ULTRA-SENSITIVE ENVIRONMENTAL MONITORING OF EMISSIONS FROM A RADIOISOTOPE PRODUCTION FACILITY

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ABSTRACT

The United States and Canada have jointly established an experimental radio-aerosol monitoring station in Vancouver, British Columbia as part of the International Monitoring System for the Comprehensive Nuclear Test Ban Treaty. The purpose of this station is to monitor the atmosphere for the presence of anthropogenic radio-aerosols that could be indicative of nuclear explosion debris. The station has been engineered to achieve detection sensitivities that are approximately three orders of magnitude higher than conventional environmental and emergency preparedness monitors. Due to its ultra-sensitive measurement capability, the station has regularly detected micro quantities of radioisotope emissions from a nearby commercial production facility that would otherwise have gone unnoticed. The major isotope, ^{123}I, was identified by spectroscopic analysis and correlated to the facility through source emission data. These measurements demonstrate the multiple uses of the monitoring stations for other areas including environmental, public health, and safety applications.

1 INTRODUCTION

The newly signed Comprehensive Nuclear Test Ban Treaty (CTBT) prohibits countries from testing nuclear weapons, whether in the hydrosphere, lithosphere or atmosphere. Verification of this treaty is achieved by an International Monitoring System (IMS) which has over 400 global sensors from multiple technologies. The four verification technologies are seismic, hydroacoustic, infrasonic and radionuclide. Radionuclide monitoring of the CTBT will be conducted through a network of 80 stations that will be deployed globally over the next 3 to 5 years. The objective of this technology is to continuously monitor the atmosphere for airborne radioaerosols and gases and detect those which could be indicative of nuclear explosive debris. All data collected at the monitoring stations are instantaneously transmitted to a central processing facility called the International Data Centre (IDC), where it is automatically processed and interactively analyzed. The radionuclide results are combined with those from other technologies and then disseminated to the signatories of the CTBT.

In March 1996, an experimental radionuclide monitoring station was established at Vancouver, British Columbia, one of the future monitoring sites of the IMS. The station was jointly commissioned by United States and Canada and involves the collaborative efforts of Health–Canada, the University of British Columbia and the U.S. Department of Defense. The Vancouver station, CA002, is one of several early monitoring sites that were established to operationally test the concept of a global radionuclide IMS. The station is linked to the prototype International Data Center (PIDC) in Arlington, Virginia, United States and transmits data as part of the experimental network that includes 23 stations in 11 different countries. When the future IMS radionuclide network is completely populated, approximately one-third of the 80 stations

will be located around the Pacific Rim. The IMS monitoring capability is expected to provide an increased level of international and environmental security in the region as well as the world.

2 EXPERIMENTAL

2.1 Station equipment

The experimental monitoring station continuously monitors the atmosphere for anthropogenic radioaerosols in discrete 24 hour samples. The major processes of the system are collection, sample preparation, assay, and communication. These processes are integrated into a single infrastructure and are conducted in a semi-automated manner. (Mason and Bohner, 1997).

Collection of large volumetric air samples is achieved through a high-volume air sampler capable of 1100 m^3/h. The flow-rate is maintained constant by a variable speed controller that compensates for decreased flow caused by filter loading. Normal operating parameters of the sampler are a flow-rate of 1000 m^3/h and a collection time of 24 hours resulting in a total volumetric air sample of 24,000 m^3. Aerosols are separated from this sample through a high-efficiency particulate filter with a square face area of 0.25 m^2 that results in a linear face-velocity, v_f, of about 1.1 m/s. The filter composition is glass fiber and its collection efficiency is rated at 99.99 percent for 1.0 micron diameter particles (v_f = 1.0 m/s). This is used to optimize particle trapping in the size range of interest for long-range nuclear test debris, 0.1 to 5 μm diameter (Glasstone, 1977).

Sample preparation is achieved through high-pressure compression of the filter into a disk with a diameter of 6.4 cm and thickness of 1.3 cm. This is done to optimize the counting geometry by reducing the solid angle between the sample and the detector that increases the intrinsic efficiency. To prevent cross-contamination between samples in the press apparatus, the filters are folded and placed in wax paper sheathing prior to compression.

Radiometric assay of the sample is achieved by using dual high-purity germanium (HPGe) detectors that are cooled by a freo-electric cryogenics system. The detectors are rated at 40 percent relative efficiency (relative to standard 7.62 cm x 7.62 cm NaI scintillator). The energy resolution (FWHM) of these detectors is less than 1.2 keV at 122 keV and less than 2.2 keV at 1332 keV. After a collection period of 24 hours, the sample is allowed to decay for 4 hours prior to radiometric assay. This 4 hour decay period is used to reduce the natural radioactivity on the filter caused by radon progeny. These natural radionuclides interfere with the detection capability of the sample due to increased background caused mostly by Compton scattering and other photon interactions. A detailed discussion of the effects of terragenous (^{220}Rn and ^{222}Rn) and cosmogenic (^{7}Be) radioactivity can be found in several references (Eisenbud, 1987). During this 4 hour decay period, a preliminary spectrum is acquired on the detector that can be used for later diagnostic analysis. The 4 hour decay period is followed by a 20 hour acquisition period for a total operating cycle time of 48 hours. At the completion of the assay period, spectral data are automatically transmitted to the IDC for processing and analysis.

2.2 Air sampling characteristics

Although a large majority of the spectra contain only background natural radionuclides, the central objective of this station is to detect and quantify minute concentrations of anthropogenic radio-aerosols that can be indicative of nuclear explosive debris. In order to quantify the concentration of a detected radionuclide, the air sampling dynamics must be understood. The interaction of an anthropogenic plume with a monitoring station begins with a *dwell* time in which the plume's air parcels are in a position to be collected by the sampler. During this period, the activity of the nuclide is governed by the following time-dependent rate equation,

$$\frac{dA}{dt} = Q(t)F(t) - \lambda A, \quad (1)$$

where A is the activity on the filter (μBq), Q is plume activity concentration (μBq/m^3), F is the sampler flow-rate (m^3/h) and λ is the nuclide decay constant. When the plume departs the sampler's field of regard, or when the discrete sampling period ends, the source term is eliminated and the activity is subjected only to radioactive decay. In this specific application, the plume dwell time is assumed to span the entire sampling period and the airborne plume concentration and the volumetric flow-rate are assumed to be constant. Using these simplifications, several straightforward approaches can be used to derive the following solution for the nuclide concentration collected on the filter,

$$C_{FP} = \frac{\varepsilon \gamma Q F}{\lambda_{FP}^2}\left(1-e^{-\lambda_{FP} j}\right)\left(1-e^{-\lambda_{FP} m}\right)e^{-\lambda_{FP}(f+r)}, \quad (2)$$

where ε is the energy dependent detector efficiency, γ is the branching ratio of the nuclide's key gamma line, T is the sampling time, m is the counting time, f is the decay time and λ is the nuclide's decay constant (Evans, 1995). It is noted that the aforementioned equation is only valid for a nuclide whose only source is from the sampled air. Figure 1 shows a temporal plot of the activity build-up and decay periods during a generic monitoring process

2.3 Data analysis

After the spectrum is acquired during the assay period, the spectral data and calibration information are instantaneously transmitted to the PIDC where they are automatically processed using a standard spectroscopy algorithm[1]. Energy, efficiency and resolution data are checked and the calibration functions are adjusted as necessary to compensate for small perturbations in these data. Gamma ray peaks are located and quantified by a Gaussian peak fit routine. The peaks are then associated to the corresponding nuclides using a specially tailored library and activities are quantified and converted into air concentrations. These measurements are characterized based on their spectral content and sent to human analysts for interactive review. A detailed description of this process has been previously reported (Bohner, Mason, et al, 1997). Figure 2 shows a typical spectrum collected at the CA002 station. The figure illustrates the resulting product of IDC data analyses that consists mostly of typical naturally occurring radionuclides. However, one atypical gamma ray peak that cannot be attributed to natural radioactivity is observed at 159.0 keV. The detection and source identification of this gamma ray is the subject of the following discussion.

[1] Spectroscopy analysis system is based on Canberra Industries Genie-UNIX system which is a special product for this program.

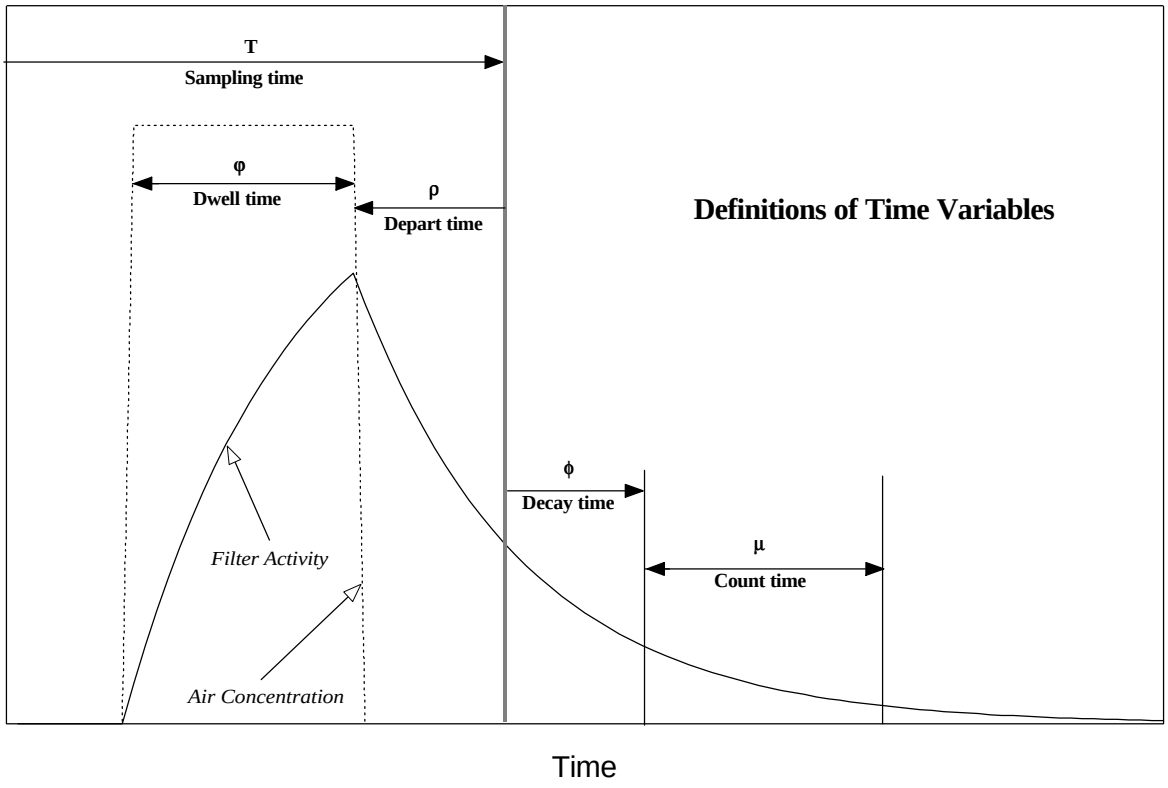

Figure 1 Time dependent activity on the filter paper during a generic monitoring cycle.

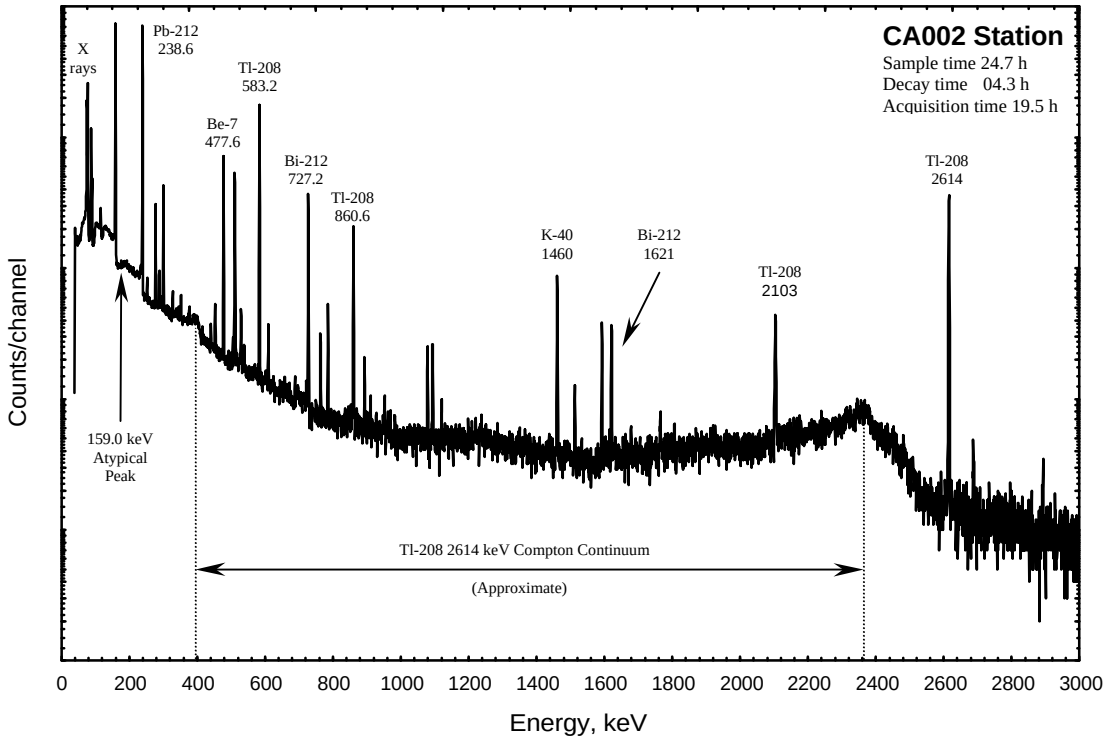

Figure 2 Typical spectrum from the station in Vancouver. Note the peak at 159.0 keV.

3 RESULTS AND DISCUSSION

3.1 ^{123}I Detection

Soon after the CA002 station became operational, the spectral analysis indicated the presence of the unusual gamma peak at 159.0 keV. The occurrence of the peak was significant in both magnitude and frequency and thus the IDC, UBC, and Health–Canada began a collaborative investigation to determine its origin. The first step was to formulate a list of candidate nuclides that had key lines near the 159.0 keV peak. This list was further truncated by eliminating those nuclides with very short half-lives that could not survive the 4 hour decay period. Two prime candidate nuclides emerged: ^{123m}Te ($t_{1/2}$ = 119.7 d) and ^{123}I ($t_{1/2}$ = 13.3 h). Both of these nuclides have key lines near 159.0 keV without other significant lines to aid in identification.

As previously mentioned, a preliminary spectrum is also acquired during the 4 hour decay period that proved very useful for this case. Using the count-rate ratio of the 159.0 keV peaks in the preliminary and full spectra for multiple measurements, it was determined that the peak source decayed at a mean half-life of 13.34 ± 0.60 h. This identified the peak as ^{123}I.

Considering the more common releases of anthropogenic radioactivity: nuclear power plants, research reactors, fuel reprocessing plants and medical facilities, ^{123}I is not typical. This is due to its extremely low yield for most fission reactions and limited applications. However, UBC researchers quickly identified its source when it was revealed that a nearby commercial facility produces ^{123}I for medical imaging applications. Figure 3 shows the time history of ^{123}I measurements at CA002 for the past 18 months.

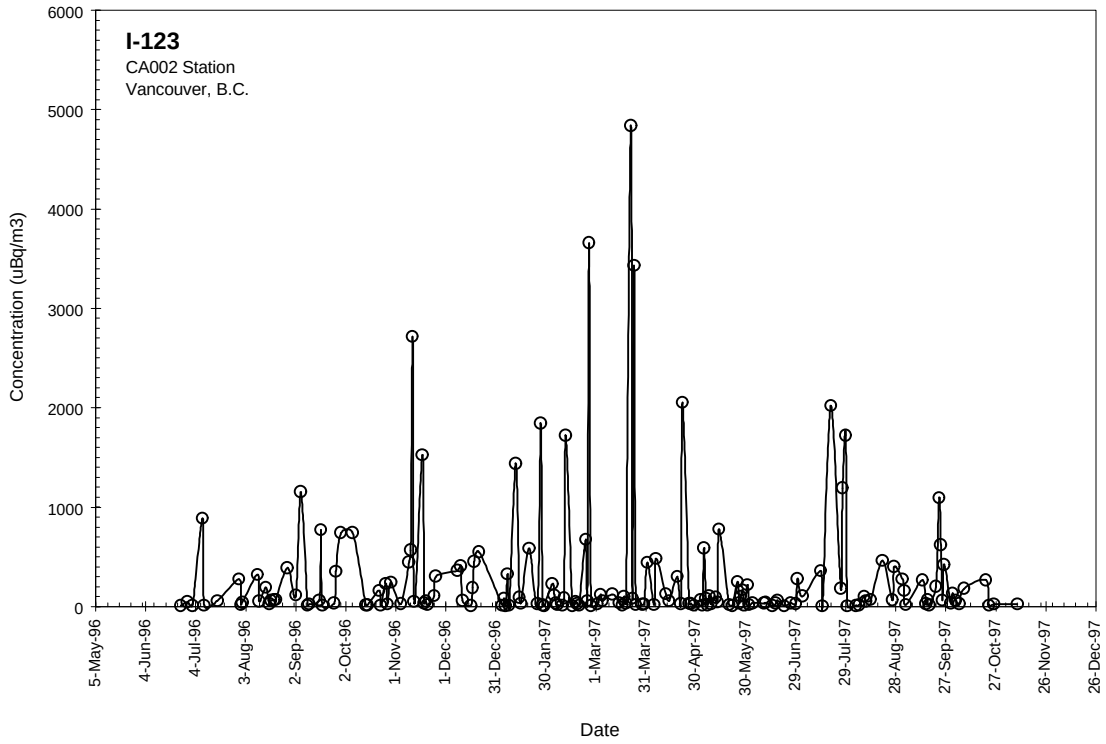

Figure 3 Time history concentrations of ^{123}I at CA002 during the last 18 months

3.2 Source production

The commercial radioisotope production facility is located approximately 2 km southeast of the CA002 monitoring site. This facility utilizes a small production cyclotron that accelerates protons to 30 MeV with a beam current of 1 mA. The facility is used to produce a variety of radioisotopes that have applications in medicine and industry. One such isotope, ^{123}I, is produced regularly and is used as a diagnostic imaging agent in brain, heart, kidney and thyroid studies.

This production method utilizes highly enriched ^{124}Xe gas as the target material that is bombarded with a beam of 30 MeV protons. The gas charge is typically irradiated for 4-6 hours followed by decay period to allow the progeny to decay to ^{123}I ($t_{½}$=13.2 h) by one of the following *p,2n* reactions:

$$ {}^{124}_{54}Xe\left(p,2n\right){}^{123}_{55}Cs \xrightarrow{5.94m} {}^{123}_{54}Xe \xrightarrow{2.08h} {}^{123}_{53}I\ , \tag{3} $$

$$^{124}_{54}Xe\left(p,2n\right)^{123m}_{55}Cs \xrightarrow{1.62s} ^{123}_{54}Xe \xrightarrow{2.08h} ^{123}_{53}I\ . \qquad (4)$$

After the irradiation and decay periods, the gas charge is transferred from the target chamber that is washed to collect the ^{123}I in solution. The ^{123}I is then concentrated by passing the solution through an ion exchange column and the final product is eluted off the column, assayed, diluted and dispensed. The overall production cycle normally takes about 18 hours from irradiation to purification.

The facility operates a high-efficiency particulate filtration system to reduce radioactive emission from the exhaust and has detectors to monitor the radioactivity leaving the stack. The stack monitors routinely detect ^{124}Xe and ^{123}I and although the amounts depend on the specific production parameters, a typical ratio of the parent to progeny is 3:1.[2] In some production cycles, the system is purged prematurely to meet other requirements and therefore a higher proportion of ^{124}Xe may be released to the stacks.

Emissions that escape the stack are believed to be an instantaneous plume release consisting of both radioxenon and radioiodine. Although the radioxenon is transported only as gas, radioiodine has a more complex profile with both aerosol and gaseous parts. For radioiodine released to the environment, the gaseous fraction typically varies from 60 to 90 percent of which both molecular (I_2) and organic (CH_3I) are formed (Borisov, Budyka, et al, 1994). Aerosol formation of radioiodine is a complex function of the nucleation rate, solubility, attachment factor, and the ambient aerosol concentration in the atmosphere. The high gaseous fraction results in an underestimation in the concentration calculations because they are not corrected for these losses. The deposition of ^{123} I on the filter has two sources: (1) direct emission of ^{123}I from the stack and (2) ^{124}Xe progeny that escapes the stack and decays enroute to the detection station.

3.3 Data Correlation

Transport from the source facility to the detection station is driven mostly by advection and the turbulent atmospheric conditions caused by the complex land-sea interface. These conditions will only be treated in a cursory manner due to the limited scope of the paper. It was previously noted that the production facility is located approximately 2 km southeast of the monitoring station. To correlate the two points, it was necessary to gain information about the mean wind conditions. An analysis of the station's wind rose showed the predominate wind parameters were between due east (90^o) and east-southeast (110^o) with a mean speed from 3 to 10 km/h. Although tenuous, these data indicate a link between the source and receptor. It also suggests that plumes would generally reach the monitoring station within 1 hour of release time.

In addition to the tenuous spatial correlation, a temporal correlation is possible due to the somewhat regular periodicity in the ^{123}I production schedule. Operational data were obtained from the facility and these data were compared to the observations when detectable amounts of ^{123}I were measured at the station. The results for this short-term correlation study are shown in Figure 4. As the plot shows, the production schedule is consistent with the observed occurrences of ^{123}I at CA002. However, some uncertainty exists in the strict source to receptor temporal correlation due to the 13.3-hour half-life, since ^{123}I can exist up to several days in environment before being detected at the station.

It should be noted that several uncertainties must be taken into account when comparing the calculated concentrations of ^{123}I. The two major areas are the gaseous fraction and the sampling interval. The previously discussed gaseous radioiodine fraction showed that the concentrations could be underestimated by as much as an order of magnitude. Additionally, the entire sampling time was assumed equal to the

[2] Personal communication from Anne Trundell, Staff Health Physicist for the UBC TRIUMF facility.

plume dwell time. If the plume interacts with the station for less time, then the concentrations could also be underestimated by as much as an order of magnitude. However, even in the worst case uncertainty scenarios, the measured concentrations (~10^3 μBq/m^3) at CA002 are still several orders of magnitude below the Canadian allowable limits for ^{123}I (~10^7 μBq/m^3) based on maximum permissible dose.

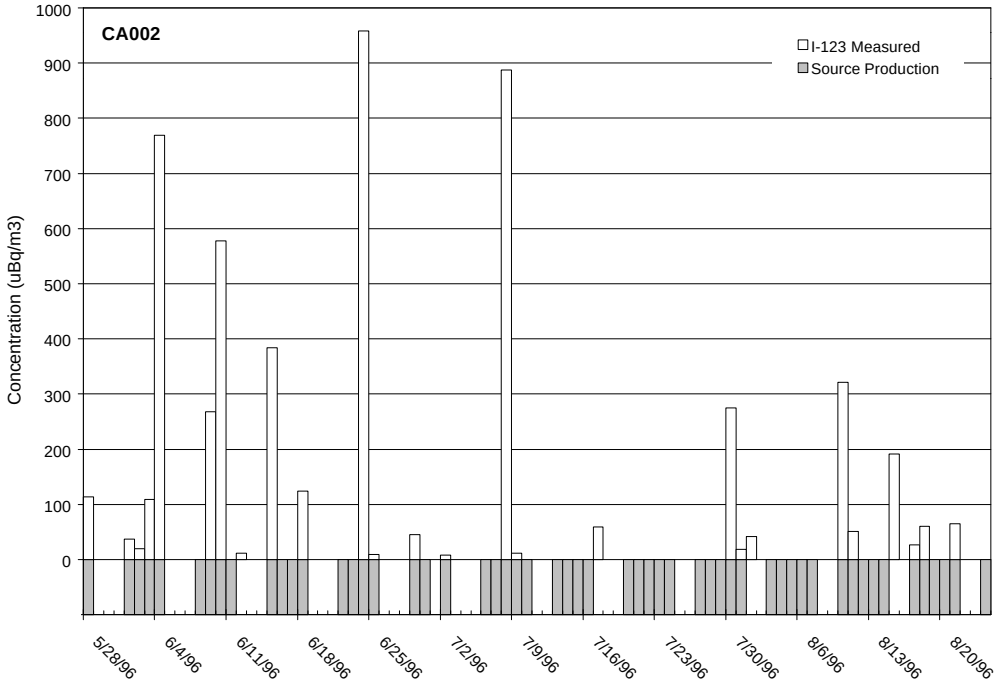

Figure4 Plot showing the dates of ^{123}I source production and detection at the Vancouver station.

4 CONCLUSIONS

An experimental monitoring station was jointly established by Canada and the United States in Vancouver, B.C. Canada to support verification of the CTBT. Its mission is to detect and identify anthropogenic radioaerosols that may be indicative of nuclear explosion debris. Measurements from this station indicated the presence of an atypical anthropogenic radionuclide, ^{123}I. Through a multi-lateral effort, the ^{123}I source was determined to be a nearby commercial production facility that produces the radioisotope for medical applications. Detection of these micro quantities of anthropogenic radioactivity demonstrates the ultra-sensitivity of the monitoring station for CTBT purposes as well as environmental and public health related applications. Rapid detection and near-real time reporting make this data particularly valuable and actionable in the international user community. The Vancouver station is one of 80 future global radionuclide monitoring stations that will operate in this capacity and provide a greater level of international and environmental security in the Pacific Rim and the world.

5 ACKNOWLEDGMENTS

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7 KEY WORDS

Radiation detection, gamma spectroscopy, environmental radioactivity, radioisotope production, atmospheric monitoring, radioiodine.