Paris, France. These systems were successfully configured in the IDC testbed, and data is automatically processed on a daily basis.

The number of samples reviewed by the IDC has increased since 2011 (Figure 5). This has included the review of over 80,000 samples with radioxenon detections observed in 47% of IMS samples. Furthermore, both detection rate and activity concentration distribution vary across a wide range depending on the IMS site location. The highest frequencies are observed at IMS sites under the influence of medical isotope production facilities (MIPFs) and NPPs (Figure 6). Examples include USX75 (MIPF at Chalk River, Canada), SEX63 and DEX33 (IRE MIPF at Fleurus, Belgium), AUX04 and NZX46 (ANSTO MIPF at Sydney, Australia) and JPX38 (local and regional NPPs). The shutdown of medical isotope production at Chalk River from November 2016 has been reflected by the decline in IMS detections at USX75 and CAX17.

Activity concentrations due to nuclear explosions may fall within the same range (or lower) than emissions from radiopharmaceutical facilities with a similar isotopic ratio. The samples that are categorized as of interest (Level C) account for 4% of samples (approximately 1 sample day<sup>-1</sup>) which is still a considerable number of samples from a verification perspective. Reducing

radioxenon emissions from medical isotope production facilities in the world is expected to mitigate the impact of noble gas background at IMS sites. This includes measuring released activities at the stack level and sharing the data with the CTBTO monitoring community. This is expected to enhance the common understanding in terms of impact on IMS noble gas systems.



Figure 5. Reviewed NG samples per month in IDC operations.



Figure 6. Detection rate (Levels B + C, %) from Sep 2012 to Aug 2018.

#### 2.6 Status of the noble gas capability at the CTBTO International Monitoring System

Nikolaus Hermanspahn (CTBTO) provided an overview of the IMS network and its role for verification of the CTBT. The network's radionuclide monitoring segment is unprecedented in its combination of global coverage, detection sensitivity, network density and temporal resolution. Of the 80 IMS radionuclide stations, which measure aerosol borne radionuclides, 40 stations will also be equipped for the measurement of radioxenon isotopes. Radioxenon monitoring systems are a key component of the IMS, because radioxenon isotopes are of particular interest for monitoring for underground nuclear explosions. The radioxenon monitoring systems have been specifically developed for the IMS. In the past few years, developers have been working on the evolution of the next generation IMS NG systems, with focus on improvements to performance and reliability. Such prototypes are now under evaluation by the PTS in preparation for their future use in the network.

### 2.7 Session photographs







#### PNNL-28870



### 3.0 Session 2: Background sources

# 3.1 How can CTBTO NG stations be impacted by radioxenon releases from an isotope production facility

Martin Kalinowski (CTBTO) discussed the impacts of MIP on the NG measurements made by the IMS. The monitoring of atmospheric radioxenon is a key aspect of the unambiguous identification of an underground nuclear explosion by the CTBTO. However, the detection capability of the CTBTO's NG network is affected by a radioxenon background produced by nuclear civil activities. MIPFs have been identified in the past as contributors to the radioxenon background. Despite their very limited number, the releases during normal routine operations can be up to several orders of magnitude above those attributed to other civil nuclear facilities. Understanding emissions from these facilities would help interpreting the data collected at the noble gas stations.

The objective of this work is to discuss the analysis results of a large data set collected at the BATEK MIPF located in Indonesia, during a joint CTBTO – BATAN – PNNL collaboration supported by EU and US voluntary contributions. For a 1-year period, about 51,000 near real-time stack measurement spectra were collected using an on-line LaBr<sub>3</sub> stack monitor system at the BATEK facility. Using this data, the temporal evolution of the estimated release was measured. This utilized a similar technique as that used for the mobile EUX10 system, which includes reconstruction of the source term using ATM and comparing the source term with the emission profile. Using this technique, two appropriate periods of time were chosen to calculate dilution (scaling) factors and these were then applied to the entire BATEK dataset (Figure 7 and Figure 8).



Figure 7. Estimated emissions during 22-31 May 2013.





This approach gave high confidence in the reconstructed source term and high confidence in the analysis of the stack measurement data. The fluctuations in the reconstructed source term were deemed significant and well correlated with observed <sup>133</sup>Xe concentrations at the mobile system. The measurement campaign affords a unique opportunity to assess how the CTBTO NG stations can be impacted by the radioxenon releases from a MIPF.

# 3.2 An overview of global atmospheric radioxenon background simulation studies

Sylvia Generoso (CEA) discussed the need to improve the understanding of the radioxenon background to facilitate the analysis of IMS data by National Data Centers (NDCs). The lack of background information makes it complex to apply numerical methods for source localization, as events of interest might not be clearly identified (for example, "what is a detection and what is a non-detection in a context of quasi-daily <sup>133</sup>Xe signal?"). Characterization of the radioxenon atmospheric background has thus become crucial for analysis of IMS detections. This has been undertaken using global atmospheric transport simulations which show that radioxenon levels are explained by variable combinations of multiple sources. This provides an update of the <sup>133</sup>Xe global background modelled during 2007-2009 (Achim, Generoso et al. 2016). The update was provided using data from 2013-2014 (Figure 9) and included a <sup>133</sup>Xe source release of 51 TBq day<sup>-1</sup>, calculated from the emissions of 9 MIPFs and 178 NPPs (383 reactors). A total of 65% of the release was in the Northern Hemisphere. The results showed large spatial, seasonal and daily variability (Generoso, Achim et al. 2018).



Figure 9. Simulated map of <sup>133</sup>Xe atmospheric background during 2013-2014.

Daily occurrences of <sup>133</sup>Xe detections show the existence of a significant <sup>133</sup>Xe background. It is important to understand this to allow discrimination of the origin of the detections. One way of doing this is through numerical simulation, which includes real-time automatic simulations for the assessment of <sup>133</sup>Xe levels. These simulations are performed using 4-day forecasts with accounting of each day for the past 30 days of industrial releases worldwide. More than 500 simulations are achieved per day using a High Performance Computing (HPC) cluster for this task. These background simulations can be subtracted from IMS measurements to enable more accurate measurements in support of the CTBT.

# 3.3 Investigation of the radioxenon background trend at noble gas IMS systems

Christophe Gueibe (SCK-CEN) discussed observed trends in the global radioxenon background using IMS data. The noble gas component of the IMS has considerably grown and improved since the certification of the first noble gas system (i.e., USX75 - Charlottesville, Virginia in 2010). Since then, additional NG systems have been certified in the IMS and are continuously sampling and measuring radioxenon in the atmosphere. During the operation of these systems, a worldwide radioxenon, especially <sup>133</sup>Xe, background, resulting from civilian nuclear applications, has been observed. This worldwide civilian radioxenon background makes the CTBT verification work of national data centers (NDCs) very challenging. In some regions of the world, the NG systems such as GBX66 (BIOT), the number of detections has decreased due to unknown reasons. A reduction has been observed at CAX17 (St. John's) but this is due to the Chalk River MIPF closing.

The high radioxenon background is blurring and limiting the detection capability for CTBT related events of the NG component of the IMS. It is thus crucial to understand the radioxenon background observed by each NG system as to allow a better discrimination between potentially CTBT related detections and the detections from the civilian background. The trend in radioxenon detections at certified NG systems was investigated over a period from 3 up to 7 years, depending on the data availability of the NG system. This was used for identifying detection increases or decreases at each station. Multiple sources could explain some detections, including those at MNX45 (Ulaanbaatar). Potential further investigations could include coupling different IMS stations for the ATM investigation.

# 3.4 ATM analysis results on radioxenons detected in Japan by JAEA/NDC

Yuichi Kijima (JAEA) discussed ATM analysis results of radioxenon measurements made using the Transportable Xenon Laboratories (TXLs). These were deployed by JAEA, in collaboration with the CTBTO, for the purpose of strengthening the IMS detection capability for nuclear tests. Two TXLs were installed at Horonobe, Hokkaido and Mutsu, Aomori, and radioxenon measurements commenced in 2018. These additional stations increase the measurements of radioxenon in Japan for Treaty verification purposes to a total of 3 locations (Horonobe TXL, Mutsu TXL and IMS Takasaki station). The <sup>133</sup>Xe isotopes with activity concentration above the background level were detected at Horonobe, Mutsu and Takasaki in the end of April 2018 (Figure 10). These detections were at a similar concentration and timing. Since there was the possibility that these isotopes were released from the same location, the estimation of emission source was conducted by ATM simulation. The nuclear facilities at the Democratic People's Republic of Korea (DPRK), South Korea and some regions of China were included as possible source regions. Assuming Nyeongbyeon (DPRK), Hongyanhe (China) or Kori (South Korea) are the emission source, there are not strong correlations between the calculated values and the observed values at Horonobe, Mutsu and Takasaki. It is therefore possible that the <sup>133</sup>Xe isotopes detected at the 3 sampling locations are released from a different location. Additional ATM analysis was performed using <sup>135</sup>Xe and did not provide any conclusive evidence of the potential source location, and further investigation is recommended.





# 3.5 Observations of radioxenon emissions from BWRs compared to stack data

Anders Ringbom (FOI) discussed correlations between the released radioxenon activity from the Forsmark nuclear power plant, Sweden, and the observed activity concentrations in Stockholm, Sweden. Whilst there are many weak sources of radioxenon, they are numerous and have potential to emit sizeable amounts of radioxenon. Operational signatures are also typically

different from a fresh nuclear explosion (although the emissions from a nuclear explosion can also be quite delayed). Perhaps surprisingly there are many exceptions from typical reactor operations, which means there is a need to know as much as possible about reactor emissions. There is also wide variation in yearly reported releases, with <sup>133</sup>Xe NPP releases in the range of 10<sup>9</sup> to 10<sup>14</sup> Bg (Kalinowski and Tuma 2009). For nuclear power plants (NPPs) there are many reactor and fuel types (e.g., pressurized water, boiling water, (PWR, BWR), and Magnox) with large similarities in radioxenon ratios. However, some differences may become important in special cases. In Sweden there are 8 operating reactors at 3 locations (Ringhals, Oskarshamn and Forsmark) generating a total of 8206 MW. The Forsmark NPP is closest to SEX63 (Stockholm) SAUNA with 3 BWRs and has resulted in many detections over the years. The facility has provided stack release data that can be compared with the SEX63 measurements. The detected <sup>135</sup>Xe/<sup>133</sup>Xe ratios agree well with modelled results. However, decay times through the Forsmark mitigation system must be taken into account in the data interpretation. Most <sup>133m</sup>Xe/<sup>133</sup>Xe ratios agree with models, but there are occasional observations that are much higher than expected (ratios of approximately 0.1) indicating fresh fission separated relatively quickly (Figure 11). A few high <sup>131m</sup>Xe/<sup>133</sup>Xe ratios are also observed.



Figure 11. Forsmark NPP Reactor 3 shutdown/startup in Mar-Apr 2017.

Measurements of radioxenon at SEX63, and a FOR-analysis associates almost all the <sup>133m</sup>Xe, <sup>133</sup>Xe and <sup>135</sup>Xe plumes to emissions from the Forsmark facility. The isotope ratios fit well with reactor shut-down/start-up scenarios, although there could be cases with unexplained high ratios from NPPs also in this case. The isotopic ratios are compatible with models of reactor cycling, with the exception of <sup>133m</sup>Xe/<sup>133</sup>Xe ratios, which sometimes are higher than expected. The results show that more refined models of reactor operation are needed in order to explain the observations. It is recommended that the data be further analyzed using more stack-data, as well as refined ATM-modelling. The results show the power of the next generation NG systems, resulting in an increased range of measurable isotopic ratios, as well as a higher number of

observations for a given time period. Furthermore, it is crucial to have good communication with the NPPs in order to gain more insight into radioxenon observations in IMS.

# 3.6 Research reactor contributions to radioxenon atmospheric inventories

Steve Biegalski (Georgia Tech) discussed the contributions of research reactors (RRs) to the global radioxenon background. There are many designs of RRs around the world, and radioxenon emissions are more from air activation than from fission due to neutron beam lines and other irradiation facilities. The High Flux Isotope Reactor (HFIR), for example, fits on the air activation decay line for radioxenon ratio measurements. Air samples were collected from the University of Texas TRIGA reactor at 6 locations and radioxenon measured using a SAUNA beta-gamma system and <sup>41</sup>Ar measured using an ultra-low background detection system (Table 1). Calculations using these measurements indicate that typical RRs emit a total of in excess of  $10^{11}$  Bq year<sup>-1</sup>for all isotopes combined. This is based on an assumption that release rates form all RRs are similar to TRIGA reactors. The main pathway for <sup>133</sup>Xe production and emission is via air activation in the reactor pool. Many RRs do not have an open pool design, so this calculation should be considered conservative. These calculations predict that the world's entire RR fleet produces below the threshold of 5 x  $10^9$  Bq d<sup>-1</sup> proposed elsewhere (Bowyer, Kephart et al. 2013).

Annual Release	Commonto
(Bd λL-1)	Comments
7.67E+06	Based on Measurement
8.16E+01	Normalized to <sup>37</sup> Ar
6.23E+10	Based on Measurement
2.07E+07	Normalized to <sup>133</sup> Xe
6.76E+00	Normalized to <sup>133</sup> Xe
4.21E+04	Normalized to <sup>133</sup> Xe
1.52E+09	Normalized to <sup>133</sup> Xe
1.53E+08	Normalized to <sup>133</sup> Xe
7.15E+07	Normalized to <sup>133</sup> Xe
2.38E+04	Normalized to <sup>133</sup> Xe
4.88E+06	Normalized to <sup>133</sup> Xe
1.30E+06	Normalized to 133Xe
3.36E+06	Based on Measurement
5.52E+05	Normalized to <sup>133</sup> Xe
9.03E+06	Normalized to <sup>133</sup> Xe
2.63E+05	Normalized to <sup>133</sup> Xe
7.41E+03	Normalized to 133Xe
	(Bq yr-1) 7.67E+06 8.16E+01 6.23E+10 2.07E+07 6.76E+00 4.21E+04 1.52E+09 1.53E+08 7.15E+07 2.38E+04 4.88E+06 1.30E+06 3.36E+06 5.52E+05 9.03E+06 2.63E+05

# Table 1. Extrapolated annual University of Texas TRIGA reactor releases based on measurements.

# 3.7 Physical characterization of filters from German and Sweden radiological monitoring networks with ruthenium-106 from 2017

Ian Hoffman (Heath Canada) presented research that aimed to characterize air filter samples from Eastern and Western Europe containing ruthenium isotopes. Such samples were detected in late September to early October 2017 by several national radiation surveillance networks in Eastern and Western Europe. These reported low levels of the airborne radionuclide contaminants <sup>106</sup>Ru and <sup>103</sup>Ru. Filters from the national networks of Sweden operated by FOI and of Germany operated by the German Meteorological Services (DWD) containing <sup>106</sup>Ru were characterized (Figure 12). This included using coincident high-resolution gamma spectroscopy, electron microscopy, autoradiography, and species solubility. Gamma-spectrometry indicated that the sample was very radiopure, with a ruthenium activity 100 to 10.000 times greater than other radionuclides considered. Electron microscopy and autoradiography indicated the ruthenium was highly dispersed in particulates likely less than one micron in size. Solubilization studies discount the possibility of ruthenium existing as tetroxide (RuO<sub>4</sub>) and demonstrated that 50 to 60% of the species present can be extracted in polar solvents including water. It was concluded that the ruthenium material was highly processed and radiopure. The ruthenium on the filter has likely had multiple species given the solubility and reaction results observed, with no  $RuO_4$  on the filter.  $RuO_2$  is a possible candidate for the insoluble, unreactive ruthenium forms on the filter. The original form was not likely pure RuO<sub>2</sub> as it is relatively inert and would not have readily converted into the different groups of species on the filter. Nanometer scale dispersion also would be difficult if it was dispersed in this pure form.



#### Figure 12. An air filter supplied by FOI.

Ruthenium metal would be difficult to disperse without high temperature oxidation through  $RuO_4$ . Pure  $RuO_4$  is very reactive but would not likely form the range of species on the filter without a relatively concentrated chemical environment. In a dilute environment, it would more likely convert to  $RuO_2$ . This cannot explain the soluble/reactive species on the filter but could lead to highly dispersed  $RuO_2$ .  $RuO_4$  is a common by-product in subsequent stages of purification in fuel reprocessing. It has a high vapor pressure and is readily dispersed. If the dispersal event included dispersion of other concentrated reagents and/or reaction products of  $RuO_4$ , it could produce a mixture of species released from the event site and at high dispersion on a final filter. No  $RuO_4$  would be present on the filter in this case but  $RuO_2$  product could be present as an insoluble portion and other species would be present.

# 3.8 European monitoring of <sup>106</sup>Ru airborne concentrations in Fall 2017

Olivier Masson (IRSN) presented a discussion of the ruthenium detection event from 2017. A study of field observations suggested a short release duration. After correction of the sampling duration the highest activity level integrated is no longer in Romania and 7-day averaging indicate a similar range from central Europe to Urals. Back-trajectories ending in Romania indicate that the air mass had travelled very close to the Mayak complex in Russia between 25-26 September 2017. A data inversion technique makes it possible to locate the <sup>106</sup>Ru release area in the South Urals and assess the source term to a few hundred TBq (Figure 13). At this stage, the link with the Mayak complex had not been fully established or recognized. However, this is the only facility that was commissioned to produce a mega source of <sup>144</sup>Ce whose production is accompanied with the collateral production of <sup>106</sup>Ru.



Figure 13. Location of the ruthenium release term.

#### 3.9 Isotopic ratios for <sup>103</sup>Ru/<sup>106</sup>Ru

Jonathan Burnett (PNNL) discussed high-precision gamma-spectrometry measurements of  $^{103}$ Ru/ $^{106}$ Ru performed on an air filter sample supplied by FOI. During 30 September to 23 October 2017 there were 38 detections of  $^{106}$ Ru at 9 IMS stations. All of the detections made by the IMS were categorized by the IDC as level 4 ( $^{106}$ Ru) with the exception of one level 5 which contained both  $^{106}$ Ru and  $^{103}$ Ru. Detections were also made in the non-IMS samples of approximately 30 countries. This included the national monitoring network of Sweden, and a sample from 2 October 2017 was sent to PNNL for gamma-spectrometry. Very high-sensitivity analysis was performed using a multidimensional gamma-spectrometer (the Advanced Radionuclide Gamma SpectrOmeter or ARGO) located in the Shallow Underground Laboratory (SUL) at PNNL. The system is optimized for trace-level measurements of fission and activation products, and includes low-background components, dual high-purity germanium (HPGe) detectors, segmented Compton suppression and a cosmic veto background subtraction system. It provides improved sensitivity for CTBT relevant radionuclides by a factor > 20. The instrument provided an extremely accurate  $^{103}$ Ru/ $^{106}$ Ru ratio of 2.5 x 10<sup>-4</sup> ± 4.5 x 10<sup>-5</sup> which indicates the age of the material is 570-670 d (Figure 14).



#### Figure 14. Age dating the ruthenium sample.

In addition to the ruthenium detections, there have been numerous other detections where multiple radionuclides have been detected. In 2017 there were 259 level 4 events at IMS stations, which is a significant increase from the 147 level 4 events reported in 2016. This included fission (<sup>106</sup>Ru, <sup>131</sup>I, <sup>99m</sup>Tc, <sup>137</sup>Cs) and activation (<sup>60</sup>Co, <sup>54</sup>Mn, <sup>24</sup>Na) products. Many of these are difficult to explain in terms of what caused them and where the release originated from. They have the potential to mask other relevant detections (i.e. releases from underground nuclear explosive tests) and are analogous to radioxenon releases from medical isotope production. There is a need to investigate and understand the impact of these increasing detections.

# 3.10 Using machine learning to estimate atmospheric sources from the 2017 ruthenium detections across Europe

Lee Glascoe (LLNL) applied source inversion techniques to the <sup>106</sup>Ru detections occurring across Central and Eastern Europe in the fall of 2017. Bayesian inverse methods are ideally suited for such data-rich problems providing a probabilistic source characterization. Machine learning techniques can be used to accelerate the Bayesian analysis through the training of the error and correlation between forward atmospheric transport modelling and observations. This includes using a Monte Carlo design model with 5184 transport simulations and 6 uncertain source parameters (latitude, longitude, start time, duration, height, amount) to compare predicted and observed release concentrations. The machine-learned Bayesian inversion of the 2017 <sup>106</sup>Ru detects, highlight an efficient process to maximize likelihood. Likely <sup>106</sup>Ru sources match the timing and amplitude of detections and Bayesian analysis was used to provide a maximum likelihood <sup>106</sup>Ru source location probability map (Figure 15). The analysis suggests that 0.1-3.0 g of <sup>106</sup>Ru was released shortly after 25 September in a localized region of Central Asia.



Figure 15. A source location probability map for the <sup>106</sup>Ru detections.

# 3.11 Contribution to NPP emissions to the <sup>133</sup>Xe measurements at the remote IMS stations

Jolanta Kuśmierczyk-Michulec (CTBTO) discussed a study of the effect of NPP emissions on the <sup>133</sup>Xe observations at two remote IMS stations at Wake Island (USX77) and Hawaii (USX79) using ATM results. The study used data from June to September 2014. During that period, an initial study based on operations source receptor sensitivity files indicated none of the major known medical facilities contributed to the measurements of <sup>133</sup>Xe at USX77 and USX79. Thus it is hypothesized that the observed detections can be associated with emissions from NPPs. To validate this hypothesis, the study compared modelled NPP releases with detections at the stations. The results of quantitative approach demonstrate that for the cumulative monthly values, the simulated values are lower than the measured values, approximately by one order of magnitude. The qualitative study done for one NPP (Figure 16), Diablo Canyon, indicates that the possible explanations for this difference in modelled and observed results, is that the assumption of continuous operation (instead of pulses) leads to such discrepancies. It is also possible that there are other unknown sources not included in this study.

#### PNNL-28870



Figure 16. Modelled plume releases from NPP Diablo Canyon.

#### PNNL-28870

### 3.12 Session photographs










# 4.0 Session 3: Research to reduce the radioxenon impact of the IMS

### 4.1 Overview of the CRP

Tom Hanlon (IAEA) discussed a Coordinated Research Project (CRP) aimed at sharing and developing protocols to further minimize radioactive gaseous releases to the environment in the manufacture of medical radioisotopes. The International Atomic Energy Agency (IAEA) <sup>99</sup>Mo activities include interactions with RRs, MIPFs, fundamental physics research and nuclear medicine. Related activities include the stability of <sup>99</sup>Mo supply in member states, conversion from highly enriched uranium (HEU) to low enriched uranium (LEU) targets and indigenous production using non-HEU targets. Other related activities include small-scale production of <sup>99</sup>Mo or <sup>99m</sup>Tc for local use and associated regulatory aspects and new alternatives to <sup>99m</sup>Tc radiopharmaceuticals. The CRP request was received from Australia, Belgium, the Netherlands, Republic of Korea and the USA in May 2014. The request proposed that the IAEA initiate a CRP on the topic of technologies to reduce emissions from MIPFs. The CRP is aimed at identifying important technical issues and is open to member states. It aims to identify the steps and factors of the MIP process that need proper gaseous emissions that could mitigate the emissions. Additionally, it aims to evaluate and develop stack measurement systems for radioactive gases and explore data exchange methodologies and formats. To support the CRP, the first research coordination meeting was held on 17-21 August 2015 in Vienna, Austria with 8 participants and 9 observers in addition to IAEA and mission staff. The second research coordination meeting was held on 6-10 March 2017 in Vienna, Austria with 7 participants and 5 observers, plus IAEA and mission staff. Both meetings aimed to define a work plan and facilitate collaborations. The final report was drafted at the third research coordination meeting on 12-16 November 2018 in Vienna, Austria, with 6 participants and 3 observers.

Under the CRP, agreements have been established with Belgium (SCK-CEN), Canada (Radiation Protection Bureau, Health Canada), Germany (University of Hamburg) and USA (PNNL). Contracts have also been established with Indonesia (PT INUKI), Poland (National Centre for Nuclear Research) and Republic of Korea (KAERI). A workplan has been developed that includes the following:

- Atmospheric transport modelling
- Xenon mitigation
- Stack monitoring data characterization (pre- and post-shutdown of <sup>99</sup>Mo production at Chalk River, Canada)
- Development of NG adsorbent materials: silver-doped zeolite
- Development of a compact adsorption module for xenon mitigation research
- Adsorption of radioxenon on organic and inorganic adsorbents
- Comparison of various adsorbents, including charcoal and silver-doped zeolite for the efficiency of <sup>131</sup>I absorption.
- Higher fidelity stack monitoring technologies for voluntary incorporation
- Assist in identification and incorporation of emissions control in facility design

The final report produced by the CRP recommends that the cooperation between the IAEA and CTBTO is productive and should be continued. The work performed during the CRP should also be shared and made accessible to Member States beyond those involved in the CRP. New absorption materials and mitigation technologies should be further explored, in combination with modelling adsorption/desorption cycles to allow cost and space efficient systems for reducing emissions, potentially in a new CRP. There should be further study of global MIP which can lead to potential releases impacting Treaty verification (e.g. radioxenon & iodine). Collection and sharing of data should also be encouraged, including datasets from stack monitoring and environmental monitoring, and atmospheric dispersion and transport modelling. A study on the worldwide background levels of <sup>131</sup>I should also be undertaken.

## 4.2 Sharing experience on mitigating radioxenon through preparation of design studies for three civilian nuclear facilities

Christophe Gueibe (SCK-CEN) presented research on radioxenon emission mitigation commissioning through the EU Council Decision V Project. The monitoring capabilities of the IMS NG systems could be improved by substantial reductions of radioxenon emissions. If these reductions are achieved at nuclear facilities that have considerable radioxenon releases in normal operation, this could significantly enhance the CTBT verification capability. In this framework, the SCK-CEN was contracted by the CTBTO under the EU Council Decision V project to realize a technical solution for the reduction of radioxenon emissions from civilian nuclear facilities. The project was divided into 3 tasks:

- Task 1: Scaling-up of the prototype system and long-term behavior analysis
- Task 2: Design studies at additional facilities
- Task 3: Stack release data monitoring

The main result of the project was the development and construction of a xenon retention prototype system. In order to share the experience accumulated during the previous study, the SCK-CEN was contracted under the EU Council Decision VI project to perform design studies at other civilian nuclear facilities. As part of Task 1, a call for voluntary applications was issued to more than 70 facilities worldwide. Three facilities were selected by the SCK-CEN, based on predetermined criteria. This included ANSTO (Australia), Nordion inc. (Canada) and PT INUKI (Indonesia). For Task 2, at each location there was a technical visit to the facility and a design study undertaken for emission reduction. The results of the design studies were delivered to each facility and included detailed calculations and analysis which could be used by the facility to develop or improve their radioxenon mitigation means. At Nordion this included a mitigation system based upon the NUSORB GXK and Ag-ETS-10 sorbents. At ANSTO it included a xenon trapping column based on the Ag-ETS-10 material that could be applied to the new ANM facility (Figure 17). Two generic design studies were performed at INUKI as identification of xenon releases could not be performed (stack monitoring data was not available). These utilized the NUSORB GXK and Ag-ETS-10 sorbents and were for prompt and continuous release scenarios. At the end of the project, the final design studies were donated to the facilities for further use and implementation. It was observed that the most efficient radioxenon trapping can only occur in well controlled conditions and close to the emission source.





## 4.3 Development of radioxenon abatement system for the fission <sup>99</sup>Mo production process

Suseung Lee (KAERI) presented research on the LEU-based fission <sup>99</sup>Mo production process that has been undertaken in preparation of the new research reactor that is scheduled to be built in Kijang, Busan, Korea. As radioxenon emissions from <sup>99</sup>Mo production have potential to interfere with nuclear explosion monitoring activities, KAERI pledged to CTBTO in 2013 that its radioxenon emissions from the planned facility will be under the 5 GBq day<sup>-1</sup> level as suggested (Bowyer, Kephart et al. 2013). To mitigate the radioxenon emissions, a lab-scale delay bed module that can control the bed temperature was fabricated and the performance was evaluated at varied bed temperatures. Iterations of the design involved the removal of radioxenon decay tanks, in preference of a direct route to a radioxenon delay bed. The absorption efficiency of the bed was shown to be improved by operating a low bed temperature, which increases the adsorption efficiency of the charcoal (Figure 18). Further improvements in radioxenon retention were investigated by designing and fabricating a system that reduces the bed temperature to 165K. This system would be suitable for processing ~ 3,000 Ci per process, with 6 productions per week, and compact enough to be installed into a hot cell.





## 4.4 Testing and validation of a radioxenon trap system under the EU Council Decision VI Project: results and outcomes

Dominique Moyaux (IRE) discussed testing and validation of a radioxenon mitigation system developed in collaboration with SCK-CEN and CTBTO as part of the EU Council Decision V project. This aimed to provide a technical solution for the reduction of radioxenon emissions from civil nuclear facilities such as IRE. The prototype used 3 columns of Ag-ETS10 zeolite material, and preliminary validation was performed at the SCK-CEN laboratory. The column configuration was deemed to be sufficient for 20 days usage or to ensure xenon adsorption from 8 or 9 successive production runs. During the implementation period, the prototype system was integrated into the LEU process line and validated through the purge of <sup>133</sup>Xe vials to establish breakthrough curves. The prototype was then assessed under different operating conditions. The influences of several major parameters (e.g. xenon mass, activity, carrier gas, adsorption competitions with other gases, etc.) on the Ag-ETS10 adsorption capacities were also investigated. In each instance, the dissolved uranium target was bubbled with an inert carrier gas (He or Ar) to carry out the noble gases, and the activity detected before and after the mitigation system using Geiger-Muller detectors. The order of the release was krypton followed by xenon. Repeatability tests were also performed using LEU runs which demonstrated high xenon trapping on the columns (Figure 19). Observed results were compared to the modelled retention, and were not always in agreement. This could indicate some co-adsorption of gases produced during dissolution. For routine usage, it is recommended that the system be combined with decay storage tanks to maximize the radioxenon mitigation.

Run #	Col #	Xe-133 Activity (Ci)	Xenon trapping (%)	Prototype Time (h)
1	3	3575	99.73	0h59
2	2	4080	95.95	0h47
3	2	3003	94.06	0h35
4	2	2950	96.04	0h47
5	2	2950	91.82	0h42
6	1	4390	99.08	1h11
7	3	4390	99.04	0h50
8	1	4390	98.95	1h12
9	1	2213	99.32	0h50
10	3	2212	99.29	0h50
11	1	2862	99.51	0h52

Figure 19. Prototype assessment under real dissolution conditions.

## 4.5 Verification and validation of atmospheric transport models for nuclear security with the University of Florida Training Reactor

Andreas Enqvist (UF) presented a study using the University of Florida Training Reactor (UFTR). This is a 100-kW ARGONAUT type reactor with a unique design such that a large volume of air passes through the core, producing and releasing significant quantities of <sup>41</sup>Ar. Once released, the radioargon is dispersed throughout the atmosphere and decays by a 1294-keV gamma-ray emission. The radioargon has similar properties to other NGs of high importance in nuclear security for tracking peaceful or clandestine nuclear activities such as reprocessing, nuclear events and nuclear weapons testing. When operating at full power the UFTR releases 15 MBq s<sup>-1</sup> of <sup>41</sup>Ar through two plume stacks. The UFTR can utilize this quantified, validated, and controlled <sup>41</sup>Ar plume to verify and validate atmospheric transport models (ATMs). This is accomplished by coupling an ATM with MCNP and comparing the results to experimental measurements. For this study, an in-house developed Gaussian dispersion model and the EPA approved Gaussian model AERMOD were coupled with MCNP to simulate the results of a six-hour measurement taken on May 8th, 2018 (Figure 20).





In-house model at 80 m elevation



AERMOD model at 80 m elevation

Figure 20. Comparisons between in-house and AEROMOD atmospheric transport models of <sup>41</sup>Ar releases from the UFTR.

For the measurements and simulations, a grid of seven Nal detectors were placed at four different rooftop locations near the UFTR plume stack. With easterly winds dominating during the morning and early afternoon, both the measurement results and simulations displayed a higher <sup>41</sup>Ar photon count rate on the detectors west of the plume stack throughout the day. Overall, the AERMOD simulations produced better results when compared to the in-house model, particularly for the detectors placed east and west of the plume stack. While some of the AERMOD data showed a strong correlation with the measurements, the results are still inconsistent. Due to the varying accuracy of the simulations results, it is theorized that the relationship between measurement results and simulations will improve with the use of localized weather information or the utilization of non-Gaussian dispersion models. Additionally the UFTR was utilized in a joint campaign with PNNL and the Remote Sensing Laboratory (RSL), where a helicopter equipped with large volume Nal detectors was used to map radioargon flows from the reactor.

## 4.6 Session photographs











# 5.0 Session 4: Molybdenum-99 production overview: Recent and planned activities at isotope production facilities

## 5.1 Radioisotopes used in nuclear pharmacy and nuclear medicine practice: Perspectives from the United States

Neil Petry (Duke University) discussed the field of nuclear pharmacy, which is a specialty practice of pharmacy that seeks to improve and promote health through the safe and effective use of radiopharmaceuticals (radioactive drugs) for diagnosis and therapy. In the United States, nuclear pharmacy facilities and nuclear medicine clinics are very common and spread geographically throughout the nation. These is an estimated 1,000 nuclear pharmacists practicing in the US. Most of the facilities and clinics are in the large cities and in the surrounding metropolitan areas (Figure 21). As in other parts of the world, nuclear pharmacy and nuclear medicine practice routinely involves the handling and clinical use of many unique radiopharmaceuticals; manufactured compounds containing radioisotopes formulated and intended for human use in the diagnosis and treatment of common and rare diseases. Radiopharmaceutical drug products are produced in sophisticated highly engineered manufacturing facilities specifically designed to yield safe radioactive drug products but also to minimize occupational radiation exposure and prevent the release of radioisotopes into the local and distant environment. Consequently, these manufacturing facilities are highly controlled at the federal, state and local levels by broad scope regulations, which include stringent limits on the release of radioisotopes by any route into the environment.



#### Figure 21. Nuclear pharmacies in the US.

These radiopharmaceutical production facilities are few in number, yet located geographically where radioisotope releases, under the right set of conditions, could potentially produce aberrant nuclear monitoring data patterns at nearby monitoring stations. Most often manufactured radiopharmaceutical distribution is by ground or air transport to nuclear pharmacy facilities that prepare and then ground deliver prescribed patient specific unit dosages to nuclear medicine

clinics. A hospital based nuclear pharmacy facility requires several operational features and controls necessary to promote the safe handling of radiopharmaceuticals and to prevent the release of medical radioisotopes into the local and global external environment. The radioxenon isotopes <sup>127</sup>Xe, <sup>133</sup>Xe and <sup>135</sup>Xe are used as a diagnostic marker for lung function (ventilation and perfusion) and blood flow (cerebral, myocardial, renal, peripheral and liver). The gases are administered by inhalation or by injection. Since the 1950s, lung ventilation studies have used <sup>133</sup>Xe (<sup>127</sup>Xe is no longer used). As known to the nuclear monitoring community, <sup>133</sup>Xe is also a primary marker for nuclear explosion detection. Therefore, there is some potential concern medical use releases of <sup>133</sup>Xe and perhaps other radioisotopes from nuclear medicine procedures could be a problem for the nuclear explosion monitoring community. There is thus a concern that <sup>133</sup>Xe and other medical radioisotopes could be released in into the environment in significant amounts and thus yield aberrant nuclear monitoring data patterns at selected monitoring stations located in geographic proximity to multiple nuclear pharmacy and / or nuclear medicine facilities.

### 5.2 <sup>99</sup>Mo production overview

Tom Hanlon (IAEA) provided an overview of <sup>99</sup>Mo production and future challenges to meet supply requirements. Technetium-99m (99mTc), the daughter of molybdenum-99 (99Mo), is the most widely used radioisotope in diagnostic nuclear medicine. It is used in approximately 80% of all nuclear medicine procedures worldwide. Following shortages in 2009-2010, the international <sup>99</sup>Mo community has been working to ensure a long-term reliable supply of this critical radioisotope for the medical community. Significant changes in the industry are occurring at this time; including conversion of <sup>99</sup>Mo production targets from HEU to LEU, cessation of production at Canada's National Research Universal (NRU) RR, and the emergence of new technologies and producers. In 2018, the Medical Isotope Supply Review identified the requirements for global demand, irradiation capacity, processing capacity and supply. This included projections for future demand (Figure 22). There have been positive developments with conversion of Curium Pharma (Netherlands) to LEU in January 2018 and the first licensing for an alternative generator by NorthStar (USA). However, the extended unplanned outage at NTP (South Africa) has pushed processing capacity below the North Energy Agency (NEA) demand of +35% outage reserve guideline. The supply situation will continue to require careful planning to minimize security of supply risks and react effectively in the event of an unplanned outage.



Figure 22. Current and future demands for <sup>99m</sup>Tc.

### 5.3 CNEA update

Eduardo Carranza (CNEA) provided an update of the radioisotope production at CNEA in Argentina. Fission <sup>99</sup>Mo is being produced at the RA-3 reactor at the Ezeiza Atomic Centre since 1985. The RA-3 reactor was inaugurated in 1967 to provide for the national demand of radioisotopes for medical use and the experience has supported the design and construction of several other reactors (Peru, Algeria, Egypt and Australia). Initially a 5-MW reactor using HEU material testing reactor (MTR) fuel elements, it was increased to 10 MW using LEU MTR fuel elements in 2000. In 2002, the commercialization of <sup>99</sup>Mo from LEU targets began, and in 2005 <sup>131</sup> production. The production process starts with the dissolution of uranium-aluminum alloy "meat" cladded with aluminum in an alkaline medium and is purified using filtration and ion exchange on four different columns. Weekly emissions from the facility have been measured for more than 20 years, and typical weekly production is 5.5 x 10<sup>13</sup> Bq of <sup>99</sup>Mo and 9.3 x 10<sup>12</sup> Bq of <sup>131</sup>I. The percentage of activity release from <sup>133</sup>Xe during weekly emissions is 35%. The annual release in 2017 was 8.7 x 10<sup>4</sup> GBg of <sup>133</sup>Xe and 3.8 x 10<sup>4</sup> GBg of <sup>135</sup>Xe (Figure 23). Research is being undertaken on radioxenon emission mitigation and includes devices to separate and capture hydrogen and devices to reduce the activity of noble gases from air emissions. Separating hydrogen from other gas elements such as xenon is useful as it reduces the gas volume required for storage (the hydrogen is returned to the hot cell), allowing longer storage and increased <sup>133</sup>Xe decay. At Ezeiza, the storage tanks used for reducing emissions have a residence time of 4 to 8 weeks. At 4 weeks, the <sup>133</sup>Xe is reduced by a factor of 35 and at 8 weeks by a factor of 1623.



Figure 23. Annual emissions from the RA-3 reactor.

### 5.4 Radioisotope production updates in Jordan Research and Training Reactor

Ahmad Malkawi (JAEC) discussed the production of radioisotopes using the Jordan Research and Training Reactor (JRTR). The JRTR is Jordan's first nuclear reactor, located 65 km north of Amman within the Jordan University of Science and Technology (JUST). Its construction was started in 2010 and criticality achieved on 25 April 2016. The JRTR project includes a reactor, radioisotope production facility, education and training building, radioactive waste facility and cold neutron facility. The reactor is 5 MW (upgradeable to 10 MW) with a maximum flux of 1.45 x 10<sup>14</sup> neutrons s<sup>-1</sup> (Figure 24). The reactor is especially important for supplying radioisotopes on a regular schedule for medical procedures and industrial applications in Jordan and the surrounding countries. The JRTR is capable of producing many radioisotopes including <sup>131</sup>I (capsule and solution), <sup>192</sup>Ir (source assembly) and <sup>99</sup>Mo (<sup>99m</sup>Tc generator). The radioisotope production facility (RIPF) contains 3 banks and 10 hot cells. The production of <sup>99</sup>Mo is through the neutron irradiation of MoO. The irradiation of <sup>235</sup>U is not used, as the current production process is sufficient for meeting supply requirements. Typically, 1 batch is prepared per week, with production of 3.7 x 10<sup>13</sup> Bq year<sup>-1</sup>. The domestic requirement is 30% of this value.



Figure 24. Cherenkov radiation within the JRTR open pool.

### 5.5 Status and challenges of the IRE LEU conversion program

Dominique Movaux (IRE) discussed the LEU conversion program at IRE. As a worldwide major radioisotope producer concerned by the reduction of proliferation risks, IRE is actively pursuing the conversion of its production processes to the use of LEU targets. This will enable the production of <sup>99</sup>Mo, <sup>131</sup>I and <sup>133</sup>Xe. All three processes currently require validation and some safety improvements are needed, including those for chemical process and production equipment modifications, and updates to the production environment. The <sup>99</sup>Mo process ramp-up is progressing well with LEU targets being irradiated in the same conditions as the future commercial productions. A total of 200 irradiated targets have been processed with 45 LEU runs over 2 years. This is not sufficient to meet <sup>99</sup>Mo production requirements, so an addition 3 HEU productions are being undertaken a week to maintain supply. The results obtained on irradiated LEU targets are significantly different from what has been observed previously during the cold commissioning and the first active tests. <sup>131</sup> has been found in various streams where it is usually excluded and thus represents a safety issue at the industrial production scale that must be overcome in priority. This has had a strong and negative impact on the daily <sup>133</sup>Xe release, which is higher than for HEU irradiations (Figure 25). An important process improvement effort is ongoing in order to address this safety issue and meet the stringent regulatory requirements. This includes modifying operating and chemical conditions, including using a new NaOH-NaNO<sub>3</sub> solution. Initial data indicates that this has had a positive effect on reducing both <sup>131</sup>I retention and on the daily <sup>133</sup>Xe release. These results require confirmation during the next ramp-up phase with an anticipated phase out of the HEU irradiation process in 2020.



Figure 25. Radioxenon production at IRE for LEU and HEU irradiations.

## 5.6 Update on Curium

Luis Barbosa (Curium) discussed <sup>133</sup>Xe production at the Curium facility at Petten, Netherlands. The <sup>133</sup>Xe is used for lung scintigraphy for patients who may have pulmonary embolisms. In the US, there are more than 600,000 people afflicted with pulmonary embolisms each year. With therapy, mortality rates fall from 30% to 5%. At Curium they have been developing a <sup>133</sup>Xe production facility using uranium irradiation. Current research and development focus on the development and optimization of a water-trapping column for xenon (Figure 26). Using this system, for which a patent application has been made, the first commercial batch of <sup>133</sup>Xe has been produced in April 2018.



Figure 26. <sup>133</sup>Xe production at Curium.

## 5.7 Update on isotope production plans in Algeria

Oualid Mahdaoui (Permanent Mission of Algeria) provided an update on peaceful uses of nuclear technologies in Algeria and its contribution to the CTBT. The Algerian Atomic Energy Commission (COMENA) is the principal governmental agency for implementing the national policy for the promotion and development of nuclear power and nuclear techniques. It acts under the authority of the Ministry of Energy since 2006, developing skills and infrastructure in the fields of nuclear fuel and materials, technology of nuclear facilities, and applications of nuclear science and technology. Algeria currently operates two RRs, including the Nour (1 MW) light-water reactor used for laboratory-scale production of radioisotopes, research and training; and the Es-Salam (15 MW) heavy-water reactor. Algeria supports multiple nuclear security, safety and safeguards protocols and nuclear activity in Algeria is strictly regulated under law. Applications for radioisotopes in Algeria, include both medical (radiotherapy and nuclear medicine) and industrial applications (non-destructive testing, gamma-densitometry, borehole logging, thickness gauging and gamma-scanning). Additional agricultural applications include plant breeding by radioeradication of insects (sterile insect technique) and progesterone mutagenesis. radioimmunoassay for enhancing the reproductive efficiency of ruminants. In each instance, these activities are developed in close collaboration with COMENA.

## 5.8 <sup>133</sup>Xe for medicine and industry

Ira Goldman (Lantheus Medical Imaging) provided an overview of Lantheus and its 60-year history of producing radiopharmaceuticals and radionuclides for medical and industrial applications. Lantheus is a commercial manufacturer of radiopharmaceutical products for molecular imaging of human disease, including FDA-approved radiopharmaceutical agents labeled with <sup>99m</sup>Tc (TechneLite® and Cardiolite®); <sup>67</sup>Ga, and <sup>201</sup>Tl for injection; Quadramet® (Sm-153 EDTMP) for bone pain palliation; and <sup>133</sup>Xe gas for inhalation (predominantly for pulmonary embolism detection). Lantheus is the longest continuous supplier of <sup>133</sup>Xe for pharmaceutical use in the US. Lantheus works with IRE to process and package <sup>133</sup>Xe produced from HEU and LEU irradiation at IRE. For lung ventilation studies the dose is typically 370 MBg or 740 MBg. Lantheus is also now producing <sup>133</sup>Xe for industrial-tracer applications, including as a tracer for NPPs (10<sup>7</sup> Bq used) and for oil and gas well-logging (10<sup>10</sup> Bq used). Industrial applications for <sup>133</sup>Xe are currently a small part of the overall <sup>133</sup>Xe market. While NPP calibration sources have existed for a relatively long-time, the use as a tracer for oil and gas operations is new business although it is currently limited to a small number of customers. Lantheus utilizes a <sup>133</sup>Xe trapping system to reduce emissions with a 18500 Bq ml<sup>-1</sup> year<sup>-1</sup> concentration limit. The stack emissions are monitored using an in-stack Geiger-Muller detector (Figure 27).





Lantheus' radiopharmaceutical product line in recent years has focused on production of FDAapproved products for medical purposes, with manufacturing and quality systems optimized to produce short half-life products within a just-in-time supply chain to meet FDA current Good Manufacturing Practices (cGMP). Lantheus operates six medium energy cyclotrons and maintains customer service, logistics, quality and regulatory affairs functions. Lantheus undertook a review several years ago to examine additional ways to use its cyclotron and associated target processing and material recycling infrastructure and related capabilities to produce additional products for which there is market demand.

## 5.9 Development of fission <sup>99</sup>Mo process and facility for the new research reactor of Korea

Suseung Lee (KAERI) discussed the development of KEARI's <sup>99</sup>Mo production facility. As <sup>99</sup>Mo and its daughter <sup>99m</sup>Tc, are one of most important medical radioisotopes for nuclear diagnostics,

the stable supply of <sup>99</sup>Mo to the market has been an important issue. This is especially true after the <sup>99</sup>Mo crisis in 2009 and 2010. In 2012, the Korean fission <sup>99</sup>Mo process development project was launched along with the new RR (Kijang New Research Reactor, KJRR) project. The Korean fission <sup>99</sup>Mo project covers development of uranium targets (UAIx) as well as chemical processing. By 2016, the target design and fabrication process had been completed and its gualification procedures had been established. However, low- and high-power irradiation test and postirradiation examination were delayed for two years due to the unexpected shut-down of the HANARO reactor. In July of 2018, target irradiation and post-irradiation tests had been completed with the resuming of the HANARO reactor. The fission <sup>99</sup>Mo process development covers dissolution, separation, purification and waste treatment (Figure 28). Caustic dissolution of the UAIx target is applied to the KAERI process. By 2016, a series of cold experiments with full-size UAIx targets were made with pilot-scale equipment. Full-size cold mock-up for the <sup>99</sup>Mo production was completed in February of 2017. In the June of 2018, the first hot production of fission <sup>99</sup>Mo from a full-size depleted uranium target was completed at the HANARO reactor. The construction permit for the KJRR has been delayed due to the earthquake events near the site in 2016 and 2017 and subsequent geological surveys. It is expected to start construction of KJRR in early 2019, aiming its first criticality by 2022.



Figure 28. The <sup>99</sup>Mo separation facility.

### 5.10 Plans by NWMI

Carolyn Haass (Northwest Medical Isotopes) gave an overview of the future Radioisotope Production Facility (RPF). The NWMI mission is to establish and deliver a domestic, reliable, securable source of at least 10<sup>14</sup> Bq of <sup>99</sup>Mo per week, steady state and without the use of HEU. The facility would use fission-based <sup>99</sup>Mo from LEU and rely upon a network of university RRs. Targets would be sent for irradiation and processed at the RPF (Figure 29). The location of the facility shall be Columbia, Mo, and final design, NRC operating license development and preconstruction is ongoing. Various research activities are also being undertaken in support of the future facility, including cold and hot small-scale irradiations, target fabrication and process testing. Work is also being undertaken in association with PNNL on abatement techniques to

reduce noble gas emissions. It is envisaged that the facility would become operational in late 2022.



Figure 29. Process flow diagram for the RPF.

### 5.11 Update at Ensterna

Yudiutomo Imardjoko (Ensterna) presented <sup>99</sup>Mo production at Ensterna in Indonesia. Xenon release reduction techniques include adsorption or absorption (difficult for the noble gas nature of xenon), increasing flow retention time, and trapping and confining. The latter is being investigated at Ensterna. The Sub-critical Assembly for Molybdenum Production (SAMOP) is a project of BATAN (the National Nuclear Agency of Indonesia) and is a neutron irradiation facility at the Indonesia Kartini reactor. Using SAMOP, the average <sup>99</sup>Mo production is 1.6 x 10<sup>10</sup> Bq per batch. The Thorium Aqueous Homogenous Reactor for Molybdenum Production System (ThAHRMOPS) is a reactor system designed to produce <sup>99</sup>Mo for medical purposes using thorium fuel. It is essentially an aqueous homogenous reactor whereby the thorium fuel is mixed with uranyl nitrate with light water as the solvent. The <sup>99</sup>Mo is produced by <sup>233</sup>U fission and extracted for processing.

### 5.12 Radioisotope production from LEU in INUKI

Bambang Herotumo (INUKI) provided an update of <sup>99</sup>Mo production. Since 2012, <sup>99</sup>Mo has been produced from <sup>235</sup>U fission using LEU targets. Previously INUKI used HEU targets. The LEU targets are prepared by electroplating on the inside wall of a stainless steel tube with a plating efficiency of 75%. The irradiated targets are dissolved with a mixture of H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub> solution. <sup>99</sup>Mo is then separated from other fission products by precipitation using alpha benzoin-oxime. The <sup>99</sup>Mo precipitate is then dissolved in NaOH/H<sub>2</sub>O<sub>2</sub>. The solution is passed through silver-coated charcoal column and zirconium oxide columns, respectively. Using this process, the separation

yield of <sup>99</sup>Mo from fission of <sup>235</sup>U was 60-70%. The quality of the <sup>99</sup>Mo product has met the required specification of United Sate Pharmacopea (USP XXI).

## 5.13 Argonne National Laboratory support for fission <sup>99</sup>Mo production

Alex Brown (ANL) discussed that the National Nuclear Security Administration's (NNSA) Office of Material Management and Minimization is currently supporting three potential US <sup>99</sup>Mo producers who plan to produce <sup>99</sup>Mo using fission-based technologies. Shine Medical Technologies, Niowave, and Global Medical Isotope Systems have developed accelerator-driven processes to produce <sup>99</sup>Mo from a uranium solution or target. Argonne has assisted or is currently assisting in the development of <sup>99</sup>Mo separation, recovery, and purification processes, target solution preparation, and solid target dissolution. This has utilized the Argonne LEAF Electron LINAC to irradiate DU targets at 35 MeV. Additional areas of support for these potential <sup>99</sup>Mo producers include, radiation effects on solution chemistry, off-gas generation, and precipitation, pH control, precipitation mitigation, and automation processes. Argonne has also been producing <sup>99</sup>Mo from <sup>100</sup>Mo disks (95.008% enriched) for the Radiogenix product. This utilizes a LANL developed and fabricated target and can produce 4 x 10<sup>11</sup> Bq of <sup>99</sup>Mo from 6 12 mm diameter disks (Figure 30).



Figure 30. <sup>100</sup>Mo enriched discs.

## 5.14 Session photographs













## 6.0 Session 5: Stack measurements

## 6.1 The STAX project overview

Lori Metz (PNNL) provided an overview of the Source Term Analysis of Xenon (STAX) project. This seeks to procure and install an experimental network of stack monitoring systems at MIPFs to assist the CTBTO PrepCom and NDCs in monitoring for signatures of nuclear explosions (Figure 31). PNNL has been tasked with specifying, purchasing, and contracting for the installation of STAX equipment. The equipment design, purchase and installation are provided at no cost to the producer. The data will be collected on a 10-15 minute time basis and stored at a central repository. It shall be disseminated to participating NDCs with explicit approval from the producers. The STAX project will take place over 5 years, with the first system being installed at IRE on November 2017 and the second system at ANSTO on October 2018. As part of the project, a STAX data pipeline is being developed. This shall include the analysis of spectra at the facility locations, and transmission of either spectra or isotope results, together with state-of-health information, to the central repository. The extent of data sharing from each facility shall be subject to negotiation, and data sharing plans established with each producer. Data collection is currently on-going at IRE, and several thousands of data points have been collected, that is useful for discrimination purposes from a nuclear explosive test.



Figure 31. The concept behind the STAX project.

## 6.2 Use of and procedures for sharing stack release data received by the Preparatory Commission for the CTBTO

Martin Kalinowski (CTBTO) provided an explanation of the categorization scheme for NG samples currently used for IMS measurements and how anomalous detections (level C) could trigger the utilization of stack release data to understand potential source terms. It is very important to consider confidentiality issues with access to stack release data, especially as under the provisions of the CTBT, all State Signatories must have access to all data for reconstruction of

IDC event analyses. The data provided must also be used for only Treaty-relevant purposes. However, there is a precedence for non-IMS data to be shared, including mining explosions and meteorological data. To facilitate possible implementation, several research and development stages are required. This includes a situation analysis phase, which has been completed, and includes scientific studies to understand the impact of MIPF releases on IMS stations. A proof-of-principle phase is currently being undertaken, which utilizes the limited amounts of historical data for developing and testing methodologies (e.g. ATM challenge). Following this should be a method development phase, that utilizes systematic historic and increasingly continuous near real-time data for scientific research into robust algorithms for network performance, contribution to sample concentrations, and source attribution. The final phase is to demonstrate the operationalization phase with continuous near real-time data for demonstration of being able to enter operations.

## 6.3 Xenon release source term estimation based on near-range monitoring and atmospheric dispersion modelling

Johan Camps (SCK-CEN) presented details of a radioxenon measurement campaign undertaken in close proximity to IRE. This combined off-site near-range monitoring with ATM to estimate the IRE source-term. This was a one-week campaign with the objective of observing the specific signature of releases for normal operation of the IRE facility. It aimed to focus on real-time data collection using in-situ spectroscopy and ambient dose rate measurements, to collect a complete data set for near-range model validation. This would enable an assessment of source term estimation based on the measurement results. The project involved collaborators from SCK-CEN, FANC-AFCN, RMI and IRE, and was undertaken under the framework of the IAEA CRP. Although IRE releases are well below any regulatory and health standards, the gamma fluency and/or gamma dose rate from the released cloud of noble gases was detectable using gamma sensors in the off-site environment. The study used both sensors from the fixed early-warning network, positioned at the fence of the institute, and mobile detectors which were installed, based on meteorological predictions, at larger distances. This included Nal, LaBr, Csl and plastic scintillation detectors combined with HPGe data from the IRE stack. A simple but robust and validated near-range atmospheric dispersion model was used in combination with meteorological data from a met-tower on-site to perform the atmospheric dispersion calculations and source term reconstruction. Using this approach there was very good agreement with modelled results based on detailed stack data and the off-site measurements undertaken during the campaign (Figure 32). The estimated source term for radioxenon isotopes fits very well with the stack monitoring source term for large releases. Future work should look to extend the data for a longer time series.



Figure 32. Backward atmospheric transport modelling to understand the stack source term.

## 6.4 Radioisotope emissions from Canada's Chalk River medical isotope production facility

Guy Jonkmans (Defence R&D Canada) discussed a 2-year monitoring program that measured the emissions from the National Research Universal (NRU) research reactor at Chalk River, Canada, prior to its closure on 31 March 2018. This MIPF was one of the world's single largest producers of medical isotopes and was a significant contributor to the world background of radioxenon. It was estimated that the NRU provided on the order of 90% of all radioxenon emissions into the northern hemisphere. The objective of the study was to improve knowledge to support the development of scientific attribution methods for activities at nuclear facilities. This would be accomplished by performing a measurement campaign of radioactive gas emissions and develop signatures across a range of emissions that are correlated with MIPF activities. This would allow an improvement, validation and expansion of atmospheric transport models and source term reconstruction techniques. Furthermore, this database of noble gas emissions, laboratory activities and field measurements would provide an improved understanding of the noble gas emissions background. During operation, the NRU reactor was responsible for approximately 1 TBg h<sup>-1</sup> of noble gas releases. Medical isotope production was responsible for the majority of the site's atmospheric emissions (>90%). It involved a complex extraction process combined with acid-based dissolution. The emissions were minimized through filtering and monitored using high (HPGe) and low (Nal) resolution stack detectors. Radioisotopes of xenon, argon, krypton, iodine and cesium were measured in the region surrounding the NRU up to a distance of approximately 200 km. The measurements comprised those made using Geiger Muller tubes, Nal detectors, air sample (xenon isotopes and <sup>85</sup>Kr) and aerosol sampling for <sup>137</sup>Cs and <sup>135</sup>Xe. Aerial measurements were also performed using a helicopter fitted with large volume Nal detectors and measurements were correlated with ground-based observations. Highresolution (250 m) atmospheric dispersion modelling was used to map the in-field radionuclide measurements back to the source term (Figure 33). The results were well-correlated, showing agreement between measured emissions, predicted emissions and medical isotope production.



Figure 33. Atmospheric dispersion modelling of NRU releases.

Measurements were also made using a SPALAX system in Ottawa for NRU normal operations when isotopes are being produced and production shutdown periods. The effect of medical isotope production is to create more dispersion in 3- and 4-isotope plots (Figure 34). Future research shall aim to understand these variations more fully. It is also the intention to produce a documented and shareable data set aligned with the Government of Canada's ambitions to stimulate innovation and improve international collaboration.



Source: I. Hoffman and R. Berg., J. Radioanal. Nucl. Chem., 318, 165 – 173, April 2018

Figure 34. Chalk River laboratory site emissions.

## 6.5 How the UK National Data Centre utilizes stack monitoring data in support of the Comprehensive Nuclear-Test-Ban Treaty

Richard Britton (AWE) presented a case study for how the UK National Data Centre (NDC) can use stack monitoring data in combination with IMS data for assessing CTBT relevant events. The UK NDC has developed a pipeline for IMS data that provides both forward and backward atmospheric transport modelling (ATM) to link detections to possible sources. The pipeline also provides automated forward simulations for MIPFs including IRE. These MIPF simulations can be used to predict the expected radioxenon activity at IMS stations (e.g. the contribution of IRE to SEX63). When combined with stack monitoring data to accurately provide a source term for the forward modelling, they have potential for improving the confidence of predictions at IMS stations. To demonstrate this potential, a fictious scenario was presented whereby a real radioxenon detection at SEX63 was ATM backtracked to a location in Wales that coincided with a seismic event with a magnitude of 2.7. Hypothetically, it was suggested that this could be a potential CTBT violation. The high uncertainty of the radioxenon ratio made it difficult to differentiate the source term of the detection, such that a nuclear explosion could not be excluded. However, using STAX data from the IRE MIPF combined with forward modelling, showed that the SEX63 detection was likely to be attributable to IRE emissions, both in terms of potential release area and isotopic measurements observed at SEX63 (Figure 35). In this manner it was possible to confirm that a nuclear explosion in Wales was not the cause of the IMS detection (despite a coincident seismic event) and that the signal was most likely from IRE emissions. Although this was a fabricated scenario, unexplained IMS detections do regularly occur and STAX data is useful for eliminating these events.



Figure 35. Constraining the radioxenon release location.

### 6.6 The STAX Project – Data processing infrastructure

Matthias Auer (ISTI) discussed the development of a data processing infrastructure in support of the Source Term Analysis of Xenon (STAX) project. ISTI is establishing a software infrastructure to collect, transmit, process, store and distribute data from stack monitors, such as those installed

at the medical isotope production facilities at IRE, Belgium and ANSTO, Australia. Three different types of data messages are compiled from the stack monitor at each facility: spectrum (PHD) files, state of health (SOH) data files and time series files. PHD and SOH files are following the IMS2.0 format that is used in the IMS network of the CTBTO. For time series data, a new format, the Automated Time Series Report (ATS), has been developed that accumulates spectra and release data from 24 hours intervals. Data are encrypted and transmitted via email to a central data server (staxdata.org), where they are stored and available for distribution to authorized users, including the International Data Centre (IDC) and National Data Centers (NDCs). In a next step, tools to interactively retrieve and view data will be developed. High emphasis is put on data security and access, during transmission from facilities to the data server as well as for retrieval of data from the data server.



#### Figure 36. The STAX network configuration.

## 6.7 CTBTO platform for handling stack release data

Abdelhakim Gheddou (CTBTO) provided an update of the development of a dedicated system under the virtual Data Exploitation Centre (vDEC) platform for handling Stack Release Data (SRD). This data would be received by the CTBTO on a voluntary basis from Medical Isotope Production Facilities and Nuclear Power Plants. The purpose of the CTBTO platform for SRD is to make these data available to experts for use in scientific studies on characterization of global radionuclide background with focus on the calibration and performance of the IMS Noble Gas (NG) systems. It may also be applied in developing and validating new methods to assist State Signatories in identifying the possible source of an event. The goal is to build an integrated environment for receiving, processing, archiving and disseminating raw spectral data and analysis results of the four CTBT relevant xenon isotopes. Sharing of data will be based on permissions from the data providers and signing of confidentiality agreements by the recipients. An SRD processing pipeline is being developed that uses an integrated database schema for storing input data, feeding processing modules, analyzing spectra and holding analysis results (Figure 37). The pipeline can be used in client/server database connection mode or in standalone mode and is compatible with the vDEC platform. The platform was tested with stack data from the medical isotope facility at IRE and successful spectra processing demonstrated.



Figure 37. SRD processing pipeline.

## 6.8 Investigating noble gas emissions from nuclear facilities in central Europe

Andreas Bollhofer (BfS) discussed how the noble gas emissions from civil nuclear facilities can affect the verification capability of the IMS of the CTBTO. Major emitters of radioxenons for instance are a few medical isotope production facilities (MIPF) worldwide, but smaller emissions from nuclear power plants, in particular those in close vicinity to monitoring stations, also impact on the radioxenon levels and isotopic composition in the atmosphere. Some stations within the BfS noble gas network show occasionally elevated radioxenon levels which can usually be traced back to local emitters, and some relevant sources include pressurized water reactors (PWR) and research reactors (RR). Measurements were performed from these facilities as part of the IAEA Coordinated Research Project CRP F23031 and in the frame of collaborative research between BfS and the Universities of Bern and Hamburg. This included measurements from the stacks and reactor halls and analysis for radioxenon (<sup>133</sup>Xe, <sup>135</sup>Xe, <sup>133m</sup>Xe, <sup>131m</sup>Xe), <sup>41</sup>Ar and <sup>85</sup>Kr (Figure 38). It was estimated that 75-80% of the radioxenon detections at the DEX33 (Freiburg) IMS station were from the IRE MIPF, and that nearby nuclear power plants were partly responsible for the remaining detections. Radioxenon emissions from a large PWR are found to be at least an order of magnitude higher than those from a high neutron flux RR. In both instances, the 4-isotope fingerprint measured 1-2 days after sample collection for both PWR and RRs falls within the civil domain defined by DEX33 detections. Emissions of radioargon were found to be an order of magnitude higher than those of radioxenon. The <sup>41</sup>Ar/<sup>37</sup>Ar ratio measured in the reactor hall resemble activation of argon in air, although it is strongly influenced by the dynamics of production and decay processes. In contrast, the <sup>37</sup>Ar emitted from MIPFs was relatively small (approximately 14 mBg m<sup>-3</sup>) and may be attributable to calcium impurities (and subsequent activation) in the target.



Figure 38. Noble gas emissions from reactors.

### 6.9 Radioxenon and radioargon emission signatures from Chalk River laboratories

Steven Biegalski (Georgia Tech) presented a sampling campaign undertaken in 2014 and 2016 at Sheenboro, QC, Canada to examine imprinting of radioxenon into the local geology. This location was chosen due to its proximity to the Chalk River Laboratories. During operation, the NRU at Chalk River emitted between 5 and 44 x 10<sup>12</sup> Bg day<sup>-1</sup>. Air samples were collected underground and above surface and subsequently measured for radioxenon isotopes at PNNL. These were the first ever measurements of radioxenon imprinting into subsurface gas and concluded that subsurface sampling (at 1 m depth) could result in detected levels of radioxenon on the order of 10% of the average atmospheric concentration. These soil gas concentrations were observed to increase following the passage of a plume, with <sup>133</sup>Xe activity of up to 48 Bg m<sup>-</sup> <sup>3</sup> being measured. This maximum subsurface concentration corresponded to 2 days of highly elevated atmospheric concentrations. To support these measurements, Chalk River Laboratories provided stack data for the measurement period so that the source term could be matched with measurements at the sampling location. While the MIPF is no longer in operation, these data help to assess the range of isotopic signatures that may be expected from the emission point and calculations are performed to determine downwind isotopic ratios. The MIPF stack provides activity concentrations for <sup>133</sup>Xe, <sup>135</sup>Xe, and <sup>135m</sup>Xe. Through activation and decay calculations, <sup>131m</sup>Xe and <sup>133m</sup>Xe activities may be estimated (Figure 39). The reactor stack data provides activity concentrations including <sup>41</sup>Ar and <sup>87</sup>Kr, the ratios of which were found to be significantly different from predicted calculations using the ORIGEN model.





#### 6.10 ANSTO experience with the stack monitoring system

Tom Loosz (ANSTO) discussed installation and operational experiences with a stack monitor installed at ANSTO as part of the STAX project. The STAX system utilizes a high-resolution HPGe gamma-spectrometer and replaced a back-up low-resolution Nal that was originally going to be installed onto the ANM facility. The installation location was carefully considered, and a risk assessment to the local infrastructure and STAX hardware undertaken prior to install. Some minor installation hurdles had to be overcome, including interfacing the system with the ANSTO stack flow monitor input signal. Pre-familiarization training would also have been useful to provide users with a better understanding of how the system works. Initial commissioning has been completed and shows good performance of the system with a detector dead-time of approximately 1% (Figure 40). This data has already proved beneficial for commissioning and operation of the ANM facility.

#### PNNL-28870



Figure 40. Initial measurements from the stack monitor at ANSTO.

### 6.11 Installation of STAX monitoring system at IRE

Benoit Deconninck (IRE) provided a performance evaluation of the stack monitor installed as part of STAX project at the IRE facility. This was the first STAX monitoring system installed in October 2017. Measurements from the system were compared with an existing stack monitor at IRE. Whilst both systems utilized a high-resolution HPGe, the STAX system undertook measurements through a Marinelli and the non-STAX system measured through a cartridge system. Despite these measurement differences, results were found to be in good agreement (Figure 41). After 1 year of operation there have been no major issues with the system and <sup>131</sup>I contamination of the system has not been observed.



Figure 41. A comparison of stack monitor measurements at IRE.

## 6.12 An autonomous spectroscopic noble gas monitor with continuous operation and analysis

James Zickefoose (Mirion Technologies) discussed the design and development of a spectroscopic stack monitoring system for the measurement of noble gases discharged from MIPF stacks (Zickefoose, Burnett et al. 2018). The system has currently been installed at IRE (Belgium) and ANSTO (Australia). The prototype is based on a Marinelli-beaker style HPGe measurement operating in a continuous acquisition mode. Continuous acquisition is accomplished with novel software/hardware which allows for unattended acquisition, analysis, and storage of data over multiple workflow definitions. Since multiple averaging times are allowed for a single data stream, it is possible to attain both swift reaction times as well as very low minimum detectable concentrations (Table 2). Furthermore, since the release of noble gases may be in short bursts of high concentration, the system was fit with a transistor reset preamplifier which allows for spectroscopic analysis up to and beyond 500,000 counts second<sup>-1</sup>. As a direct result of the multiple averaging times and the use of the transistor reset preamplifier, the dynamic range of the system covers nearly 8 orders of magnitude, with a measurable concentration range of  $10^2$  to  $6.3 \times 10^9$  Bq m<sup>-3</sup>.

Nuclide	Key line (keV)	Yield (gamma/dis)	Efficiency (cts/gamma)	ROI BKGD Rate (cps)	MDC (Bq/m <sup>3</sup> )
Kr-85	514	0.00434	4.06E-03	9.59E-03	6.91E+04
Kr-85m	151.2	0.7519	9.77E-03	1.25E-02	1.85E+02
I-131	364.5	0.8122	5.44E-03	5.35E-03	2.20E+02
Xe-131m	163.9	0.01953	9.59E-03	1.31E-02	7.41E+03
Xe-133	80.99	0.3699	6.45E-03	1.27E-02	5.74E+02
Xe-133m	233.2	0.1013	7.86E-03	1.01E-02	1.56E+03
Xe-135	249.8	0.8999	7.46E-03	1.03E-02	1.87E+02
Xe-135m	526.6	0.808	3.99E-03	3.06E-03	2.46E+02

Table 2. Minimum detectable concentration achievable using the stack monitoring system	n for a
10 m acquisition.	

### 6.13 Tailored designed stack monitor for STAX project

Mariana Di Tada (INVAP) discussed the development of a stack monitor for the STAX project called the Air Effluent Monitor (AEMi). The system has a modular design and is tailored for the emissions of individual MIPFs. The system has evolved from systems installed at RRs and MIPFs, including those located in Algeria, Egypt, Australia and India. The system intended for the STAX project has been modelled using MCNP for normal and incident operating conditions. It consists of an air sampling system with a lead shielded mechanically cooled Ortec 10% efficiency HPGe coupled with a PC processing unit. This shall provide a dynamic range of 10<sup>4</sup> Bq m<sup>-3</sup> to 10<sup>8</sup> Bq m<sup>-3</sup>. The system is designed to meet the requirements of the STAX project, including facilitating the transfer of stack data to a central database. The first system is currently being built at INVAP for PNNL.

## 6.14 Simulating <sup>133</sup>Xe concentrations at IMS noble gas stations using stack emission data from medical isotope production facilities

Andy Delcloo (Royal Meteorological Institute of Belgium) discussed how radioxenon stack emission data from civilian nuclear facilities, combined with ATM, can help to discriminate potential Treaty violations from legitimate civil nuclear facilities. Modelling was performed of the radioxenon background on IMS NG stations using <sup>133</sup>Xe stack emission data sets from two MIPFs: IRE in Belgium, and the Canadian Nuclear Laboratories in Canada. The Lagrangian particle dispersion model Flexpart was used to calculate the transport and dispersion of <sup>133</sup>Xe, using numerical weather prediction data from the European Centre for Medium Range Weather Forecasts. The results were in good statistical agreement with observations of the IMS.

## 6.15 IPF gaseous effluent stack monitor calibration with customized detection geometries and tailored gas-like sources

Mariana Di Tada (INVAP) presented details on the calibration of a new stack monitor. The design is based on the individual monitoring requirements of a nuclear facility, and in the instance of an MIPF, needs to take into account the possible high activity concentration levels from a sudden release. In the INVAP system, there is a CdTe and NaI detector for the measurement of <sup>133</sup>Xe and <sup>135</sup>Xe, each contained within a modular shielding assembly (Figure 42). Calibration of the two NG chambers was performed using tailored gas-like radioactive sources manufactured with the same geometry as the measuring chambers. These typically consist of a low-density material spiked with representative gamma-emitting radionuclides such as <sup>133</sup>Ba and <sup>152</sup>Eu. Whilst these are not as realistic as actual gas samples, they can be used for a representative calibration at relatively low-cost. Actual gas calibration samples are also often difficult to obtain and handle. The efficiency and energy calibrations from these sources compared well with those predicted using MCNP modelling techniques and was within a 15% error.



Figure 42. The INVAP stack monitor.

### 6.16 Design of the 3<sup>rd</sup> ATM-Challenge 2019

Christian Maurer (ZAMG) discussed the design of the 3rd ATM Challenge in 2019. Two ATM challenges were successfully performed within the last three years (2015 & 2016) and were useful for drawing some general scientific conclusions on the current status of ATM forecasting. However, these challenges did not address the more practical aspect of estimating radioxenon background at selected IMS stations. This estimation is needed for calibration and performance assessment of the verification system as described in the CTBT. Estimating the radioxenon background is the main goal of a 3<sup>rd</sup> Challenge. This will be accomplished in collaboration with the European Joint Research Center (JRC) in Ispra (Italy) through a multi-model ensemble modelling approach. A training approach will be used to define the optimal set of members, specific to each station, needed to estimate the radioxenon background. The training will be applied to the four selected IMS stations CAX17 (St. John's), DEX33 (Schauinsland/Freiburg), SEX63 (Stockholm) and USX75 (Charlottesville). <sup>133</sup>Xe stack emission data for the time period June - November 2014 from the IRE (Belgium) and CRL (Canada) radiopharmaceutical plants will be used for this challenge. In addition, recent publicly available emission estimates for NPPs and RRs distributed over the northern hemisphere will be considered. The annual emissions from the Mallinckrodt facility (The Netherlands), the NIIAR facility (Russia) and the Karpov Institute (Russia) will be also used to refine predictions. Participants will calculate contributions to the signal at CAX17, DEX33, SEX63 and USX75 for up to 6 months based partly on a unit emission approach. The ultimate goal of the ATM exercise is to provide an ensemble analysis of radioxenon background levels at IMS stations frequently impacted by industrial emissions.

## 6.17 Session photographs











Session 5: Stack measurements


## 7.0 Conclusions

WOSMIP VII was attended by 91 experts from 19 countries and discussed the impact of manmade radioisotopes on nuclear explosion monitoring in support of the CTBT. During the 4-day meeting, sessions were held on background sources of man-made isotopes, research to reduce the radioxenon impact on the IMS of the CTBT, current and planned <sup>99</sup>Mo production activities, and radioxenon stack measurements. At WOSMIP VII, the workshop host ANSTO, reaffirmed its commitment to mitigating the impact of <sup>99</sup>Mo production emissions on nuclear explosion monitoring. This included the installation of a radioxenon stack monitor at the ANM <sup>99</sup>Mo facility in support of the STAX project. This demonstrates the value of WOSMIP in making a difference to the verification regime of the CTBT. The contribution of MIPFs to the global radioxenon background was recognized as one of the most important topics for the nuclear explosion monitoring community. While WOSMIP has facilitated a significant improvement in the understanding of background sources, more research is needed as only approximately 50% of IMS detections are fully understood.

While RRs are not generally considered to be a significant background source, significant gaps in understanding their releases were identified, including spurious releases and potential air activation signals. These may become more important as radioxenon monitoring systems become more sensitive and the WOSMIP community's understanding of the background improves. The contribution of NPPs may also sometimes be important depending on meteorological conditions. A new background source was also identified in the use of radioxenon in tracer operations by the oil and gas industry. Additionally, anomalous particulate IMS detections were discussed with focus on detections of ruthenium isotopes (<sup>103</sup>Ru, <sup>106</sup>Ru) in Europe during 2017. As with radioxenon measurements, a need to more fully understand these background fluctuations and source terms was recognized.

WOSMIP VII identified new focus areas, including understanding the impacts of new medical isotope producers and advancements in emissions mitigation and stack monitoring technology. Collaborations with the MIP and nuclear explosion monitoring communities are encouraged, to facilitate the development of more suitable xenon abatement technologies, that are smaller, passive, cost effective, robust, and more selective. Such benefits have been demonstrated by the partnerships of ANSTO and IRE with PNNL for the installation of STAX stack monitors for radioxenon emissions.

### 8.0 References

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## Appendix A – Workshop program

Tuesday 4<sup>th</sup> December 2018

8.00 am	Delegate Registration – Level 1, Pullman Sydney Hyde Park	
All Conference Sessions will be held in the Ibis Room on the 1 <sup>st</sup> Floor		
	Session 1: Welcome and Workshop Overviews	
	Chair: Judah Friese, Pacific Northwest National Laboratory	
9.00 am	Welcome to WOSMIP VII	
	Adrian Paterson, CEO, Australian Nuclear Science and Technology Organisation	
9.10 am	WOSMIP VII Kick-off	
	Emmy Hoffmann, Australian Nuclear Science and Technology Organisation	
9.20 am	The History of WOSMIP	
	Paul Saey, International Atomic Energy Agency	
9.35 am	WOSMIP VII Overview	
	Ted Bowyer, Pacific Northwest National Laboratory	
9.50 am	Morning Tea in the Ibis Foyer - Sponsored by INVAP, S.E. and Group Photo	
10.35 am	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and	
10.35 am	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and Mobile Systems Tammy Taylor, Director, International Data Centre, Preparatory Commission for the	
	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and Mobile Systems Tammy Taylor, Director, International Data Centre, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization	
10.35 am	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and Mobile Systems         Tammy Taylor, Director, International Data Centre, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization         Status of the Noble Gas capability at the CTBTO International Monitoring System         Nikolaus Hermanspahn, Preparatory Commission for the Comprehensive Nuclear-Test-Ban	
10.35 am 10.55 am	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and Mobile Systems         Tammy Taylor, Director, International Data Centre, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization         Status of the Noble Gas capability at the CTBTO International Monitoring System         Nikolaus Hermanspahn, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization         How Can CTBTO NG Stations Be Impacted y Radioxenon Releases from an Isotope	
10.35 am 10.55 am	Updated Worldwide Background of CTBT Relevant Xenon Isotopes Based on IMS Data and Mobile Systems         Tammy Taylor, Director, International Data Centre, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization         Status of the Noble Gas capability at the CTBTO International Monitoring System         Nikolaus Hermanspahn, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization         How Can CTBTO NG Stations Be Impacted y Radioxenon Releases from an Isotope Production Facility         Jonathan Baré, Preparatory Commission for the Comprehensive Nuclear-Test-Ban-Treaty	

11.55 am	Investigation of the Radioxenon Background Trend at Noble Gas IMS Systems Christophe Gueibe, Belgian Nuclear Research Centre		
12.15 pm	ATM Analysis Results on Radioxenons Detected in Japan by JAEA/NDC Yuichi Kijima, Japan Atomic Energy Agency		
12.35 pm	Working Lunch and Introductions in Windows On The Park		
	Session 2: Background Sources Chair: Martin Kalinowski, Preparatory Commission for the Comprehensive Nuclear-Test-Ban- Treaty Organization		
2.00 pm	<b>Observations of Radioxenon Emissions from BWRs Compared to Stack Data</b> Anders Ringbom, Totalförsvarets Forskningsinstitut		
2.20 pm	Research Reactor Contributions to Radioxenon Atmospheric Inventories Steven Biegalski, Georgia Institute of Technology		
2.40 pm	Physical Characterization of filters from German and Sweden radiological monitoring networks with Ruthenium-106 from 2017 Ian Hoffman, Health Canada		
3.00 pm	<b>European Monitoring of <sup>106</sup>Ru Airborne Concentrations in Fall 2017</b> Olivier Masson, Institut de Radioprotection et de Sûreté Nucléaire		
	Olivier Masson, Institut de Radioprotection et de Sûreté Nucléaire		
3.20 pm	Olivier Masson, Institut de Radioprotection et de Sûreté Nucléaire Afternoon Tea in the Ibis Foyer - Sponsored by INVAP, S.E.		
	Afternoon Tea in the Ibis Foyer - Sponsored by INVAP, S.E.		
3.20 pm	Afternoon Tea in the Ibis Foyer - Sponsored by INVAP, S.E.		
3.20 pm 3.50 pm	Afternoon Tea in the Ibis Foyer - Sponsored by INVAP, S.E. INVAP, S.E. Isotopic Ratios for Ru-103/Ru-106 Jonathan Burnett, Pacific Northwest National Laboratory Using Machine Learning to Estimate Atmospheric Sources from the 2017 Ruthenium Detections across Europe		

5.50 pm	Poster Session with End of Day Drinks sponsored by Mirion Technologies (Canberra)		
		MIRION TECHNOLOGIES	
	CANBERRA		
7.30 pm	Close of Day 1		

## Wednesday 5<sup>th</sup> December 2019

8.30 am	Delegate Registration – Tea and Coffee on Arrival – Ibis Foyer		
	Session 3: Research to Reduce the Radioxenon Impact on the International Monitoring System		
	Chair: Johan Camps, Belgian Nuclear Research Centre		
9.00 am	Welcome to Day 2		
	Johan Camps, Belgian Nuclear Research Centre		
9.05 am	Overview of the CRP		
	Tom Hanlon, International Atomic Energy Agency		
9.25 am	Sharing Experience on Mitigating Radioxenon Through Preparation of Design Studies for Three Civilian Nuclear Facilities		
	Christophe Gueibe, Belgian Nuclear Research Centre		
9.45 am	Development of Radioxenon Abatement System for the Fission Mo-99 Production Process		
	Suseung Lee, Korea Atomic Energy Research Institute		
10.05 am	Testing and Validation of a Radioxenon Trap System under the EU Council Decision VI Project: Results and Outcomes		
	Dominique Moyaux, Institut National des Radioéléments		
10.25 am	Verification and Validation of Atmospheric Transport Models for Nuclear Security with the UF Training Reactor		
	Andreas Enqvist, University of Florida		
10.45 am	Morning Tea in the Ibis Foyer sponsored by Scienta Sensor Systems		
	scienta sensor systems		
	Session 4: Current Mo-99 Production Overview: Recent and Planned Activities at Isotope Production Facilities		
	Chair: Paul Saey, International Atomic Energy Agency		
11.15 am	Radioisotopes Used in Nuclear Pharmacy and Nuclear Medicine Practice: Perspectives from the United States		
	Neil Petry, Duke University		

	Tom Hanlon, International Atomic Energy Agency		
11.50 am	<b>CNEA Update</b> Eduardo Carranza, Comisión Nacional de Energía Atómica		
12.05 pm	Radioisotope Production Updates in Jordan Research and Training Reactor           Ahmad Malkawi, Jordan Atomic Energy Commission		
12.20 pm	Status and Challenges of the IRE LEU Conversion Program Dominique Moyaux, Institut National des Radioéléments		
12.35 pm	Update at Curium Luis Barbosa, Curium Pharma		
12.50 pm	Working Lunch in Windows On The Park		
2.20 pm	<b>Update on Isotope Production Plans in Algeria</b> Oualid Mahdaoui, Permanent Mission of Algeria to the United Nations and Other International Organisations in Vienna		
2.35 pm	Xe-133 for Medicine and Industry Ira Goldman, Lantheus Medical Imaging		
2.55 pm	<b>Development of Fission Mo-99 Process and Facility for the New Research Reactor of Korea</b> Suseung Lee, Korea Atomic Energy Research Institute		
3.10 pm	Plans by NWMI Carolyn Haass, Northwest Medical Isotopes		
3.25 pm	Afternoon Tea in the Ibis Foyer sponsored by Scienta Sensor Systems		
	scienta sensor systems		
3.55 pm	Roundtable Discussion – See Instructions on Page 15		
4.55 pm	Update at Ensterna Yudiutomo Imardjoko, Ensterna		
5.10 pm	Radioisotope Mo-99 Production from LEU In INUKI Bambang Herutomo, Indonesia Nuclear Technology and Services		
5.25 pm	Argonne National Laboratory Support for Fission Mo-99 Production		

	Alex Brown, Argonne National Laboratory		
5.45 pm	Close of Day 2		
6.45 pm	Delegates to Meet in the Lobby of the Pullman Sydney Hyde Park for short walk to the Australian Museum, corner of College Street and William Street, Sydney.		
7.00 pm	Workshop Dinner – Australian Museum (Pre-registration Required), Corner of College Street and William Street, Sydney Sponsored by ANSTO, ARPANSA, ASNO, GeoScience Australia, and the Pacific Northwest National Laboratory		
	Australian Government Australian Government Australian Government Australian Radiation Protection and Nuclear Safety Agency Australian Covernment Australian Radiation Protection and Nuclear Safety Agency		
	Australian Government Australian Safeguards and Non-Proliferation Office		
	Australian Government Geoscience Australia		

### Thursday 6<sup>th</sup> December 2019

8.30 am	Delegate Registration – Tea and Coffee on Arrival in the Ibis Foyer
	Session 5: Stack Measurements
	Chair: Ian Hoffman, Health Canada
9.00 am	Welcome and The STAX Project Overview
	Lori Metz, Pacific Northwest National Laboratory
9.20 am	Use of and Procedures for Sharing Stack Release Data Received by the Preparatory Commission for the Comprehensive Nuclear-Test-Ban-Treaty Organization
	Martin Kalinowski, Preparatory Commission for the Comprehensive Nuclear-Test-Ban-Treaty Organization
9.40 am	Xenon Release Source Term Estimation Based on Near-range Monitoring and Atmospheric Dispersion Modelling
	Johan Camps, Belgian Nuclear Research Centre
10.00 am	Radio-isotope Emissions from Canada's Chalk River Medical Isotope Production Facility
	Guy Jonkmans, Defence R&D Canada
10.20 am	How the UK National Data Centre utilises Stack monitoring data in support of the Comprehensive Nuclear Test-Ban Treaty
	Richard Britton, Atomic Weapons Establishment
10.40 am	Morning Tea in the Ibis Foyer Sponsored by ISTI
	isti
11.10 am	<b>The STAX Project – Data Processing infrastructure</b> Matthias Auer, Instrumental Software Technologies
11.30 am	CTBTO Platform for Handling Stack Release Data
	Abdelhakim Gheddou, Preparatory Commission for the Comprehensive Nuclear-Test-Ban- Treaty Organization
11.50 am	Investigating Noble Gas Emissions from Nuclear Facilities in Central Europe
	Andreas Bollhöfer, Bundesamt für Strahlenschutz
I.2.10 pmRadioxenon and Radioargon Emission Signatures from Chalk River LaboratoriesSteven Biegalski, Georgia Institute of Technology	

12.30 pm	Working Lunch in Windows On The Park – Discussion on the Use of Stack Data	
2.00 pm	ANSTO Experience with the Stack Monitoring System	
	Tom Loosz, Australian Nuclear Science and Technology Organisation	
2.20 pm	Installation of STAX Monitoring System at IRE – Performances Evaluation	
	Benöit Deconninck, Institut National des Radioéléments	
2.40 pm	An Autonomous Spectroscopic Noble Gas Monitor with Continuous Operation and Analysis	
	James Zickefoose, Mirion Technologies (Canberra)	
3.00 pm	Tailored Designed Stack Monitor for the STAX Project	
	Mariana Di Tada, INVAP S.E.	
3.20 pm	Afternoon Tea in the Ibis Foyer sponsored by ISTI	
3.50 pm	Simulating Xe-133 Concentrations at IMS Noble-Gas-Stations Using Stack Emission Data from Medical Isotope Production Facilities	
	Andy Delcloo, Royal Meteorological Institute Of Belgium	
4.10 pm	IPF Gaseous Effluent Stack Monitor Calibration With Customized Detection Geometries and Tailored Gas-like Sources	
1 20 nm	Mariana Di Tada, INVAP S.E.	
4.30 pm	Design of the 3rd ATM-Challenge 2019 Christian Maurer, Zentralanstalt für Meteorologie und Geodynamik, Austria	
4.50 pm	Roundtable Discussion – See Instructions on Page 15	
-		
5.50 pm	Announcement of the Wozzie Award	
6.10 pm	Close of Day 3	

## Friday 7<sup>th</sup> December 2019

8.00 am	Delegate Registration – Tea and Coffee on Arrival in the Ibis Foyer
	Session 6: ANSTO
	Chair: Emmy Hoffmann, Australian Nuclear Science and Technology Organisation
8.30 am	Welcome to Day 4
8.35 am	ANSTO ANM Facility
	Michael Druce, Australian Nuclear Science and Technology Organisation
9.05 am	ANM Mo-99 Active Ventilation System
	Stuart Brink, Australian Nuclear Science and Technology Organisation
9.25 am	Workshop Wrap Up
10.00 am	Morning Tea in the Ibis Foyer
10.20 am	Board coach and depart the Sydney Pullman Hyde Park by 10.30 am
	Please be sure to bring your passport & wear closed shoes.
11.30 am	Arrive at ANSTO. Security check & bio-break
11.40 am	ANSTO Overview Presentation
11.50 am	ANSTO Tour – Part 1
	Split into groups for the tour of the ANSTO Nuclear Medicine facility:
	<ul> <li>Production &amp; QC Labs</li> <li>Production hot cells</li> </ul>
	- STAX monitoring system
1.00 pm	Working Lunch and Quiz
2.00 pm	ANSTO Tour – Part 2
	OPAL multi-purpose reactor
2.22	- Australian Centre for Neutron Scattering
2.30 pm	ANSTO Tour – Part 3 (Optional)
	Delegates not attending Part 3 will depart on a bus for return to Pullman Sydney Hyde Park at 2.30 pm. Delegates wishing to attend the extended tour will depart (additional \$28 AUD cost
	for this tour to be paid on arrival at Symbio. Credit cards accepted at the Ticket office.
	Sunscreen and hat recommended).
5.00 pm	Delegates depart for return to Pullman Sydney Hyde Park
6.30 pm	Arrive at Pullman Sydney Hyde Park

## **Appendix B – Roundtable discussions**

The following instructions were provided to facilitate the roundtable discussions.

#### Session 2 Roundtable: Background

The International Monitoring System (IMS) continually monitors the world for radionuclides in the environment. This network involves both particulate and noble gas measurements. The IMS is a unique international capability and understanding detections in the network is an important aspect in a robust treaty verification regime.

As the detections in the IMS becomes more numerous, our understanding of these detections must also continue to grow. Questions to discuss at this round table involve where we think the state of the science for understanding environmental detections currently is and our future challenges.

Starting questions:

- What are the uses of <sup>106</sup>Ru in such large quantities as observed?
- Do we have a good enough understanding of radioactivity in the environment?
- Will nuclear reactor emissions be important to understand in the future?
- Looking to the future, what are future man-made sources that we may observe in the IMS?

#### Session 3 Round table: Xenon Abatement Research

The abatement of radioxenon releases is a well-known challenge, and this challenge has been successfully met for lowering emissions to the point where health and safety is not a concern. However, with the deployment of the International Monitoring System (IMS), even this safe level of radioxenon releases are causing routine detections. The current industrial methods for xenon abatement can be expensive to implement and not practical at some facilities. Questions to consider during this roundtable include: How can future research that lower the cost and difficulty of adding additional abatement impact the IMS? How can partnerships between communities help in understanding and solving this difficult issue? As new facilities for production that could release radioxenon are proposed and built, how can radioxenon abatement well below health and safety be implemented successfully?

Starting Questions:

- What is the need for new/better xenon absorption materials?
- What are the industrial limitations of low xenon emissions?
- How can the WOSMIP community be an advocate for continued work in xenon abatement?

#### Session 4 Roundtable: Stack Monitoring

All facilities do routine monitoring of their emissions for regulatory purposes. This is robust and typically required by the regulating body in each country. In previous WOSMIP workshops, the use of emission data from the stack of nuclear facilities was proposed as a method to lower the impact on the International Monitoring system (IMS) by understanding civilian radioxenon sources in the environment. By knowing the source of radioactivity entering the environment, these sources can be subtracted from monitoring detections. This proposed method for limiting the impact on the IMS has generated scientific work in understanding how this could be implemented. Questions to consider include what type of data would be useful and how would it routinely be used in understanding detections? This round table discussion will begin with comments from experts involved in stack monitoring and their thoughts on the prospect of this being used for treaty monitoring.

Appendix B

# Appendix C – Conference posters

1	Jonathan Baré	Impact of CRL shutdown on CTBTO North-American noble gas stations
2	Steven Bell	Radioactive Gas Metrology at NPL
3	Pierre Bourgouin	Evaluating different alternative sites for IMS stations
4	Jagoda Crawford	Radon-222 detection in outdoor air and applications in atmospheric transport and mixing studies
5	Christophe Gueibe	Physical experiments and modelling work on xenon capture
6	Bambang Herutomo	INUKI experience with stack monitoring
7	lan Hoffman	Medical Isotope Production and Research Reactors and the Global Radioxenon Background
8	Miroslav Hyza	Radiological Monitoring of the Atmosphere Using an Autonomous Aerosol Sampler
9	Martin Kalinowski	What do we need to know to access how radioxenon emissions from nuclear power plants interfere with nuclear explosion monitoring?
10	Stanislav Kocvara	NGM-2000 Spectrometric Noble Gas Monitor with HPGe Detector
11	Jolanta Kusmierczyk- Michulec	Influence of emission time resolution on the Atmospheric Transport Modelling (ATM) results
12	Olivier Masson	European-scale detection of I-131 in early 2017
13	Lori Metz	Emissions Mitigation R&D for the US Government
14	Blake Orr	Summary of East Asia Regional National Data Centre Workshop 2018
15	Yves Pelletier	The Judge: A Statistical Validation Tool for Atmospheric Transport Modelling
16	Gary Perkins	Automated Gas Extraction System for the handling of radioactive waste gases from routine carbon-11 production
17	Eduardo Quintana	Chimney Emission Monitoring of RA-3 Research Reactor at EZEIZA Atomic Center
18	Kristin Shannon (Presented by Lori Metz)	Progress Toward an LEU Fuel Cycle for Domestic Radioisotope Production from Fission Fragments
19	Pouneh Tayyebi (Presented by Martin Kalinowski)	Can radioxenon emissions from nuclear research reactors interfere with nuclear explosion monitoring?
20	Momtaz Waheed	Current Status of Medical Radioisotope Production in Bangladesh
21	Matt Watrous	Radioactive Test Materials
22	Kassoum Yamba	On the usability of event zero time determinations using radioxenon isotopic activity ratios given the real atmospheric background observations

First Name	Last Name	Organization
Uwe	Ackermann	Austin Health, USA
Moshiul	Alam	Australia's Nuclear Science and Technology Organisation, Australia
Rainer	Auer	Instrumental Software Technologies, Inc, USA
Steven	Bell	National Physical Laboratory, UK
Helena	Berglund	Scienta Sensor Systems, Sweden
Steven	Biegalski	Georgia Institute Of Technology, USA
Andreas	Bollhöfer	Bundesamt Für Strahlenschutz, Germany
Theodore	Bowyer	Pacific Northwest National Laboratory, USA
Stuart	Brink	Australia's Nuclear Science and Technology Organisation, Australia
Richard	Britton	Atomic Weaspons Establishment, UK
Michael	Brown	Argonne National Laboratory, USA
Haryono	Budisantosa	Universitas Gadjah Mada, Indonesia
Jonathan	Burnett	Pacific Northwest National Laboratory, USA
lan	Cameron	Pacific Northwest National Laboratory, USA
Johan	Camps	Belgian Nuclear Research Centre (SCK•CEN)
Eduardo	Carranza	National Atomic Energy Commission of Argentina
Michael	Corry	Australia's Nuclear Science and Technology Organisation, Australia
Jagoda	Crawford	Australia's Nuclear Science and Technology Organisation, Australia
Ashley	Davies	Atomic Weaspons Establishment, UK
Andy	Delcloo	Royal Meteorological Institute Of Belgium, Belgium

## Appendix D – Workshop participants

Mariana	Di Tada	INVAP S.E., Argentina
Michael	Druce	Australia's Nuclear Science and Technology Organisation, Australia
Andreas	Enqvist	University of Florida, Florida
Timothy	Evans	U.S. National Nuclear Security Administration, USA
Judah	Friese	Pacific Northwest National Laboratory, USA
Sylvia	Generoso	French Alternative Energies and Atomic Energy Commission, France
Abdelhakim	Gheddou	Preparatory Commission for the Comprehensive Nuclear-Test- Ban Treaty Organization, Austria
Lee	Glascoe	Lawrence Livermore National Laboratory, USA
Ira	Goldman	Lantheus Medical Imaging, USA
Marcus	Grzechnik	Australian Radiation Protection And Nuclear Safety Agency, Australia
Christophe Rudy L.	Gueibe	Belgian Nuclear Research Centre, Belgium
Carolyn	Haass	Northwest Medical Isotopes, USA
Robert	Hague	Idaho National Laboratory, USA
Thomas	Hanlon	International Atomic Energy Agency, Austria
David	Hardman	Australian Radiation Protection And Nuclear Safety Agency, Australia
Sidney	Hellman	Instrumental Software Technologies, Inc, USA
Ryan	Hemsley	Australian Radiation Protection And Nuclear Safety Agency, Australia
Nikolaus	Hermanspahn	Preparatory Commission for the Comprehensive Nuclear-Test- Ban Treaty Organization, Austria
Bambang	Herutomo	Indonesia Nuclear Technology And Services, Indonesia
Ben	Heshmatpour	Defense Threat Reduction Agency, USA

lan	Hoffman	Health Canada, Canada
Emmy	Hoffmann	Australia's Nuclear Science and Technology Organisation, Australia
Miroslav	Hyza	National Radiation Protection Institute of the Czech Republic, Czech Republic
Yudiutomo	Imardjoko	Ensterna, Indonesia
Deconninck	Jacques R.	Institute for radioelements, Belgium
Guy	Jonkmans	Defence R&D Canada, Canada
Martin	Kalinowski	Preparatory Commission for the Comprehensive Nuclear-Test- Ban Treaty Organization, Austria
John	Kalish	Australian Safeguards And Non-Proliferation Office, Australia
Elizabeth	Keegan	Australia's Nuclear Science and Technology Organisation, Australia
Yuichi	Kijima	Japan Atomic Energy Agency, Japan
HYUNIL	КІМ	Korea Institute of Nuclear Safety, Korea
Steven	Kreek	Lawrence Livermore National Laboratory, USA
Jolanta	Kusmierczyk- Michulec	Preparatory Commission for the Comprehensive Nuclear-Test- Ban Treaty Organization, Austria
Gilbert	Le Petit	French Alternative Energies and Atomic Energy Commission, France
Denise	Lee	Oak Ridge National Laboratory, USA
Suseung	Lee	Korea Atomic Energy Research Institute, Korea
Thomas	Loosz	Australia's Nuclear Science and Technology Organisation, Australia
Oualid	Mahdaoui	Permanent Mission Of Algeria To The United Nations And Other International Organisations In Vienna, Austria
Ahmad	Malkawi	Jordan Atomic Energy Commission, Jordan
Olivier	Masson	Institut De Radioprotection Et De Sûreté Nucléaire, France

Christian	Maurer	Zentralanstalt fuer Meteorologie und Geodynamik, Germany
Lori	Metz	Pacific Northwest National Laboratory, USA
Luis António	Miguel Marques Barbosa	Curium Pharma, Netherlands
Dominique	Moyaux	Institute for radioelements, Belgium
Kylie	Olufson	Australia's Nuclear Science and Technology Organisation, Australia
Blake	Orr	Australian Radiation Protection and Nuclear Safety Agency, Australia
Giuseppe	Ottaviano	Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Italy
Yves	Pelletier	Meteorological Service Of Canada, Canada
Gary	Perkins	Australia's Nuclear Science and Technology Organisation, Australia
Neil	Petry	Duke University Medical Center, USA
Eduardo	Quintana	Autoridad Regulatoria Nuclear, Argentina
Terrill	Ray	U.S. Department of State, USA
Anders	Ringbom	Swedish Defence Research Agency, Sweden
Luis	Rudenas	Australia's Nuclear Science and Technology Organisation, Australia
Paul	Saey	International Atomic Energy Agency, Austria
Amanda	Sandifer	U.S. Air Force, USA
Jaclyn	Shafer	U.S. Air Force, USA
Marius	Stein	Mirion Technologies (Canberra), Inc., USA
Ulf Martin	Sundstrom	Scienta Sensor Systems, Sweden
Tammy	Taylor	Preparatory Commission for the Comprehensive Nuclear-Test- Ban Treaty Organization, Austria
Julio	Villafuerte	U.S. National Data Centre, USA

Momtaz	Waheed	Bangladesh Atomic Energy Commission, Bangladesh
Matthew	Watrous	Idaho National Laboratory, USA
Tami	Weber	Pacific Northwest National Laboratory, USA
Kassoum	Yamba	le Centre National de la Recherche Scientifique et Technologique, Burkina Faso
James	Zickefoose	Mirion Technologies (Canberra), Inc., USA

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