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Source term estimates of radioxenon released from the BaTek medical isotope production facility using external measured air concentrations

Paul W. Eslinger [a, *], Ian M. Cameron [a], Johannes Robert Dumais [b], Yudi Imardjoko [b], Pujadi Marsoem [b], Justin I. McIntyre [a], Harry S. Miley [a], Ulrich Stoehlker [c], Susilo Widodo [d], Vincent T. Woods [a]

[a] Pacific Northwest National Laboratory, 902 Battelle Boulevard, P.O. Box 999, Richland, WA 99352 USA
[b] P.T. BATAN Teknologi, Puspiptek, Serpong 15310, Indonesia
[c] Federal Office for Radiation Protection, Rosastr. 9, D-78098, Freiburg, Germany
[d] National Nuclear Energy Agency of Indonesia (BATAN), Jl. Kuningan Barat, Mampang Prapatan Jakarta, 12710, Indonesia

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ABSTRACT

BATAN Teknologi (BaTek) operates an isotope production facility in Serpong, Indonesia that supplies ^{99m}Tc for use in medical procedures. Atmospheric releases of ^{133}Xe in the production process at BaTek are known to influence the measurements taken at the closest stations of the radionuclide network of the International Monitoring System (IMS). The purpose of the IMS is to detect evidence of nuclear explosions, including atmospheric releases of radionuclides. The major xenon isotopes released from BaTek are also produced in a nuclear explosion, but the isotopic ratios are different. Knowledge of the magnitude of releases from the isotope production facility helps inform analysts trying to decide if a specific measurement result could have originated from a nuclear explosion. A stack monitor deployed at BaTek in 2013 measured releases to the atmosphere for several isotopes. The facility operates on a weekly cycle, and the stack data for June 15–21, 2013 show a release of 1.84×10^{13} Bq of ^{133}Xe. Concentrations of ^{133}Xe in the air are available at the same time from a xenon sampler located 14 km from BaTek. An optimization process using atmospheric transport modeling and the sampler air concentrations produced a release estimate of 1.88×10^{13} Bq. The same optimization process yielded a release estimate of 1.70×10^{13} Bq for a different week in 2012. The stack release value and the two optimized estimates are all within 10% of each other. Unpublished production data and the release estimate from June 2013 yield a rough annual release estimate of 8×10^{14} Bq of ^{133}Xe in 2014. These multiple lines of evidence cross-validate the stack release estimates and the release estimates based on atmospheric samplers.

1. Introduction and background

1.1. International monitoring system

The International Monitoring System (IMS) is part of the verification regime for the Comprehensive Nuclear-Test-Ban-Treaty Organization (CTBTO, 2014) that is designed to detect nuclear explosions no matter where they occur on the earth. When complete, eighty of the IMS stations located around the world will have aerosol measurement systems sensitive enough to detect releases from nuclear explosions at great distances. A primary method used in the IMS is the detection of four radioactive xenon isotopes (^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe) produced in a nuclear explosion (Bowyer et al., 2002; Kalinowski et al., 2010). When complete, half of the 80 stations will have xenon measurement systems.

1.2. ^{99}Mo production

A number of studies of the release and transport of radioxenon from nuclear explosions, nuclear power plants, and medical isotope production facilities have been published in the last decade (Becker

* Corresponding author. Pacific Northwest National Laboratory, MSIN K7-76, 902 Battelle Boulevard, P.O. Box 999, Richland, WA, USA.
E-mail addresses: paul.w.eslinger@pnnl.gov (P.W. Eslinger), Ian.cameron@pnnl.gov (I.M. Cameron), dumais@batan.go.id (J.R. Dumais), yudi@batan.go.id (Y. Imardjoko), pujadi@batan.go.id (P. Marsoem), Justin.McIntyre@pnnl.gov (J.I. McIntyre), harry.miley@pnnl.gov (H.S. Miley), Ulrich.Stoehlker@CTBTO.ORG (U. Stoehlker), swidodo@batan.go.id (S. Widodo), Vincent.Woods@pnnl.gov (V.T. Woods).

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et al., 2010; Eslinger et al., 2014; Hoffman et al., 2009; Kalinowski et al., 2008; Saey et al., 2010; Wotawa et al., 2010, 2003). These studies confirm that fission-based production of ^{99}Mo for medical purposes contributes measurable quantities of radioxenon to worldwide background levels. The ^{99}Mo (half-life of 66 h) then decays into ^{99m}Tc (half-life of 6 h) and the resulting ^{99m}Tc is used in about 30 million medical procedures per year. The global demand for ^{99}Mo production in 2014 is estimated to be on the order of 10^4 Ci (3.7×10^{14} Bq) (OECD, 2014), and the demand is expected to increase over the next few years.

1.3. DPRK 2013 nuclear test

The only atmospheric indication of the underground nuclear test performed by the Democratic People's Republic of Korea in 2013 was the detection of ^{131m}Xe and ^{133}Xe (Ringbom et al., 2014). Taken individually, the detected concentrations of these two isotopes were within the typical range detected by current IMS samplers. However, the ratio of the two isotopes is different for a nuclear explosion than for releases from either a nuclear power plant or a medical isotope production facility (Kalinowski et al., 2010). The isotopic ratios from a medical isotope production facility are more similar to those from a nuclear explosion than those from a nuclear power plant.

Knowledge of releases from medical isotope production facilities can help to correctly flag many samples as containing routine or periodic background emissions not related to nuclear testing. For example, ^{133}Xe has been detected in about 64% of the samples taken since the start of 2010 at the IMS station in Charlottesville, Virginia, USA. Most, but not all, of the ^{133}Xe in these samples comes from the medical isotope production facility in Chalk River, Ontario, Canada. Reducing the releases of radioxenon (Bowyer et al., 2013) from medical isotope production facilities to 5×10^9 Bq/d of ^{133}Xe will make it easier to identify releases caused by nuclear explosions.

1.4. International noble gas experiment

The International Noble Gas Experiment (INGE) was formed in 1999 as an informal group of expert developers of radioactive xenon measurement systems (Auer et al., 2010; Bowyer et al., 2002; CTBTO, 2014). INGE has continued to meet on an annual basis and it is now in a third phase. A recent paper discusses detailed monitoring designed to measure the releases of radioxenon (Saey et al., 2012). One of the radioxenon measurement systems used for INGE was deployed in Pasar Jumat, Indonesia, near the BaTek medical isotope production facility associated with the G.A. Siwabessy Multipurpose Research Reactor. The reactor is a 30 MW open-pool-type reactor, cooled and moderated by light water, fueled by 19.75% enriched uranium (Sunaryo et al., 2007), with a thermal neutron flux of up to 2.5×10^{14} n cm^{-2}s^{-1}. The BaTek medical isotope production facility has the capacity to produce 100 6-day curies of ^{99}Mo per week, but historically it has operated at about 40% of capacity.

1.5. Stack monitoring at Batek

A stack monitor deployed at the BaTek medical isotope production facility provided nearly a year of atmospheric release estimates for several xenon isotopes. The purpose of this work was to cross-validate the stack release estimates for ^{133}Xe with air concentrations measured at a sampler in Pasar Jumat at 14 km from Batek and provide a rough estimate of annual releases. The work reported here was based on collaboration among BATAN (the Indonesian National Nuclear Energy Agency), the Comprehensive Nuclear Test Ban Treaty Organization (CTBTO), and Pacific Northwest National Laboratory (Richland, Washington, USA) to determine effective ways to use medical xenon stack monitoring knowledge in improving the verification regime of the treaty.

2. Data and models

The activity concentration (Bq m^{-3}) of a radionuclide in the air after an atmospheric release can be expressed as the product of the release (Bq) and a time- and location-dependent effective dilution factor (m^{-3}). The general modeling approach in this paper has three steps. First, effective dilution factors between the BaTek facility and the sampler in Pasar Jumat were calculated for a sequence of times when releases were occurring. Second, the computed dilution factors were combined with measured values at the sampling location in an optimization process to estimate the release amounts. Third, the estimated release amounts were compared to the stack monitor data.

The BaTek facility typically operates on a weekly cycle. One set of uranium targets is irradiated in the reactor and the targets are then processed to remove and purify the ^{99}Mo. A week later the same cycle is repeated. As a consequence, it is useful to use the atmospheric sampling data to estimate the total release from the production facility over a week. Xenon isotopes are produced in the reactor at the same time the ^{99}Mo is produced, but significant releases to the atmosphere do not start until the beginning of the fuel dissolution step.

2.1. Sampling data

The xenon sampler used was a Swedish Automatic Units for Noble gas Analysis (SAUNA) developed at the Swedish Defense Research Agency (Ringbom et al., 2003). Sixteen of the IMS noble gas stations use SAUNA samplers. The sampler was located in Pasar Jumat for more than 18 months.

Several things must occur simultaneously to obtain air concentration data useful for making facility release estimates. First, the BaTek facility must be operating. The facility operated about 40% of the time the SAUNA was in place. Second, the SAUNA must be operating. The SAUNA delivered data for about 75% of the time it was deployed. Third, the local winds must blow from the BaTek facility towards the sampler for a contiguous period of a week or longer. Finally, the stack sampler must be operating. Of the entire time period the SAUNA was deployed in Pasar Jumat, only the short time period of June 15–21, 2013 met all of the conditions for making a release estimate that can be compared to the stack monitor. From March 28 to April 3, 2012, the BaTek facility was operating and the wind blew in a favorable direction, but the stack monitor was not operating.

Source estimates are provided here for both the time periods in 2012 and 2013. Only the estimate based on 2013 data can be compared directly to the stack monitor. However, comparing two weekly source estimates with each other is useful because the facility was attempting to produce approximately the same amount of ^{99}Mo both weeks.

Ground-level air concentrations of ^{133}Xe for the 2012 data set are provided in Table 1 and the concentrations in 2013 are provided in Table 2. The collection period for all of the samples was 12 h. Unfortunately, we do not have error estimates for the 2013 data in Table 2, but most of the sample concentrations are well above the minimum detectable concentration (MDC). Whether or not ^{133}Xe was detected in a sample is determined by the critical limit for that particular measurement, and the critical limit varies from sample to sample. The MDC is an operational parameter describing the sensitivity of the detector and normally MDC is about twice the critical limit.