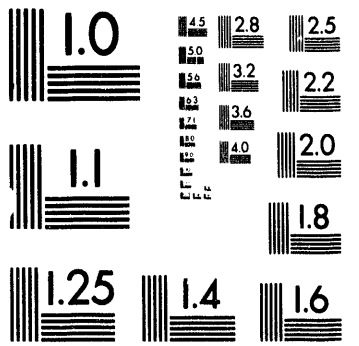
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Assessment of Unsaturated Zone Radionuclide Contamination in the 200 Areas of the Hanford Site, Washington

J. R. Brodeur
C. D. Wittreich

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Westinghouse Hanford Company
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ASSESSMENT OF UNSATURATED ZONE RADIONUCLIDE CONTAMINATION IN THE 200 AREAS OF THE HANFORD SITE, WASHINGTON

John R. Brodeur and Curtis D. Wittreich
Westinghouse Hanford Co.
P.O. Box 1970, Richland, WA 99352

ABSTRACT

The 200 East and 200 West Areas at the Department of Energy's Hanford site in southeastern Washington, contain chemical and nuclear fuel processing facilities that disposed of large volumes of chemical and radionuclide effluents to the ground via various structures such as ponds, cribs and ditches. A geophysical logging investigation was implemented in 1992 to assess the nature and extent of contamination beneath select liquid disposal sites in the 200 Areas.

The borehole geophysical logging was accomplished with a recently developed spectral gamma-ray logging system called the Radionuclide Logging System (RLS). This system has a high-resolution, intrinsic germanium detector mounted in a downhole probe and is calibrated and operated specifically for use in a borehole environment. It provides a means to develop in-situ, gamma-emitting radioelement concentration profiles. Approximately 50 boreholes were logged in this study.

The RLS log data provided information about the migration and deposition patterns of specific radionuclides in the unsaturated zone and their impacts on the groundwater. Approximately 10 radionuclide species were detected and quantified. Results of the field investigation are being used to refine site specific conceptual models, support Hanford Site remediation decisions and focus future characterization activities.

INTRODUCTION

A limited field investigation was conducted to assess the nature and extent of radionuclides released into the unsaturated zone sediments in the 200 Areas of the Hanford Site. The Hanford Site, operated by the U.S. Department of Energy (DOE) occupies about 1,450 km^2 of the southeastern part of Washington State. The 200 Areas including the 200 West and East Areas which are located near the center of the Hanford Site (Figure 1).

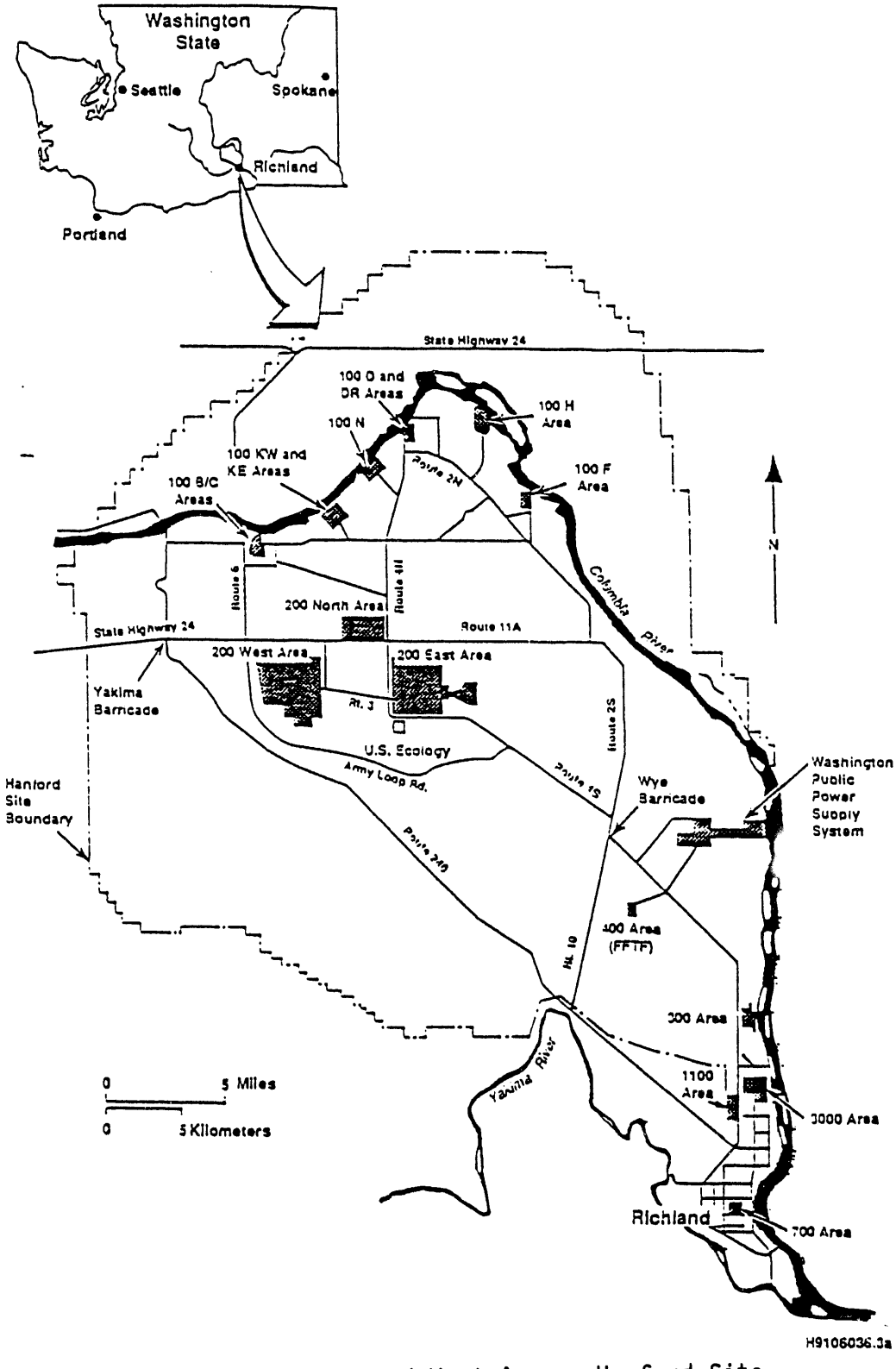

Figure 1. Location of the 200 East and West Areas, Hanford Site.

Operations in the 200 West and East Areas were primarily related to chemical and nuclear fuel processing. The 200 Areas contain close to 300 liquid effluent waste sites, a number of which received effluent containing radionuclides. Typically, low level liquid wastes were allowed to infiltrate into the ground through ponds, cribs and ditches. However, some high level liquid waste had been disposed of in certain cribs and trenches. The 200 Areas was estimated to have discharged 6.33×10^{11} L of liquid waste to the soil column between 1943 and 1980 (Zimmerman et al., 1986) A limited number of liquid effluent sites are still in use today, however they are operated under strict environmental controls and effluents contain only trace concentrations of radionuclides.

A number of relatively immobile radionuclides were retained within the sediment in the unsaturated zone, and as a result, the unsaturated zone is considered to be a significant contaminant source. The purpose of this investigation was to assess the nature and extent of gamma emitting radionuclides in the unsaturated zone in the 200 Areas using an intrinsic germanium, spectral gamma-ray logging system. The primary objective of the investigation was to develop radioelement specific concentration profiles for high priority liquid effluent waste sites using existing boreholes and to assess if radionuclides had reached groundwater. The project was considered to be a screening level effort.

SITE GEOHYDROLOGY

The 200 East and West Area liquid effluent sites are situated in the Hanford formation which comprises the vadose zone through most of the area. The Hanford formation is a product of cataclysmic flooding in the Pleistocene from glacial lake Missoula. It is comprised of 150 to 350 feet of unconsolidated glaciofluvial sediments consisting of pebble to boulder size gravels, fine- to coarse-grained sand, and silt and therefore is divided into three facies; gravel dominated, sand dominated and silt dominated.

Beneath the Hanford formation at 200 West Area, the discontinuous early "Palouse" soil and the Plio-Pleistocene unit occur. These strata are composed of carbonate-cemented silt or sand, or a calcic silty paleosol. They can be important to the migration of radionuclides in the subsurface because these units act as impermeable barriers and can create localized perched water zones.

The late Miocene to Pliocene Ringold Formation occupying the lower vadose zone and unconfined aquifer is up to 600 feet thick but pinches out in the 200 East area. The Ringold consists of fluvial gravel, fluvial sand, overbank deposits, lacustrine deposits and alluvial sand deposits. It is consolidated relative to the Hanford formation. The Columbia River Basalt Group occurs beneath the Ringold Formation and forms the bottom of the unconfined groundwater. Additional information on the geology of the Hanford Site can be found DOE-RL (1992,1993).

METHODOLOGY

Because of the large number liquid effluent waste sites, a selection process was developed to identify high priority sites which have a high probability of significant contamination. A majority of the liquid effluent sites have unsaturated zone monitoring boreholes historically used for gross gamma-ray logging, and/or groundwater monitoring wells. A extensive library of gross gamma-ray logs, waste disposal records and groundwater data were used to identify sites that had received and/or contain in the subsurface, significant inventories of radionuclides. For example, waste sites with gross gamma-ray logs that showed elevated gross activity throughout the unsaturated zone and into the groundwater were preferentially selected for spectral gamma-ray logging.

The investigation was limited to one borehole per waste site or waste site group. Only those boreholes which were had evidence of contamination (from gross gamma-ray logs),and optimally configured for passive gamma-ray logging were selected. Boreholes that had grout or bentonite seals behind the casing were often rejected for logging because the seals attenuated the gamma radiation in an unquantified manner which would result in inaccurate concentration profiles. As a result of the site and borehole selection process, thirty eight waste sites were identified for spectral gamma-ray logging of forty seven boreholes.

Equipment

The spectral gamma-ray logging system, referred to as the Radionuclide Logging System (RLS), was adapted from standard laboratory detection equipment to operate in a downhole environment. The primary component of the system is an 18% efficiency, high-purity intrinsic germanium detector mounted in a downhole probe along with the associated electronics and a liquid nitrogen dewar. The entire downhole assembly is water tight, allowing it to be used below the groundwater table.

The surface electronics equipment consists of a suite of standard laboratory rack mounted spectroscopy amplifiers and multi-channel analyzers interfaced to a personal computer. The probe depth is computer controlled by utilizing a servo-controlled hydraulic winch system and associated depth indicating equipment. This allows completely automated movement of the probe and very accurate probe depth positioning. The logging system is mounted in a cabin on a 2-ton logging truck.

Data Acquisition, Reduction and Interpretation

Gamma-ray spectra were recorded about every 0.5 feet in the borehole. A sample spectrum is shown in Figure 2. Each energy peak represents the gamma radiation from an individual radioisotope and the area under the peak provides a count rate which is used to calculate the in-situ radioelement concentration. The difficulty in detecting or quantifying the isotopes

depended on the concentration of the isotope in the subsurface, it's specific activity and the photon energy emitted during decay. The higher the concentration, the higher the specific activity and the higher the photon energy, the easier it was to quantify.

Custom data reduction software was prepared for reducing the individual spectra to radioelement concentrations. This software calculated the individual photon peak intensities in the spectrum, identified the parent man-made or natural radionuclide source, applied the calibration conversion to the radioelement concentration and corrected the data for any borehole environment factors. Finally, the data were output to a file in a format that allowed it to be plotted with the data from spectra recorded at other depths in the borehole. The data were graphically presented as spectral gamma-ray log plots of radioelement concentration versus depth as shown in Figure 3. Information on the calibration and calculation of radioelement concentration is provided in Brodeur et al. (1991).

- The left plot in Figure 3 shows the total number of gamma-ray photons detected in the entire spectrum versus depth, similar to a common gross gamma-ray log. To the right of the total gamma plot are individual plots for man-made radionuclides identified in the borehole. Plots similar to Figure 3 were prepared for all boreholes logged under the investigation.

Spectral gamma-ray log plots were reviewed to determine the location and deposition patterns of specific radionuclides. The radionuclides and the deposition pattern were correlated with the release records to see if the major constituents released were detected and to see if the deposition patterns observed could be explained by the type or quantity of effluent released. For boreholes that penetrated to groundwater, a determination as made as to whether or not the radionuclides had reached and impacted groundwater.

RESULTS

Man-made isotopes detected or measured with the spectral gamma-ray system include 60 Co, 125 Sb, 154 Eu, 152 Eu, 137 Cs, 238 U, 235 U, 241 Am, 233 Pa (237 Np), 238 Pu, 90 Sr. Cesium, cobalt, europium, antimony and protactinium were rather easy to measure down to a few picocuries per gram. Processed 238 U was quantified by the decay of the second daughter nuclide, 234 Pa which emits a 1001 keV gamma-ray photon. This was shown to be processed uranium as opposed to naturally occurring 238 U because it was not in secular equilibrium with it's eighth and ninth daughter nuclides.

241 Am, 235 U and 238 Pu were more difficult to quantify because they emit low energy gamma-rays which are significantly attenuated by the steel casings of boreholes. 90 Sr does not emit a gamma-ray, but when found in very high concentrations, it is detected by a bremsstrahlung radiation spectrum. Because the system was operated in a screening mode, the low concentrations of naturally occurring K, U and Th were not quantified.

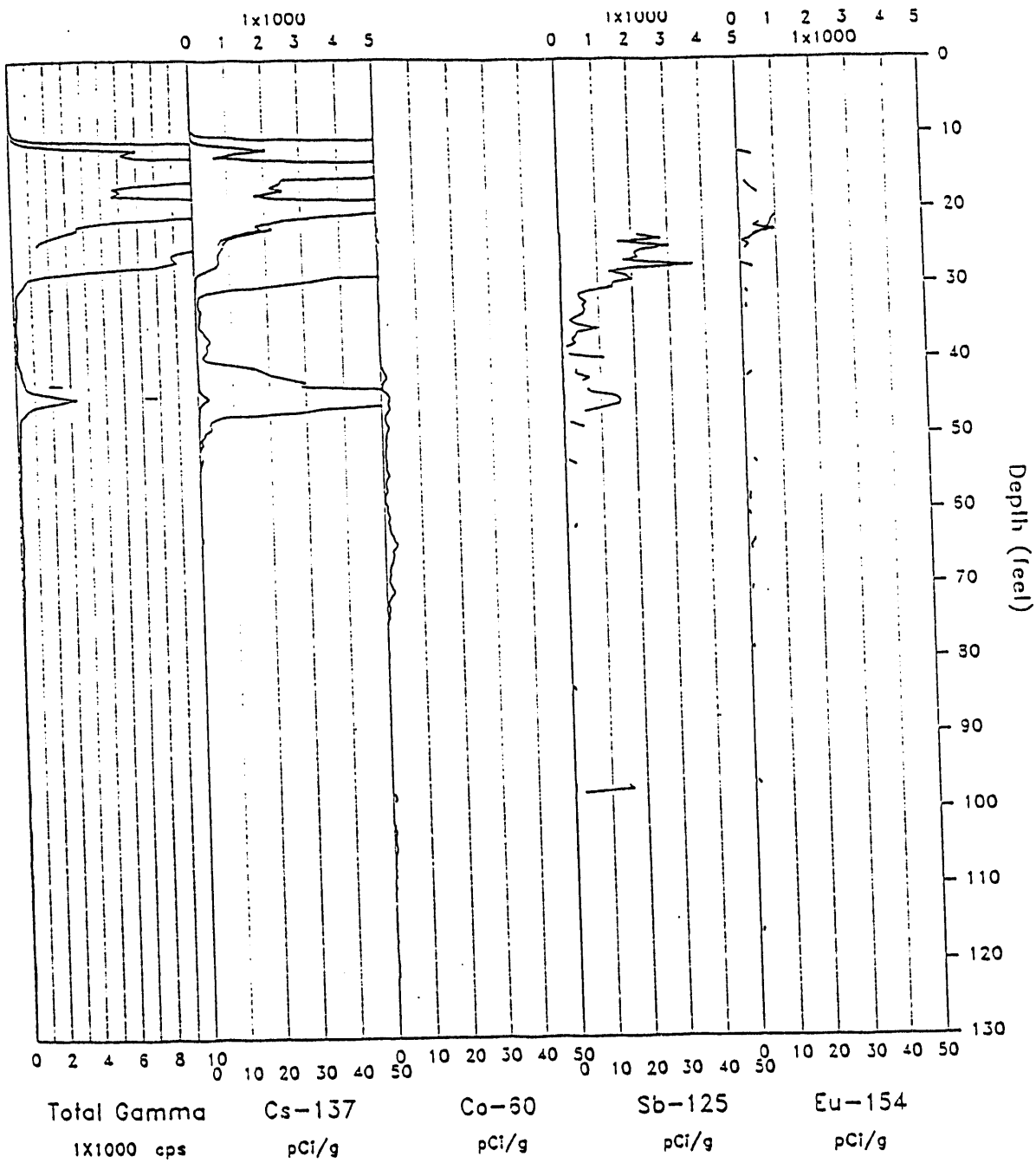

Figure 3. Typical borehole profile of radioelement concentration versus depth.

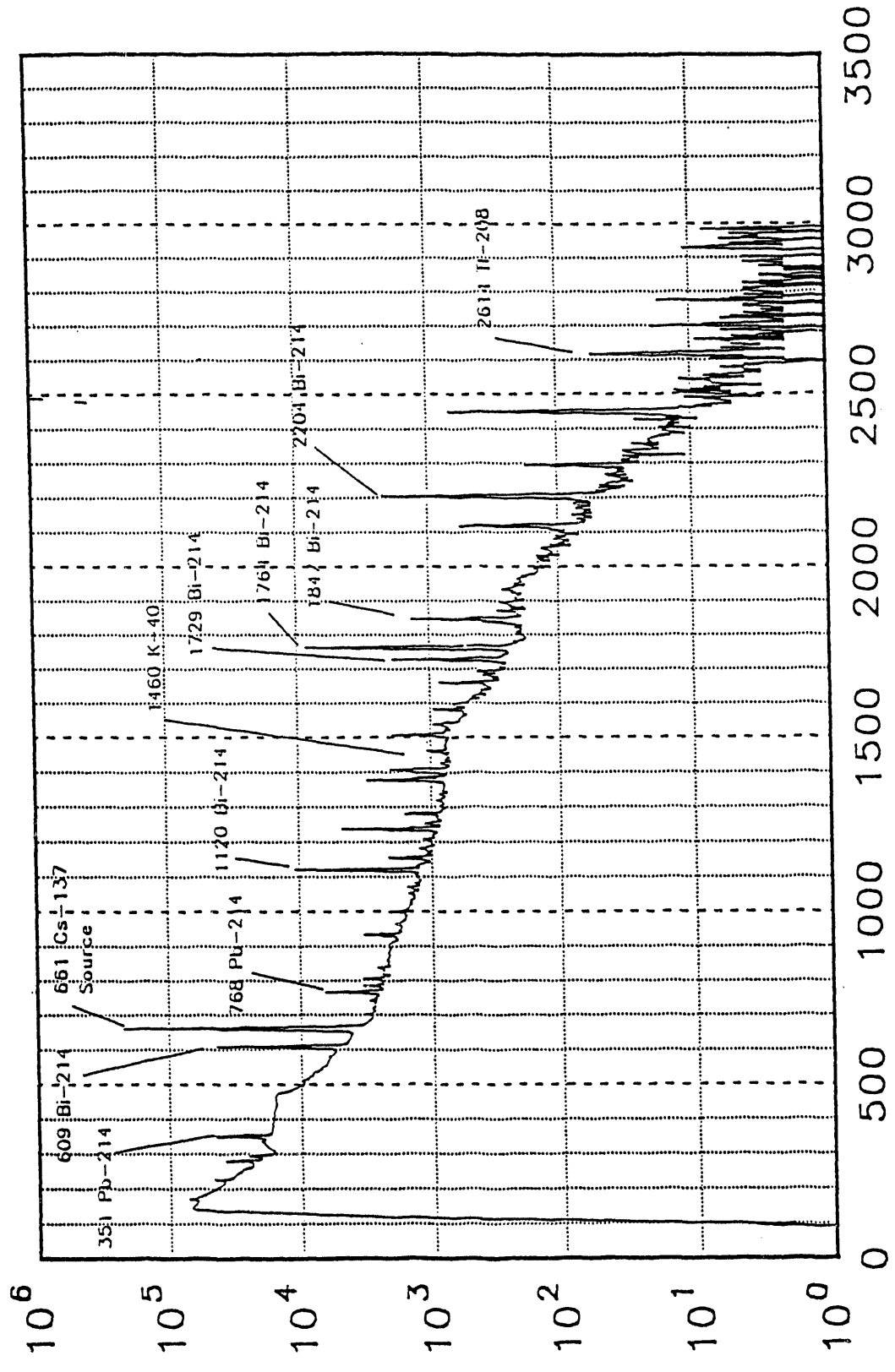

Figure 2. Typical gamma-ray spectra.

Radionuclide release inventory did not always agree with the radionuclides detected during the logging investigation. In some cases, specific isotopes were detected when operational records showed no evidence of the release. In others cases, records would indicate a relatively large release of a particular radionuclide, however none was detected. In the later case, it is possible that the radionuclides were not intercepted by the monitoring borehole. Geologic features may be present which could create preferential migration pathways away from a particular monitoring borehole.

Radionuclide Migration

Figure 3 illustrates typical radionuclide migration patterns found at the waste sites. Chemical cesium was generally found in a zone of very high concentration just beneath the liquid effluent structure and quickly decreased in concentration with depth. It does not tend to migrate very far. The exception to this is where cesium laden acidic effluent was released. The low pH effluent increased the mobility of cesium in the unsaturated zone. Figure 4 shows a radioelement concentration profile for a crib where effluent with pH as low as 2 was released.

Cobalt, antimony and europium have higher mobilities in the unsaturated zone sediments than cesium and are often found further from the release point. Uranium was the most mobile radionuclide detected in the study, especially when the effluent was even slightly acidic. Uranium was not found in any locally high concentration zone, indicating that it probably traveled vertically down with the moisture front.

Historical gross gamma-ray logs were used to assess if there had been any appreciable migration of the unsaturated zone radionuclides. Although there was no way to quantify the rate of migration, it was shown that at most waste sites studied there had not been any significant movement of the radionuclides observed in the investigation.

Groundwater Contamination Sources

Radionuclide concentration profiles indicated that a significant portion of gamma-emitting radionuclides released at the waste sites were retained in the unsaturated zone sediments. Six waste sites were identified as probable sources of groundwater contamination. When contaminants were detected below the water table, concentrations were several orders of magnitude less than those in the unsaturated zone. This is likely the result of an increase in the mobility of the radionuclide after reaching the groundwater, and the resulting dilution. However, it was apparent that the thick unsaturated zone retarded the migration of many gamma-emitting radionuclides and retained a large mass of contaminants high up in the unsaturated zone.

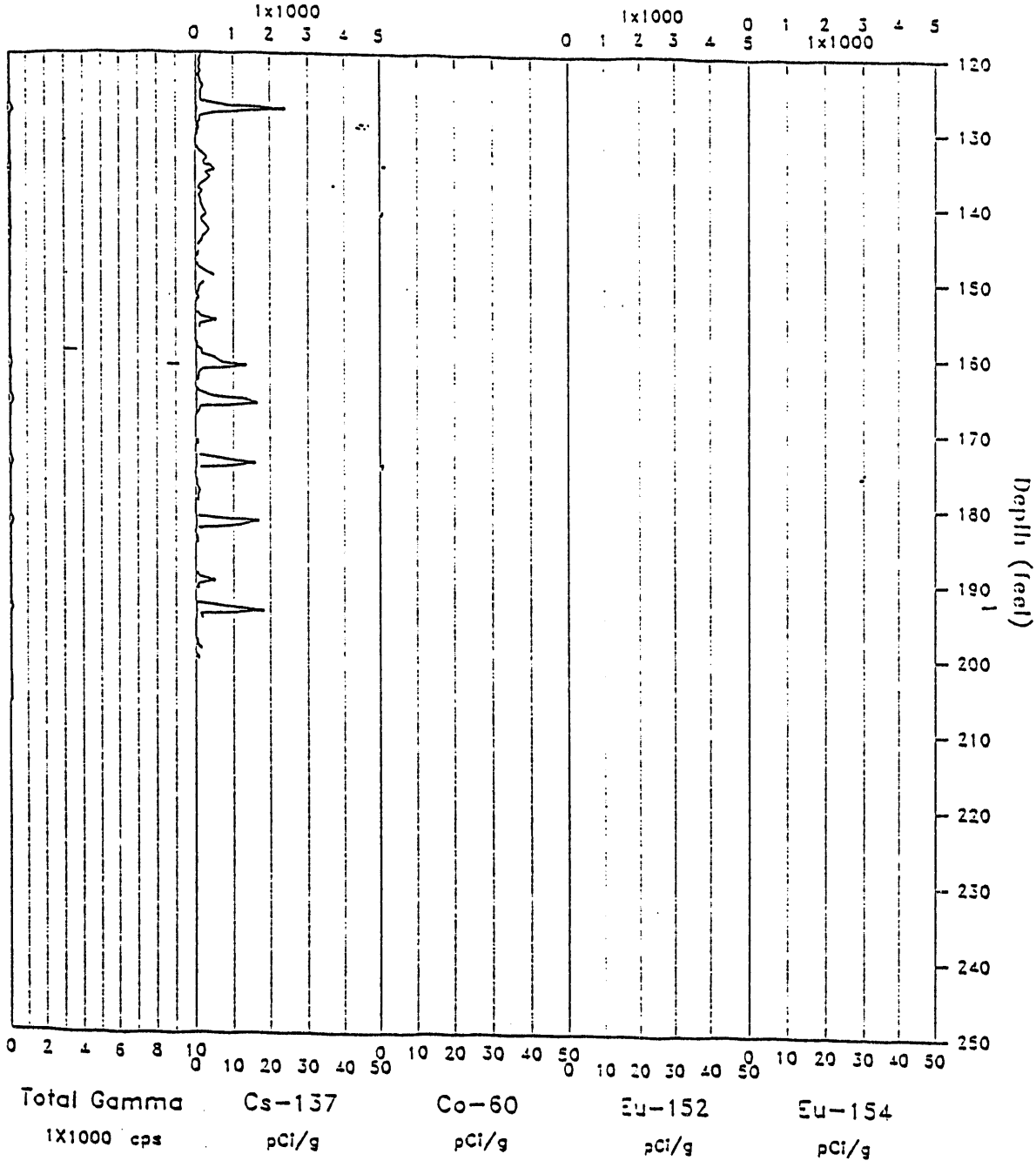

Figure 4. Profile showing the distribution of cesium in a borehole.

SUMMARY

This investigation utilized a spectral gamma-ray logging system to provide gamma-ray-emitting radionuclides concentration profiles of the subsurface at thirty eight radioactive liquid waste effluent discharge sites. From these profiles, it was possible at many of the sites to assess if the site had contributed to the groundwater contamination. During the course of individual site assessments, observations were made about the characteristic behavior of the various radionuclides in the unsaturated zone and a better understanding was obtained of the role played by the effluent chemistry (primarily pH) in the final deposition of the radionuclides. Data also indicated the potential for individual radionuclides to migrate.

The RLS was shown to be a cost-effective tool to provide initial site characterization data on gamma-ray emitting radionuclides in the subsurface. The data will be used to refine conceptual models of the saturated and unsaturated zones, and focus future remedial activities in the 200 Areas.

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