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Detection in subsurface air of radioxenon released from medical isotope production

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ABSTRACT

In order to better understand potential backgrounds of Comprehensive-Nuclear Test-Ban Treaty on-site inspection relevant gases, a sampling campaign was performed near Canadian Nuclear Laboratories in the Ottawa River Valley, a major source of environmental radioxenon. First of their kind measurements of atmospheric radioxenon imprinted into the shallow subsurface from an atmospheric pressure driven force were made using current on-site inspection techniques. Both atmospheric and subsurface gas samples were measured and analyzed to determine radioxenon concentrations. These measurements indicate that under specific sampling conditions, on the order of ten percent of the atmospheric radioxenon concentration may be measured via subsurface sampling.

1. Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) includes the ability for member states to request an On-Site Inspection (OSI) as part of the verification process. The purpose of an OSI is to gather local evidence of a possible nuclear event; the presence of radioactive noble gases, particularly radioxenon, is a primary indicator of recent nuclear activity and hence is relevant to an OSI. As a noble gas, xenon is chemically inert resulting in a relatively high leakage probability from an underground test and has previously been measured at the surface after a nuclear test (Carrigan et al., 1996; Dubasov, 2010; Schoengold et al., 1996).

On-Site Inspections will include the option to employ both above ground and subsurface soil gas monitoring in the search for nuclear explosion signatures. The radioxenon isotopes ^{131m}Xe, ^{133m}Xe, and ^{133}Xe and the radioargon isotope ^{37}Ar are the primary targets of subsurface sampling (Carrigan and Sun, 2014).

As advances in detector technology and analysis methods lead to lower detection limits, the presence of radioxenon from sources other than nuclear explosions, both natural and anthropogenic, becomes a complicating factor in the assessment of environmental samples. The current OSI capability for radioxenon detection is in the range of 1 mBq/m^3 for ^{133}Xe using mobile measurement systems (Ringbom et al., 2015; Wieslander and Khrustalev, 2014). More significantly, in the case of low-level detection the presence of these sources may also bias the isotopic ratios used to discriminate significant events.

1.1. Medical isotope production as a radioxenon source

With an estimated total annual release of 11 × 10^{15} Bq of ^{133}Xe, medical isotope production facilities are the most significant contributor to the global radioxenon background (Saey, 2009). While the current distribution of facilities has resulted in elevated concentrations primarily in the mid-latitudes of the Northern Hemisphere, new facilities are causing an increased background level in certain Southern Hemisphere locations as well (Saey et al., 2010a; Wotawa et al., 2010). An additional complication is that the isotopic ratios of radioxenon released from medical isotope production may be similar to that of a nuclear explosion, specifically in facilities which use highly-enriched uranium targets (Saey et al., 2010b). Other sources of background radioxenon, including

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nuclear power plants and natural production, are more easily distinguished but they still serve to complicate the overall background (Hebel, 2010; Saey et al., 2010b).

One of the largest producers of ^{99}Mo globally is Canadian Nuclear Laboratories' Chalk River Laboratories in Ontario, Canada. At this facility, highly-enriched uranium targets are irradiated to produce ^{99}Mo, which is then extracted on-site using chemical dissolution. During this dissolution process and the later cementing of the resultant waste, ^{133}Xe is released at an estimated rate of up to 10^{13} Bq/day (Saey, 2009).

1.2. Imprinting

Measurement of radioxenon during an OSI is heavily dependent on underground transport processes and the geology through which transport occurs (Carrigan et al., 1996; Carrigan and Sun, 2014; Lowrey et al., 2013; 2012). While transport in homogenous media may be dominated by diffusion at small scales, in fractured geologies barometric pumping is predicted to dominate the bulk movement of air (Nilson et al., 1991). Barometric pumping involves changes in atmospheric pressure acting to impress or withdraw gas from the subsurface, with it generally resulting in an increase in the rate of vertical mixing of air in the subsurface (Carrigan and Sun, 2011; Lowrey, 2013). Because trace gas lost to the atmosphere is not replaced, barometric pumping primarily acts as an upward transport mechanism for signatures of underground nuclear explosions. However, it is predicted that barometric pumping may have an additional effect of driving atmospheric gas into the shallow subsurface, which is the realm of an OSI. Simulations have shown that during periods of increasing barometric pressure, detectable quantities of atmospheric radioxenon could potentially be imprinted into the shallow subsurface (Carrigan and Sun, 2014; Lowrey, 2013; Lowrey et al., 2015a, 2012). This process can both serve to contaminate the soil-gas environment if a plume of radioactive noble gas passes over sampling locations, which could complicate the interpretation of radioxenon detections in an OSI, but it could also dilute potential signature-containing subsurface air with atmospheric, thus reducing concentrations of the gases being measured.

In order to validate the predictions of imprinting, a sampling campaign was performed near the Canadian Nuclear Laboratories medical isotope production facility, the world's largest source of atmospheric ^{133}Xe. An analysis of the stack release data and atmospheric transport of the radioxenon released from this site can be found in Johnson et al. (Johnson et al., 2015a).

1.3. Infiltration

Another method by which atmospheric intrusion might occur is through infiltration. This differs from imprinting only in the method by which the atmospheric gas is introduced to the subsurface. While the term imprinting implies the natural process of barometric pumping, infiltration signifies that the atmospheric gas was drawn into the subsurface by the sampling process. This infiltration could be the result of a poorly sealed hole which allows atmospheric air to be drawn directly into the subsurface sample through the hole itself. However, assuming a well-sealed sampling hole, the main source of infiltration is assumed to be due to the sampling process itself. Samples are obtained by continuously pumping on a single point for 12–24 h, creating a substantial pressure drop and subsequent flow of gas from the surrounding subsurface area. This can also serve to pull atmospheric gas into the subsurface, acting almost as an enhanced imprinting mechanism (Lowrey et al., 2015a).

2. Experimental

From 10 to 19 September 2014, daily air samples were collected from both above-ground (atmospheric) and from the subsurface. While previous work had been performed to select an ideal sampling location (Johnson et al., 2015b), complications led to the need to choose an alternate site upon arrival. The site utilized was a clearing in Sheenboro, Quebec, 14 km southeast of the Canadian Nuclear Laboratories medical isotope production stack; the locations of both the sampling site and the stack are shown in Fig. 1. This site was co-located with a permanently emplaced NaI(Tl) detector belonging to Health Canada as part of their fixed-point surveillance network (Grasty et al., 2001).

2.1. The hole

The sampling hole was drilled using a 2-in auger to a depth of 0.90 m. A screen was then attached to sampling tubing and lowered to the bottom of the hole, which was then backfilled with the removed soil to a depth of 0.49 m. It was then verified that this backfill completely covered the screen at the bottom of the hole. The remainder of the hole was filled with a bentonite mixture which consisted of a mixture of 1 part bentonite to 4 parts water. The bentonite was then capped with construction grade sand. A top view of the completed sampling hole can be seen in Fig. 2.

2.2. The sampler

Subsurface gas sampling was carried out using a Sub-Surface Gas Sampler (SSGS) designed and built by Pacific Northwest National Laboratory (PNNL). The sampler was programmed to provide a constant flow rate to the collection bags over the sampling period. Within the sampler, a small fraction of the air was diverted to a Durridge RAD7 radon detector that integrated measurements of the radon concentration in the sampled air in intervals of 5 min. Additionally, the SSGS contained a barometer to measure local atmospheric pressure as well as humidity and temperature sensors.

2.3. Sample collection

Samples of the atmospheric air at ground level were collected simultaneous to subsurface sampling. A sampling tube was emplaced just above ground level, pointed downward to prevent water from entering the system in case of rain. The tubing was then connected to a separate pump which was set to the same flow rate (generally 1.3–1.5 L/min) as that of the subsurface gas sampler and fed into a separate sample collection bag. The setup is shown in Fig. 3 with arrows indicating the general direction of flow. The atmospheric bag was marked to distinguish it from the subsurface sample bag to prevent cross-contamination in case of any memory effect in the bag.

The gas samples were collected inside a 2 m^3 grey water bag during sampling. Both bags were placed 3–4 m away from the sampling hole to avoid acting as a seal on the atmosphere-subsurface interface. After the sampling period, the samples were removed from the collection bags and compressed into scuba bottles at pressures ranging from 1000 to 2500 psi for transport, a process which generally took 20–30 min. During compression, the subsurface samples were always compressed first in an effort to limit any potential memory effects (i.e., cross contamination between samples) within the compressor. The times of compression were also notated for future comparison with NaI(Tl) measurements of atmospheric radioxenon concentrations.