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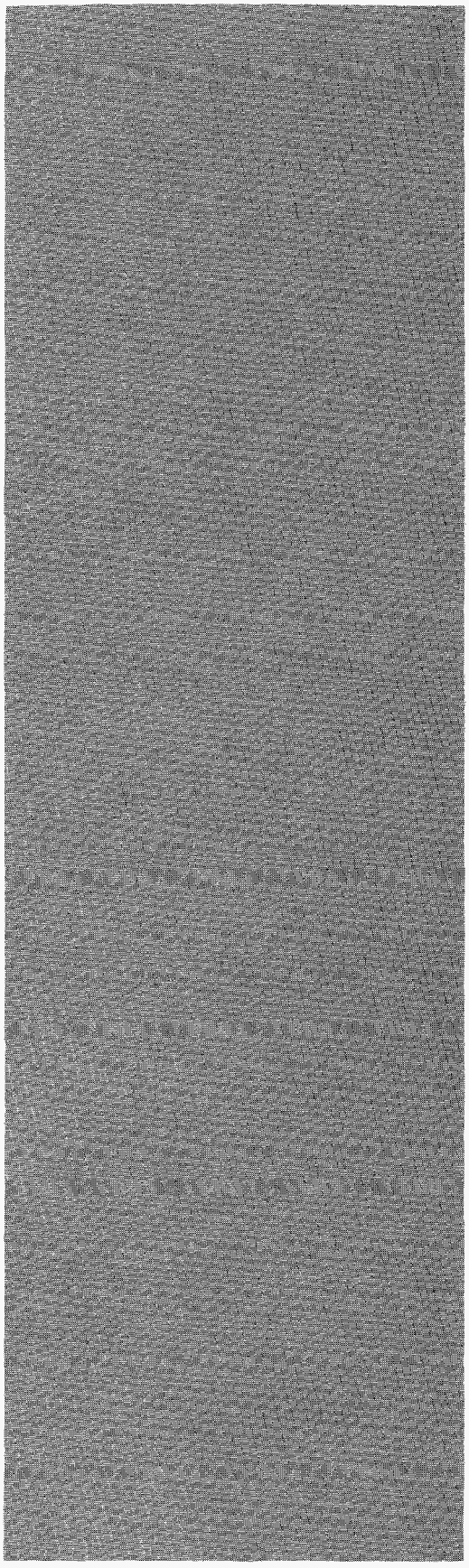

Radionuclides Identified at a U.S. Customs Service Site

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ABSTRACT

The movement of radionuclides through the U.S. Customs Service port of entry at Blaine, Washington, has been studied using a high-resolution gamma-ray spectrometer and an automated data logging system. Data covering about 10 weeks of operation were obtained and analyzed. The data-acquisition system and the site of the measurement are described. Results are reported and interpreted in light of the known traffic of radioisotopes produced at the Canadian TRIUMF facility and imported into the United States for use in radiopharmaceuticals.

I. INTRODUCTION

In the summer of 1996, the U.S. Department of Energy (DOE) and the U.S. Customs Service (USCS) embarked on a series of joint efforts to study the problem of radioactive materials entering the United States through USCS-controlled ports of entry. The relevance of this problem to the broader problem of "nuclear smuggling" should be apparent: successful interdiction of special nuclear materials (SNM) being transported across national borders would reduce dramatically the opportunities for malevolent use of SNM escaping from the weapons complex of former Soviet Union states, which has been alleged to have certain security deficiencies.[1]

Relatively little is known about the movement of *non*-SNM radionuclides through USCS ports. Some degree of legitimate commerce certainly exists; many of the radiopharmaceuticals used in the United States are produced in Canada, as are some other isotopic sources. These materials are brought into the country via land and air, usually in heavily shielded containers that reduce the intensity of the emitted radiation by many orders of magnitude and that may render the radiopharmaceuticals utterly undetectable. Additionally, radiopharmaceuticals *in vivo* certainly enter the country, "carried" by individuals who have had recent nuclear-medicine procedures. The extent of inadvertent or surreptitious introduction of radioactive materials into the United States is not known, but such incidents are known to have occurred, the most famous one being the "radioactive-rebar" episode of 1984, in which ^{60}Co came into the United States from Mexico in shipments of construction material that was contaminated with the radionuclide from a teletherapy unit in Juarez, Mexico.[2] A study of known incidents involving radioactive materials in recycled metals through 1994 has been compiled by Lubenau and Yusko.[3] Most of these incidents, however, involved materials from domestic sources that did not cross the US border inbound.

During the summer of 1996, Sandia National Laboratories (SNL) emplaced a radiation-detection system at the USCS facility at Dulles International Airport in Washington, D.C. One goal of that project was to gain a better understanding of the movement of radioactive materials through Dulles and to demonstrate that an instrument could be used, at least in principle, to detect such materials at a

port of entry. The gamma-ray detector used in the Dulles work was an NaI scintillator and thus had limited energy resolution, although sophisticated analysis methods were used to extract as much information from the spectra as possible. Identification of the detected radionuclides was therefore problematic in some cases, at least compared to the relative ease of isotope identification possible with a high-resolution gamma-ray spectrometer. However, low- and high-resolution gamma-ray spectrometers are in many regards complementary: the former can be built to be very large and efficient and therefore to provide high-statistics (if low-resolution) spectra in comparatively short times. Consequently, when preparations were made for installation of an SNL system at another USCS site at Blaine, Washington, it was recommended that a high-resolution gamma-ray spectrometer also be installed to facilitate identification of the radionuclides. Los Alamos National Laboratory was asked by DOE to provide an instrument for this purpose and to analyze the resulting data.

In this report we describe the system used for high-resolution γ-ray spectroscopy at Blaine and report the results of its use during the period 12 September–25 November 1996. In Section II of the report we describe the hardware and software used, and in Section III we discuss the operating environment at Blaine. Procedures for analyzing the acquired data are described in Section IV. While the great majority of the more than 18,000 spectra acquired at Blaine showed nothing but background radiation (^{40}K, uranium and its daughters, thorium and its daughters), a few spectra revealed the presence of other radionuclides, and these are discussed in Section V. The Appendix to this report gives dates, times, peak energies, and isotope identifications for all spectra showing the presence of radionuclides other than the background activities.

II. INSTRUMENTATION

The primary function of the control and data-acquisition system was to store onto a hard disk the spectrum generated by the spectrum analyzer. A block diagram of the hardware is shown in Fig. 1. The primary units of the system are the detector, the analog/digital electronics, and the computer. The converter and uninterruptible power supply (UPS) are secondary units. The manufacturer and model of the units used are listed in Table 1.

Table 1. Primary and secondary units of the control and data-acquisition system, listed with manufacturer.

System Unit	Manufacturer Part Number
Detector	PGT Lab Cooler IGC2019
MCA/ADC	EG&G ORTEC DSPec
Computer	DELL Latitude Xpi P120ST
Converter	Allied Telesis MR122T
UPS	APC SU1000XL

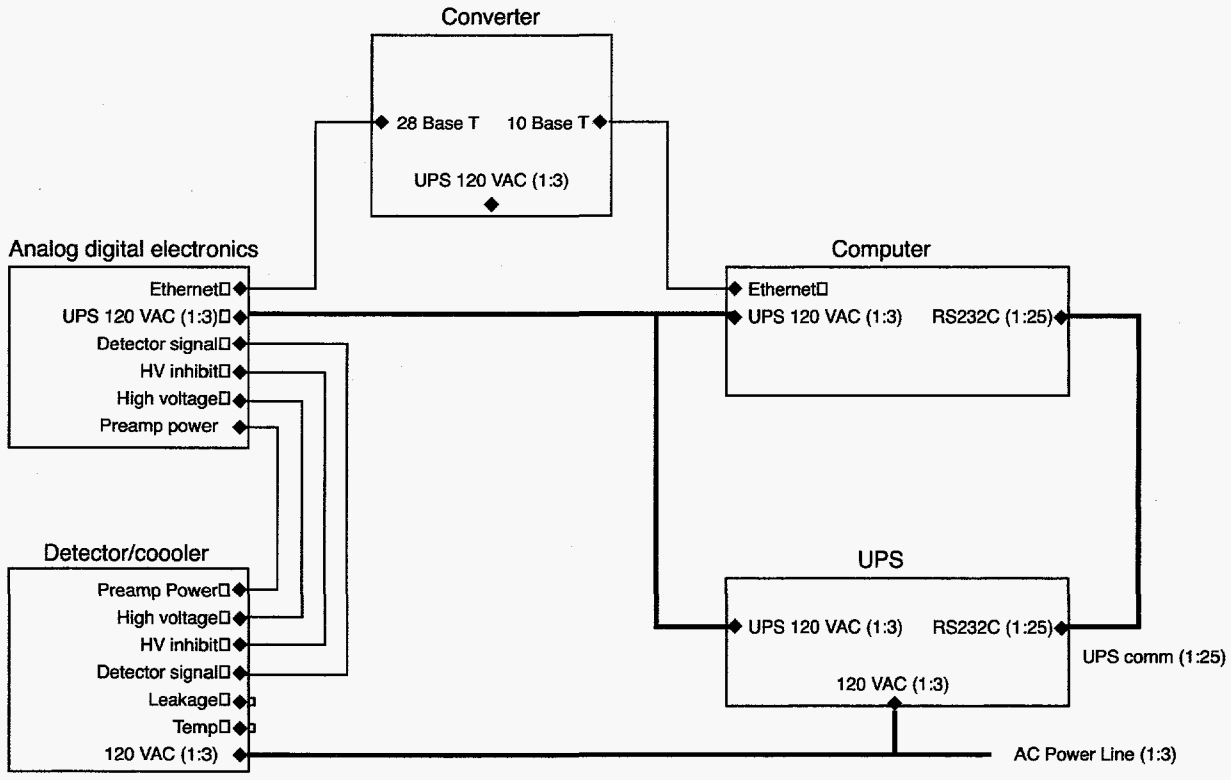

Fig. 1. System Block Diagram

The detector was a mechanically cooled HPGe spectrometer with a crystal having an efficiency of 19% relative to the canonical 3"x3" NaI detector at 1.332 MeV. This relatively small crystal was used because it was integral to the mechanical cooler and was available at the time DOE requested that the project be undertaken. A larger crystal would have provided better count rates for most of the sources encountered; however, instantaneous count rates in a few cases were apparently so high already that significant dead time existed, a condition that would have worsened if a larger detector had been used. System optimization, in any event, was beyond the scope of the present project.

The computer permanently stored the data accumulated by the detector and analog electronics on hard disk. Data were periodically transferred to floppy diskettes for analysis at Los Alamos. Communications between analog/digital electronics and the computer were mediated by Ethernet between the two units. The computer also provided the operator with tools for setting up the parameters of the analog electronics (gain, high voltage, etc.).

The UPS was used to permit the system to operate during momentary dropouts of the 120-Vac power line. Prior warnings of "brownouts" at the site suggested the wisdom of including the UPS. The UPS used in this application had the capability of recording ac voltage fluctuations.

The converter provided compatibility between the Ethernet format of the computer and that of the spectrometer.

Software for integrating this system was developed specially for this project, drawing on commercially available modules and locally written code. The software served to (a) periodically transfer data from the electronics to the computer, (b) create a new directory each day for the data to

be stored during that day, (c) present an interface that allowed the operator to set up the parameters of the system and look at the data being acquired, and (d) gracefully shut down the system in the event of a prolonged power failure.

Windows NT 3.5.1 was the operating system. The Maestro application from ORTEC provided the operator interface. Powerchute from APC would have provided the system shutdown had power failed for more than 2 minutes; during operation, however, no unexpected power outages occurred. Los Alamos-designed software provided the data storage and organization function.

While operating, the system continuously acquired spectra, each having a count time of five minutes as selected by the operator interface when the system was initially set up at Blaine. The choice of five minutes for the count time represented a compromise between the operational conditions described in the next section, for which shorter count times would have been preferable, and the need to accommodate data storage on a disk of limited size. No provisions were included in the present system for real-time analysis of the data, as the objective of the project was solely to perform data logging and subsequent off-line analysis.

III. THE SITE

The site at which this system was installed is a port of entry at Blaine, Washington, specifically intended to service commercial traffic entering the United States from Canada on Canadian highway 91, which connects to US Interstate 5. Another port of entry to service noncommercial traffic (passenger cars, etc.) is nearby. Figure 2 is a photograph taken "downstream"* of the facility, showing the location of the booths and lanes. Although the physical plant allows as many as four lanes to be open for traffic, each with a booth for a USCS official, only one or two lanes are used under normal conditions. The lane nearest the office structure visible at the right of Fig. 2 (hereinafter referred to as the "inboard" lane, occupied by a truck in the photograph), and sometimes the lane farthest from the office structure (the "outboard" lane, not visible in the photograph). Information from the USCS indicated that perhaps 1300 to 1600 trucks might pass through this facility in a given day, the majority of them passing through the inboard lane.

Both the detector system described in this report and the SNL low-resolution system were placed in the normally unused booth denoted by the arrow in Fig. 2. This booth is a few meters upstream of the booth used by USCS officials when they interview drivers of vehicles in the inboard lane, so that the driver would normally be approximately 7m from the detector during the interview. When a large tractor-trailer truck was being examined in the inboard lane, the detector was located nearest to some point in the trailer, while if an automobile, van, etc., was being examined, the entire vehicle may have been downstream of the detector by some small distance. The detector was placed on a small table so that the crystal was centered approximately 1.5 m above the ground and pointing toward the inboard lane, as shown in Fig. 3, which was the best configuration for measuring possible sources in trailers. The computer and analog electronics were placed under this table. The entire assembly was then surrounded with a wire mesh (removed in the photograph for clarity) to keep out birds and small animals that could enter the booth through the door or small openings in the roof.

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* In the following text, use of the term "downstream" will indicate that the described location is reached by a vehicle after it passes through the inspection area or other reference point; "upstream" has the opposite sense.

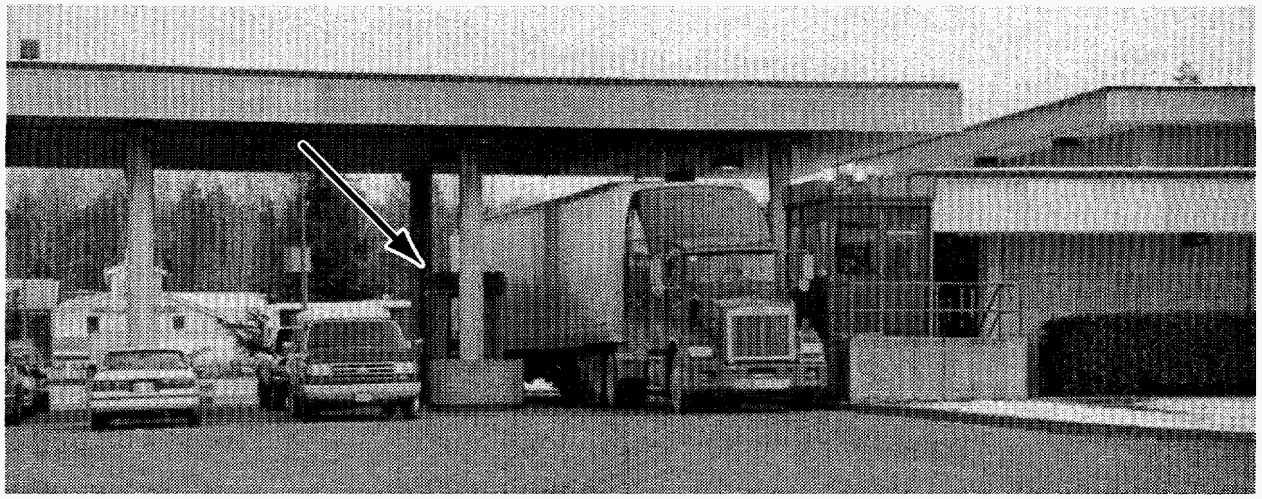

Figure 2. Configuration of the Blaine port of entry. The arrow denotes the booth in which the radiation detection system was installed.

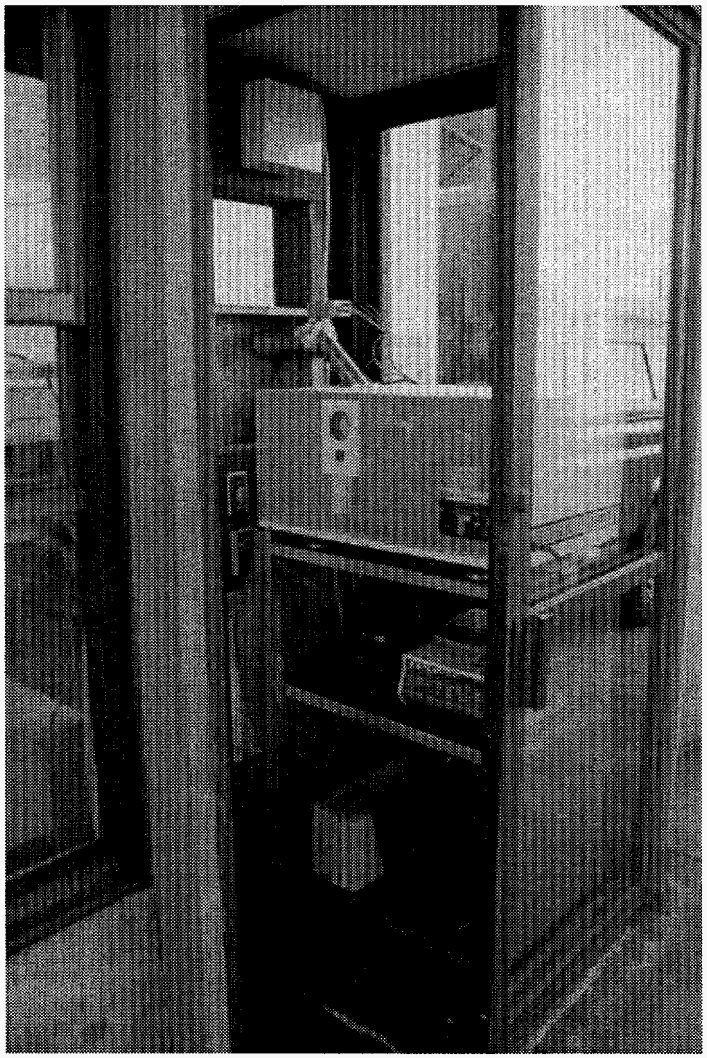

Figure 3. Radiation detection system in the booth at Blaine.

Observation of routine operations at the Blaine facility showed that in most cases, a vehicle would spend about 45 to 60 seconds at the booth while the USCS official interviewed the driver. Additionally, a vehicle might sit in line upstream of the booth for times ranging from a few seconds to a few minutes depending on the traffic volume, which fluctuates during the day. During off hours, there might be no line at all. Only rarely would a vehicle not exit the facility directly as soon as Customs clearance was obtained. Therefore a representative "experimental condition" for measurements would be a source-detector distance of 3 meters* and a measurement time of one minute, each parameter possibly varying by a factor of two, depending on the size of the vehicle and the time taken to do the interview. Note that spectra were accumulated for five minutes, so that data from multiple vehicles might be contained in a single spectrum.

The USCS operates a mobile baggage-inspection system that emits low-energy X rays and that is carried in a van normally parked near the office building. The distance between this van in its usual location, just past the right edge of the photograph in Fig. 2, and the detector system was about 15 m. Since the baggage-inspection system incorporates shielding to render its X-ray beam directional, and since the beam was not normally directed toward the LANL detector system, it was believed that no X-rays would be detected from the baggage-inspection system under normal circumstances. Experiments in which USCS personnel operated the van for several minutes at a time appeared to validate this belief; no X-rays were visible in the spectra obtained during these experiments.

Because of the location of the Blaine facility, it was speculated prior to installation of the equipment that some traffic of radioisotopes would be seen originating at the TRIUMF research facility in Vancouver, BC. Table II shows the isotopes that are commonly produced at the TRIUMF facility and the prominent lines that these isotopes might contribute to a γ-ray spectrum. Discussions with USCS personnel revealed that shipments of TRIUMF-produced radioisotopes pass through the Blaine facility several times a week, but did not provide any insights into what might be seen in the spectra collected at Blaine, there being no information available regarding shielding, etc., in the vehicles used to transport the radioisotopes. Information from Nordion International, Inc., the commercial distributor of radiopharmaceuticals from TRIUMF and other Canadian medical-isotope production facilities, indicates that other isotopes are produced on rare occasions, but no information was available on the less frequently produced isotopes.

* Sufficiently strong sources in trucks in the outboard lane, during the occasional periods when it was in use, could also have been recorded by the system. A source-detector distance of ~15-20 m would be appropriate in analyzing data attributed to sources in the outboard lane, but the system lacked portal monitors or other means for noting which lane was used by the truck harboring the source if one was detected.

Table II. Radioisotopes produced at TRIUMF for nuclear-medicine applications (information from Nordion International, Inc.).

Isotope	Key γ rays (keV)[a]	Notes
^{57}Co	122.1, 136.5, 692.0	b
^{67}Ga	93.3, 184.6, 300.2, 393.5	
^{111}In	536.7	
^{123}I	159.0, 346.3, 440.0, 528.9	
^{82}Rb	776.5	c
^{201}Tl	Hg K x-rays, 167.4	d

a Energies rounded to nearest 0.1 keV; from Browne and Firestone (Ref. 4). Some isotopes also emit low-energy X rays and γ rays and/or γ rays with very low branching ratios, that could not be detected with this system.

b ^{56}Co present as a contaminant; prominent lines at 846.8, 1037.9, 1238.3, 1771.5, and 2598.6 keV, many other weak lines.

c Short-lived ($t_{\frac{1}{2}}$=1.27 min) daughter of ^{82}Sr, which emits no detectable γ rays. ^{82}Rb is used in medical procedures and is "milked" from a ^{82}Sr "cow."

d ^{200}Tl present as a contaminant; prominent lines at 368.0, 579.32, 828.4, and 1205.7 keV, many other weak lines.

IV. ANALYSIS METHOD

Over 18,000 five-minute gamma-ray spectra were acquired. The spectra were recovered at intervals of one to three weeks and returned to LANL for analysis. A batch file then processed the data. First, each spectrum was analyzed using the peak fitting routines in the commercial software program Maestro. The peak-fitting threshold was set so that typically one to three background peaks were fit in each spectrum. This threshold marked visually obvious peaks as well as occasionally fortuitous alignments of background data points. Overall, a large number of false peaks were found, but it ensured that we missed a minimum number of small-but-significant real events.

Next, the batch process listed the peaks to a file, did a one point energy recalibration using the ^{40}K peak at 1460.75 keV, and filtered the peak list by eliminating background lines. The remaining lines were listed to an event log file. If the energy of the peak corresponded to a medical radioisotope, that isotope was listed in the file. In addition, any spectrum with overall high count rates was flagged. The batch process could handle up to 3000 files at a time, limited only by the available hard disk space.

After the first month's data, the filter program was modified so that unusually high background count rates would be flagged as well. One hundred background spectra were summed to determine average background count rates. The filter program then marked as events any peaks with count rates exceeding five standard deviations from normal. The number of peaks marked as events increased from an average of one event peak per ten spectra to one event per six spectra, with the increase mostly due to the poor counting statistics in five minutes for the background lines. However, several instances of abnormally high background line count rates were detected with this additional filtering.

Once the batch file completed, a gamma spectroscopist examined the events to determine the true hits. The event log file was checked to see if any spectrum had multiple peaks marked as events. An

event was indicated if the count rate of the logged peak was greater than 0.2 counts per second, if there was any peak above 500 keV, if the filter program identified the line as belonging to a medical isotope, or if the dead time of the spectrum was over one percent The dead times associated with no-event spectra were typically less than 0.05 percent. In the case of medical isotope shipments, all of these conditions were true. In other cases, only one of these conditions was satisfied.

V. RESULTS AND DISCUSSION

A total of 26 spectra were observed to contain statistically significant peaks not attributable to naturally occurring isotopes. Table III summarizes the findings of the analysis of these spectra; a more complete listing of individual line energies and intensities is given in the Appendix. In addition to these spectra, another 8 spectra showed either anomalously high count rates in lines associated with naturally occurring isotopes or increases in the continuum count rate without any obviously anomalous lines. Data from these spectra are also presented in the Appendix.

Of the isotopes seen, ^{123,131}I, ^{82}Rb, ^{67}Ga, ^{99m}Tc, and ^{111}In are all commonly used in nuclear medicine. All of these but ^{131}I and ^{99m}Tc are produced at the TRIUMF facility. The events with ^{131}I probably involved radiopharmaceuticals *in vivo* rather than material produced at TRIUMF; the relative intensities of the lines seen in the spectra were consistent with an absence of massive shielding, in contrast to the spectra associated with the other radiopharmaceuticals. The ^{99m}Tc event was probably also due to a radiopharmaceutical *in vivo*. ^{57}Co, while not normally used in radiopharmaceuticals, also has a connection to nuclear medicine in that large surface-area "flood" sources of this isotope are used to test and calibrate Anger cameras and other imaging instruments. It is thought that the events of 2 October/1501, 21 October/1427 and 24 October/1108 were due to material intended for production of a flood source being brought into the country after having been produced at TRIUMF. This explanation is consistent with the observation that ^{56}Co was also present and that large quantities of shielding surrounded the sources of both isotopes, as deduced from the ratios of peak intensities. ^{56}Co is produced as an impurity in the process used to produce the ^{57}Co.

Several other events appear also to involve shipments from TRIUMF, although some analysis is required to understand them. The isotope ^{200}Tl is not commonly used in medical practice or in industrial applications that we know of. However, for the multiple events involving this isotope, an explanation is at hand similar to that for the cobalt isotopes. The isotope ^{201}Tl is one of the most widely- used medical isotopes and is produced at TRIUMF by (p,xn) reactions that also make ^{200}Tl. In contrast to ^{200}Tl, ^{201}Tl emits only low-energy γ rays (see Table II). The relative intensities for the ^{200}Tl lines and the elevated continuum at low energies in spectra where ^{200}Tl is present imply that this material was heavily shielded, possibly by as much as 3 cm of lead. The transmission of the highest-energy (167.4 keV) ^{201}Tl γ ray through even 1 cm of lead is less than 10^{-7}, so up to 100 Ci of ^{201}Tl may have been present with the ^{200}Tl and gone undetected. This observation is consistent with the shipment of ^{201}Tl produced at TRIUMF and destined for use in the United States (warranted by Nordion to contain not more than 1.5% ^{200}Tl), and has obvious and unfortunate implications for the possibilities of detecting ^{235}U (the gamma radiation from which is all low-energy) through passive means at ports of entry. By contrast, the event of 6 October/1818, in which a small quantity of ^{201}Tl was detected (apparently *in vivo*), did not involve observable amounts of ^{200}Tl. The event involving ^{203}Pb (the data do not allow one to distinguish between this isotope and ^{203}Hg, although the former is considered more likely) is presumed, but not known, also to be attributable to TRIUMF production of this isotope. Information from Nordion indicates that this isotope is produced, albeit rarely, at TRIUMF.

Table III. Non-background nuclides observed.

Date	Time (PST)	Nuclides	Approximate source strength (mCi)	Notes
9/19	1851	^{203}Pb	0.03	
9/22	1726	???	???	Isotope unidentified; peak at 204.9 keV
9/22	1836	???	???	Isotope unidentified; peak at 241.3 keV
9/23	1429	^{123}I ^{200}Tl	1000 1000	
9/28	0839	^{131}I	0.2	
10/2	1401	^{111}In ^{56}Co ^{57}Co ^{123}I	100 300 2000 400	
10/4	1236	^{131}I	1	
10/4	1241	^{131}I	0.2	Same source as previous event
10/6	1818	^{201}Tl	0.2	
10/7	1420	^{123}I	300	
10/7	1425	^{123}I	2000	Same source as previous event
10/8	1352	^{67}Ga ^{123}I	2000 300	
10/9	1318	^{111}In ^{123}I ^{201}Tl	100 40 60	
10/18	1356	^{111}In	100	
10/20	1100	^{67}Ga ^{99m}Tc	20 0.3	
10/21	1427	^{56}Co ^{57}Co ^{82}Rb ^{123}I	20 large 15000 1000	
10/22	1133	^{137}Cs	0.011	
10/22	1418	^{67}Ga ^{123}I	10 200	
10/22	1423	^{67}Ga ^{123}I	1700 30	Same source as previous event
10/22	2355	^{99m}Tc	0.8	Doubtful, weak peak
10/24	1108	^{56}Co ^{57}Co	1 1000	
10/28	1536	^{123}I	500	

Table III. Non-background nuclides observed. (cont.)

Date	Time (PST)	Nuclides	Approximate source strength (mCi)	Notes
10/29	1458	^{67}Ga ^{123}I	2000 4	
10/30	1310	^{111}In ^{123}I	100 300	
11/11	1409	^{123}I	100	
11/11	1414	^{123}I	300	Same source as previous event
11/12	1026	^{137}Cs	7	
11/18	1531	^{123}I	1000	
11/22	1456	^{131}I	1	

Of the other events, the spectra of 22 October/1133 and 12 November/1026 that reveal ^{137}Cs cannot be clearly attributed to production at TRIUMF. ^{137}Cs is used in medical facilities as an external irradiation source, but this application cannot explain the apparent passage of a small source through Blaine. (The Cs events are presumed to have involved small sources rather than heavily shielded, large ones because no increase in the continuum below the peak is evident in the spectrum.) Small ^{137}Cs sources are used in various other industrial applications, and such a source may have been responsible for the 22 October and 12 November events. Contamination incidents involving this isotope have also been reported, the most notorious one being the episode in Brazil during 1987, in which a source used in nuclear medicine broke open and the escaping material caused severe to fatal radiation exposures in a few individuals.[5] There is no information in the present spectra whereby contamination could be distinguished from passage of small, properly contained but unshielded check sources. The exact cause of the cesium events, therefore, remains unknown.

The two events for which we have no even marginally satisfactory explanation are the events of 22 September/1726 and 1836 hours. A portion of the latter spectrum, as well as the one taken immediately before it, is reproduced in Fig. 4. A moderately strong peak at 241.4±0.5 keV is evident that is not present in the previous spectrum. No other lines are visible apart from background, and there is no evidence of a raised continuum at energies lower than the peak energy, implying that little if any shielding was present. A similar peak, of about equal magnitude, was seen in the spectrum of 1726 hours at an energy of 204.9±0.7 keV. Both peaks are highly statistically significant; in the case of the 241.4-keV peak, it is estimated that the likelihood of even one spectrum, of the 18,000+ obtained at Blaine, containing a "peak" of greater or equal intensity solely as a result of random fluctuations in the background is considerably less than 10^{-8}.

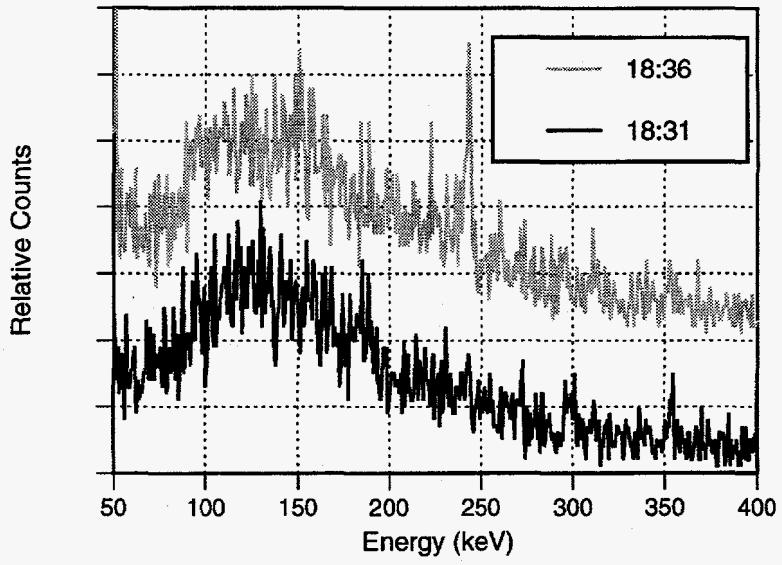

Figure 4. Data from the event of 9/22/96 1836 and the previous spectrum (9/22/96 1831), the latter offset to emphasize the absence of the peak at 241 keV.

A search of the literature, encompassing both the Browne-Firestone[4] and Erdtmann-Soyka[6] compilations of decay information, reveals no commonly used isotope that emits only (or predominantly) a 204.9- or 241.4-keV gamma ray. The isotope ^{77}Br, which is used in some experimental procedures in nuclear medicine, emits a 239.0-keV gamma ray that might produce the observed peak if a slight gain miscalibration was assumed; however, another ^{77}Br line at 520.6 keV, which should surely be visible in any spectrum with such a strong 239-keV line, is absent in the Blaine spectrum. The isotope ^{125}Xe emits a 243.4-keV γ ray, but can also be ruled out through the non-observation of lines at 188.4 and 453.8 keV; shielding material thick enough to remove the former from the spectrum would cause the latter to be prominent compared to the 243.4-keV line, and in any event, no evidence of shielding is seen in the spectrum. Similar arguments preclude the presence of ^{151}Gd and ^{189}Ir. One remote possibility is the isotope ^{86}Zr ($t_{\frac{1}{2}}$=16.5 h) with a line at 242.8 keV and another, somewhat weaker line at 612.0 keV that might be obscured by the nearby 609-keV background line. However, other lines from ^{86}Y, produced by the decay of ^{86}Zr, would be expected to be present unless the Zr had been produced extremely recently. No such lines are seen, the absence of the prominent ^{86}Y line at 1076.7 keV (branching ratio 82.5%) being a particularly strong argument against the presence of ^{86}Zr more than a few hours old. The identity of the radioisotope producing this spectrum therefore remains unclear. Similar arguments rule out simple explanations for the 204.9-keV line.

An attempt was made to quantify the amounts of material present in the various events through calculation of detector efficiency and estimation of attenuation effects. Results are included in Table III. Extraction of quantitative information from these spectra is complicated by the variations in measurement conditions. The "representative" conditions described above (source-detector distance = 3 m, vehicle present near detector for 60 seconds) might be incorrect for any single measurement by amounts large enough to cause order-of-magnitude errors in derived source strengths. Additionally, correction for attenuation by shielding materials, while straightforward in the cases of isotopes for which more than one line was seen in the spectrum, could be done only approximately for isotopes for which only one line was seen. The values shown in Table III must therefore be

considered uncertain by a factor of 10 in either direction. Note that the events of 7 October/, 22 October/1418+1423 and 11 November produced lines in two consecutive spectra, and that analysis of the two spectra in each case led to inferred shielding and source strengths that differed, the latter typically by a factor of three or so. This observation is consistent with the presumed factor-of-10 uncertainty.

It is of interest to compare the isotopes seen in the present study to those seen in the Dulles measurements. The isotopes ^{99m}Tc, ^{201}Tl, ^{131}I, and probably ^{67}Ga were seen at Dulles in conditions suggestive of radiopharmaceuticals *in vivo*. All of these except ^{131}I were rare *in vivo* at Blaine. The difference is interpreted as stemming from the relative remoteness of the Blaine facility, the lower density of human traffic through the portal, and the short persistence of radiopharmaceuticals other than those containing radioiodine in the body (the biological half life of iodine may be as nearly great as the radioactive half life). While ^{67}Ga was seen at Blaine, the absence of low-energy lines suggests that the material was *in vitro* (shielded) rather than *in vivo*. By contrast, the common radiopharmaceuticals ^{123}I and ^{111}In were seen at Blaine (*in vitro*) but not at Dulles. These differences suggest that every port of entry may have a unique traffic in radionuclides.

The significance of the spectra with anomalously high count rates in lines from naturally occurring radioisotopes is unclear. The Sandia team also reported events attributed to naturally occurring radioisotopes, and attributed some of the events involving ^{40}K and ^{232}Th (and its daughters) to shipments of fertilizer and thorium-bearing helicopter parts, respectively. Interestingly, however, the spectra accumulated with the present system at times when the Sandia system saw excess ^{40}K or ^{232}Th do not appear to show anomalously high rates in lines attributable to these isotopes. Conversely, the high-background events recorded by the present system do not appear in the logs kept by the Sandia system. The Sandia system only preserved data at times when the count rate was anomalously high; therefore, it is not possible to examine data from other times to look for high count rates in background lines. This discrepancy might be explained either by dismissing all of the high-background events as statistical fluctuations, and/or by noting that the fields of view of the Sandia and Los Alamos detectors differed: for example, possibly some of the events seen by the Los Alamos system but not the Sandia system were from materials in the "outboard" lane (which would have been invisible to the collimated Sandia detector). This explanation may also be appropriate for some of the events in Table III that were not reported by Sandia. Similarly, it is possible that the events seen by the Sandia system but not the Los Alamos system would not have been observed above background in the present spectra since the backgrounds were higher than in the Sandia system.

VI. ACKNOWLEDGMENTS

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APPENDIX
DETAILS OF EVENTS RECORDED DURING SYSTEM OPERATION.

Key to columns:

Date, Time: Date and time (PDT through 10/27, PST thereafter) when the five-minute spectrum containing the listed peaks first began acquiring counts

% DT: Percentage dead time associated with the spectrum; high dead times are diagnostic of the presence of intense sources

Cont.: General magnitude of the continuum at energies below that of the reported peak(s); increased continuum is generally diagnostic of Compton scattering, in turn indicative that a heavily shielded source is present

Isotope: Identity of isotope inferred from the peak energies. Listing of the isotope as "511" indicates that some positron emitter was present. Identification of positron emitters cannot be made simply from the presence of the 511-keV peak. The isotopes ^{56}Co, ^{82}Rb, and ^{200}Tl, discussed in the main text, all emit positrons; the other isotopes discussed in the main text do not.

E (keV): Energy in keV of unusual peaks present in the spectrum, including normal background peaks with unusually high count rates. Energies are typically uncertain by about ±1 keV.

Counts: Net number of counts in the indicated peak. Uncertainties in the number of counts vary considerably but in any event are less significant in determining source strength than uncertainties in source-detector distance (as discussed in the text), hence are omitted. Detector efficiency for the detector used in these measurements has been found to obey the relation

$$\ln \varepsilon = -0.06673*x^6 -0.4001*x^5 -0.8685*x^4 -0.6009*x^3 +0.1849*x^2 -0.8148*x -9.528,$$

where ε = detector efficiency at source-detector distance = 25 cm ($1/r^2$ correction is adequately accurate for other distances)

$x = \gamma$ energy in MeV.

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
9/19	18:51	0.63	normal	Pb-203	278.43	85
9/22	17:26	0.63	normal	?	204.94	130
9/22	18:36	0.53	normal	?	241.30	128
9/23	14:29	7.20	10 x	I-123	439.81	724
				I-123	505.23	331
				511	511.00	52
				I-123	528.90	2605
				I-123	539.00	626
				Tl-200	579.25	438
				I-123	624.52	251
				I-123	735.92	202
				I-123,Tl-200	784.19	196
				Tl-200	828.28	881
				Tl-200	886.08	234
				Tl-200	1205.70	4900
				Tl-200	1225.37	542
				Tl-200	1252.00	149
				Tl-200	1260.00	15
				Tl-200	1273.30	569
				Tl-200	1290.94	163
				Tl-200	1362.96	819
				Tl-200	1407.54	246
				Tl-200	1475.00	45
				Tl-200	1514.88	744
				Tl-200	1570.40	62
				Tl-200	1604.31	217
				Tl-200	1630.00	14
				Tl-200	1690.00	8
				Tl-200	1718.33	59
				Tl-200	1758.85	38
				Tl-200	1904.00	17
				Tl-200	1971.00	15
				Tl-200	2270.00	8
				Tl-200	2296.52	10

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
9/28	08:39	0.70	1.5 x	I-131	364.24	276
				I-131	637.35	21
				I-131	721.00	17
10/02	14:01	1.10	3 x	In-111	245.32	67
				I-123	346.19	105
				I-123	439.89	190
				I-123	505.32	276
				I-123	528.91	1106
				I-123	538.00	301
				I-123	624.63	54
				Co-57	692.19	53
				I-123	735.59	49
				I-123	783.33	77
				Co-56	846.00	13
				Co-56	1236.00	34
				Co-56	1766.00	14
				Co-56	2598.00	9
10/04	12:36	0.60	2 x	I-131	364.27	483
				I-131	636.75	53
				I-131	720.00	9
10/04	12:41	0.57	2 x	I-131	364.35	337
				I-131	637.00	21
10/04	19:03	0.37	normal	511	511.00	36
10/05	12:55	0.33	normal	511	511.00	19
10/05	23:16	0.33	normal	K-40 excess	1460.00	107
10/06	18:18	0.53	normal	Tl-201	167.29	71
				annihil	511.00	28

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
10/07	14:20	0.90	3 x	I-123	439.86	254
				I-123	505.18	155
				I-123	528.90	1032
				I-123	538.00	258
				I-123	624.64	43
				I-123	687.82	19
				I-123	735.77	62
				I-123	783.59	60
10/07	14:25	3.00	12 x	I-123	346.50	572
				I-123	439.82	1679
				I-123	505.16	1671
				I-123	528.82	7425
				I-123	538.00	1979
				I-123	624.51	522
				I-123	687.74	141
				I-123	735.65	387
				I-123	783.40	398
				I-123	1067.18	24
10/07	14:51	0.33	0.75 x	511	511.00	19
10/08	13:52	1.10	3 x	Ga-67	393.31	174
				I-123	439.84	124
				I-123	505.31	208
				I-123	528.90	724
				I-123	538.00	190
				I-123	624.00	17
				I-123	687.00	3
				I-123	735.64	49
				I-123	782.00	56
				Ga-67	794.38	100
				Ga-67	887.62	358

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
10/09	13:18	1.47	3.5 x	In-111	245.22	178
				511	511.00	17
				I-123	528.70	110
				Tl-200	579.21	78
				Tl-200	661.00	47
				Tl-200	827.88	257
				Tl-200	886.68	99
				Tl-200	1205.32	1016
				Tl-200	1225.16	99
				Tl-200	1254.00	47
				Tl-200	1262.00	9
				Tl-200	1273.08	139
				Tl-200	1362.66	143
				Tl-200	1406.87	56
				Tl-200	1514.77	156
				Tl-200	1603.00	54
				Tl-200	1717.50	18
10/18	13:56	0.47	1.25 x	In-111	245.34	92
				511	511.00	18
				K-40 excess	1460.00	105
10/19	04:02	0.50	normal	511	511.00	12
10/19	08:47	0.47	normal	K-40 excess	1460.70	133
10/19	09:02	0.50	normal	511	511.00	14
				K-40 excess	1460.70	132
10/20	11:00	0.43	normal	Mo-99	140.00	50
				Ga-67	186.00	30
				Ga-67	300.00	0

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
10/21	14:27	3.70	12 x	I-123	439.92	345
				I-123	505.35	856
				511	511.00	263
				I-123	528.96	4764
				I-123	538.00	1203
				I-123	624.70	322
				Co-57	692.40	201
				I-123	735.73	234
				Rb-82	776.52	323
				I-123	782.00	248
				Co-56	846.65	87
				Tl-200	1205.51	20
				Co-56	1238.02	106
				Rb-82	1395.44	167
				K-40 excess	1460.00	107
				Rb-82	1474.00	43
				Rb-82	2479.00	12
				Co-56	2598.00	16
10/22	11:33	0.43	normal	Cs-137	661.80	15
				Bi-214 excess	1760.00	19
10/22	14:18	1.07	4 x	I-123	439.97	262
				I-123	505.15	282
				I-123	528.86	1323
				I-123	538.00	302
				I-123	624.54	82
				I-123	687.44	68
				I-123	735.56	69
				I-123	783.49	49
				Ga-67	792.85	5
				Ga-67	887.44	129

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
10/22	14:23	0.70	2.5 x	I-123	440.39	79
				I-123	505.25	63
				I-123	529.02	285
				I-123	538.00	83
				I-123	623.00	13
				I-123	783.00	14
				Ga-67	793.00	6
				Ga-67	887.62	33
10/22	23:55	0.47	normal	Mo-99	143.00	73
				Mo-99	181.00	27
				511	511.00	24
10/23	22:17	0.50	normal	511	511.00	16
				Bi-214 excess	1760.00	17
10/24	00:47	0.47	normal	K-40 excess	1460.70	129
10/24	11:08	0.50	1.3 x	Co-57	692.02	39
				Co-56	846.26	44
				Co-56	1237.71	60
				Co-56	2596.76	20
10/28	08:45	0.37	normal	bkg	350.00	74
				511	511.00	27
				bkg	609.00	43
				bkg	1120.07	18
10/28	12:46	0.47	normal	bkg	295.00	31
				bkg	352.00	52
				511	511.00	26
				bkg	609.00	56
				bkg	1120.00	11
				bkg	2204.00	8

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
10/28	15:36	1.00	3 x	I-123	439.82	392
				I-123	505.34	358
				I-123	528.84	1619
				I-123	538.00	354
				I-123	624.75	123
				I-123	688.21	30
				I-123	735.75	105
				I-123	783.41	90
10/29	14:58	0.83	3 x	I-123	439.00	84
				I-123	505.42	0
				I-123	528.76	110
				I-123	538.00	22
				Ga-67	794.25	33
				Ga-67	887.65	126
10/30	13:10	0.83	3 x	In-111	245.32	64
				I-123	439.93	320
				I-123	505.17	220
				511	511.00	10
				I-123	528.90	1138
				I-123	538.00	281
				I-123	624.52	60
					685.00	20
				I-123	735.79	28
				I-123	783.45	57
11/11	14:09	0.67	1.8 x	I-123	439.59	83
				I-123	505.13	63
				511	511	10
				I-123	528.91	365
				I-123	538	71
				I-123	625	14
				I-123	688	5
				I-123	735.88	27

Date	Time	% DT	Cont.	Isotope	E (keV)	counts
11/11	14:14	1.07	3 x	I-123	346.23	99
				I-123	439.92	354
				I-123	505.23	294
				511	511	20
				I-123	528.91	1430
				I-123	538	339
				I-123	624.68	115
				I-123	688.14	42
				I-123	735.8	61
				I-123	783.38	83
11/12	10:26	0.87	3 x	Cs-137	661.54	399
				K-40 excess	1460	105
11/18	15:31	1.67	5 x	I-123	346.14	241
				I-123	439.72	705
				I-123	505.17	634
				I-123	528.75	3249
				I-123	538	946
				I-123	624.43	248
				I-123	687.65	86
				I-123	735.49	132
				I-123	783.39	214
11/22	14:56	0.27	1.25 x	I-131	364.29	177
				511	511.00	17