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Estimates of radioxenon released from Southern Hemisphere medical isotope production facilities using measured air concentrations and atmospheric transport modeling

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ABSTRACT

The International Monitoring System (IMS) of the Comprehensive-Nuclear-Test-Ban-Treaty monitors the atmosphere for radioactive xenon leaking from underground nuclear explosions. Emissions from medical isotope production represent a challenging background signal when determining whether measured radioxenon in the atmosphere is associated with a nuclear explosion prohibited by the treaty. The Australian Nuclear Science and Technology Organisation (ANSTO) operates a reactor and medical isotope production facility in Lucas Heights, Australia. This study uses two years of release data from the ANSTO medical isotope production facility and ^{133}Xe data from three IMS sampling locations to estimate the annual releases of ^{133}Xe from medical isotope production facilities in Argentina, South Africa, and Indonesia. Atmospheric dilution factors derived from a global atmospheric transport model were used in an optimization scheme to estimate annual release values by facility. The annual releases of about 6.8×10^{14} Bq from the ANSTO medical isotope production facility are in good agreement with the sampled concentrations at these three IMS sampling locations. Annual release estimates for the facility in South Africa vary from 2.2×10^{16} to 2.4×10^{16} Bq, estimates for the facility in Indonesia vary from 9.2×10^{13} to 3.7×10^{14} Bq and estimates for the facility in Argentina range from 4.5×10^{12} to 9.5×10^{12} Bq.

1. Introduction and background

The aim of the Comprehensive Nuclear-Test-Ban Treaty (CTBT, 1996) is to eliminate nuclear explosions. One of the main verification provisions of the CTBT (CTBTO, 2013) is the International Monitoring System (IMS), which includes a network of stations that have radioactive xenon monitoring capability. As of 2013, 31 IMS stations have xenon capability in provisional operation to study and improve the performance of the verification provisions in preparation for the entry into force of the treaty.

In addition to nuclear explosions, nuclear power plants and medical isotope production facilities release radioxenon (Hoffman et al., 2009; Kalinowski et al., 2008; Saey et al., 2010; Wotawa et al., 2010). Medical isotope production facilities may release orders of magnitude more radioxenon than nuclear power plants. A typical order of magnitude release from a nuclear power plant is $\sim 10^9$ Bq/d of ^{133}Xe, while medical isotope production facilities may release $\sim 10^{11} - \sim 10^{13}$ Bq/d (Saey, 2009). These sources are large enough that they form a background of radioxenon that can be detected at many places around the world, and those releases could be confused with evidence of a nuclear explosion. Thus, releases from these sources must be factored into verification activities. Reduction of future releases of radioxenon from medical isotope production facilities is one way to mitigate potential impacts of background radioxenon on treaty verification activities (Bowyer et al., 2013) where one may be seeking to identify the location of a single release of the same size as the average release from the South Africa medical isotope production facility on any given day (Ringbom et al., 2009). Other methods include estimating the impact of known releases on each verification measurement in an attempt to separate the influence of background radioxenon from

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radioxenon potentially released in a nuclear explosion. This work shows that it is possible to estimate long-term releases from medical isotope facilities, but it also illustrates the difficulty in accurately matching any single measurement with the release or releases that caused the measurement.

The Australian Nuclear Science and Technology Organisation (ANSTO) operates a reactor and medical isotope production facility in Lucas Heights, Australia. The medical isotope production facility (hereafter called the ANSTO facility) produces more than 500,000 patient doses of radiopharmaceuticals each year (ANSTO, 2013) and currently has the capacity to produce 1000 6-d Ci of ^{99}Mo per week (OECD, 2012). A recent study (Schöppner et al., 2013) modeled atmospheric transport of time-based releases of ^{133}Xe from the ANSTO facilities over a several-year period and explored the effect of the releases on samples collected at two IMS stations. One of the stations is in Melbourne, Australia (denoted by AUX04), about 700 km southwest of the production facility. The other station, on Chatham Island in New Zealand (denoted by NZX46), is 3000 km southeast of the ANSTO facility. One of the conclusions in the study by Schöppner et al. (2013) is that these two stations routinely detect ANSTO emissions, but the ANSTO emissions do not account for all of the ^{133}Xe in the samples.

Atmospheric transport runs performed for the work reported here indicated that releases of ^{133}Xe from the ANSTO facility can also influence samples collected at the IMS station in Darwin, Australia (denoted by AUX09), even though this station is about 3100 km northwest of the ANSTO facility. Transport model results indicate that samples taken at AUX09 can also be influenced by releases from three other medical isotope production facilities in the Southern Hemisphere. Although contaminants released to the air in the northern hemisphere can mix across the equator into the southern hemisphere, such mixing is general slow enough that releases of isotopes of interest in the northern hemisphere generally decay to negligible levels before reaching sampling stations in the southern hemisphere. Thus, this study only considers releases from medical isotope facilities in the southern hemisphere. One of these facilities is in Batan Serpong, Indonesia. It has the capacity to produce 100 6-d Ci of ^{99}Mo per week (personal communication), but some news reports indicate it is seeking to raise production to 900 6-d Ci of ^{99}Mo per week (Prasetyo, 2012). There are no published release numbers for this facility, but (Schöppner et al., 2013) suggest that releases from this location are less than 20% of the releases from ANSTO. Another facility in Pelindaba, South Africa, is run by the Nuclear Technology Products (NTP) division of the South African Nuclear Energy Corporation. It is a major supplier of radiopharmaceuticals and has a production capacity of 3000 6-d Ci of ^{99}Mo per week (OECD, 2012). Some researchers have estimated this facility released about 4.1×10^{15} Bq of ^{133}Xe in 2008 (Saey, 2009). The third facility, run by the National Atomic Energy Commission (CNEA) of Argentina, is near located Buenos Aires. It has a capacity of 900 6-d Ci of ^{99}Mo per week (OECD, 2012), and has reported emissions of 6.36×10^{12} Bq of ^{133}Xe in 2010 and 5.65×10^{12} Bq in 2011 (Carranza et al., 2013). The locations of these medical isotope production facilities and the IMS sampling locations with operational radioxenon samplers are shown in Fig. 1. The operators of the ANSTO facility have shared detailed stack-monitoring data with the treaty-monitoring community. The purpose of this study was to estimate the releases from the three Southern Hemisphere facilities that do not publish stack-monitoring data.

2. Data and models

2.1. Sampling data

Estimation of the long-term releases from medical isotope production facilities using transport modeling is predicated on the availability of long sequences of sampled data. This study used ^{133}Xe concentration data collected between August 30, 2009 and January 31, 2012, at three sampling locations. The stations are at Melbourne, Australia, (AUX04), Darwin, Australia, (AUX09) and Chatham Island, New Zealand (NZX46). The xenon samplers at these locations are Swedish Automatic Units for Noble gas Analysis (SAUNA) units developed at the Swedish Defense Research Agency (Ringbom et al., 2003). These samplers extract xenon from the atmosphere and convert it into liquid form before determining the amount of radioactivity in the sample. They use a 12-h xenon collection period and report two concentration estimates per day.

The prevailing winds general move releases from the four medical isotope production facilities towards the east. This suggests that measurements taken at four other sampling locations (FRX29, FRX30, GBX66 and GBX68) shown on Fig. 1 might be useful in this analysis. Of these four stations, no data were available from FRX30 and GBX68 in the time frame of interest, and only 14 samples were available from FRX29. After initial quality screening, there were 492 samples available from GBX68 from December 2010 through December 2011. These data were excluded from the analysis because they did not cover the entire period of interest.

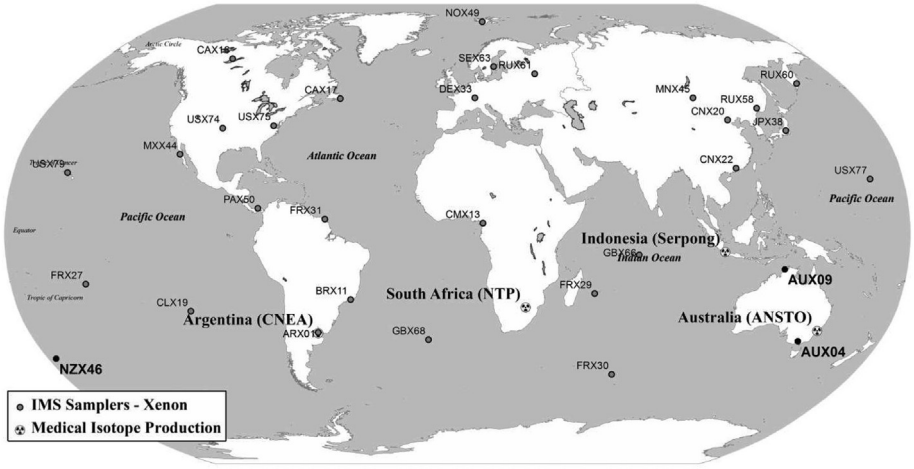

Fig. 1. Global operational IMS noble gas sampling locations and Southern Hemisphere medical isotope production facilities.