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Improved performance comparisons of radioxenon systems for low level releases in nuclear explosion monitoring

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ABSTRACT

The Comprehensive Nuclear-Test-Ban Treaty bans all nuclear tests and mandates development of verification measures to detect treaty violations. One verification measure is detection of radioactive xenon isotopes produced in the fission of actinides. The International Monitoring System (IMS) currently deploys automated radioxenon systems that can detect four radioxenon isotopes. Radioxenon systems with lower detection limits are currently in development. Historically, the sensitivity of radioxenon systems was measured by the minimum detectable concentration for each isotope. In this paper we analyze the response of radioxenon systems using rigorous metrics in conjunction with hypothetical representative releases indicative of an underground nuclear explosion instead of using only minimum detectable concentrations. Our analyses incorporate the impact of potential spectral interferences on detection limits and the importance of measuring isotopic ratios of the relevant radioxenon isotopes in order to improve discrimination from background sources particularly for low-level releases. To provide a sufficient data set for analysis, hypothetical representative releases are simulated every day from the same location for an entire year. The performance of three types of samplers are evaluated assuming they are located at 15 IMS radionuclide stations in the region of the release point. The performance of two IMS-deployed samplers and a next-generation system is compared with proposed metrics for detection and discrimination using representative releases from the nuclear test site used by the Democratic People's Republic of Korea.

1. Introduction and background

The Comprehensive Nuclear-Test-Ban Treaty (Comprehensive Nuclear-Test-Ban Treaty, 1996) bans all nuclear tests and mandates the implementation of verification measures to detect treaty violations. One verification measure is the detection of radioactive xenon isotopes produced in the fission of actinides. The International Noble Gas Experiment (INGE) was initiated in 1999 to determine the feasibility of building and deploying automated systems to detect the four main radioxenon isotopes of interest: ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe (Auer et al., 2010; Bowyer et al., 2002; CTBTO, 2016).

Four radioxenon detection systems were developed for the INGE: 1) Automatic Radioanalyzer for Isotopic Xenon (ARIX), from the Khlopin Radium Institute, Russia (Dubasov et al., 2005), 2) the Automated Radioxenon Sampler-Analyzer (ARSA), from the Pacific Northwest National Laboratory (PNNL), United States (McIntyre et al., 2001), 3) the Swedish Automatic Unit for Noble Gas Acquisition (SAUNA), from Totalförsvarets Forskningsinstitut (FOI), Sweden (Ringbom et al., 2003), and 4) the Système de Prélèvement d'Air Automatique en Ligne avec l'Analyse radioXénons atmosphériques (SPALAX) from Departement Analyse, Surveillance, Environnement du CEA (CEA/DASE), France (Fontaine et al., 2004). These prototype systems were tested in a comparison exercise in 2002 (Auer et al., 2004) and production versions of ARIX, SAUNA and SPALAX make up the current International Monitoring System (IMS) noble gas network. The IMS noble gas network is currently planned to consist of 40 stations with the possibility of expanding

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to 80 stations.

When the INGE was started, developers had little information on atmospheric radioxenon backgrounds and designed the systems with a focus on detecting ^{133}Xe because it was the isotope most likely to be detected from a nuclear test. The design basis for the radioxenon network performance was a 90% probability of detecting a 10^{14} Bq release of ^{133}Xe from a 1 kt TNT equivalent explosion (CD/NTB/WP.224, 1995). Since ^{133}Xe is produced with high fission yields for ^{235}U and ^{239}Pu, one of the requirements set for these systems was to be able to detect ^{133}Xe in concentrations of 1 mBq/m^3 in the atmosphere. All of the systems met this requirement, which is now recognized by the monitoring community as being not sufficiently discriminating. Subsequent measurements at locations around the world showed frequent detections above the respective system minimum detectable concentrations (MDCs) which are now known to be background and not nuclear explosions.

Investigation into the sources of these detections found that the production of medical isotopes at a few facilities around the world caused the majority of the radioxenon background, not nuclear power plants as expected (Bowyer et al., 2013; Saey, 2009; Saey et al., 2010a, 2010b, 2010c, 2012, Stocki et al., 2008). This is due to the method of irradiating uranium targets for a few days and dissolving them in order to chemically isolate the isotopes needed for medical diagnostics and treatments, such as ^{99}Mo, which decays to ^{99m}Tc. During this process, fresh gaseous fission products - including radioxenon isotopes - are released into the atmosphere. The understanding of the scope of the background of radioxenon isotopes led to efforts to use ratios of different isotopes to distinguish between those produced through medical and industrial applications and those produced in a nuclear test (Hoffman et al., 2013; McIntyre et al., 2006).

The use of isotopic ratios of radioxenon to identify detections as resulting from nuclear tests, or nuclear reactor related sources is well documented in the literature (Kalinowski et al., 2010). Radioxenon detections and the isotopic ratios measured following the 2010 Fukushima nuclear accident were consistent with the failure of cladding of nuclear fuel from the power reactors operating until the time of the earthquake and tsunami that triggered the accident (Bowyer et al., 2011; Eslinger et al., 2014a). Other detections of radioxenon and their isotopic ratios in the region were consistent with potential releases from the underground nuclear tests declared in 2006 and 2013 by the Democratic People's Republic of Korea (DPRK) (Ringbom et al., 2009, 2014).

Releases of radioactivity to the atmosphere from small underground nuclear explosions can be within the range of routine releases from nuclear power plants or medical isotope production facilities (Becker et al., 2010; Bowyer et al., 2013; Eslinger et al., 2014b; Saey, 2009). Although there is a large and growing body of literature about releases of radioxenon to the atmosphere from civilian nuclear facilities, there have been few nuclear explosions since the INGE was initiated. Atmospheric radioxenon was detected after the underground nuclear tests in 2006 and 2013 by the DPRK (Ringbom et al., 2009, 2014); however, no radioxenon was detected above normal background levels following the announced tests in 2009 and 2016. The magnitude of the 2006 test by the DPRK was estimated to be on the order of 0.5 kt (Zhao et al., 2008), while the magnitude of the 2013 test was estimated at 12.2 kt (Zhang and Wen, 2013). Other yield estimates have been published (Barth, 2014; Zhao et al., 2016). These facts imply that the original design basis for the noble gas network of 10^{14} Bq ^{133}Xe produced in a 1 kt nuclear test is not sufficiently challenging for a modern network to monitor real-world nuclear explosion releases in terms of sensitivity or discrimination. In this paper we propose an alternative release profile and metrics to evaluate system performance. Then we evaluate the relative performance of three sampling and measurement systems using these metrics in conjunction with a release profile that is more realistic than the original design basis release profile.

2. Approach and models

Analysis of the radioxenon data collected around the times of the four underground nuclear tests announced by DPRK seeks to address several questions that can be posed in the following general form: 1) Did a nuclear event occur? 2) What was the event type (e.g., reactor operations, medical isotope production, or explosion)? 3) What was the magnitude of the release? 4) What was the location of the release? Comparison metrics between different types of radioxenon samplers should be designed to help elucidate these general questions.

2.1. Proposed comparison metrics

We propose the following metrics:

1. Detection – the probability that at least one sampling station detects at least one of the four xenon isotopes
2. Discrimination – (a) the probability that multiple xenon isotopes are detected in at least one sample and (b) the relative uncertainty in isotopic ratios used to distinguish the type of release event
3. Magnitude – the probability the estimated release magnitude for ^{133}Xe is within a factor of 5 of the modeled release magnitude
4. Location – the probability the estimated release location is within 100 km of the modeled release location.

We suggest that comparison of the performance of sampling systems should be based on one or more representative releases. The initial release scenario for underground testing for the International Monitoring System (CD/NTB/WP.224, 1995) was venting of 10^{14} of ^{133}Xe over a 12-h period (a 10% release of a 1 kt test).

In our analysis, the representative release assumes: 1) an underground nuclear explosion occurs with a 10 kt TNT equivalent yield from fission-spectrum neutron induced fission of ^{235}U, 2) radioxenon isotope release to the surface begins 1 h after the explosion and continues for 1 h, and includes ingrowth from iodine prior to release,3) 1% of the radioxenon isotopes that are produced vent to the atmosphere in the 1 h period, and 4) no background sources of radioxenon interfere with the signal from the representative release. Typical release amounts to the atmosphere for four xenon isotopes are provided in the column of Table 1 titled “1% Vent” using these assumptions. Values for a 0.1% vent and a 10% vent of xenon isotopes of a 10 kt explosion are also included for analysis purposes. The release values for the 1% vent beginning 1 h after the explosion and ending 2 h after the explosion are a factor of

Table 1
Representative release amounts of xenon isotopes as a function of the fraction of radioxenon vented to the atmosphere from a 10 kt TNT equivalent explosion after a 1 h holdup. Values based on independent yield of ^{235}U fission (from (Chadwick et al., 2011)).

Isotope	0.1% Vent (Bq)	1% Vent (Bq)	10% Vent (Bq)
^{131m}Xe	1.15×10^{8}	1.15×10^{9}	1.15×10^{10}
^{133}Xe	6.00×10^{11}	6.00×10^{12}	6.00×10^{13}
^{133m}Xe	6.72×10^{10}	6.72×10^{11}	6.72×10^{12}
^{135}Xe	7.02×10^{13}	7.02×10^{14}	7.02×10^{15}