Christine Johnson*^a, Steven R. Biegalski^a, Edward. J. Artnak^a, Ethan Moll^a, Derek A. Haas^{a,b},
Justin D. Lowrey^b, Craig E. Aalseth^b, Allen Seifert^b, Emily K. Mace^b, Vincent T. Woods^b, Paul
Humble^b

Production and release rate of ^{37}Ar from the UT TRIGA Mark-II research reactor

Abstract

Air samples were taken at various locations around The University of Texas at Austin's TRIGA Mark II research reactor and analyzed to determine the concentrations of ^{37}Ar , ^{41}Ar , and ^{133}Xe present. The measured ratio of $^{37}\text{Ar}/^{41}\text{Ar}$ and historical records of ^{41}Ar releases were then utilized to estimate an annual average release rate of ^{37}Ar from the reactor facility. Using the calculated release rate, atmospheric transport modeling was performed in order to determine the potential impact of research reactor operations on nearby treaty verification activities. Results suggest that small research reactors (~ 1 MWt) do not release ^{37}Ar in concentrations measurable by currently proposed OSI detection equipment.

Keywords

CTBT; Effluent monitoring; ^{37}Ar ; TRIGA research reactor; Treaty monitoring; On-Site Inspection

Introduction

Radioargon isotopes, particularly ^{37}Ar , have been proposed for use during an On-Site Inspection (OSI). Activation of calcium in the local geology is predicted to produce ^{37}Ar in quantities large enough that it may be measurable by inspectors on site and with a 35.04 day half-life it is expected that concentrations will remain elevated for a sufficient period to allow for samples to be taken after a suspected test (Haas et al., 2010). The estimated detection limits of ^{37}Ar for a field system is 20 mBq m^{-3} and for a laboratory system is 0.02 mBq m^{-3} (Haas et al., 2010).

Work is ongoing to establish the natural background of ^{37}Ar in various geologies and locations (Johnson et al., 2015; Riedmann and Purtschert, 2011; Wilson et al., 2015). Anthropogenic sources of ^{37}Ar , however, are less well studied. Measurements performed by (Matuszek et al.,

1975) indicate that ^{37}Ar released from nuclear power plants may be measurable at considerable distances from the release stack. However, more recent theoretical work indicates that research reactors should not be considered a significant source of ^{37}Ar (Fay and Biegalski, 2012).

Argon constitutes approximately 0.93% of dry air by volume. The primary isotope, ^{40}Ar , accounts for 99.6% of natural argon, with ^{36}Ar (0.34%) and ^{38}Ar (0.06%) present in much smaller quantities. Activation of air in a reactor, both room air and air dissolved in water, results in the production of ^{37}Ar , ^{41}Ar , and other argon isotopes. These isotopes can then be released from the facility via one of two pathways. First, room air is released through the facility's HVAC system. Additionally, a separate argon purge system is designed to remove air that may contain larger quantities of ^{41}Ar and other argon isotopes. This system pumps air from locations such as near the reactor pool surface and from the neutron beamports to confine and isolate it before release. These two streams are eventually recombined at the exhaust stack and released to the environment. The release rate of ^{41}Ar is monitored and reported annually, both for this facility and for many other nuclear facilities. Activity concentrations of ^{37}Ar are below current monitoring detection limits and special efforts are required to measure concentrations via low background counting facilities.

The NETL Reactor

The reactor at The University of Texas at Austin's Nuclear Engineering Teaching Laboratory (NETL) is a 1.1 MW TRIGA MARK II research reactor. The reactor utilizes uranium-zirconium hydride fuel elements which contain uranium enriched to 19.7% ^{235}U . The reactor is an open pool design, containing approximately 40 m³ of demineralized water which serves as both

shielding and coolant. During normal operation, the pool is covered to prevent access; however, the lid is not airtight. Surrounding the pool is a concrete bioshield and concrete structures which shield the beamport termini. The purpose of this study is to experimentally measure ^{37}Ar at various locations around the reactor and use the results to estimate the potential impact of research reactor operations on the local concentrations of ^{37}Ar .

Methods

Air samples were collected at The University of Texas at Austin's TRIGA MARK II reactor and compressed to 3000 psi into 3 m³ SCUBA bottles. Samples were taken from six locations within the reactor bay during operation at 950 kW and then repeated for five of the six locations approximately 2.5 days after shutdown as shown in Table 1. Each sample was counted in front of an HPGe detector which had been calibrated for SCUBA bottle counting. The samples were then shipped to Pacific Northwest National Laboratory (PNNL) where they were processed to analyze both the concentration of radioxenon and radioargon in the samples.

Sample Collection

Sample collection began with a Jordair PE-100-G portable compressor, which was located just outside the NETL building. In order to reach air sampling locations around the TRIGA reactor (see Table 1), 60 meters of flexible 'food grade' PVC hosing was attached to the compressor intake. Air samples were then compressed into standard aluminum 80 (AL80) SCUBA tanks, which were placed inside a blast deflector cylinder to redirect momentum in case of a catastrophic tank failure. For each air sample the collection was preceded by purging and priming of the intake hose and the compressor. In order to ensure that only air from the location

of interest was sampled, the compressor was started and run until the pressure-maintaining valve (PMV) opened and expelled air through the high pressure outlet. After the PMV opened the compressor was run for an additional 20 seconds, purging old air from the intake hose and the compressor and priming the system with air from the current sample location. Once the system was purged and primed, the AL80 SCUBA tanks were then attached and compressed to 3000 psi.

Table 1 Sample collection locations and reactor power status.

Tank #	Location	Reactor Status	Other
1	In Ar purge system stack	950 kW – End of Day	Stack exhaust is on
2	In HVAC system stack	950 kW – End of Day	Stack exhaust is on
3	Inches above reactor pool	950 kW – End of Day	
4	Top deck of reactor	950 kW – End of Day	
5	Reactor bay ground floor	950 kW – End of Day	
6	In front of beamport 3	950 kW – End of Day	Beamport 3 turned on all day
7	In Ar purge system stack	0 kW – No operation for days	Stack exhaust is on
8	In HVAC system stack	0 kW – No operation for days	Stack exhaust is on
9	Inches above reactor pool	0 kW – No operation for days	
10	Top deck of reactor	0 kW – No operation for days	
11	Reactor bay ground floor	0 kW – No operation for days	

Radioxenon Analysis

The concentration of radioxenon in the samples was measured using a SAUNA (Swedish Automatic Unit for Noble Gas Acquisition) system (Ringbom et al., 2003). The system was reconfigured to pull samples from a SCUBA bottle rather than through the atmospheric sampling

stack. The system was also capable of measuring other xenon isotopes, however, ^{133}Xe was the only isotope consistently present in measurable quantities.

Radioargon Analysis

Once samples were compressed into the SCUBA bottles they were placed in front of an HPGe detector which had been calibrated using quantified radioxenon and radioargon (Haas et al., 2008; Klingberg and Biegalski, 2015) for this sample geometry. The efficiency calculation and calculation of the minimum detectable concentration of ^{41}Ar for this geometry can be found in the work of Biegalski et al., 2015. Each bottle was counted for 30 minutes in front of the detector in the configuration shown in Figure 1 and analyzed using the 1293.6 keV peak to calculate the ^{41}Ar concentration in each sample. Additionally, the same HPGe detector (a portable model) was used to directly measure a spectrum at the top of the reactor pool. This spectrum was used to verify the method used to acquire the ^{41}Ar concentrations in the compressed samples.

Fig. 1 HPGe detector configuration for measuring compressed air samples.

Analysis of ^{37}Ar was performed at PNNL using a low-background internal-source gas proportional counter. The detector utilizes high-purity electroformed copper and a niobium anode wire to reduce the background due to detector materials (Seifert et al., 2013). The design and construction of the detectors is detailed in Aalseth et al., 2009. The measurements of ^{37}Ar were made in PNNL's Shallow Underground Laboratory using the Ultra-Low-Background Counting System detailed in Aalseth et al., 2012 and Seifert et al., 2013.

Results

The concentrations of ^{37}Ar and ^{41}Ar measured in various locations around the reactor are given in Table 2, while the concentrations of ^{133}Xe are shown in Table 3. Due to the short half-life of ^{41}Ar (109.34 minutes), no ^{41}Ar was measured after the shutdown period.

Table 2 Concentrations of ^{37}Ar and ^{41}Ar measured at various locations around The University of Texas TRIGA reactor during operation at 950kW and after several days of no operation. No ^{41}Ar was measured post-shutdown.

Location	^{37}Ar [mBq m ⁻³]				^{41}Ar [mBq m ⁻³]	
	950kW	+/-	no operation	+/-	950kW	+/-
Argon purge stack	12050	45	2691	15		
HVAC stack	89.3	3.3	14	0.6		
Inches above reactor pool	7709	24	4344	20		
Top deck of reactor	21.1	1.0	1	0.4		
Reactor bay ground floor	30.2	1.9	26	0.9	<	-
In front of beamport 3	14.2	1.2	-	-		

Table 3 Concentrations of ^{133}Xe measured at various locations around The University of Texas TRIGA reactor during operation at 950kW and after several days of no operation.

Location	^{133}Xe [mBq m ⁻³]			
	950kW	+/-	no operation	+/-
Argon purge stack	2216	1174	8387	2293
HVAC stack	508.3	869	1528	1149
Inches above reactor pool	4337	1713	4007	1990
Top deck of reactor	< 93.4	-	< 13240	-
Reactor bay ground floor	< 3626	-	< 197.8	-
In front of beamport 3	3092	2265	-	-

At first glance, the increase in ^{133}Xe concentration in the argon purge stack and HVAC stack after several days of no operation seems counterintuitive. During the days of no operation, the stack exhaust for both systems was shutdown, allowing for some buildup in the system, but the stack was purged for approximately 15 minutes before sampling was started. Another possible explanation for this increase in concentration is that the ^{133}Xe present in dissolved air within the pool had additional time to transport to the surface during the shutdown period.

The locations of the highest argon concentrations, the argon purge stack and inches above the reactor pool, support the conclusion of Fay, 2014 that the main source of radioargon in a research reactor is from activation of air dissolved within pool water. If activation of calcium in concrete was a significant source of ^{37}Ar , elevated concentrations of argon would be expected near the beamport and on the reactor bay ground floor.

Discussion

Estimated Release Rates

In order to expand these measurements to an estimate of the total annual releases of ^{37}Ar , we assume that these measurements are representative of a typical $^{41}\text{Ar}/^{37}\text{Ar}$ activity ratio in the argon purge system of the University of Texas reactor. While the ^{37}Ar concentrations are not regularly monitored, releases of ^{41}Ar are measured by an in-line continuous air monitor. The monitor is calibrated annually and the measured values are reported for regulatory purposes.

Using the measured concentrations of ^{41}Ar and ^{37}Ar during full operation at 950kW from Table 2, a ratio of expected $^{41}\text{Ar}/^{37}\text{Ar}$ release activity concentrations was calculated as . This ratio was calculated using the concentrations from the argon purge stack which is the primary exhaust point for argon from the reactor and is also the location of the argon monitor used to acquire the averaged ^{41}Ar values.

The maximum release rate of ^{41}Ar was determined for each year between 2006 and 2015 and the total range was used to provide an upper bound on the release rate of ^{41}Ar from the UT TRIGA reactor. The annual average release rate was also calculated for the same time period. The range covered by these values is given in Table 4.

Table 4 ^{37}Ar and ^{41}Ar concentrations as measured in the argon purge stack and calculated annual maximum and annual average release rate ranges for ^{37}Ar and ^{41}Ar . Cells shaded in grey indicate calculated values.

	^{41}Ar	^{37}Ar	
Measured Release			mBq m^{-3}
Annual Maximum Release Rate			Bq hr^{-1}
Annual Average Release Rate			Bq hr^{-1}

Atmospheric Transport Cases

Atmospheric transport modeling was performed using the National Oceanic and Atmospheric Administration Air Resource Laboratory (NOAA ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) atmospheric transport model (Draxler and Hess, 1998). An operational day was assumed to last for 8 hours and run from 9 AM to 5 PM Central Time. Archived meteorological data from the North American Mesoscale Forecast System (NAM 12 km) was used for the test cases. The averaging period was 15 minutes.

Six cases were run spanning various weather conditions typical of Austin, Texas. The release rate was assumed to be the average annual maximum release rate (Bq h^{-1} of ^{37}Ar). In these cases, the highest recorded concentration was 0.06 mBq m^{-3} , shown in Figure 2, however this concentration was measured in the cell containing the source. The highest concentration recorded outside of the grid cell containing the source was 0.03 mBq m^{-3} in a grid cell 0.4 km from the UT TRIGA reactor as shown in Figure 3. Assuming a minimum detectable concentration of 20 mBq m^{-3} for a

field ^{37}Ar system, a release of this magnitude would not be detectable during an on-site inspection conducted under the Comprehensive Nuclear-Test-Ban Treaty. However, concentrations do exceed the detection limits of a laboratory system (0.02 mBq m^{-3}) in all but one case.

Fig. 2 ^{37}Ar plume assuming a maximum release rate from the UT TRIGA Mark-II research reactor. The white square represents the location of the highest concentration (0.06 mBq m^{-3}) in the cell containing the source.

Fig. 3 ^{37}Ar plume assuming a maximum release rate from the UT TRIGA Mark-II research reactor. The white square represents the location of the highest concentration (0.03 mBq m^{-3}) in a cell not centered on the source location (0.4 km distance).

Conclusions

Results from the measurements of ^{37}Ar concentrations around the UT TRIGA Mark-II research reactor suggest that small research reactors ($\sim 1 \text{ MWt}$) do not release ^{37}Ar in concentrations measurable by currently proposed field OSI detection equipment. However, concentrations of ^{37}Ar within a radius of $\sim 1.5 \text{ km}$ were predicted to exceed the estimated detection limit of a laboratory system in certain cases. While this work does rule out smaller research reactors as a significant source of ^{37}Ar , it does not rule out both larger research reactors or power reactors,

suggesting that future work is needed to accurately quantify potential releases from these sources.

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