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A Review of Global Radioxenon Background Research and Issues

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Abstract—Among the most important problems for the worldwide nuclear explosion monitoring is the interference of naturally occurring and man-made radionuclides. The International Monitoring System (IMS) of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) frequently detects these interferences using sensitive radionuclide measurement equipment. We commonly refer to the presence of radionuclides that are relevant to the CTBT but do not originate from a nuclear explosion as "background". Backgrounds are highest near the sources but are known to have regional and global effects on the IMS. This review paper summarizes much of the relevant work in the area of background and discusses issues of interest for nuclear explosion detection.

Keywords: CTBT, radioxenon, nuclear explosion.

1. Introduction

The radionuclide monitoring component of the IMS is comprised of both radioactive particulate and radioactive xenon (radioxenon) noble gas measurements (see Fig. 1) (Schulze et al. 2000; Weiss et al. 2000; Auer et al. 2004). Radioxenon isotopes are the most highly sought signatures because they are easily vented and detected even from underground nuclear testing, they are produced in copious quantities from nuclear explosions, and are non-reactive in the environment. The radioxenon isotopes of interest for the CTBT: ^{131m}Xe ($\tau_{1/2}$ = 11.8 d); ^{133}Xe ($\tau_{1/2}$ = 5.2 d); ^{133m}Xe ($\tau_{1/2}$ = 2.2 d); ^{135}Xe ($\tau_{1/2}$ = 9.1 h) (ENSDF 2019) have half-lives in the range of several hours to nearly 2 weeks making them ideal for detection because they do not build-up in the atmosphere but are long enough lived to allow transport of thousands of kilometers before their decay. Atmospheric radioxenon measurement equipment has been especially designed for CTBT monitoring and several articles have written on the subject (Auer et al. 2010; Haas et al. 2017; Sivels et al. 2017; Cagniant et al. 2018; Ringbom et al. 2018).

Atmospheric radionuclide measurements for the IMS were developed starting in the 1990s to supply evidence that a suspect seismic event was of nuclear origin and to detect nuclear explosions, even absent a seismic trigger. During negotiations of the design of the IMS, however, concentrations of radionuclides and in particular radioxenon isotopes in the environment were not well understood or studied. In addition, it is likely that the airborne concentration and distribution of radioxenon isotopes have changed since that time due to evolving activities and locations associated with fission-based medical isotope production and nuclear power generation. However, the Protocol to the CTBT did explicitly refer to backgrounds and implied an issue that needed to be addressed. Therefore, the writers of the Treaty composed text referring to backgrounds of "man-made" (anthropogenic) radionuclides.[1] Following negotiations on the implementation of the verification regime specified in the Treaty and during the development and testing of radionuclide monitoring technology, it was found that radioxenon backgrounds were non-negligible and persistent in parts of North America and Europe despite their short half-lives (Bowyer et al. 1997, 2002; Weiss et al. 2000; Auer et al. 2004; Le Petit et al. 2008). After performing atmospheric transport model calculations (ATM) in the early 2000s, it was determined that sources were consistent

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[1] Annex 2 to the Protocol of the CTBT States "*For events detected by the International Monitoring System radionuclide component, the following parameters, inter alia, may be used: concentration of background natural and man-made radionuclides;…*".

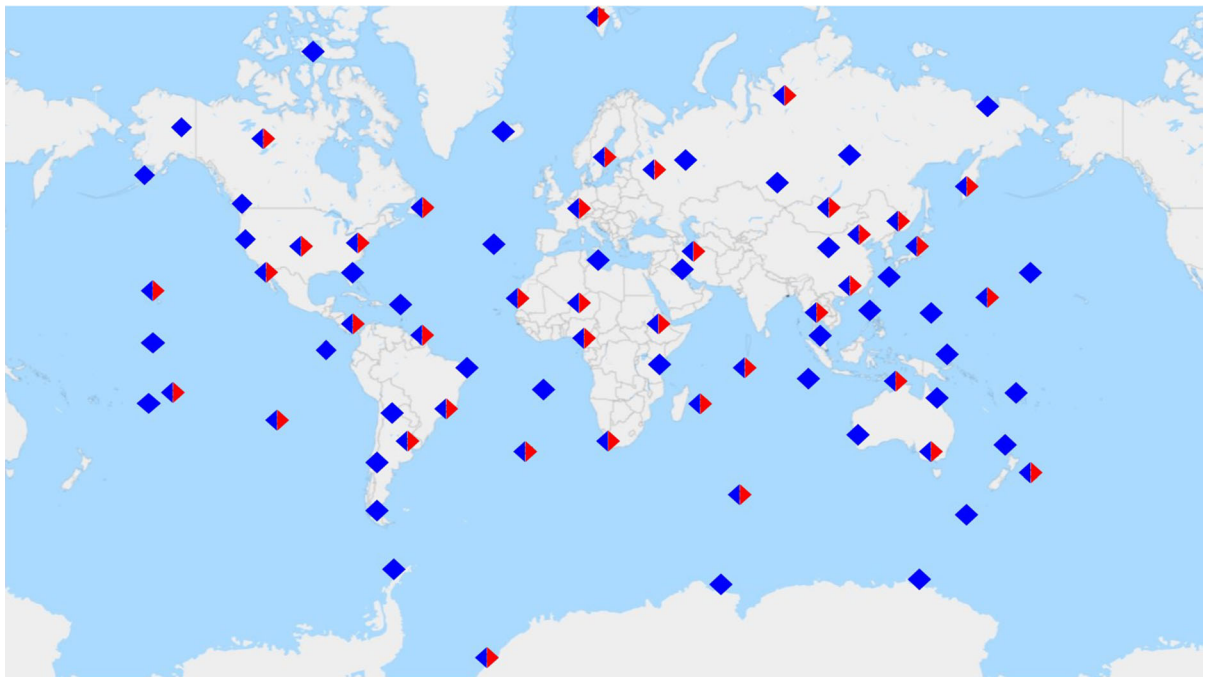

Figure 1
Schematic illustration of the current and planned locations of the particulate (blue diamonds) and combination radioxenon noble gas/particulate monitoring stations (blue/red diamonds) in the international monitoring system

with both nuclear power reactors and more distant sources which were hypothesized at the time to be medical isotope production facilities. As noble gas systems completed initial testing and were located at several locations around the world for further testing, it was soon realized that backgrounds were complex and largely originated from isotope production facilities. In some cases, the levels of background radioxenon isotopes were, and to this day remain, high enough to make detection of nuclear tests challenging (Hoffman et al. 2009; Saey 2009; Zahringer et al. 2009; Matthews et al. 2012; Saey et al. 2013).

2. *Mechanisms for Creating Background*

Studies in the early 2000's showed that radioxenon isotopes originated from fission-based isotope production and to a lesser degree, from nuclear power generation and research reactors operations. Xenon noble gases and other fission products are created and liberated into the environment primarily during the production of ^{99}Mo, which is used extensively across the world; see for example (Lee et al. 2016). In practice, in order to "screen-out" events detected by the IMS that are of peaceful origin, several techniques have been investigated to both distinguish types of events using isotopic signatures, and to mitigate the effect of the background in other ways. In 2009 a dedicated series of workshops entitled, "the Workshop on Signatures of Medical Isotope Production" (WOSMIP),[2,3] (see Fig. 2) was established and continues to this day (Matthews et al. 2010, 2012, 2013; Metz et al. 2014; Doll et al. 2015; Bowyer et al. 2017; Burnett 2018). The workshop was designed to bring the nuclear explosion monitoring and isotope production communities together

[2] After WOSMIP-5, the title of the workshop was renamed to the "*Workshop on Signatures of Man-made Isotope Production*" to better reflect the fact that many types of sources contributed to the background.

[3] WOSMIP reports can be found at http://www.wosmip.org/summary-reports.

Figure 2
Photo of participants at the WOSMIP meeting in Brussels

to better understand the radioactive signatures associated with man-made isotope production and to discuss ways to mitigate their effect on the IMS.

Following identification of the large effect that fission-based isotope production has on the IMS, dedicated studies and calculations (Kalinowski et al. 2010; Saey et al. 2010) have been performed to better understand the isotopic signatures from fission-based isotope production and better understand the emissions from these sources. It was quickly realized that the isotopic signatures from medical isotope production were different from those from nuclear power reactors, but unfortunately similar to nuclear explosions, depending on the specific and unique conditions in fission-based isotope production. It also became clear that more information was necessary to understand the isotopic signatures from medical isotope production so that the effect might be removed or accounted for in some way. In fact, not all fission-based isotope production locations are well known or documented to this day, nor are average emission levels from some key known facilities documented, leading to measurements that were (and remain to this day) confusing to understand.

In fission-based isotope production, the most frequently produced isotope used for a variety of medical procedures is ^{99m}Tc, the daughter of ^{99}Mo a high-yield fission product. It is created when a uranium target is irradiated in a nuclear reactor. Chemical processing of the U targets is performed as soon as practicable and the ^{99}Mo is extracted for subsequent extraction of ^{99m}Tc for medical procedures (Saey 2009). During the dissolution of the U targets used to create ^{99}Mo, xenon fission gases are released generally through an emission control system. In nuclear power generation and research reactors a similar process occurs in that U is irradiated (though usually for a much longer period), however, most of the radioxenon (Bowyer et al. 1998; Kalinowski and Pistner 2006) is retained within encapsulated fuel rods and hence the amount of emissions are far less even though the number of fissions and hence radioxenon creation is much higher.

In addition to persistent sources of radioxenon, a number of groups also measured the impact to the radioxenon background from events such as Chernobyl and Fukushima that released in excess of 10^{18} Bq into the environment (Pakhomov and Dubasov 2010; Bowyer et al. 2011; Masson et al. 2011; Sinclair et al. 2011; Biegalski et al. 2012; Orr et al. 2013; Wang et al. 2013; Woods et al. 2013;

Zhou et al. 2013; Achim et al. 2014; Eslinger et al. 2014; Xie et al. 2014). Since accidents are by their very nature intermittent and the half-lives of the radioxenon isotopes are short, the background caused by these events do not contribute to a permanent background that needs to be addressed.

3. *Effects of Background on Nuclear Explosion Detection*

The state-of-the-art for determining whether an explosion is of a nuclear nature via radioxenon isotopic measurements is to compare the ratios of several isotopes in a multi-isotopic plot, as shown in Fig. 3 derived from Kalinowski et al. (2010). The dashed nearly vertical line denotes a theoretical discrimination line between regions where events originating from nuclear reactors (left of the dashed line), nuclear explosions (grey shaded area), and fission-based isotope production (green shaded area) lay. Figure 3 assumes that at least three isotopes would be detected by an IMS station, and that the events are "pure", hence it is assumed in the calculation that measured samples originate from a single source. Of course, it is impossible for this to occur if there is persistent background, which is known to exist at many locations on earth. Where there are significant admixtures of types of events—air masses with isotopes originating from more than one source, the corresponding measured data will be altered.

Figure 4 shows the effect of an arbitrary amount of additional ^{133}Xe—presumably from an unknown background source—into the multi-isotope discrimination plot. Excess and unaccounted radioactive ^{133}Xe in measured samples will cause data points to be shifted from their true value (shown in blue) to a different value (shown in red) and so could cause a false positive if the amount of ^{133}Xe unaccounted for is large enough to cross the discrimination line shown in the figure. In other words, if a sample is collected and measured at an IMS station that is suspected as originating from a nuclear explosion, but the sample also has ^{133}Xe from other sources, that will tend to give false positives. Similarly is the case for the

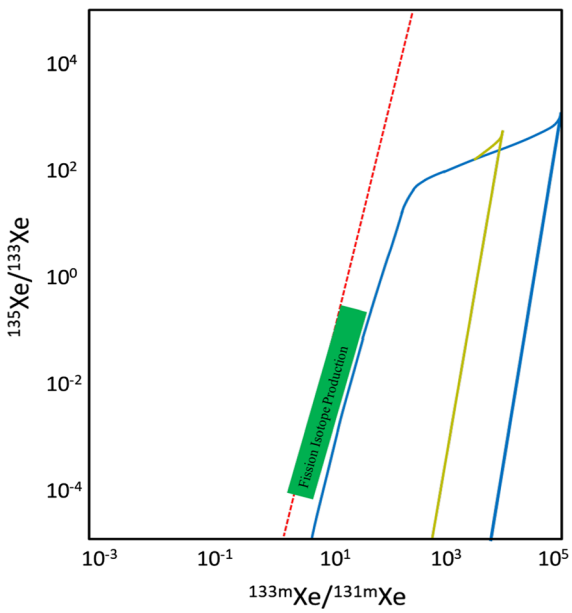

Figure 3
Theoretical calculation showing a dotted discrimination line separating the expected xenon ratios that would be measured following a nuclear explosion (between the blue lines), fission-based isotope production (green), and nuclear reactors close to equilibrium (left side of dotted line) using ^{133m}Xe/^{131m}Xe and ^{135}Xe/^{133}Xe ratios (from Kalinowski et al. 2010)

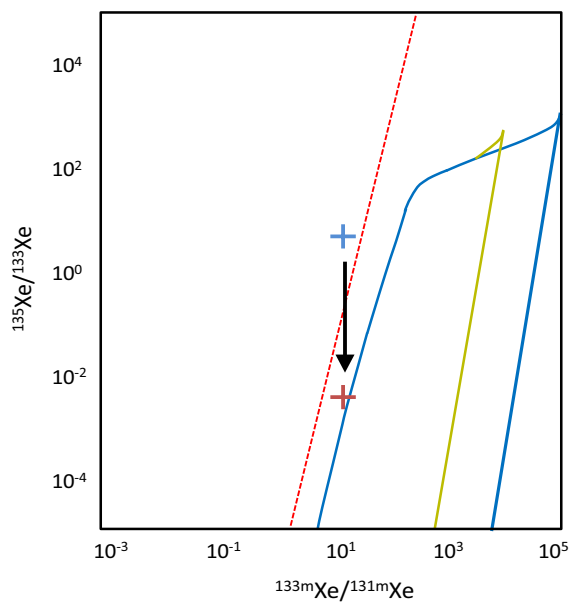

Figure 4
Calculation schematically (from Kalinowski et al. 2010) showing how a set of measured xenon ratios orginating from a nuclear explosion could be affected from the presence of a single background source of Xe-133. This could give rise to a false positive

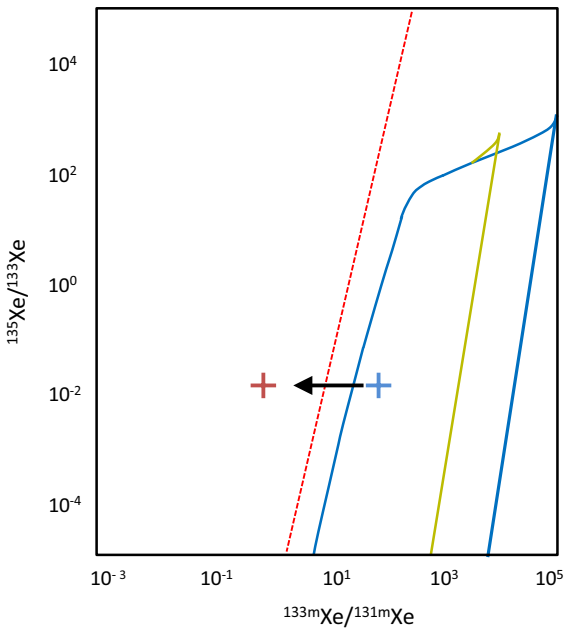

Figure 5
Calculation schematically (from Kalinowski et al. 2010) showing how a set of measured xenon ratios originating from a nuclear reactor could be affected from the presence of a large fission-based isotope production signal containing Xe-131m. This could give rise to a false negative

longer-lived ^{131m}Xe; Fig. 5 shows that data that may fall into the "nuclear explosion-like" side of the discrimination line could move into the reactor-like part of the discrimination plot if there is a large admixture of fission-based radioxenon (from ^{131m}Xe) that is measured at the same time, potentially causing a negative.

4. Emission Sources

Studies from several sources have shown that emission levels from nuclear power plants average around 10^9 Bq/day. In contrast fission-based isotope production levels vary between approximately 10^9 and 10^{13} Bq/day depending on a number of factors such as the dissolution chemistry, the amount of ^{99}Mo produced, off-gas treatment technology and procedures, and other factors (Kalinowski and Tuma 2009; Saey 2009; Saey et al. 2010a, b; Wotawa et al. 2010; Bowyer et al. 2013; Kalinowski et al. 2014; Achim et al. 2016; Gueibe et al. 2017). These levels are as large or larger than recent and historical releases from underground nuclear explosions which vary from less than 10^9 to 10^{15} Bq (Saey et al. 2007; Ringbom et al. 2009; Becker et al. 2010; De Meutter et al. 2017; Kurzeja et al. 2018). The effect of new man-made sources can be of particular concern in locations such as around the Korean peninsula as outlined in Bowyer et al. (2014) and Lee et al. (2016).

Background levels are known to fluctuate on several timescales. First, fission-based isotope production which dominates background radioxenon isotopes, produces plumes of radioxenon daily over the time period of around a week. Second, seasonal changes in weather patterns effect the backgrounds at most locations as weather patterns change. Thirdly, as the annual demand for ^{99m}Tc for medical procedures changes, the production levels change and hence the radioxenon background changes. Lastly, the locations of fission-based production changes on a longer timescale, such as the 2018 closing of the world's largest emission source at Chalk River, Canada from the Nordion isotope production company. In addition to fluctuations, it has been noted that the annual production of ^{99}Mo (^{99m}Tc) currently is increasing at a level of approximately 1.5% per year over 2015–2018 (OECD 2018).

5. Modeling Atmospheric Transport of Radioxenon Isotopes

Several groups have worked to improve the understanding of the sources of transport of radioxenon isotopes from the sources mentioned above. Global maps of the average concentrations expected across regions and across the world have been produced with varying levels of success in predicting the observed levels with the limited information on the actual releases from isotope production facilities (Matthews et al. 2010, 2012; Wotawa et al. 2010; Metz et al. 2014; Doll et al. 2015; Achim et al. 2016; Bowyer et al. 2017; Generoso et al. 2018).

Figure 6 shows a representative calculation of the *yearly average concentration* of radioxenon across the globe due to industrial sources (Achim et al. 2016). The actual concentration of the radioxenon isotopes at any location could be significantly

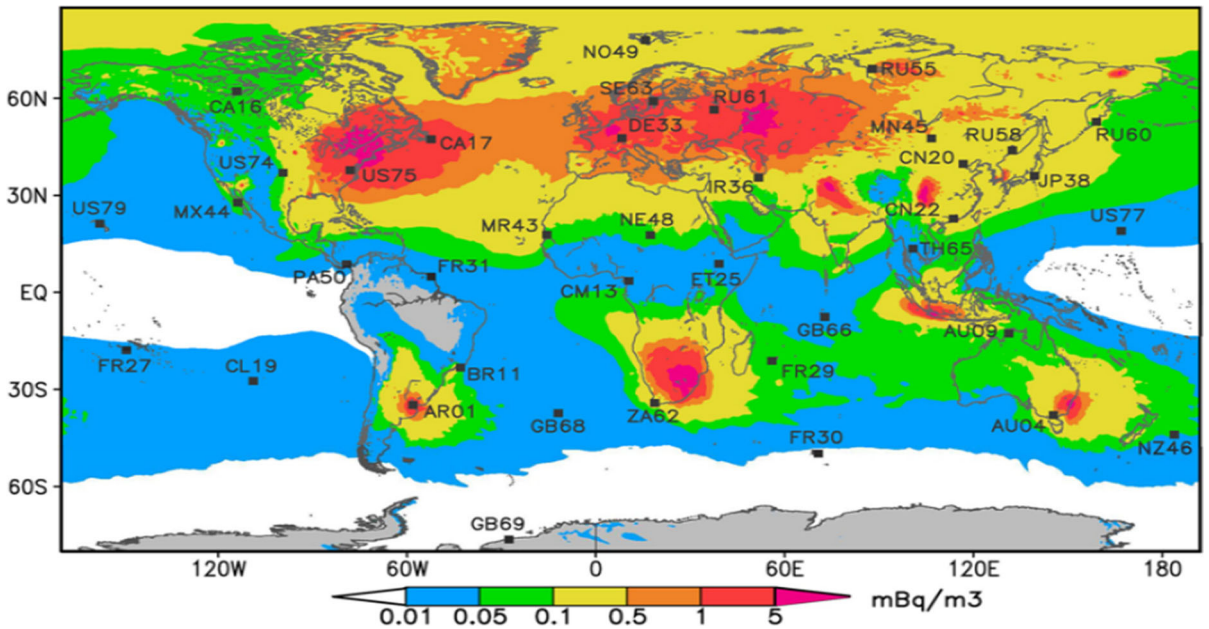

Figure 6
Plot of the average global concentration of Xe-133 from anthropogenic sources (Achim et al. 2016)

different on any 1 day and hence average values, such as those theoretically calculated and provided in the figure are used only to understand gross features, such as where the placement of sensors could be problematic. "True" concentrations of the xenon isotopes at a specific location, and the amount that might originate from a peaceful source and those from a nuclear explosion would require other information to determine the separate effect of a nuclear explosion.

While in the 1990s some may have believed that emissions from nuclear facilities were largely a localized effect, there have been a number of more recent studies showing the truly global effect of emissions, nominally due to the fact that emissions from isotope production can be large and radioxenon monitoring can be very sensitive. For example, (Hoffman et al. 2009; Kalinowski and Tuma 2009; Saey 2009; Zahringer et al. 2009; Wotawa et al. 2010; Matthews et al. 2012; Saey et al. 2013; Kalinowski et al. 2014; Achim et al. 2016; Hoffman and Berg 2018) have all studied the global effects of isotope production, including atmospheric transport modeling.

During discussions of the effect of fission-based medical isotope production on the IMS, calculations were completed (Bowyer et al. 2013) to answer the question posed by the isotope production community: "What is the maximum level of emissions that could be made by a production facility and have *no* appreciable affect on the IMS?" This work showed that on average emissions in the range of 1–10 GBq/day could be observed by the IMS in most circumstances and so a value of 5 GBq/day has subsequently been adopted by the community as a rule of thumb.

Additional work has begun to determine whether local measurements performed at facilities could provide high enough fidelity data to allow for the emissions from known facilities to be subtracted through ATM calculations. In 2015 the first "Atmospheric Transport Modeling Challenge" was created and held to determine whether stack monitoring would prove a successful solution (Eslinger et al. 2016), with follow-on exercises also conducted (Maurer et al. 2018). This first exercise showed that stack monitoring could allow for subtraction of the effect of even large background sources at IMS

stations. Following this exercise, a project, "Source Term Analysis of Xenon (STAX)" was initiated to purchase and install high-resolution stack monitoring systems to measure emissions locally and make this data available to improve IMS measurements (Friese 2019).

6. Mitigation Technology

Generically speaking and shown schematically in Fig. 1, the technique used for discrimination of nuclear explosions from other phenomena is the measurement of more than one radioxenon isotope and comparing the result to historically observed and theoretical calculations (Bowyer 1998; Finkelstein 2001; Biegalski et al. 2010; Kalinowski and Liao 2014; Gueibe et al. 2017; Galan et al. 2018). However, emissions from isotope production can exceed those from nuclear explosions and the isotopic signatures are similar (Saey 2010b; Bowyer et al. 2013). The most powerful technique is currently thought to be the comparison of the isotopic signatures from all four radioxenon isotopes (^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe) (Kalinowski et al. 2010). Unfortunately, due to the detection sensitivities of the current IMS measurement technology, often the concentration of one or more of the radioxenon isotopes present in the sample is below the systems detection limit. This means that in practice usually the four- and often the three-isotope discrimination plots shown in Fig. 3 cannot be used.

When measurements are made and we cannot use the multi-isotope discrimination technique, we must rely on the use of extensive atmospheric transport models to determine whether the detection of a signal is consistent with other information, such as a seismic trigger or the radioxenon emissions from known sources. As mentioned above, the STAX project allows for this information to be made available so that the effect of radioxenon emissions from isotope production can be subtracted from IMS measurements.

However, there will always be an intrinsic limit to how well the effect of a background signal can be subtracted from a gross signal, due in part to uncertainties in atmospheric models and other independent measurements of a background signal. For example, even if measurement of the emission at a facility is known perfectly and an "exact" model that precisely predicts the effect at a station were possible, subtraction of the effect will have some uncertainty due to statistical and systematic uncertainties of the nuclear detectors used in IMS stations. Therefore, high backgrounds will always cause sizable uncertainties in measurements made at IMS stations. Because of this, studies have begun to design systems that could be used to reduce emissions at the production location (Gueibe 2015; Lee et al. 2016), and other studies have been done such as exploring the use of tracers that could be deployed at facilities (Biegalski et al. 2013). Research and development is continuing in this area, but if mitigation technology is successfully implemented at one or more fission-based isotope production location, the effect on the IMS should be very beneficial.

Fortunately, although there can never be an exact solution, emissions can be subtracted from IMS measurements with an acceptable accuracy, even given the uncertainties mentioned above. The first ATM challenge showed that the effect of isotope emissions could be subtracted with stack monitoring and ATM models to an accuracy of 10–15%, significantly decreasing the effect of those emissions from peaceful, non-nuclear explosive sources (Eslinger et al. 2016).

7. Background Measurements Locations

Background measurements at various locations commenced well before the first noble gas system was installed at an IMS location. Some of the first background measurements completed include the Northeast United States and Europe (Bowyer et al. 1997; Auer et al. 2004) and important lessons were learned that started a major change to the thinking of radioxenon monitoring of the early 1990s. In these background measurement studies, it was learned that there was a large diversity of the combination of radioxenon isotopes that were detected in the IMS, and other specific examples of issues that were learned include:

- Emissions from large sources could be detected across the entire globe,
- Xe-135 could occasionally be detected with no other radioxenon isotopes detectable (likely arising from nuclear reactor start-up conditions, when Xe-135 concentrations can be 600 times greater than the other xenon isotopes), and that reactor start-up was not as rare a condition as expected in reactor-dense areas of the globe),
- Xe-131m could occasionally be detected with no other radioxenon isotopes detectable (possibly from old sources of radioxenon), and the production and use of I-131 for medical purposes.
- Theoretical calculations of sources such as fission-based isotope production facilities and nuclear power reactors did not agree with measurements at IMS locations, possibly due to either admixtures of many types of signatures and/or wrong assumptions about the inventories and release mechanisms of radioxenon from large sources.

These scientific discoveries and other questions lead to an interest in the scientific community in performing more measurements to improve the capability of the IMS in detecting nuclear explosions. Measurements were made, among other locations in:

- North America (Bowyer et al. 1997; Stocki et al. 2005, 2008; Milbrath 2007),
- Europe (Auer et al. 2004; Saey et al. 2006, 2010c; Bieringer et al. 2009; Achim et al. 2016; De Meutter et al. 2016, 2018),
- The South Pacific (Stocki et al. 2005),
- Russia (Dubasov and Okunev 2010),
- South Asia (Saey et al. 2013; Eslinger et al. 2015),
- Kuwait (Saey et al. 2013),
- South Korea (Bowyer et al. 2014),
- Western Africa.

8. *Other Effects of Background*

While the primary focus for the measurement of backgrounds are to understand and mitigate the effect of background sources on IMS measurements, several studies have also been conducted on the effect of high levels of background on gases that could emanate from subsurface during an on-site inspection (OSI). These measurements found that radioxenon that is present in the atmosphere could diffuse into the subsurface and contaminate measurements of samples meant to be collected from the subsurface (Johnson et al. 2015, 2017; Lowrey et al. 2016). This would suggest that surface measurements during OSIs are needed to subtract the imprinting effect of surface air into subsurface measurements especially at high background level locations.

9. *Summary and Next Steps to Address the Problem of Backgrounds in the Environment*

The "holy grail" of radionuclide detection would be to have full knowledge of the background of every isotope in the atmosphere in each cubic meter of air at any time and these values compared against actual IMS measurements, and a residual calculated for each measurement that would be indicative of a nuclear explosion. In theory, this goal could be achieved if all sources of radionuclides (and for the purpose of this paper, we consider radioxenon) were known, including the temporal profile of releases, their isotopic concentrations, continuous and accurate IMS measurements, and a near perfect atmospheric transport model. In practice, there are only a few anthropogenic sources that dominate the background, and so it is not out of the question that a useful model could be created and used operationally, if computational power continues to improve.

In order to reach some kind of usable tool, a number of political, operational and research issues would need to be addressed including:

- A rather complete knowledge of every sizeable emitter of atmospheric radioxenon would be required;
- Stack monitoring such as the STAX project should be implemented at more locations with sufficient accuracy and the temporal resolution clearly understood;
- Continued R&D in atmospheric transport modeling, especially to understand uncertainties in the models;

- A recursive model should be created to fold IMS measurements to adjust source emissions or ATM calculations in a self-consistent way; and
- An operational tool that is not overly computationally intensive needs to be developed and tested.

While the bulleted list above is a goal, some uncertainty exists on whether all the political hurdles can be overcome to improve IMS measurements, as stack monitoring is not a compulsory activity at (usually) commercial facilities. Until all these hurdles can be overcome, an alternate list of items can be addressed to help improve the monitoring situation:

- Operators of facilities that emit large amounts of radioxenon should be encouraged to voluntarily report their emissions using a stack monitoring system;
- Better understanding of the expected isotopic releases from a variety of sources is needed;
- Improved temporal and sensitivity measurements at IMS stations should be explored;
- Better ATM models should be explored to reduce uncertainties;
- Known emitters of large concentrations of radioxenon isotopes should be encouraged to voluntarily reduce emissions to levels that do not appreciably affect the IMS; and
- Measurements of backgrounds of radioxenon isotopes should be pursued across the globe.

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