ADVANTAGES AND DISADVANTAGES OF GAMMA-RAY SPECTRAL LOGGING FOR SITE CHARACTERIZATION OF LOW-LEVEL RADIOACTIVE WASTE

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ABSTRACT

The Borehole Geophysics Section of the Geological Survey of Canada (GSC), in collaboration with the Low-Level Radioactive Waste Management Office (LLRWMO) of Atomic Energy of Canada Limited (AECL) recently investigated the application of borehole gamma-ray spectral logging to the characterization of sites contaminated with low-level radioactive waste (LLRW). Studies were conducted at two sites in Canada: the municipal landfill in the Town of Port Hope, Ontario which contains primarily radium contamination; and a temporary storage facility in Surrey, British Columbia which houses a sand/gravel/slag mixture with elevated thorium concentrations. In both cases, the gamma-ray spectral logs were successfully used to identify, delineate and quantify the radioactivity; however, a problem related to the movement of radon gas (from the uranium/radium decay series) into boreholes was encountered at the Port Hope site.

In this paper, examples from both sites which demonstrate the advantages of gamma-ray spectral logging for characterizing LLRW are presented. The "radon problem" encountered at Port Hope is discussed, including its effect on the logging results and a procedure that was developed to reduce this effect.

INTRODUCTION

The Borehole Geophysics Section of the Geological Survey of Canada (GSC) conducts research into the application of borehole geophysics to minerals, geotechnical and environmental problems. Recently, the GSC collaborated with the Low-Level Radioactive Waste Management Office (LLRWMO) of Atomic Energy of Canada Limited (AECL) in investigations of the use of gamma-ray spectral logging for characterizing low-level radioactive waste (LLRW) at two sites: one in Port Hope, Ontario; and another in Surrey, British Columbia.

The LLRWMO was established by the federal government in 1982 to resolve historic waste problems that are a federal responsibility (those for which the original producer can no longer reasonably be held responsible), to ensure that a user-pay service is established for the disposal of LLRW produced on an on-going basis, and to address public information needs concerning LLRW. The LLRWMO is operated by AECL through an agreement with Natural Resources Canada (formerly, the Department of Energy, Mines and Resources (EMR)), the federal department which provides the funding and establishes national policy. The LLRWMO attends to problems encountered across Canada.

In the characterization of a site contaminated with LLRW, it is desirable to identify the various radionuclides present, to measure their concentrations, and to determine their distribution both laterally and with depth. With some limitations, this information can be acquired rapidly and cost-effectively from gamma-ray spectral logs. Gamma-ray spectral logging provides a continuous record of the distribution with depth, of radioactivity in the soil surrounding the borehole. Compared to soil sampling, gamma-ray probes sample a larger (more representative) volume (up to a 30 cm radius in most conditions) around the borehole, which is in-situ (undisturbed), and, in addition, preliminary results can be obtained immediately in the field. Gamma-ray logs also allow more flexibility than soil samples regarding varying criteria and depth filters. The ability to identify the sources of radioactivity (via their characteristic energy spectra) and to measure them quantitatively (via calibration) gives spectrometry an added advantage over gross-count logging.

A limitation of gamma-ray spectral logging is that some radionuclides must be detected indirectly through gamma-radiation produced by one or more of their daughter products. In these cases, the method relies on the assumption of radioactive equilibrium between the parent and daughter. However, this equilibrium may be disrupted by, for example, the removal of one of the intermediate radionuclides in the series, such as by chemical processing or through the loss of a gaseous member of the decay series.

PORT HOPE

From 1933 until the 1950's, a radium refinery operated in the town of Port Hope, Ontario. LLRW from the refinery, consisting of radium (^{226}Ra) along with uranium series radionuclides in various states of equilibrium, was dumped at the present site of the municipal landfill beginning in the 1940's. Since the 1950's, the site has received municipal and commercial refuse from the town.

The current situation at the landfill site is depicted as a cross-section in fig. 1. The fill unit, which ranges in thickness up to 12 m overlies native sand and silt (1). It consists of three main components: refuse, sand fill and LLRW (2). The LLRW is composed primarily of ash, cinders and contaminated soil which is generally encountered in a lower sand fill layer.

In the fall and winter of 1991/92, the LLRWMO contracted a drilling and gamma-ray spectral logging survey to

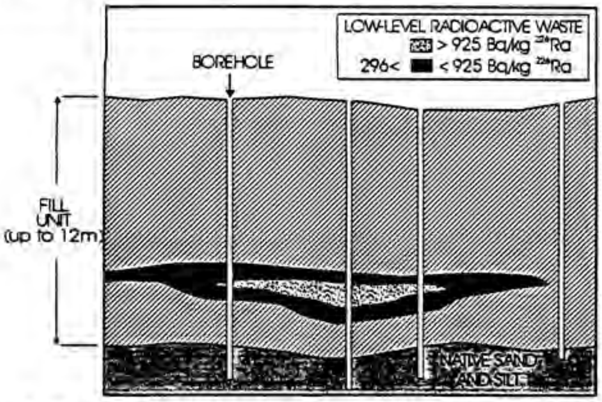

Fig. 1. Typical vertical section at the Port Hope landfill site, showing the fill unit which consists of refuse and sand fill layers, and pockets or layers containing LLRW. The LLRW is shown for two clean-up criteria.

delineate the LLRW at the landfill site. The focus of the survey was radioactive contamination using radium and uranium as prime indicators.

^{226}Ra Distribution From Gamma-Ray Spectral Logs

^{226}Ra belongs to the ^{238}U decay series shown in Fig. 2. When this series is in radioactive equilibrium (i.e., the rate of decay of each element is equal to its rate of production by decay of its parent), the measured (reference) gamma-ray energy spectrum has a characteristic shape as illustrated in Fig. 3. As this figure shows, the gamma rays emitted by ^{214}Bi dominate the spectrum. However, when the series is in equilibrium, the relative intensities of the gamma rays emitted by the radionuclides in the series are fixed, as are the relative abundances of the radionuclides themselves. Therefore, the ^{226}Ra concentration can be estimated reliably from the ^{214}Bi gamma-ray count rate. For this reason, the ^{226}Ra logs are called equivalent ^{226}Ra logs and represent true ^{226}Ra distributions only if equilibrium exists between ^{226}Ra and ^{214}Bi.

Due to the chemical separation at the radium refinery, equilibrium throughout the entire ^{238}U series would not be expected in much of the waste at the landfill site. However, it is expected that radionuclides in the decay series from ^{226}Ra through to ^{214}Po, which produce most of the ^{238}U series gamma rays, generally exist in equilibrium proportions because of their short half-lives. Thus, spectra from radium-contaminated zones will resemble the ^{238}U series reference spectrum shown in Fig. 3 and the above method of determining the ^{226}Ra concentration is applicable, except for the complication of radon losses explained below.

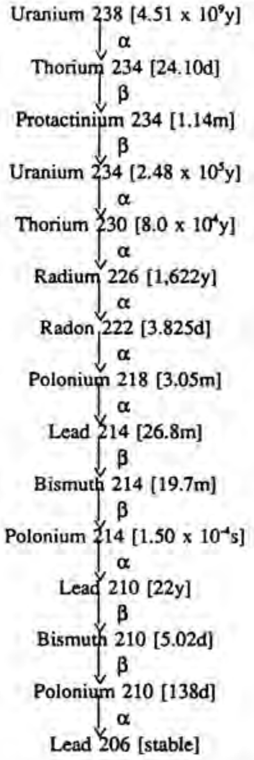

Fig. 2, ^{238}U decay series.

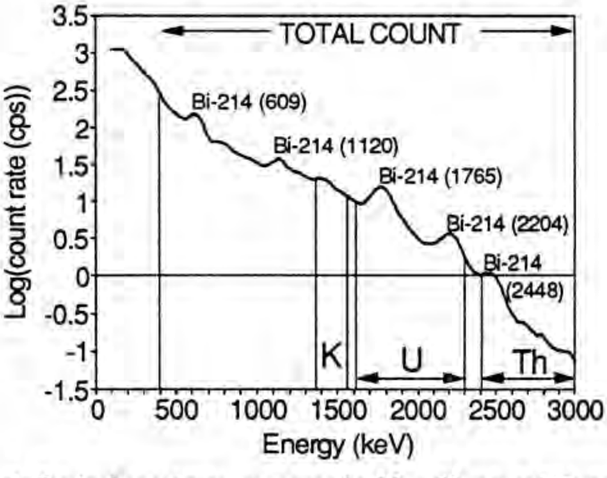

Fig. 3. Typical spectrum, measured with a 32 mm x 127 mm cesium iodide scintillation detector, from uranium in equilibrium with its daughters. Typical potassium (K), uranium (U) and thorium (Th) windows are shown.

Figure 4 illustrates the principle of gamma-ray spectral logging, in which the gamma rays from radionuclides within the rock or soil surrounding a borehole are counted and their energies are measured. Gamma-ray energy spectra, containing characteristic peaks which can be used to identify the source of radioactivity, are thus acquired continuously or step-wise (continuous was chosen for Port Hope) as the detector moves along the borehole. By defining energy windows encompassing specific peaks in the spectra and computing the count rates in these windows, the vertical distribution of radioactivity (or gamma-ray logs) due to selected radionuclides can be plotted. Four commonly chosen energy windows include a total count (TC) window covering the entire spectrum, and three windows centered on gamma-ray peaks related to the three most abundant naturally occurring radionuclides; potassium, uranium and thorium (see Fig. 3). As shown in Fig. 4, either the total count or uranium window can be used to produce equivalent ^{226}Ra logs, if the probe has been calibrated and the only source of radioactivity is ^{226}Ra. In such cases, the total count provides better counting statistics. If other sources are present, the total count will not be proportional to the ^{226}Ra content.

Data Acquisition and Analysis

A portable gamma-ray spectral logging system manufactured by IFG Corporation of Brampton, Ontario was used to characterize the radioactive waste at the Port Hope landfill. The system, which uses a 25 mm x 76 mm cesium iodide detector measures natural gamma radiation in the energy

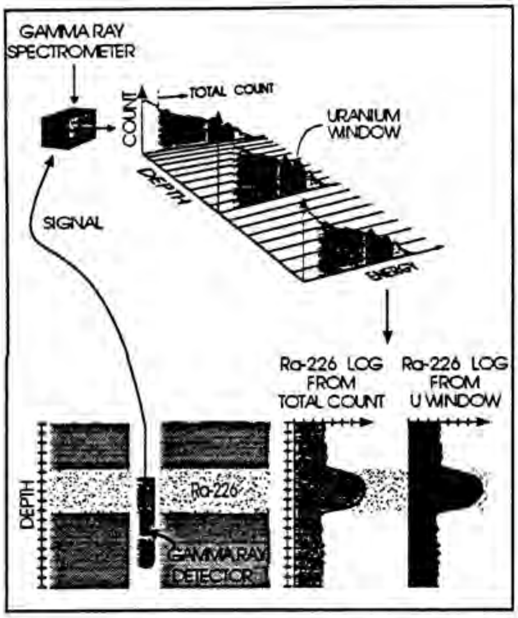

Fig. 4. Diagram of the concept of spectral gamma-ray logging. In this example, logs of the equivalent ^{226}Ra content are computed from either the total count or uranium window logs.

range from 100 to 3000 keV in 256 channels. Measurements were also made at Port Hope with the truck-mounted GSC R&D logging system. The GSC spectral gamma-ray probe used at Port Hope contains a 32 mm x 127 mm cesium iodide detector and also measures gamma-rays with energies up to 3000 keV in 256-channel spectra. With a logging speed of 0.5 or 1 m/minute and a sample interval (counting time) of 1 second, the resulting depth interval between readings was 8 mm at 0.5 m/minute and 16 mm at 1 m/minute.

At the landfill site, 203 mm diameter boreholes were drilled with hollow-stem augers. The holes were dry and were generally fitted with 76 mm diameter open-bottom PVC pipes. The space between the borehole wall and the PVC pipe (a 64 mm annulus) was left air-filled and covered at the top of the hole with plywood.

In order to convert the gamma-ray logs to equivalent radium logs, the GSC probe was calibrated by logging in model boreholes at the GSC's Bells Corners calibration facility near Ottawa, Ontario (3). The equivalent radium logs can then be used to estimate the volume of radium-contaminated fill using any clean-up criteria above background.

Advantages of Spectrometry

The main advantage of spectral logging over total count logging is source identification, an example of which is the anomaly at a depth of 4.4 m in hole GLM05 (the "protactinium anomaly"). Logs, acquired by the GSC, of the count rate in the total count and uranium windows for this hole are shown in fig. 5. The total count log contains two anomalies with similar amplitudes at 4.4 m and 8.0 m. From the total count log alone, both anomalies would be attributed to radium contamination. However, in the uranium log the absence of an anomaly at 4.4 m indicates that this anomaly is due primarily to lower energy gamma rays. This is clearly evident in the spectrum from this anomaly, shown in Fig. 6 with the ^{238}U series reference spectrum from Fig. 3. Therefore, this anomaly is not a typical radium anomaly.

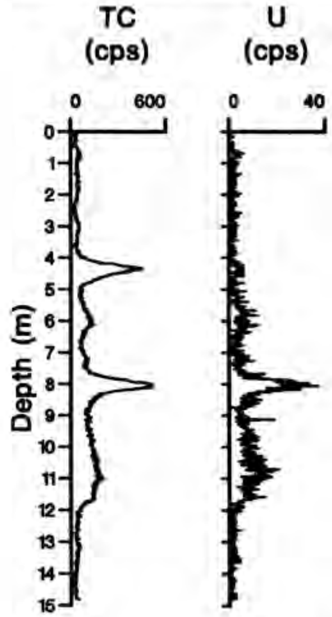

Fig. 5. Total count and uranium window logs from hole GLM05 at the Port Hope landfill site.

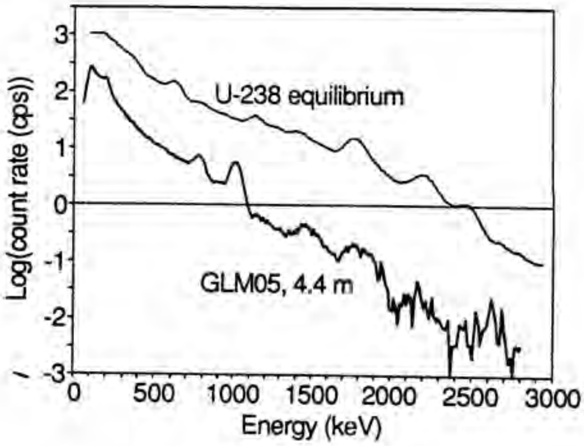

Fig. 6. Spectrum from the anomaly at 4.4 m in hole GLM05 (Port Hope) shown with the ^{238}U series reference spectrum from Fig. 3.

Investigation into the history of the Port Hope refinery revealed that several experimental methods of uranium extraction were tested, some of which produced waste containing up to 20% uranium and high levels of protactinium (Duncan Moffat, personal communication). This waste would today be expected to contain elevated levels of the first four radionuclides in the ^{238}U decay series (Fig. 2), but insignificant amounts of the remainder of the ^{238}U series decay products due to the 80000 year half-life of ^{230}Th, the next nuclide in the series. The resulting spectrum would contain two prominent peaks at 766 keV and 1001 keV from protactinium (^{234}Pa) and the count rate would decrease at higher energies. These features are observed in the spectrum from the anomaly at 4.4 m in hole GLM05. The source of the anomaly was thus determined to be ^{234}Pa which is supported by anomalous concentrations of ^{238}U. A more in-depth discussion of this anomaly is presented in Killeen and Pflug (4).

Radon Migration Problem

During the first phase of drilling and logging at the landfill site it was noted that, in some of the holes in the eastern section of the property where the largest inventory of ^{226}Ra is known to exist, the spectral gamma-ray logs were not reproducible from one day to the next.

This variability was attributed to significant and varying levels of radon (^{222}Rn) in the boreholes. ^{222}Rn, a gas, is a short-lived (3.825 day half-life) nuclide in the ^{238}U decay series between ^{226}Ra and ^{214}Bi (Fig. 2). If radon from a remote source with a higher radium concentration than the area being measured migrates into and spreads along the boreholes at the landfill site, an excess of ^{214}Bi will develop and plate onto surfaces within the boreholes. This excess ^{214}Bi in the boreholes causes an increase in the measured count rate which results in calculated equivalent radium concentrations that are higher than the true radium content at the measurement location. The thicknesses of the contaminated zones will consequently be overestimated.

To confirm the presence of radon in boreholes in the eastern section of the landfill site, a series of experiments was conducted by the GSC with the assistance of the LLRWMO and Gartner Lee Limited (the contractor responsible for the delineation of radioactive wastes at Port Hope for the 1991/92 investigation) during March 1992. These experiments are discussed by Killeen et al (5).

Air Flushing Technique

After reviewing the results of the radon experiments, the GSC and the LLRWMO developed a new protocol for drilling and logging at the landfill site to reduce the effects of radon on the gamma-ray logs. In addition to data from the radon experiments, the results of Hilpert and Bunker (6) suggested flushing radon-contaminated holes with compressed air before and during logging to remove radon from the holes and to allow time for most of the radon daughters on surfaces in the boreholes (i.e., the borehole walls and PVC pipes installed in the holes) to decay (about 2 to 3 hours). Holes were fitted with 76 mm diameter, sealed-bottom PVC pipes to confine radon to the annulus between the PVC and the borehole wall. Air flushing of the space around the PVC pipe was accomplished by pumping compressed air to the bottom of the hole through a 25 mm diameter PVC tube inserted between the borehole wall and the 76 mm PVC pipe.

Example results of the application of air flushing of the annulus around a sealed-bottom PVC pipe were presented in Killeen et al (5). In this example, the thickness of the contaminated layer as computed from the gamma-ray log acquired after 3 hours of air flushing was close to that which would have been predicted from the soil samples. However, the radium values computed from the gamma-ray logs were higher than the soil sample values in the anomalous zone. This discrepancy could be due to the different volumes sampled by the two methods or to the inability to completely flush the hole and the surrounding soil (5). It is the subject of further investigation. Generally, the air flushing technique was considered adequate in removing radon effects from the results.

SURREY

Niobium slag mixed with sand and gravel is located in a temporary concrete storage facility (bunker) at the site of a former smelter in Surrey, British Columbia. The waste consists of 30 parts soil to 1 part slag. The slag, which contains approximately 1% thorium and 0.026% uranium, originated from niobium smelting on site and was later used for site improvement and grading. Since 1984 the material has been contained in a storage bunker at the site. The bunker sits on an asphalt base, which overlies an additional layer of fill about 1 meter thick. The soil/slag mixture in the bunker is capped with a geomembrane and is covered by a layer of clean sand and gravel. A cross-section through the bunker is illustrated in Fig. 7.

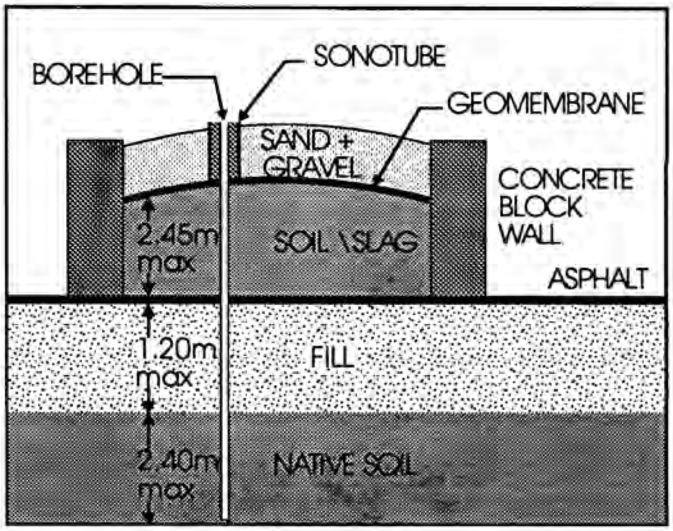

Fig. 7. Typical vertical cross-section through the temporary storage facility at the Surrey site, showing the three layers of interest (soil/slag, fill, native soil).

In the fall of 1992, a drilling and soil sampling program to evaluate the waste in the bunker was completed at the request of the Surrey Siting Task Force (SSTF), the group responsible for finding a final disposal site for the waste. One of the goals of the SSTF program was to determine if any of the three layers of material in and below the bunker (soil/slag, fill, and native soil) contained more than 500 ppm thorium and uranium, in which case it would be classified as radioactive waste. As part of the SSTF program, the LLRWMO conducted a gamma-ray spectral logging survey to determine the spatial distribution of uranium and thorium in the bunker and to provide a second estimate (in addition to the soil samples) of the average concentrations of uranium and thorium in the material in and below the bunker. The GSC was consulted regarding the spectral gamma-ray logging protocol and data analysis, and reviewed the data collected.

Detection of ^{232}Th

The ^{232}Th decay series is shown in fig. 8. As for radium, thorium is measured indirectly by counting gamma-rays from one of its daughter products because ^{232}Th does not emit any useable gamma rays itself. An energy window straddling the 2614 keV gamma ray from ^{208}Tl is generally chosen as the preferred thorium window. As shown in Fig. 4 for radium, logs of the thorium distribution with depth can be computed from the thorium-window count rate. This assumes the spectral gamma-ray probe has been properly calibrated and the ^{232}Th series is in equilibrium, which is generally expected due to the relatively short half-lives in the decay series. Once the radionuclides in the series are separated, equilibrium will be re-established in about 40 years.

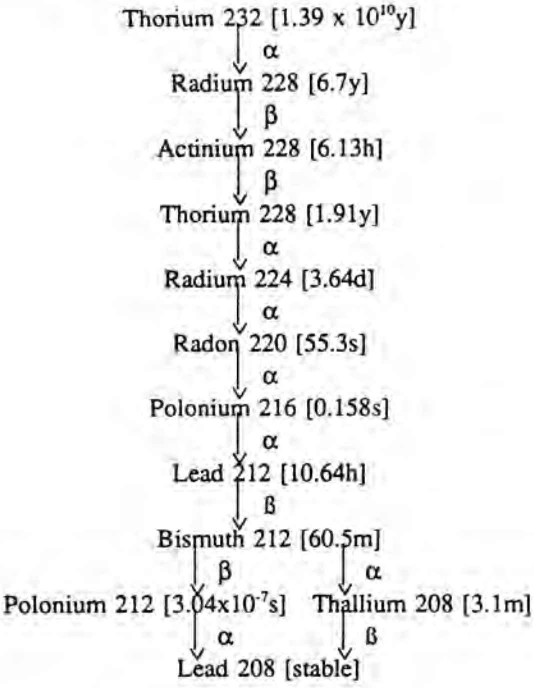

Fig. 8. ^{232}Th decay series.

Drilling and Logging

Sixteen boreholes were drilled into the material in and below the bunker for soil sampling and spectral gamma-ray logging. Using a statistical approach, a sampling plan was developed which called for dividing the bunker into eight cells of equal volume above the asphalt and drilling two randomly positioned boreholes in each cell. The boreholes were 100 mm in diameter, cased with 76 mm diameter sealed-bottom PVC pipes, and extended at least 0.5 m into the native soil. More details regarding the drilling and sampling program can be found in Gartner Lee Limited (7).

The spectral gamma-ray logs were acquired with an Exploranium GR-256 portable gamma-ray spectrometer and GP-200 probe with a 35 mm x 100 mm sodium iodide detector. The equipment, which records 256-channel gamma-ray spectra with an upper energy limit of 3000 keV, was controlled by an IBM compatible notebook computer connected to the spectrometer, using software written by the LLRWMO (8). This software is also capable of performing a preliminary analysis of the data during logging.

Stationary gamma-ray spectra were acquired in the boreholes at 0.5 m intervals in uncontaminated zones and at 0.1 m intervals where the holes were known to intersect radioactive contamination based on quick profiles of the radiation acquired previously with a gross count logging system. The counting time for each spectrum was controlled by the software to achieve pre-determined levels of uncertainty due to counting statistics.

The boreholes were checked for radon contamination (as experienced at the Port Hope landfill site) by relogging four of the holes on different days with the gross count equipment. As was predicted, no radon migration problem was observed in the Surrey investigation, due to the short half-life of ^{220}Rn.

Data

Potassium, uranium and thorium concentrations were computed from the spectral gamma-ray data using calibration parameters determined at the GSC's Bells Corners facility. Average potassium, uranium, and thorium concentrations for each of the three layers were then computed from the concentrations averaged over each borehole. The details of the data analysis are discussed in McCallum and Huffman (8).

A typical set of logs is shown in Fig. 9. Although there is some uranium present (less than 25 ppm), comparison of the thorium log to the gross count log confirms that most of the radioactivity is due to thorium. Potassium is present in normal background concentrations. In this case, where uranium is present in much lower concentrations than thorium, the total or gross count could be used to estimate the thorium concentration. However, gamma-ray spectrometry was needed to estimate the uranium concentration.

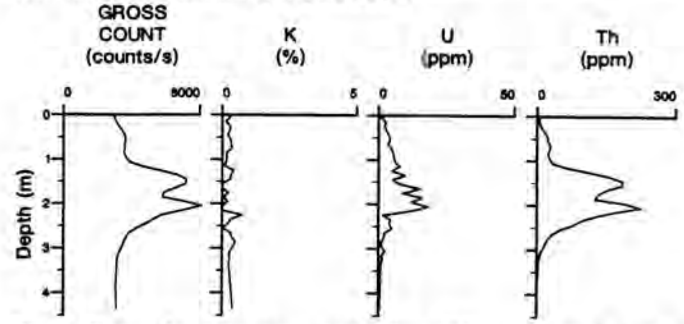

Fig. 9. Example gross count, potassium (K), uranium (U), and thorium (Th) logs from a hole at the Surrey storage site. The gross count is the sum of counts in all 256 channels.

In Fig. 10, the spectrum acquired in the main anomaly at 2 m in this hole is plotted along with a thorium-series reference spectrum (acquired in the thorium zone of a model borehole). Aside from the peak at 661.6 keV in the Surrey spectrum, which is due to a Cs-137 stabilization source in the GP-200 probe, the two spectra are similar, confirming that the source of the anomaly at 2 m is predominantly thorium contamination. (Note: the reference spectrum is smoother due to better counting statistics.)

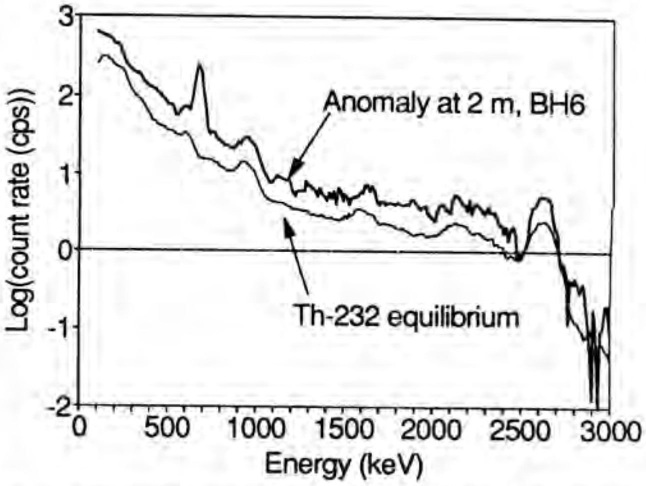

Fig. 10. Spectrum from anomaly at 2 m in the hole shown in Fig. 9, shown with a ^{232}Th series reference spectrum. The peak at 661.6 keV in the Surrey spectrum is due to a ^{137}Cs stabilization source in the probe.

Results

The results of the gamma-ray spectral logging survey agreed with the results of the separate soil sampling survey (7). Both showed that the average concentration of uranium plus thorium in each of the three layers in and below the bunker did not exceed 500 ppm. Therefore, none of the layers would be classified as radioactive waste.

CONCLUSION

Gamma-ray spectral logging has been used to measure radionuclide concentrations at two sites in Canada in waste contaminated with naturally occurring radionuclides. At the Port Hope landfill site, the radioactive contamination is primarily radium, whereas, at the Surrey site, the concern is thorium contamination. In both cases, gamma-ray spectral logging was found to be a good technique for delineating contamination. It also provides a continuous record with depth of the identity and concentration of radioactive contamination.

The main difficulty encountered in using spectral gamma-ray logging to delineate radium contamination at the Port Hope landfill site was with possible inaccuracies caused by the presence of high concentrations of radon which migrated to some boreholes from remote sources. A technique for reducing the effects of radon on the gamma-ray logs was developed. The technique involved air flushing of the space between the borehole wall and a sealed-bottom PVC pipe installed in the hole, both before and during logging in radon-contaminated holes. Other techniques, such as limited soil sampling, could be applied depending on the requirements of the project.

The importance of being able to identify the source of the contamination was demonstrated with the spectra from hole GLM05 at Port Hope, where the source of the anomaly at 4.4 m was found to be ^{234}Pa rather than ^{226}Ra as would have been concluded if only the total radioactivity had been measured. At the Surrey site, where both uranium and thorium contamination are found, spectrometry was necessary to separately determine the uranium and thorium concentrations.

In the Surrey investigation, both an extensive soil sampling, compositing and analysis process and spectral gamma-ray logging were used to provide independent measurements of the uranium and thorium concentrations in the material in the bunker. In both cases, it was concluded that the uranium plus thorium average concentration in each of the three layers considered did not exceed 500 ppm. However, the sampling program used was considerably more expensive for the same results. Radon migration was not encountered in the thorium waste.

ACKNOWLEDGEMENTS

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REFERENCES

1. T. RANNIE and HYDROTERRA LIMITED, "Town of Port Hope, Ontario hydrogeological report, Highland Drive landfill site," Report prepared for the Town of Port Hope, Ontario, Canada (1991).

2. GARTNER LEE LIMITED and MACLAREN ENGINEERS (1991) INC., "Port Hope Contamination Delineation/Removal Study, 1991/1992 Investigation," Report prepared for the Low-Level Radioactive Waste Management Office, Atomic Energy of Canada Limited (1992).

3. P.G. KILLEEN, "A system of deep test holes and calibration facilities for developing and testing new borehole geophysical techniques," in Borehole Geophysics for Mining and Geotechnical Applications (Ed. Killeen, P.G.), Geological Survey of Canada, Paper 85-27, pp. 29-46 (1986).

4. P.G. KILLEEN and K.A. PFLUG, "Detection of ^{234}Pa in low-level radioactive waste by gamma-ray spectral logging," Current Research, Part E, Geological Survey of Canada, Paper 93-1E, pp. 307-312 (1993).

5. P.G. KILLEEN, K.A. PFLUG, C.J. MWENIFUMBO, R.L. ZELMER and B.A. McCALLUM, "Application of spectral gamma-ray logging to low-level radioactive waste management studies at Port Hope, Ontario, Canada," Nuclear Geophysics, Vol. 7, No. 4, pp. 501-514 (1993).

6. L.S. HILPERT and C.M. BUNKER, "Effects of radon in drill holes on gamma-ray logs," Economic Geology, Vol. 52, No. 4, pp. 438-455 (1957).

7. GARTNER LEE LIMITED, "Testing of Waste Soil and Slag at 7800 Anvil Way, Surrey," Report prepared for the Surrey Siting Task Force, British Columbia, Canada (1993).

8. B.A. McCALLUM and D. HUFFMAN, "A Borehole Gamma Spectroscopy Investigation of the Uranium and Thorium Content of the Niobium Slag/Soil Mixture in the Bunker at 7800 Anvil Way, Surrey, British Columbia," Report prepared for the Surrey Siting Task Force, British Columbia, Canada (1993).