

MEDICAL INDUSTRY INTERFERENCE WITH CTBT MONITORING OF ATMOSPHERIC RADIONUCLIDES

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ABSTRACT

The prototype International Data Center (pIDC) has been processing radionuclide data since 1995 to support the development of technology required for verification of the Comprehensive Nuclear-Test-Ban-Treaty (CTBT). This data contains gamma-ray spectra of environmental aerosol and gas samples collected from over 20 radionuclide stations. The gamma-ray spectra are analyzed resulting in the quantification of nuclide-specific atmospheric activity concentrations. Most of the radionuclides detected originate from natural sources. However, some result from nuclear weapons tests and releases from nuclear reactors, nuclear accelerators, and medical isotope production facilities. For nuclear-test-ban verification, it is important to be able to identify the type of source for anthropogenic radionuclide measurements.

Medical industry use of radioisotopes in the diagnosis and treatment of diseases has grown dramatically in the past 20 years. Medical radioisotopes are currently used in over 13 million procedures a year. As therapeutic isotope technology advancements reveal new modalities of treatment, this number may increase further resulting in a higher probability of the detection of such nuclides in environmental samples.

The pIDC has detected a number of radionuclides originating from the medical industry. These radionuclides include ^{123}I , ^{131}I , $^{99\text{m}}\text{Tc}$, and ^{201}Tl . Other radionuclides used in the medical industry have been detected (e.g., ^{137}Cs and ^{133}Xe), but are believed to originate from other sources. Some of these nuclides are the same as those considered to be highly indicative of a nuclear weapons test. Since high accuracy is desired in monitoring compliance to the CTBT, it is crucial to understand the potential for radionuclides released from medical facilities to interfere with signals from nuclear weapons tests.

Key Words: radionuclide monitoring, aerosol, prototype International Data Centre, medical radionuclides, ^{123}I , ^{131}I , $^{99\text{m}}\text{Tc}$, and ^{201}Tl

OBJECTIVE

The purpose of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) is to cease “all nuclear weapons test explosions and all other nuclear explosions” thereby preventing nuclear weapons proliferation and enhancing international peace and security [CTBT]. Verification of the Treaty is achieved by a network of seismic, hydroacoustic, infrasound, and radionuclide stations that continuously monitor for indicators of a nuclear weapons blast.

The prototype International Data Center (pIDC) plays a pivotal role in the development and deployment of the required technology for the International Monitoring System (IMS). Since the beginning of 1996, the pIDC has received data from at least eight radionuclide monitoring stations [Mason et al., 1996]. Today, that number has increased to twenty [Bohner and Mason, 1998]. (See Table 1 for a listing of the current radionuclide monitoring stations and Figure 1 for a map showing their locations.) As a result, the pIDC has a large, comprehensive database of international atmospheric radionuclide data. The complete future radionuclide monitoring system (RMS) will contain 80 stations [CTBT].

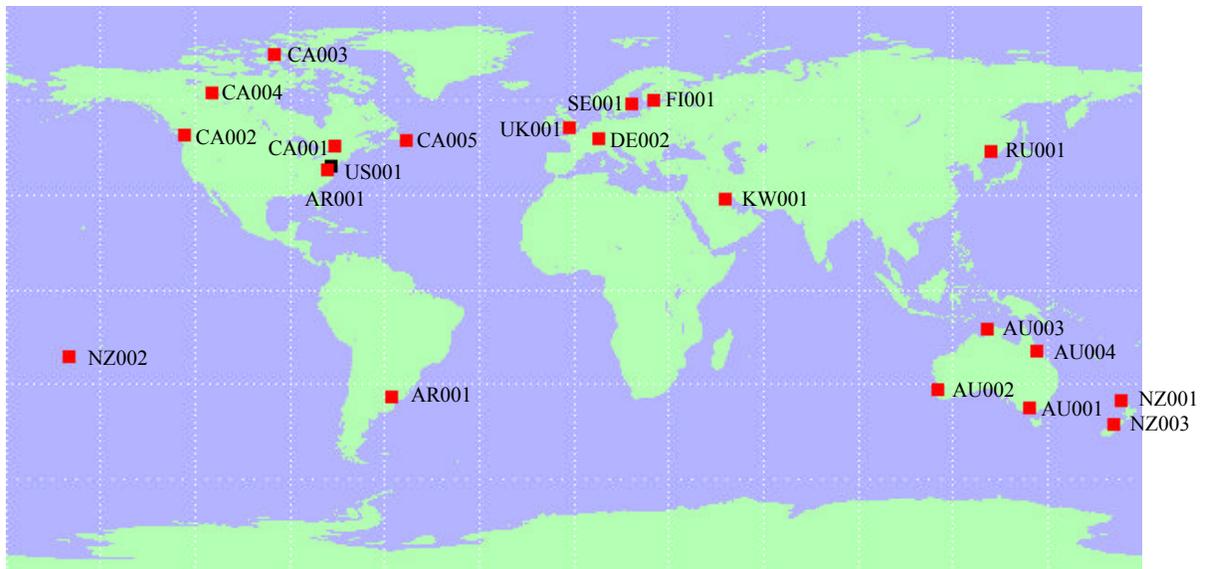


Figure 1. Current Operating Radionuclide Monitoring Stations of the pIDC

Challenges to the successful monitoring of compliance to the CTBT can be identified by studying the aerosol data stored at the pIDC. Radionuclides from various sources other than nuclear weapons tests have been detected by the Radionuclide Monitoring System (RMS) including nuclear power reactors, accelerators, medical isotope production facilities, factories, re-entrainment of fall-out, and natural sources. The objective of this paper is to identify the radionuclides detected by the RMS that have been determined to originate from the medical industry, and to discuss the effects of these interference signals in the context of CTBT verification.

RESEARCH ACCOMPLISHED

Radioactive isotopes are used in more than 38,000 diagnostic medical procedures each day in the United States [DOE, 1999]. Over 100 different radionuclides are currently administered for such purposes [NMRC, 1999]. The production of radioactive medical isotopes and the waste generated from their use are potential sources of atmospheric radioaerosols and gases. Although there are strict procedures in place for the disposal of radiopharmaceuticals and generated refuse, some of the activity is not properly disposed

because it is carried internally by patients outside of the appropriate medical departments [Beretta et. al, 1997]. Incineration of medical wastes is also a potential source for radionuclides in the atmosphere.

Table 1. Radionuclide Stations Currently Contributing Data to the pIDC

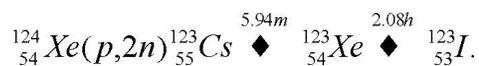
Station Code	Location	Start of Data Processing in pIDC Operations
AR001	Buenos Aires, Argentina	January, 1996
AU001	Melbourne, Australia	September, 1995
AU002	Perth, Australia	January, 1997
AU003	Darwin, Australia	February, 1997
AU004	Townsville, Australia	February, 1997
CA001	Ottawa, Canada	December, 1995
CA002	Vancouver, Canada	April, 1996
CA003	Resolute, Canada	May, 1996
CA004	Yellowknife, Canada	May, 1996
CA005	St. John's, Canada	June, 1996
DE002	Schauinsland, Germany	January, 1996
FI001	Helsinki, Finland	January, 1996
KW001	Kuwait City, Kuwait	September, 1995
NZ001	Kaitaia, New Zealand	May, 1995
NZ002	Rarotonga, New Zealand	May, 1996
NZ003	Hokitika, New Zealand	May, 1996
RU001	Ussuriysk, Russia	November, 1995
SE001	Stockholm, Sweden	September, 1995
UK001	Chilton, England	June, 1996
US001	Charlottesville, USA	December, 1995

The pIDC has identified signals from the following medical radioisotopes in atmospheric aerosol samples: ^{123}I , ^{131}I , $^{99\text{m}}\text{Tc}$, ^{201}Tl , ^{137}Cs , and ^{133}Xe . Although both ^{137}Cs and ^{133}Xe are used in the medical industry and have been detected numerous times by the RMS, the sources of these nuclides have been determined to be resuspension of fallout from nuclear weapons and Chernobyl as well as from nearby nuclear power reactors, respectively. Some detection instances for the other radionuclides listed have been directly correlated to releases from medical facilities. The implications of this phenomenon on the efficacy of monitoring compliance to the CTBT are addressed in this paper.

^{123}I ($t_{1/2} = 13.2$ hours)

This medical radioisotope is used mainly for diagnosis purposes. Depending upon test protocols, ^{123}I may be ingested or injected. By observing the absorption of ^{123}I in the body, malfunctions of the brain, thyroid, and kidney can be determined. The predominate γ -line of ^{123}I at 159 keV (83.3%) can also be utilized for myocardial and cerebral imaging.

^{123}I is cyclotron-produced by proton bombardment of ^{123}Cs by the reaction:



Although ^{123}I has been observed at only one station in the RMS, signal detection of this radiopharmaceutical occurs on a frequent basis as shown in Table 2 below.

Table 2. ^{123}I Detections at RMS Station CA002 in Vancouver, Canada from 1996 Until 14 June 1999

^{123}I							
Station	Total Spectra Reviewed at Station	Number of Spectra with ^{123}I	Percent of Spectra with ^{123}I	Average (Bq/m^3)	Standard Deviation (Bq/m^3)	Minimum (Bq/m^3)	Maximum (Bq/m^3)
CA002	1104	342	30.98	334.4	744.1	5.09	6202

Due to the high detection frequency and periodicity of ^{123}I , a source term investigation for this nuclide was initiated at the pIDC. It was found that a commercial radioisotope production facility is located 2 km southeast of the CA002 monitoring site [Mason and Williams, 1999]. Operational data was obtained from the facility and compared to the detections of ^{123}I . The results are displayed in Figure 2. A temporal correlation between the facility's production schedule and the detection of ^{123}I is evident.

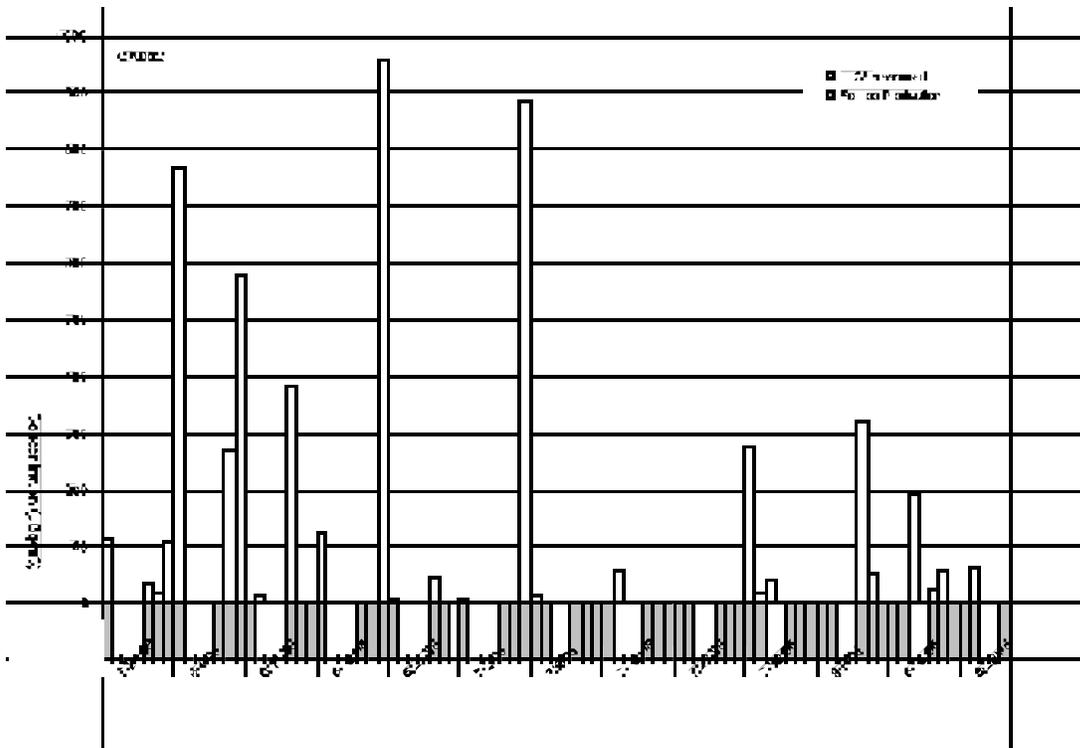


Figure 2. Dates of ^{123}I Production and Detection at CA002 [Mason and Williams, 1997]

Because ^{123}I is not considered a relevant isotope for CTBT monitoring, and because it doesn't interfere with signals from relevant CTBT isotopes, the detection of this medical isotope should not impact the efficacy of the RMS in achieving its monitoring goals.

^{131}I ($t_{1/2} = 8.020$ days)

This radiopharmaceutical is utilized in many radiotherapy protocols and diagnostic procedures. One of the most popular medical uses of ^{131}I is for the treatment of hyperthyroidism. Others include various imaging techniques; use for antibody lab biochemistry in mental illness; locating infections, tumors, and metastatic lesions; radiolabeling; and treatment of the following medical conditions: carcinoma of the thyroid, graves disease, goiters, prostate cancer, hepatocellular carcinoma, melanoma, spinal tumor, neuroblastoma [NMRC, 1999].

^{131}I is a fission product and is produced in reactors for the medical industry. Unlike ^{123}I , this isotope is considered a relevant fission product in monitoring compliance of the CTBT. Its release from sources other than a nuclear weapons test will reduce the sensitivity of the RMS to this particular radionuclide.

Table 3 displays incidences when ^{131}I was observed in RMS atmospheric samples. Some of these detections are more likely from reactor releases, especially those from stations FI001 in Helsinki, Finland and SE001 in Stockholm, Sweden.

Table 3. ^{131}I Detections at RMS Stations from 1996 until 14 June 1999

^{131}I							
Station	Total Spectra Reviewed at Station	Number of Spectra with ^{131}I	Percent of Spectra with ^{131}I	Average (Bq/m ³)	Standard Deviation (Bq/m ³)	Minimum (Bq/m ³)	Maximum (Bq/m ³)
AR001	363	17	4.7	12.3	13.0	2.57	49.4
CA002	1104	1	0.1	3.51	-	-	-
DE002	187	1	0.5	0.305	-	-	-
FI001	415	12	2.9	0.403	0.232	0.151	0.945
KW001	999	6	0.6	5.27	3.10	2.30	9.95
NZ001	143	1	0.7	1.05	-	-	-
NZ003	151	2	1.3	1.04	0.315	0.819	1.27
SE001	718	4	0.6	1.37	0.538	0.681	1.98
UK001	162	1	0.6	0.411	-	-	-
US001	1132	2	0.2	21.6	7.46	16.3	26.8

There are several ways to determine the source of ^{131}I . First, the station history must be researched and studied. If ^{131}I has never been observed at a station, its detection at that station should arouse suspicion, especially when observed in conjunction with other relevant fission products. If other fission products are identified and quantified within the same γ -ray spectrum, the source is likely not medical and radionuclide ratios may be utilized to discriminate between nuclear power emissions and a nuclear weapons test. If ^{131}I has been detected on occasion or frequently at a station, a source investigation like the one described for ^{123}I is extremely useful. In most cases, this results in the identification of a legitimate radionuclide source. The placement of stations upwind or far from possible sources of ^{131}I can greatly reduce interferences from this isotope.

 $^{99\text{m}}\text{Tc}$ ($t_{1/2} = 6.01$ hours)

$^{99\text{m}}\text{Tc}$ is used in over 80 percent of medical procedures requiring the administration of radionuclides [DOE, 1999]. It is estimated that 675 MBq of $^{99\text{m}}\text{Tc}$ were released to the environment in 1994 alone [Beretta et al., 1997].

Many characteristics of this isotope add to its versatility. For example, the half-life is long enough to examine metabolic processes yet short enough to minimize the patient's dose. The patient's dose is further reduced because there are no high-energy beta emissions to damage surrounding tissues. In addition, the low energy γ -rays of ^{99m}Tc easily escape the body and can be accurately detected by a γ -camera. The chemistry of technetium allows it to be incorporated into a range of biomolecules, which concentrate in different organs such as the brain, heart, liver, lungs, bones, thyroid, and kidney.

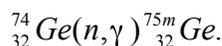
^{99m}Tc is produced on-site in generators. Hospitals and other medical facilities are supplied with such generators directly from the production reactor. A ^{99m}Tc generator is comprised of a lead container enclosing a chromatography column to which ^{99}Mo (the parent of ^{99m}Tc) is adsorbed [MDS Nordion S.A., 1997]. ^{99}Mo is a fission product with a half-life of 65.9 hours. ^{99m}Tc is eluted from the generator by a saline solution when required for use in a procedure. After two weeks or less, the generator is spent and must be returned to the production facility for recharging.

As shown in Table 4, this radiopharmaceutical has been detected at seven RMS stations in fifty spectra. The predominant γ -line of ^{99m}Tc at 140.5 keV (89.0%) is difficult to differentiate from that of ^{75m}Ge (139.7 keV at 38.8%). Before Feb. 4, 1998 the pIDC would generally identify signals at ~ 140 keV as ^{75m}Ge . This is because the key γ -line energy used by the pIDC for ^{75m}Ge (139.9 keV) was slightly higher than the value in the most recent decay data library from Brookhaven National Laboratory (139.7 keV). This problem has since been corrected, but earlier spectra have not yet been reanalyzed. Therefore, it is possible that ^{99m}Tc is present in other samples but not identified.

Table 4. Positive ^{99m}Tc Detections at RMS Stations from 4 Feb. 1998 Until 14 June 1999

^{99m}Tc							
Station	Total Spectra Reviewed at Station	Number of Spectra with ^{99m}Tc	Percent of Spectra with ^{99m}Tc	Average (Bq/m^3)	Standard Deviation (Bq/m^3)	Minimum (Bq/m^3)	Maximum (Bq/m^3)
AR001	363	10	2.8	756	508	287	1980
AU001	799	14	1.8	227	120	89.9	448
CA002	1104	16	1.5	189	126	101	609
DE002	187	4	2.1	1.62E+05	1.88E+05	142	4.01E+05
KW001	999	4	0.4	169	102	34.3	279
RU001	498	1	0.2	4.44E+04	-	-	-
US001	1132	1	0.1	339	-	-	-

^{75m}Ge is produced by neutron bombardment of ^{74}Ge within the detector by the following reaction:



There are several ways to differentiate between ^{75m}Ge and ^{99m}Tc when a peak is observed near 140 keV. The first is to attempt nuclide identification by peak centroid energy alone. The difference in the line energies, as provided in the Brookhaven National Laboratory library, is 0.8 keV. However, this method is not always reliable due to uncertainties of peak centroid locations and uncertainties of the energy calibration. A second approach includes utilizing both the preliminary (4 hour) spectrum and the full spectrum (~ 24 hours) of a particular sample. By using both spectra, the half-life of the radionuclide at ~ 140 keV can be determined. Since ^{75m}Ge would be continually produced throughout the gamma-ray acquisition, the decay rate would be nearly constant (assuming a constant cosmic-ray flux). The calculated half-life of ^{75m}Ge would consequently be very large. In comparison to the half-life of ^{99m}Tc at 6.01 hours or its parent, ^{99}Mo , at 2.75 days, it is easy to discriminate between the nuclides by employing this test.

Once ^{99m}Tc has been positively identified by one or both of the methods above, the most probable source of the nuclide may then be determined. The sample is first checked for the presence of ^{99}Mo . The detection limit for this nuclide is lower than that of ^{99m}Tc because its primary γ -line at 739.5 keV is only 12.2% abundant. However, if ^{99}Mo is present, then the likelihood of the ^{99m}Tc originating from a medical source is very low. This is because ^{99m}Tc is chemically separated from its parent during elution from a generator. Determination of the nuclide's half-life, as previously described, can also be used for nuclide source determination. If the half-life matches that of ^{99}Mo , then the two nuclides are in secular equilibrium and possibly originate from the direct fission product release of a nuclear reactor or weapons blast. If the half-life matches that of ^{99m}Tc , then the nuclide is likely from a medical industry source.

There is one station in Table 4 that does not send preliminary spectra: AR001 in Buenos Aires, Argentina. Therefore, the half-life test cannot be performed on samples from this station. However, samples from this station have been checked for ^{99}Mo and none show any evidence of this nuclide. Correspondence with station personnel indicates the presence of five potential nearby sources of ^{99m}Tc , all of which are medical centers and laboratories. The list includes:

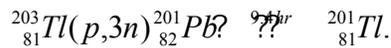
1. Tecnuar (3 km W),
2. Bacon (5 km NW),
3. Vilela Medical Center (3.3 km S),
4. Diagnostico Maipu (2.3 km N), and
5. Fernandez Hospital (5.6 km S).

Consequently, the likelihood of ^{99m}Tc detected at AR001 originating from a medical source is very high.

^{99m}Tc is a relevant fission product for monitoring CTBT compliance. The presence of this nuclide from medical sources will decrease the station sensitivity towards ^{99m}Tc from other sources. However, methods are available to determine the source of the nuclide.

^{201}Tl ($t_{1/2} = 3.04$ days)

This radionuclide is used for myocardial scintigraphy in the evaluation of coronary perfusion and cellular viability. It is also used for scintigraphy of muscles in cases of peripheral vascular disorders, parathyroid scintigraphy, and tumor visualization in different organs, especially the brain and thyroid. ^{201}Tl is cyclotron-produced by proton bombardment of ^{203}Tl by the reaction:



Only one RMS station, AR001, has had confirmed detections of this radiopharmaceutical. Considering the location of the station near so many medical centers, this phenomenon is not surprising. However, earlier detections of ^{201}Tl could have been missed by the pIDC because it was initially not included in the software radionuclide library. This problem has since been corrected, but earlier spectra have not yet been reanalyzed. Table 5 summarizes data on the detection instances at AR001.

Table 5. ^{201}Tl Detections at RMS Station AR001 from 1996 Until 14 June 1999

^{201}Tl							
Station	Total Spectra Reviewed at Station	Number of Spectra with ^{201}Tl	Percent of Spectra with ^{201}Tl	Average (Bq/m^3)	Standard Deviation (Bq/m^3)	Minimum (Bq/m^3)	Maximum (Bq/m^3)
AR001	363	4	1.1	212	206	22.5	466

^{201}Tl is not considered a relevant anthropogenic nuclide for monitoring compliance to the CTBT, nor does any of its decay photons interfere with any major lines of relevant nuclides. Therefore, the presence and detection of this nuclide by RMS stations should not impact the analysis of spectra.

CONCLUSIONS AND RECOMMENDATIONS

Based on international aerosol data compiled at the pIDC from the current RMS network, it is evident that several stations have detected radionuclides originating from medical sources. It has been shown that the detection of ^{123}I and ^{201}Tl have little effect on the ability of the RMS to monitor compliance of the CTBT. In contrast, the detection of other radiopharmaceuticals, like $^{99\text{m}}\text{Tc}$ and ^{131}I , can complicate the analysis of RMS data. To address this problem, methods have been devised to aid in nuclide and source identification.

When the RMS becomes fully operational with 80 international stations, the detection instances of medical isotopes will undoubtedly increase. It is also expected that radiopharmaceuticals other than those previously identified will be detected. (Table 6 lists radionuclides that are both radiopharmaceuticals and potentially indicative of a nuclear weapons test.) Depending on station location and local meteorological conditions, stations will observe varying amounts of interference from medical isotopes.

To reduce the impact of medical isotopes on RMS operations, local siting of stations should be optimized. When available, historical sample data should be used as an important source of information for nuclide and source identification. By determining the sources of radionuclides, we develop a good understanding of the environment and maintain awareness of possible interferences with nuclear weapons test detections.

Table 6. Radiopharmaceuticals that Are Also Potentially Indicative of a Nuclear Weapons Test

Nuclide	Half-Life	Medical Uses [NMRC, 1998]
Am-241	432 y	Osteoporosis detection, heart imaging.
As-74	17.8 d	Biomedical applications.
Au-198	2.69 d	Cancer treatment using mini-gun ; treating ovarian, prostate, and brain cancer.
Ce-141	32.5 d	Gastrointestinal tract diagnosis, measuring regional myocardial blood flow.
Co-57	272 d	Gamma camera calibration, radiotracer, and source for X-ray fluorescence spectroscopy.
Co-60	5.27 y	Destroy cancer cells, disinfect surgical equipment and medicines, external radiation cancer therapy.
Cr-51	27.7 d	Cell labeling and dosimetry.
Cs-137	30.2 y	Blood irradiators, PET imaging, tumor treatment.
Cu-64	12.7 h	PET scanning, planar imaging, SPECT imaging, dosimetry studies, cerebral and myocardial blood flow, treating colorectal cancer.
Eu-152	13.4 y	Medical.
Eu-155	4.73 y	Osteoporosis detection.
Fe-59	44.5 d	Medical.
I-131	8.04 d	See text, page 3.
Ir-192	73.8 d	Treatment of cancers of the prostate, brain, breast, gynecological cancers.
Mo-99	65.9 h	Parent for $^{99\text{m}}\text{Tc}$.
Nb-95	35.0 d	Myocardial tracer and PET imaging.
Pb-203	2.16 d	SPECT and PET planar imaging (used with Bi-212), monoclonal antibody immunotherapy, cellular dosimetry.
Pd-109	13.4 h	Potential therapeutic agent.
Rh-105	35.4 h	Potential therapeutic agent.
Ru-103	39.3 d	Myocardial blood flow, radiolabeling microspheres, PET imaging.
Sc-46	83.8 d	Regional blood flow studies, PET imaging.

Table 6. CTBT-Relevant Radionuclides Used in the Medical Industry (continued)

Nuclide	Half-Life	Medical Uses [NMRC, 1998]
Sc-47	3.34 d	Cancer treatment and diagnostics, monoclonal antibodies, radioimmunotherapy.
Sm-153	2.00 d	Cancer treatment and diagnosis, monoclonal antibodies, bone cancer pain relief, treatment of leukemia.
Tc-99m	6.01 h	See text, page 4.
Tm-170	129 d	Portable blood irradiations for leukemia, lymphoma treatment, power source.
Xe-133	5.25 d	Lung imaging, regional cerebral blood flow, liver imaging, SPECT imaging of brain, lesion detection.
Y-88	107 d	Cancer tumor therapy.
Y-91	58.5 d	Cancer treatment, cellular dosimetry.
Zn-65	244 d	Medical.
Zr-95	64.0 d	Medical.

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