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# Potential impact of releases from a new Molybdenum-99 production facility on regional measurements of airborne xenon isotopes

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ABSTRACT

The monitoring of the radioactive xenon isotopes $^{131m}Xe$, $^{133}Xe$, $^{133m}Xe$, and $^{135}Xe$ is important for the detection of nuclear explosions. While backgrounds of the xenon isotopes are short-lived, they are constantly replenished from activities dominated by the fission-based production of $^{99}Mo$ used for medical procedures. At present, one of the most critical locations on earth for the monitoring of nuclear explosions is the Korean peninsula where the Democratic People's Republic of Korea (DPRK) has announced that it conducted three nuclear tests between 2006 and 2013. This paper explores the backgrounds that would be caused by the medium to large scale production of $^{99}Mo$ in the region of the Korean peninsula.



## 1. Introduction

The International Monitoring System (IMS), designed for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), is used to detect nuclear explosions no matter where they occur on earth. One of the primary methods used in the IMS (Bowyer et al., 2002; Kalinowski et al., 2010; Wernsperger and Schlosser, 2004) is the detection of radioactive xenon isotopes ($^{131m}Xe$, $^{133}Xe$, $^{133m}Xe$, and $^{135}Xe$).

As stated in several papers over the last decade (Becker et al., 2007; Kalinowski et al., 2010; Matthews et al., 2012; Saey et al., 2010a,b), there is a growing concern over the radioxenon emissions caused by the production of fission-based $^{99}Mo$, which is milked for $^{99m}Tc$. Technetium-99m is used in approximately 30 million medical procedures per year, or approximately one per second. Fission-based production of $^{99}Mo$ also produces $^{133}Xe$ and other radioxenon isotopes that are also useful for the detection of nuclear explosions. Since the cumulative fission yields of both $^{99}Mo$ and $^{133}Xe$ are approximately 6%, and the half-lives are also similar (2.75 versus 5.2 days), their activity a few days following fission is comparable. According to the Nuclear Energy Agency Organisation for Economic Co-operation and Development (OECD), in 2010 the demand for $^{99}Mo$ production was on the order of $1.2 \times 10^4$ Ci ($3.7 \times 10^{14}$ Bq) (OECD, 2010). At this level of $^{99}Mo$ production via fission, about $8.1 \times 10^{14}$ Bq of $^{133}Xe$ would also be produced per week.

To determine the impact on monitoring for nuclear explosions, the weekly production of $^{133}Xe$ from isotope production source should be compared with the expected amount of $^{133}Xe$ vented from an underground nuclear explosion. Unfortunately for this calculation, the amount of $^{133}Xe$ that may be vented from a 1-kiloton test can vary from 0 to approximately $10^{16}$ Bq, depending on the success of the stemming of the containment scheme. However, we do know that the International Monitoring System detected radioxenon following announced DPRK nuclear tests in 2006 and 2013. Assuming that in these cases a test occurred and vented from the Punggye-ri claimed nuclear test site, the amount of vented material would have been in the range of $10^{12}$ Bq of $^{133}Xe$ (Ringbom et al., 2014). It is, therefore, evident from this calculation and from the papers from other authors (Bowyer et al., 2013; Saey et al., 2010b) that the release of radioactive xenon from fission-based $^{99}Mo$ production can cause significant backgrounds to the detection of nuclear explosions. It is also evident that the backgrounds close to the source will be much higher than those from a distance.

In a recent publication by some of the authors of this publication (Bowyer et al., 2013), we estimated that for the monitoring

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community the maximum tolerable amount of emissions from fission-based medical isotope production is likely to be in the range of $5 \times 10^9$ Bq/d. Since that publication, we have become aware of a planned fission-based medical isotope production operation in Pusan, Republic of Korea. Due to the importance of the monitoring efforts on the Korean peninsula, we have studied the potential impact that radioxenon emissions from fission-based $^{99}$Mo production in Republic of Korea could have on the detection of nuclear explosions using radioxenon isotopes.

## 2. Methods

In order to calculate the possible effect of fission-based $^{99}$Mo medical isotope production on the $^{133}$Xe background on the Korean peninsula, we have had to make a set of assumptions. The current plans are for the production of approximately 2000 6-day Ci per week $^{99}$Mo using fission at a new facility in Pusan, Republic of Korea (35.3° N, 129.3° E) using an alkaline chemistry target dissolution technique (Jung, 2013). Radioxenon releases from fission-based $^{99}$Mo medical isotope production operations vary by orders-of-magnitude. From Table 2 in Saey (2009), it is evident that radioxenon emissions per Ci of $^{99}$Mo created can vary by a factor of approximately $10^4$. In this paper, no assumption is explicitly made about the *likely* emissions from fission-based $^{99}$Mo production in Republic of Korea. Instead, we evaluate the effect of emissions of magnitudes varying between approximately $10^9$ and $10^{13}$ Bq per day, which represents the range of emissions observed in the industry so far. We used historical meteorological data to estimate the concentrations at various grid (map) points by tracking hypothetical emissions from a location in Republic of Korea using standard atmospheric transport modeling codes (see below).

While the calculations of the concentrations at each grid point in and around the Korean peninsula were made using a straightforward application of widely available transport code, the data post processing, display, and interpretation of the results, however, were not as straightforward. One of the challenges was to present results of this analysis in a way that conveyed the verification challenges associated with the detection of radioxenon monitoring for nuclear explosions. Listed below are the options we considered for a figure-of-merit for this analysis:

(a) Daily, weekly, monthly, or annual average concentrations at each grid point,
(b) Maximum calculated daily concentrations of $^{133}$Xe for various releases,
(c) Number of detections per year at the IMS station at various release levels, and
(d) Number of times per week, per month, or per year a grid point has concentrations over a threshold at various release levels.

Each of the data post processing options listed above has advantages and disadvantages in visualizing the effect of fission-based $^{99}$Mo production. But in reality, the impact on monitoring efforts on the Korean peninsula is partially subjective.

In option (a), where an average concentration is computed for each grid point such as a monthly or annual average, values may tend to be washed out, especially if the location has highly variable winds. This option may be more useful if averages are computed seasonally, but does not directly give any information on the frequency of events being detected. Although option (b) has been used in our previous assessments (Bowyer et al., 2013) of the effect of medical and industrial isotope production, for this effort, we found that determining the maximum concentration only tells part of the verification story. It does not give the reader any information on frequency of detections, which from the perspective of explosion detection, can be as significant.

We have found that options (c) and (d) seem to be the most useful from the perspective of monitoring, although both options ultimately require some value judgment on the number of medical isotope emission background events one is willing to accept. While option (c) is most relevant to the IMS, it is in fact a subset of option (d), which is a more general analysis of the effect of emissions from fission-based $^{99}$Mo production. In our final product, we have chosen to present results in terms of the number of times per year a grid point (in the form of a map) has concentrations over a threshold at the detection levels typical for IMS radioxenon systems ($\leq$0.2 mBq/m$^3$ for $^{133}$Xe) (Auer et al., 2004).

Additionally, to show the effect of fission-based medical isotope production in the region at a specific IMS location, we used historical weather to calculate the expected concentration from medical isotope production at the Takasaki, Japan radioxenon station JPX38 and compared it to the historical radioxenon levels. The Takasaki station has a SAUNA radioxenon sampling system installed and it has measured radioxenon concentrations for a continuous period over several years and was used in the detection of a radioxenon event 55 days following the announced 2013 DPRK nuclear test (Ringbom et al., 2014).

## 3. Our calculations

In our model, we employed the HYSPLIT model (Draxler and Hess, 2010; Draxler et al., 2012) available at http://www.arl.noaa.gov/HYSPLIT.php. We performed a 1° × 1° grid spacing atmospheric transport calculation for 20-m height hypothetical releases 5 days per week at various release levels, with 3-h time resolution, a 3-h assumed emission duration, and with concentrations calculated in the lower 100 m of the atmosphere. From this series of calculations, we determined the expected concentrations every day for 14 days following the release at grid points around the Korean peninsula. The atmospheric data used for our calculations are based on three years of observations (2008–2010) using the GDAS1 global data assimilation system weather database. Although no fission-based $^{99}$Mo production facility currently exists, we have calculated the effect from production in both Taejon and Pusan. For the purposes of this paper, we have only shown results for emissions from Pusan since the difference in remote monitoring between the two locations is minimal outside of the immediate vicinity.

Fig. 1 shows a map of the number of days per year in which either of the 12-h intervals during a day had a calculated $^{133}$Xe concentration exceeding a concentration of 0.2 mBq/m$^3$ for a release size of $10^{12}$ Bq/d and five batches per week. It is clear that at many locations as many as 50% of the days have detections using our set of assumptions, although the station at Takasaki is affected much less severely. The effect on the Takasaki station, however, may be explained by the local mountainous topography in Japan which incidentally also makes the monitoring location less sensitive to nuclear tests on the Korean peninsula.

In our analysis, we have also used several locations around the peninsula to estimate the number of annual detections, using the same criteria as above, except as a function of the emission strength. The results shown in Fig. 2 display an interesting trend; the number of detections increases linearly with exponentially increasing emission strengths over the range of $10^{10}$ to $10^{12}$ Bq/d. This effect may be expected. However, the expected cumulative lognormal frequency distribution of concentrations of pollutants at a distant location using the assumption of successive random dilutions is exponential (Ott, 1995).

At $^{133}$Xe release levels of $10^{12}$ Bq/d, our calculations show that much of the region outside of the peninsula appears to be affected