Radionuclide Laboratory Support for the Comprehensive Nuclear-Test-Ban Treaty International Monitoring System

Pamela D. Greenlaw, Colin G. Sanderson
Environmental Measurements Laboratory

Sponsored by The Defense Threat Reduction Agency
Arms Control Technology Division
Nuclear Treaty Branch

ABSTRACT

The protocol to the Comprehensive Nuclear-Test-Ban Treaty (CTBT) provides for the designation of a number of radionuclide laboratories to support the radionuclide monitoring network of the International Monitoring System (IMS). These laboratories, after being certified and when requested by the Technical Secretariat, are to provide additional analytical services, such as confirmatory gamma-ray spectrometry, quality control and intra laboratory exercises. The Environmental Measurements Laboratory (EML) has been designated as the U. S. CTBT Radionuclide Laboratory. EML is developing a separate secure facility for performing the required CTBT analyses and is currently set up for gamma-ray spectrometry with two, 120%, HPGe detectors. The laboratory will undergo a preliminary certification exercise with the CTBT Technical Secretariat which will included verification of the laboratory's quality assurance plan as well as it's current analytical capability. EML's CTBT laboratory is in the process of expanding its radiochemistry and counting capabilities to include both alpha and beta emitting radionuclides. This paper presents EML's continuing development of a radionuclide laboratory to corroborate the results of the routine analysis of a sample from an IMS radionuclide station, achieve a greater sensitivity for fission product analysis than can be achieved at the radionuclide stations and clarify the presence or otherwise of fission products in the case of a suspect or irregular analytical result from a particular radionuclide station.

KEY WORDS: Radionuclide Laboratory, International Monitoring System, Laboratory Certification

OBJECTIVE

The objective of this project is to establish the United States Comprehensive Nuclear-Test-Ban Treaty Radionuclide Laboratory to support the radionuclide monitoring network of the International Monitoring System.

Introduction

Since 1963 EML has maintained a world wide network of surface air sampling stations to monitor and inventory nuclear fallout. After collection, these filters are returned to EML and are analyzed for gamma-ray emitting radionuclides. Because of this long history of measuring aerosol particulates for radioactivity and the laboratory's active role in providing quality assurance for the Department of Energy's (DOE) Environmental Management programs, EML was selected by AFTAC to provide the United State's laboratory support for the CTBT radionuclide International Monitoring System (IMS) stations.

Requirements called forth in the treaty specify that a the network of radionuclide monitoring stations shall be supported by laboratories, which shall be certified by the Technical Secretariat in accordance with the relevant operational manual for the performance, on contract to the Organization and on a fee-for-service basis, of the analysis of samples from radionuclide monitoring stations. These laboratories shall, as required, also be drawn upon by the Technical Secretariat to perform additional analysis of samples from radionuclide monitoring stations. Additional requirements have been developed by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), the Radionuclide Task Group 11/5 of Working Group B of the Preparatory Commission and at informal radionuclide workshops.

The US CTBT Laboratory

In order to meet the requirements of the treaty and the Operational Manual for Radionuclide Monitoring and the International Exchange of Radionuclide Data (CTBT/WGB/TL-11/5/Rev.8), EML chose to dedicate space within the laboratory for a secure CTBT laboratory comprised of three existing interconnecting laboratories. Each laboratory has a designated function; the first is used for sample receipt, log in and sample preparation for gamma counting; the second is a radiochemistry laboratory; and the third will be the counting laboratory.

Security

In order to maintain security, access to the laboratory will require a personalized card key. Entry into laboratory with the card key will be recorded in the CTBT laboratory's communications and analysis computer.

Five identical workstations will exist on their own independent LAN and will only be accessible after an operator has had the bar code on their DOE ID badge read and verified.

The Laboratory has documented plans and facilities for receipt of samples 24 hours a day, 7 days a week. SOPs to maintain sample chain of custody from time of receipt until final dispatch are in place and bar code readers are installed at appropriate locations in order to automate the chain of custody.

VSAT Communications with the Technical Secretariat and International Data Center
The CTBT requires secure and authenticated communications between the CTBTO Technical Secretariat, the International Data Center (IDC) and the IMS supporting Radionuclide Laboratories. The types of notifications from the Technical Secretariat and IDC to the laboratory and the required acknowledgments from the laboratories to the Technical Secretariat and IDC are listed below. These messages are authenticated and transmitted via Technical Secretariat installed VSAT e-mail and FTP. A backup communication system is also be available in the event that the GCI VSAT in unavailable. For either incoming or out going messages a dial-up connection to the EML LAN or a dial-up connection to a commercial ISP will be used to communicate with the Technical Secretariat/IDC.

Types of Messages

From the Technical Secretariat or IDC to the Laboratory	From the Laboratory to the Technical Secretariat or IDC
1. Initial notification of intent to dispatch a sample	1. Acknowledgment of intent to dispatch a sample and agreement to accept the sample
2. Notification of sample dispatch	2. Acknowledgment of sample dispatch notification
3. Sample collection information	3. Acknowledgment of sample receipt
4. Analysis required and any other information available about the sample	4. Report of preliminary results
5. Acknowledgment of receipt of preliminary results report	5. Report of intermediate counting results
6. Acknowledgment of receipt of intermediate counting results and possible request to terminate analysis	6. Final report of sample analysis

Sample Receipt, Inspection and Log-in
All samples sent to the CTBT Laboratory are received, inspected and logged in by the CTBT sample custodian. As an aid for this process each sample will have a unique printed bar code. The EML guard or the GSA Building guard notifies the sample custodian when shipping containers arrive.

Initial inspection of the shipping containers takes place in the CTBT Sample Receipt Laboratory. The shipping containers are placed in a hood and screened for external radiation before being opened. After all sample information has been verified and the sample bar code read into the computer, the samples are logged in, then transferred to the sample preparation area and either prepared for gamma counting or transferred to a locked storage cabinet.

Filter Sample Preparation (including sample splitting)
The CTBT Sample Preparation Laboratory is equipped with a heated manual hydraulic press capable of achieving over 10 metric tons and 650 degrees F. With the availability of a low temperature muffle furnace aerosol filter

samples can be pressed, heat pressed or melted into standard geometries for gamma counting. Sample identification is maintained with the use of the sample preparation bar code reader.

Tests performed with the US RASA filters indicated that equivalent counting efficiencies are achieved with either the unprocessed filters wrapped around the side of the detector or the filter melt in a 600 ml beaker on the detector end cap.

If required, the laboratory will be prepared to split any samples received

System Requirements for Gamma-Ray Sample Analysis
The radionuclide operational manual gives these gamma-ray analysis specifications for the IMS stations:

1.	*Air flow*	500 m^3 /hour
2.	*Collection time*	24 hours
3.	*Measurement time*	>20 hours
4.	*Measurement mode*	HPGe high-resolution gamma spectrometry
5.	*HPGe relative efficiency*	Equal to or greater than 40%
6.	*HPGE resolution*	< 2.5 keV at 1332 keV
7.	*Baseline sensitivity*	10 to 30 □Bq/m^3 for ^{140}Ba
8.	*Calibration range*	88 to 1836 keV

The requirements in the radionuclide operational manual for laboratories supporting the IMS require lower limits of detection for ^{140}Ba. These minimum laboratory requirements are:

Property	**Minimum Requirement**
Detector type	High resolution HPGe
Detector relative efficiency	40 %
MDA for ^{140}Ba for cylindrical sample geometry with a diameter of 70 mm and height of 5 mm with decay correction to start of spectral acquisition (not to exceed a 7 day count)	24 mBq
FWHM at 1333 keV	2.3
FWHM at 122 keV	1.3
Report format	IMS current version

The radionuclide operational manual specifies the requirement for a minimum detectable concentration (MDC) at radionuclide stations as a concentration range for ^{140}Ba of 10 to 30 □Bq m^{-3} for a 24-hour count and a laboratory requirement for a better minimum detectable activity (MDA). Accordingly, the MDA requirement for Laboratories is calculated from the low end of the station requirement range using a total air sample flow of 12,000 m^3. Laboratory measurement systems must, therefore be capable of achieving a MDA of 24 mBq for ^{140}Ba using the counting efficiency for a compressed cylindrical geometry of 70mm diameter and 5mm height and a count time of 7 days or less.

In order to minimize the possibility of down tine, two high efficiency p-type HPGe detectors have been installed in the EML CTBT counting laboratory. One detector has a relative efficiency of 120% and a FWHM resolution of 1.92 keV at 1333 keV. The second detector has a relative efficiency of 114% and a FWHM resolution of 1.82 keV at 1333 keV . Both detector are housed in low background cryostats and are shielded with copper, cadmium and 10 cm of very old lead. The ^{140}Ba MDAs for these detectors with different count times are shown below:

Gamma Detection MDA (^{140}Ba @ 537.27 keV)

Detector	MDA and Count Time		
CTBT 1	19.4 mBq - 7 days	25.6 mBq - 4 days	24 mBq - 4.55 days
CTBT 2	20.4 mBq - 7 days	27.0 mBq - 4 days	24 mBq - 5.06 days

The data above show that the CTBT Laboratory HPGe gamma-ray detectors can achieve the required ^{140}Ba MDA in less than the limiting 7 day count time restriction.

Radiochemical Analysis and Counting
The following excerpts from the Treaty can be interpreted to indicate that the CTBT Laboratories may be called upon to perform additional analyses beyond the initial nondestructive gamma-ray analysis. These additional analyses could be for alpha or beta emitting radionuclides and possibly noble gasses.

11. ...as required, also be drawn upon by the Technical Secretariat to perform additional analysis of samples from radionuclide monitoring stations.

98. ...The inspection team shall have the right to transfer samples for off-site analysis at laboratories designated by the Organization only if it demonstrates that the necessary sample analysis cannot be performed on-site.

101. The designated laboratories shall conduct chemical and physical analysis of the samples transferred for off-site analysis....

The EML CTBT Laboratory has been equipped with a fully functional radiochemistry laboratory, containing a chemical fume hood, muffle furnace, centrifuges, balance, glass ware and filtering apparatus. The counting laboratory has been equipped with a bank of 8 solid state alpha detectors and 4 EML beta scintillation counters. A beta-gamma coincidence gas counting system is planned for the future.

Chain of Custody
All samples received by the CTBT Laboratory shall have a legal chain of custody (COC) established, which records a continuous record of the physical possession, storage and analysis of samples. The COC at EML begins when the samples arrive at the fifth floor guard station. The three laboratory work stations (in the sample receipt, sample preparation and sample counting/communications area) have bar code readers which provides input to the CTBT laboratory information management system (LIMS). The LIMS provides a means for sample tracking and COC.

Laboratory Operational Manual
All laboratory operations, written as standard operating procedures (SOP), are maintained in the Laboratory Operational Manual. This manual also contains copies of the EML Institutional Quality Assurance Plan, CTBT Laboratory Quality Assurance Plan, ISO 17025, all equipment manuals and calibration certificates.

The Certification Process
The Comprehensive Nuclear-Test-Ban-Treaty states in its:
Protocol to The Comprehensive Nuclear Test-ban Treaty, Part I, A. General Provisions,
C. Radionuclide Monitoring, Section 11;

The network of radionuclide monitoring stations shall be supported by laboratories, which shall be certified by the Technical Secretariat...

These laboratories are certified in accordance with the radionuclide operational manual. The process of certification takes place in several steps, involving preparation and evaluation at both the Technical Secretariat in Vienna and the Radionuclide Laboratory and can be summarized in the following steps:

1. Formal Arrangement:
All certification procedures begin with a formal Memorandum of Understanding (MOU) and/or other arrangements as initiated by the respective National Authority with the Technical Secretariat to commence the process of

certification.

2. Document Review:
The laboratory supplies the Technical Secretariat with all quality documentation including the quality manual, SOPs and equipment manuals (with certification, specification and QC data sheets) , and all quality control documentation.

3. Confirmation of the Quality of Documented Analysis Results by Gamma Spectrometry:
The Technical Secretariat reviews the documentation of any intercomparison exercises in which the laboratory has participated.

4. Functional Test of Communications:
The laboratories GCI terminal should successfully test their communication link by exchanging information text messages described in the IDC Operations Manual.

5. Functional Test of Data Authentication:
The radionuclide laboratories conduct a test to ensure authentic data is transferred from the analysis system to the communication system with the IDC.

6. Test of Data/Report Format and Transfer to the IDC:
The radionuclide laboratories must send sample test data and sample reports to the IDC to allow review for correctness of format.

7. Certification Visit by Technical Secretariat Staff to Examine Equipment and Laboratory:

7.1. Physical Facilities
The Technical Secretariat will confirm that the conditions at the laboratory correspond to the information provided and reviewed prior to the visit and confirm that the Quality System is executed as documented.

7.2. Equipment and Reference Materials
Sample preparation equipment including laboratory press, balance and oven will be inspected. Review calibration and decontamination procedures and maintenance record. Inspect reference materials to confirm traceability documentation and storage adequacy.

7.3. Gamma Spectrometer
Instrument serial numbers will be compared to serial numbers on manufacturer's specification sheets. A Technical Secretariat radionuclide source in calibrated geometry will be counted to compare results as reported by same gamma spectrometer and software used to determine CTBT samples.

A background filter supplied by Technical Secretariat will be counted in same geometry as the Technical Secretariat source to confirm MDA calculation.

8. Issue of Certificate
When the laboratory meets IMS specifications and certification criteria the Technical Secretariat will issue a certificate to the Laboratory to formally recognize it as a laboratory supporting the IMS. If the laboratory does not fulfil the certification criteria on the first visit of the Technical Secretariat, the Technical Secretariat will issue a report delineating deficiencies and revaluation criteria. The Technical Secretariat will schedule a reinspection and/or review required corrective action documentation to complete the certification process.

9. Long Term Quality Assurance
The Technical Secretariat will establish a long-term program of quality assurance including intercomparison exercises between laboratories. Criteria will be established to evaluate the results of the intercomparison exercises. The results of Technical Secretariat sponsored and other international intercomparison exercises will be used by the Technical Secretariat to monitor the laboratory performance as available.

When changes occur in the laboratory to facilities, personnel or equipment, the Technical Secretariat shall be

notified and supplied with new documentation. If the change is deemed substantial (i.e., relocation of laboratory, replacement of gamma detectors) the Technical Secretariat will perform a re-certification of the laboratory. The results of other international intercomparison exercises will be used by the Technical Secretariat to monitor the laboratory performance.

To be certified a laboratory must establish and maintain a quality system based on ISO 17025. However, if a particular Radionuclide Laboratory is nationally or internationally accredited, then meeting the quality system requirements should require no more than a review of the existing quality documentation related to CTBT operations.

All measuring and testing equipment having an effect on the accuracy or validity of sample analyses should be calibrated so as to ensure measurements are traceable to national standards.

The requirements for the physical topology of the Global Communications Infrastructure (GCI) are the same for the laboratories as they are for the radionuclide stations. This means each laboratory will be connected to the IDC via the GCI using the basic topology or other permitted configuration.

During the certification process the laboratory is required to demonstrate to the Technical Secretariat that the environment in which IMS samples are processed and analyzed is maintained to sufficiently high standards.

SOPs for preparation of samples for gamma analysis have been prepared and are available. If different procedures are to be used for samples of different matrices/ geometries, these SOPs shall be available with validation data. If requested and authorized by the Technical Secretariat, other concentration, separation and radiochemical analysis methods can be used. SOPs and validation data for other preparation methods will be prepared and made available to the Technical Secretariat.

EML will work with the Technical Secretariat to finalize implementation plans for lab certification during the coming months.