

PERFORMANCE EVALUATION AND OPERATIONAL EXPERIENCE WITH A
SEMI-AUTOMATIC MONITOR FOR THE RADIOLOGICAL CHARACTERIZATION
OF LOW-LEVEL WASTES

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ABSTRACT

Chalk River Nuclear Laboratories (CRNL) have undertaken a Waste Disposal Project to co-ordinate the transition from the current practice of interim storage to permanent disposal for low-level radioactive wastes (LLW). The strategy of the project is to classify and segregate waste segments according to their hazardous radioactive lifetimes and to emplace them in disposal facilities engineered to isolate and contain them. To support this strategy, a waste characterization program was set up to estimate the volume and radioisotope inventories of the wastes managed by CRNL. A key element of the program is the demonstration of a non-invasive measurement technique for the isotope-specific characterization of solid LLW.

This paper describes the approach taken at CRNL for the non-invasive assay of LLW and the field performance and early operational experience with a waste characterization monitor to be used in a waste processing facility.

INTRODUCTION

For the past 41 years at the Chalk River Nuclear Laboratories (CRNL), low-level waste (LLW) has been placed into interim storage facilities awaiting disposal at a later date. The volume of LLW delivered to interim storage facilities over this period is in excess of 90,000 m³ (1). As annual waste volumes have increased, it has become evident that this form of LLW management is not cost-effective and land-use efficient. By the mid-seventies, LLW handling experience had matured and annual waste volumes (~3500 m³/year) had become sufficiently large to provide the necessary incentives for alternative LLW management practices.

At that time, a two-step program was identified for improving LLW management at CRNL. First, where practical, the volume of future LLW would be reduced prior to storage. This step was realized with the commissioning of incineration and compaction techniques resulted in waste volume reduction factors of 187 (31% of site LLW) and 7.8 (20% of site LLW) respectively in 1984. Thus, use of volume reduction techniques has significantly reduced the need for new interim storage facilities for LLW (2,3).

Second, as a long-term solution, techniques and facilities for the safe and economic disposal of LLW would be developed. As part of this step, a Waste Disposal Project was established in 1982 to pioneer the transition of CRNL managed waste from interim storage to permanent disposal. Since the bulk of LLW presents only a short-lived hazard, the intent is to optimize disposal procedures by segregating wastes according to their potentially hazardous lifetimes. Individual LLW waste segments would then be matched to disposal facilities appropriately designed to isolate and contain the wastes until they no longer pose a radiological hazard to man and the environment (1,4). From the outset, a key element of the project has been the demonstration of measurement techniques for the radiological characterization of waste items.

Throughout North America, it is common practice for LLW to undergo volume reduction and standardized packaging near the point of generation to minimize overall storage and shipment costs. Consequently, the radiological characterization of these wastes is most commonly performed with the compacted waste form. At CRNL, an alternative approach has been taken where wastes are characterized prior to volume reduction and standardized packaging. Such an approach offers the potential to

segregate wastes according to hazard duration and thus minimize overall disposal costs by ensuring that the containment properties of each disposal facility are matched to the hazard duration of the waste placed in it. In addition, characterization procedures are simplified since self-attenuation and density variation effects are minimal for low density wastes. However, a potential disadvantage of this approach is the need to assay a greater volume of waste.

The first LLW characterization study at CRNL was carried out in 1982 on waste feed to the WTC incinerator (5). Typically, this LLW consists of disposable paper, plastics, wood and cleaning materials. Waste bags, on average 0.04 m³ volume and 4 kg weight, were monitored for total gamma-ray activity and gamma-ray isotopic composition prior to incineration. The results of these assays were then compared with the results of radiochemical assay of incinerator ash for verification purposes. The principal radionuclides, their half-lives and relative abundance in CRNL LLW are listed in Table I. On average, about eighty percent of the activity is associated with only five radionuclides: Nb-95, Zr-95, Co-60, Cr-51 and Cs-137.

TABLE I
Gamma-ray Emitting Radionuclides
Identified in CRNL LLW

RADIONUCLIDE	HALFLIFE (DAYS)	PERCENTAGE OF TOTAL ACTIVITY (%)
Nb- 95	35	30.9
Zr- 95	65	16.4
Co- 60	1921	15.0
Cr- 51	28	10.7
Cs-137	11042	7.1
Ce-144	284	5.0
Zn- 65	244	3.5
Ce-141	33	2.6
La-140	1.7	1.9
Ag-110m	253	1.7
I-131	8.1	1.1
Ru-103	40	0.9
Co- 58	71	0.7
Ba-140	13	0.6
Cs-134	749	0.5
Ru-106	367	0.5
Fe- 59	45	0.5
Sb-125	986	0.1
Mn- 54	303	0.1

Three main conclusions were drawn from this study. First, a large fraction of the total gamma-ray activity (>90%) is concentrated in a small number of waste items (<10%).

Second, an acceptable isotopic signature for shipments of low density waste items can be obtained from the monitoring of only the most active waste items using gamma-ray spectroscopy.

Third, non-gamma-ray emitting radionuclides can be estimated with acceptable confidence limits from the gamma-ray isotopic signature of current waste feed, that is, wastes associated with fission cycles.

Based on these experimental findings, CRNL has adopted a LLW radiological characterization concept based on passive gamma-ray spectroscopy and the use of radiochemically established correlation factors to estimate the concentrations of non-gamma-ray emitting radionuclides.

The monitoring concept implemented combines both gross and spectral gamma-ray measurements. An initial gross gamma-ray measurement is used to determine the activity level of each item and only the most active items are selected for spectral gamma-ray characterization. At the completion of a monitoring session, the gamma-ray spectra are analyzed and the radionuclide content of the gamma-ray emitting isotopes are quantified. Next, an estimate of the radionuclide inventory for all items in a given batch is estimated by scaling up the radionuclide content determined for the most active items by a correction factor determined from gross gamma-ray activity measurements of all batch items. Finally, estimates of the activity levels of non-gamma-ray emitting radionuclides are made using radiochemically determined correlation factors between non-gamma-ray and gamma-ray emitting radionuclides for the type of LLW monitored.

MONITORING REQUIREMENTS

To satisfy the passive gamma-ray spectroscopy requirement, a dedicated monitor was developed for the routine assay of LLW feed to the CRNL WTC. At the onset of the monitor development program, the following criteria were established to guide the developers. The monitor should:

- be capable of a measurement resolution of <370 Bq/g (10 nCi/g) for the major isotopes of interest in conformance with the United States (US) Nuclear Regulatory Commission (NRC) directive 10CFR61,
- be flexible so that wastes of various physical forms, shapes and sizes can be easily monitored,
- permit short assay times per waste item in order not to impede normal waste handling and processing procedures,
- be capable of being operated by non-specialist personnel,
- provide convenient data accumulation and storage facilities compatible with the current conversion of site waste management records to a computer database,

- be designed as a transportable instrument package to allow convenient use at a number of assay sites, and
- be assembled from commercially available equipment wherever possible to minimize development effort and to simplify future maintenance requirements.

MONITOR DESCRIPTION

The CRNL waste characterization monitor consists of a partially shielded enclosure that contains the gamma-ray detection equipment, weigh scale platform and liquid nitrogen storage dewar, and a single instrumentation cabinet containing nuclear counting and weigh scale electronics, a computer and an interactive terminal. A block diagram of the equipment configuration is shown in Fig. 1 and a photograph of the monitor during preliminary testing at the WTC is shown in Fig. 2.

The partially shielded enclosure consists of two 2 cm thick lead walls mounted on moveable steel frames. The walls are normally positioned 90 cm apart for monitoring small-sized waste items such as waste bags but can easily be reconfigured with a greater separation to accommodate larger-sized waste items. A wall separation of approximately 1.5 m as shown in Figure 2 accommodates the monitoring of a compacted bale of waste. Such a shielding enclosure was judged necessary to suppress the high background gamma-ray fields of $\sim 1 \mu\text{Gy/h}$ ($100 \mu\text{rad/h}$) encountered at the WTC monitoring location. The background gamma-ray field is primarily due to the storage of active waste items awaiting processing nearby the monitor location.

For the measurement of the gross gamma-ray activity of a waste item, two collimated and back-shielded NaI detectors are mounted, one in each enclosure wall. The spectral gamma-ray measurement is made with a hyper-pure germanium semiconductor detector. The detector is normally placed at one end of the enclosure on its own stand.

An industrial weigh scale is placed between the shielding walls to measure the weight of each item to be assayed. This scale has a range of measurement from 0 to 100 kg with a resolution of 0.01 kg.

A Digital Equipment Corporation MICRO-11 computer controls the system. It is interfaced to the gross gamma-ray counting equipment via the IEEE-488 interface standard and to the weigh scale and spectral gamma-ray measurement electronics via RS-232 serial interfaces. Two additional serial interfaces are used for connection of the operator console terminal and a data link via standard telephone lines to the site computing facility.

The computer is used to perform three main functions. First, it controls the acquisition and temporary local storage of data during a monitoring session in accordance with operator commands. Second, data analysis programs for gamma-ray spectral peak identification, background subtraction, nuclide identification and quantification are executed with the

spectral data from individual waste items. These data reduction functions are performed in an off-line mode once the daily monitoring session is complete. Third, the computer is used to establish a communication channel to the site central computer during the evening and to control the uploading and archival of daily monitoring data in the CRNL waste management database.

The monitor operates under direct control of process operators who must manually insert and remove waste items from the counting position, enter waste identification information and designate the waste processing option to be selected for each item. The operator interface for the monitor consists of a CRT display for status information and operator prompting messages, and a keyboard for command and data input. This terminal is placed on top of the instrumentation cabinet at waist level to facilitate convenient equipment control from a standing position. The instrumentation cabinet is normally positioned adjacent to one of the shielding walls for ready operator access to the monitoring area for insertion and removal of waste items.

The sequence of operations performed in monitoring a single waste item is as follows. First, the operator is prompted to load a waste item onto the weigh scale in the monitor position. Following acknowledgement from the operator that this action has been completed, the computer initiates both a gross and spectral gamma-ray assay of the item and a measurement of the item weight. After a five-second counting period, the count recorded with the gross gamma-ray assay system is compared to a predetermined counting limit identifying those items for which a spectral assay is required. If the gross gamma-ray assay count is greater than the predetermined limit, the spectral assay for the item is permitted to continue to completion, otherwise the spectral assay for the item is terminated and the partially collected spectral data is erased. During the assay count period, the operator is prompted to enter additional information such as the waste item type and source and reference labelling to identify the assay data collected. Next, the operator must select a waste category in which to route the monitored item. Current categories in use for bagged waste are incinerable, compactible, rejected on physical or activity properties and too active to monitor. Once a category is selected, the assay and reference data for the monitored item are stored in a temporary data file according to waste category. The operator is then offered the options of monitoring more waste items or terminating the monitoring session.

During a monitoring session, measurement of the gross gamma-ray background is updated automatically at a regular time interval via computer control or manually by the operator via keyboard command. Measurement of the spectral gamma-ray background is updated following the spectral gamma-ray assay for each waste item.

Once the monitoring session is complete, data reduction of the spectral data and formatting of data for archival is performed by the computer off-line, independent of the operator. Finally, transmission of the daily

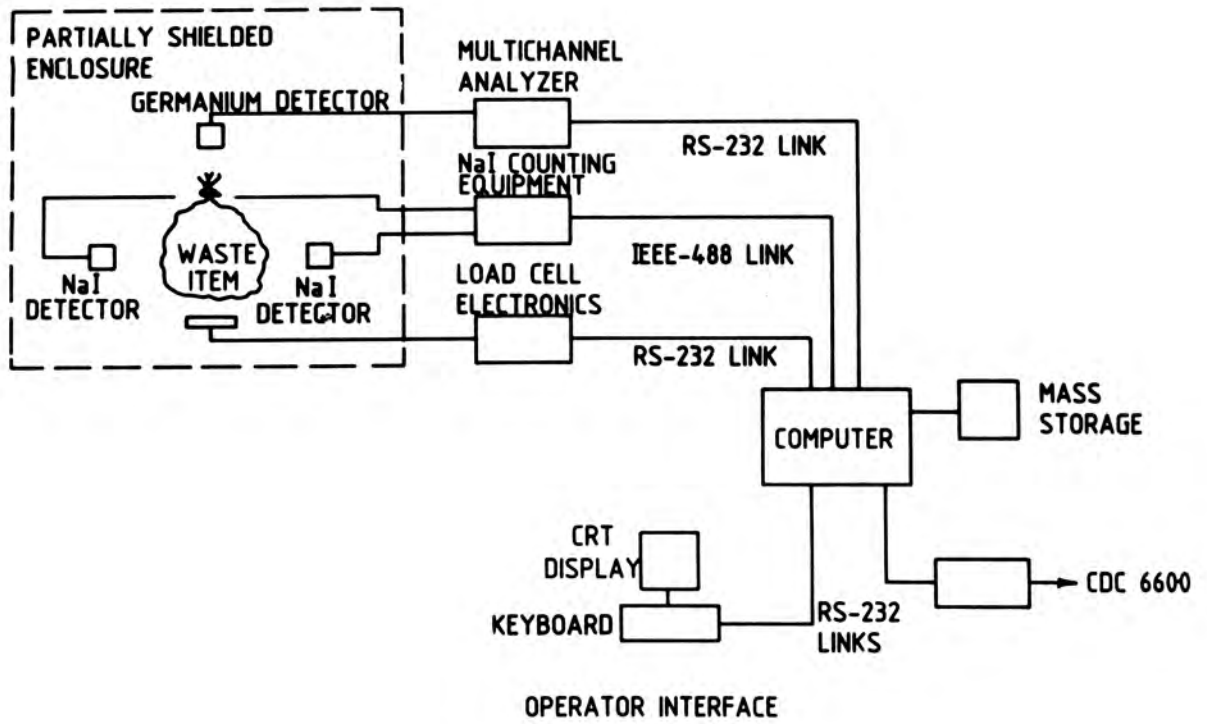


Fig. 1. Configuration of Waste Characterization Monitor.

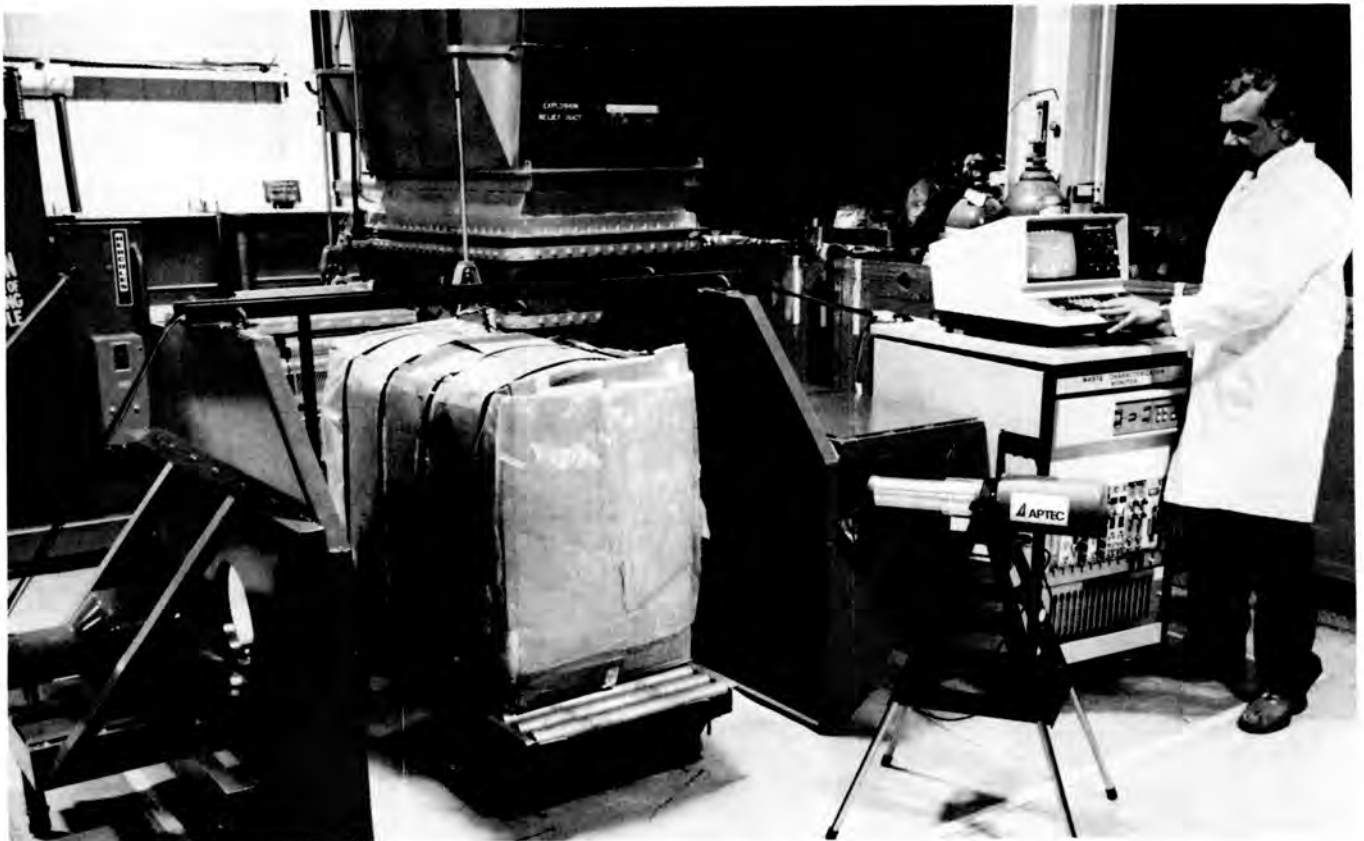


Fig. 2. Waste Characterization Monitor.

acquired assay data to the CRNL central computer for archival into the waste management database can be performed interactively by operators on a file (waste category) by file basis or automatically under unattended computer control.

PERFORMANCE

Prior to operational commissioning, the performance of the monitor was evaluated through a series of field trials in 1986 August and September. These performance trials were conducted at the CRNL WTC receiving area where LLW generated at the CRNL site is sorted and temporarily stored prior to volume reduction. This will be the operational location of the monitor when it is commissioned for routine use in early 1987. For these trials, the monitor was configured to assay LLW packaged in polyethylene bags.

The overall performance of the monitor and the range of waste item activities that the monitor can accommodate is a function of the source-to-detector separation, background gamma-ray activity level, assay count durations and counting electronics capabilities. For these trials, a source-to-detector separation of 45 cm and assay count durations of 5 seconds and 30 seconds for the gross and spectral gamma-ray measurements respectively were used.

A complicating factor in assessing the monitor's performance at the WTC is the fact that the radiological background is large and variable. This background is created by the storage and movement of LLW within the receiving area. Normally, background activity levels of 0.1 to 2 $\mu\text{Gy/h}$ are experienced.

The first test assessed the monitor accuracy as a function of waste item activity for both means of gamma-ray measurement.

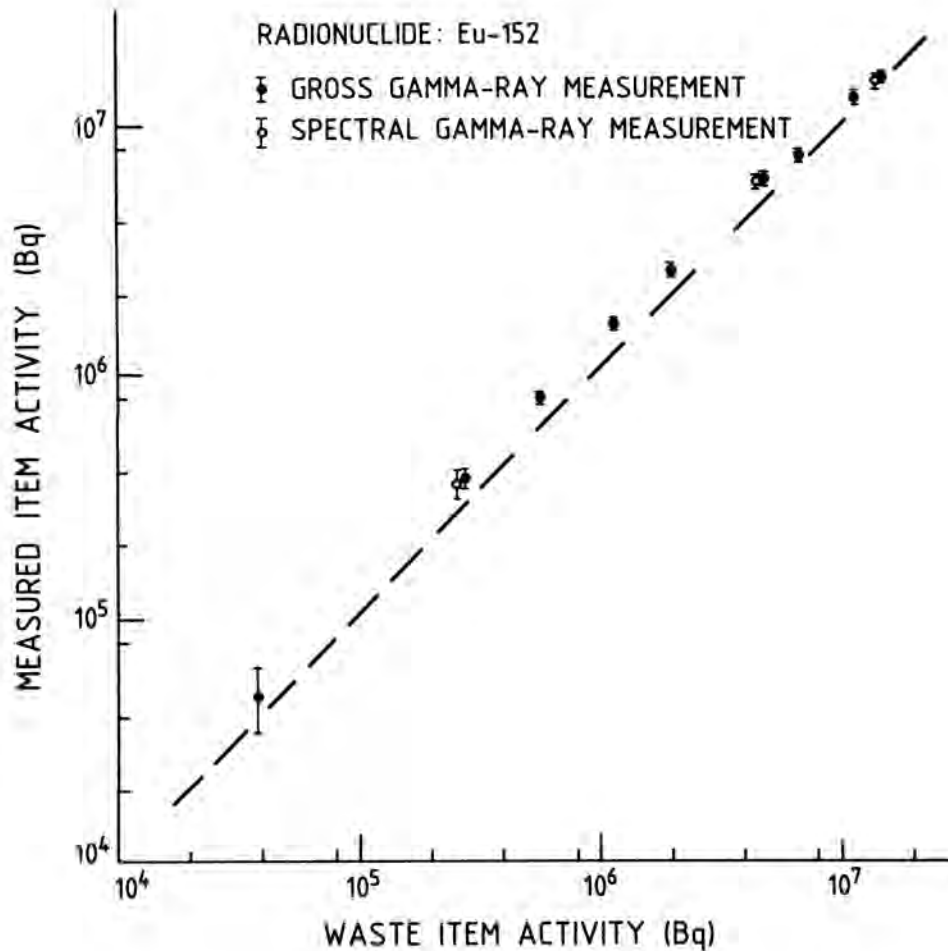


Fig. 3. Measured Item Activity Versus True Item Activity.

In this test, a series of Eu-152 radioactive sources of varying source activity were monitored. The results of this evaluation are illustrated in Fig. 3 where measured item activity is plotted as a function of true waste item activity. Error bars indicate one standard deviation in the series of measurements for a given waste item activity. In general the relationship between measured and true item activity is linear within an error margin of $\pm 25\%$.

The lower range limit or resolution for the monitor can be best expressed by an estimate of the minimum detectable activity for a given radionuclide. This quantity is defined as the minimum activity that can be detected with a probability of 95% greater than the pseudo-activity due to random natural variation of the background. For the assay and background conditions stated previously, the monitor is capable of minimum detectable activities of 8 to 80 kBq (0.2 to 2 μ Ci) per waste item for the five most common radionuclides found in CRNL LLW. For a typical 4 kg waste bag, this implies the capability to measure radionuclide activity concentrations of less than 2 to 20 Bq/g (50 to 500 pCi/g) which is more than an order of magnitude lower than the US NRC 10CFR61 standard of 370 Bq/g (10 nCi/g).

The upper range limit for the waste bag monitoring configuration is approximately 50 kBq/g (1.4 μ Ci/g). Waste items with greater specific activity contents can easily be accommodated for assay by increasing the source-to-detector separation through the movement of the shielding assembly walls.

Next, the repeatability of monitoring the same waste item multiple times was evaluated in the following way. A polyethylene bag was filled with clean lab wear and from one to five sources (Co-60, Ba-133, Eu-152, Cs-137, Mn-54). In each case, the waste bag was monitored a number of times and the mean and standard deviations for each source configuration are listed in Table II. In both the gross and spectral gamma-ray assays, the measured mean value agrees within plus/minus one standard deviation of the true item activity.

The throughput capability of the monitor is determined by the assay duration, requirements for operator parameter keyboard input, and the

number of waste items requiring full spectral characterization in a given monitoring batch. For gross and spectral gamma-ray assay and parameter entry durations of 10 and 86 seconds per waste item respectively and a waste distribution where only 10% of the waste items require full spectral characterization, a monitor throughput of approximately 100 waste items per hour is achievable.

Operator experience with the monitor is limited to date since the monitor has not been commissioned and has only spent a limited time being evaluated in the WTC environment. The primary feedback from operational personnel on the monitor has been obtained during training sessions where several WTC operational staff were trained to use the monitor and perform trial batch assays on simulated active waste bags. To date, operator acceptance of the monitor is high. An especially attractive feature is the replacement of manual inventory cataloguing by a computer-generated database of daily monitored items.

The versatility of the monitor to assay other waste forms was demonstrated in 1986 January when the monitor was used to assay five drums of WTC incinerator ash. In this trial, each drum (210 ℓ) contained approximately 80 kg of three-year-old incinerator ash with average gamma-ray specific activity of approximately 2 to 6 kBq/g (50 to 150 nCi/g). As well as non-invasive monitoring measurements, samples of the incinerator ash were analysed radiochemically. For the seven long-lived radionuclides identified via non-invasive monitoring, the agreement of radionuclide activity estimates between non-invasive monitoring and radiochemical assay varied between a factor of one to four. In general, the level of agreement was better for the most active radionuclides.

PROJECT STATUS

The CRNL waste characterization monitor is at an advanced stage of development. Current work is focussed on completing monitoring software modifications that were identified as desirable as a result of the WTC field trial 1986 September. Completion of the instrument commissioning for daily use at the CRNL WTC is currently scheduled for 1987 April.

TABLE II

Monitor Assay Repeatability

GAMM-RAY MEASUREMENT	NUMBER OF ASSAYS	SOURCE	ACTIVITY (10^5 Bq)		STANDARD DEVIATION (%)
			TRUE	MEASURED MEAN	
GROSS	20	MIXED	11	11	5.1
SPECTRAL	7	Ba-133	2.7	2.3	18
	8	Eu-152	2.8	2.9	25
	8	Cs-137	1.8	1.7	12
	7	Mn-54	0.46	0.52	30
	8	Co-60	2.1	2.1	13

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