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FIELD DEMONSTRATION OF TECHNOLOGIES FOR CHARACTERIZATION OF URANIUM CONTAMINATION IN SURFACE SOILS*

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ABSTRACT

One means of improving the efficiency of studies associated with CERCLA and RCRA investigations of sites contaminated with uranium is to introduce new field screening technologies capable of quickly delineating the contamination distribution in surface soils. To this end, the performance of four technologies suitable for field measurement of uranium concentrations in soils was evaluated at the Fernald Environmental Management Project (FEMP). The four technologies tested were high-resolution gamma spectroscopy, wide-area beta scintillation counting, laser ablation-inductively coupled plasma-atomic emission spectroscopy (LA-ICP-AES), and long-range alpha detection (LRAD). The performance of each technique was assessed by comparing of the results obtained in the field and by comparing the field measurements to data obtained from laboratory analysis of soil samples. The data indicate that, while the results obtained using the four technologies are generally consistent, there is considerable scatter. Also, systematic differences were observed, particularly when the gamma spectroscopy results were compared to the beta scintillation and LRAD results. These descrepancies are attributed to inhomogeneities in the uranium distribution within the measurement locations and to effects of these inhomogeneities on properly calibrating each technique. A preliminary conclusion is that, although demonstration of a new field screening technologies is of critical importance, such efforts must be coupled with a means of interpreting the new data in light of information acquired from other characterization methods and with clarification of the measurement sensitivity requirements based on applicable regulatory cleanup levels.

INTRODUCTION

One of the major problems facing the U.S. Department of Energy (DOE) Environmental Restoration Program is the remediation of uranium-contaminated soils. In response to this problem, DOE's Office of Technology Development has initiated an Integrated Demonstration (ID) program to evaluate and compare the versatility, efficiency, and economics of various technologies for the characterization and remediation of uranium-contaminated soils. The FEMP, located 29 km northwest of Cincinnati, Ohio, has been selected as the host site for this ID program. This selection was based on known environmental problems stemming from past production of uranium metal for defense-related applications at the Fernald site.

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Current costs and schedules associated with the characterization of soils bearing radionuclide contamination represent a significant obstacle in the remediation of such sites. One means of addressing this problem is the development of real-time field screening technologies capable of providing data at high spatial resolution. Use of such techniques would minimize the total number of soil samples that must be collected, as well as reduce the number of sampling phases. Along these lines, four field-screening technologies for delineating the distribution of uranium contamination in surface and shallow subsurface soils have been tested and evaluated relative to their suitability for use in CERCLA and RCRA investigations.[1] These technologies are high-resolution gamma-ray spectroscopy, wide-area beta scintillation counting, laser ablation-inductively coupled plasma-atomic emission spectroscopy (LA-ICP-AES), and long-range alpha detection (LRAD). Each of these techniques was tested under field conditions at selected locations at the FEMP during the late summer and early fall of 1992. This paper describes the field testing program, presents an initial evaluation of the results obtained, and discusses the conclusions drawn concerning the performance of each of the four technologies.

DESCRIPTION OF SCREENING TECHNOLOGIES AND FIELD DEMONSTRATION PROGRAM

Each of the four screening technologies was used to measure the uranium concentration (expressed in pCi/g) on a sampling grid with 18 m centers. Two sites were selected at the FEMP, the drum bailing area and the incinerator area. Uranium contamination at the drum bailing area stems from the past washing of contaminated drums and other articles, as well as air-borne deposition from milling and washing activities conducted in a nearby production area. Contamination at the incinerator area is primarily the result of emissions from the incineration of uranium-contaminated combustibles. Soil samples were also collected from each area, and analyzed in the laboratory to determine the uranium concentration. Because of the intended usage of these sampling data as benchmark comparisons for the field measurements, they will be referred to as "validation data" for ease of reference. Sampling grids for the soil samples were established according to standard FEMP protocols and do not correlate directly with the sampling grids for the field screening techniques. The goal of the demonstration was not to make a point-by-point comparison, which would be of little use given the vast differences in the scales of measurement. Rather, the desire was to compare the site-wide distribution of uranium contamination as determined by the four screening technologies with that for the soil sampling effort. Since the focus of this paper is on the field testing campaign, a brief summary of each of the field measurement techniques follows.

Field demonstration of the high-resolution gamma-ray spectroscopy system was conducted by personnel from the Pacific Northwest Laboratories (PNL). The system is based on the use of a small germanium diode that may be collimated by specially shaped heavy metal shields. For the field demonstration, spectrometers were configured for both surface and downhole measurements. In the surface monitoring mode, the detector was suspended one meter above the ground by a tripod, and a large concave shield was placed over the detector to collect data from an area of approximately 100 m. In both configurations, the spectrometer detects uranium in the exposed surface and subsurface soils to a depth of approximately 40-50 cm, but the sensitivity decreases with depth due to gamma-ray attenuation.

The wide-area beta scintillation counter was developed at PNL to measure uranium concentrations in surface soils on a real-time basis. The system consists of multiple layers of plastic scintillating material for the measurement of beta particles from surficial soils (~the top 1 cm). The plastic scintillating layers are designed to measure uranium-238 surface concentrations by detecting the 2.29 MeV (maximum energy) beta particles from 234m-protactinium, a daughter product of uranium-238 decay. The system

is designed to discriminate between these high-energy beta particles and other interfering background radiation by using coincidence counting techniques, which identify high-energy beta particles by the depth to which they penetrate into the phosphor layer stack. The device used in the field tests was designed to monitor a surface area of approximately 0.1 to 0.2 m.

The LA-ICP-AES technique is a proven laboratory analytical method that has been adapted by Ames Laboratory for field applications.[2] A neodymium YAG laser is used to ablate a small sample of insitu soil (~10-20 µg), while an argon gas stream entrains the ablated sample particles and transports them directly into the ICP. The atomic emission from the ICP is transferred by fiber optics to a spectrometer for quantitative analysis of total uranium. During the course of an individual measurement, the ablating laser beam is rastered over a sampling area of about 6.5 cm.

LRAD system, which was developed at Los Alamos National Laboratory, detects alpha particles (and other ionizing radiation) by collecting and measuring the ions that are produced when alpha particles are stopped in air.[3] Because the ambient air is the "detector gas," the field LRAD system was configured to be set directly on the ground. In this configuration, it detects the uranium in the surface soil by monitoring the air ionization near the soil surface. The LRAD system tested at FEMP was designed to monitor contamination present on an ~1m^2 of surface.

From August to September 1992, the four field-screening technologies described above were tested under field conditions at FEMP. The wide-angle gamma-ray spectrometer was demonstrated in the surface detection mode only at the drum bailing area. Measurements were not made at the incinerator area due to equipment difficulties. In contrast to gamma-ray spectrometry, the LA-ICP-AES tests were conducted only at the incinerator area. The LRAD and beta counter were used to measure surface concentrations at both areas. However, due to muddy and irregular ground surface conditions, measurements were not obtained from all pre-selected sampling locations.

RESULTS AND DISCUSSION

Available data from the field demonstration studies are compiled in Figures 1a and 1b for the drum bailing and the incinerator areas, respectively. These figures also compile the soil sampling validation data. Corresponding concentration isopleth maps, based on the results obtained from the validation samples, are shown in Figures 2a and 2b for the drum bailing and incinerator areas, respectively. These data are compiled here as a basis for discussion of the performance of each technology in measuring uranium concentrations under actual field conditions. More detailed discussions of the results obtained with the individual technologies will be published elsewhere.

The assessment of each technology is based on intercomparison of the analytical results obtained by the individual techniques and comparison of the field measurement results with the laboratory validation data (Figures 1a and 1b). Although factors such as cost and reliability will be very important in final evaluations of the suitability of any of these techniques for specific applications, the focus here is on how well each technique is able to measure the uranium contamination distribution. To facilitate comparisons, the measured uranium contamination in concentrations were expressed in units of pCi/g. The calibrations required to convert the detector responses into such concentration units introduced errors, which are discussed later.

We begin our analysis of the results by comparing the data in terms of the decision that they will be used to defend, that is, which soils exceed regulatory standards for uranium contamination. Although

much uncertainty exists concerning the actual standard, probable limits will likely fall somewhere in the range of 100 pCi/g to background. Regardless of the actual standard, the results of this study are the same. In reviewing the field and validation data for the drum bailing area, each of the characterization methods consistently predicts that the soils will exceed the assumed range for the regulatory standards, except along the far north boundary of the site (Figure 1a). However, in the incinerator area, where uranium concentrations are much lower (Figure 1b), significantly different decisions would be made concerning the soils requiring remediation, depending on which field screening technology data set is used (unless the standard were set at or near background). This is a very important finding, because sites, such as the incinerator area, where contamination is widely distributed at levels near the regulatory standard, would benefit the most from field screening techniques. As such, development of novel sensor systems must be done in conjunction with gaining an understanding of how the acquired information relates to other field screening data, laboratory data, and the regulatory standard. In the paragraphs that follow, we outline in a preliminary fashion some of the steps that can be taken in interpreting the various similarities and differences noted in the five data sets.

One-to-one comparisons between the results obtained by the various techniques are illustrated by the scatter plots shown in Figures 3 and 4. Figure 3 shows comparisons of results between gamma spectroscopy and beta scintillation counting (Figure 3a), gamma spectroscopy and LRAD (Figure 3b), and beta scintillation counting and LRAD (Figure 3c) for the drum bailing area. Corresponding plots for the LA-ICP-AES technique are not provided because measurements were not made at the drum bailing area. Similar scatter plots for the incinerator area data obtained using beta scintillation counting, LRAD, and LA-ICP-AES are shown in Figure 4. As mentioned earlier, gamma spectroscopy data were not obtained for the incinerator area because of some operational problems encountered during the field measurements. The data in Figures 3 and 4 show significant scatter, particularly for the incinerator area, and the slopes of the regression lines differ significantly from one. Interpretations of these observations are discussed below.

Although the compared for the four technologies measurements were made at the same grid locations, the observed scatter in the data may be due to either local inhomogeneities in the actual uranium concentration or to measurement artifacts. Unfortunately, the available data are not sufficient to discriminate between these possibilities. The measured concentrations are not point concentrations but average concentrations for probed volumes that vary significantly for the individual measurement techniques. As pointed out above, the surface areas to which the techniques are sensitive vary from a few square centimeters to several tens of square meters. Also, the LA-ICP-AES and LRAD techniques are sensitive to uranium contamination on the surface of the exposed soil particles; the beta scintillator, perhaps to a depth of a few millimeters from the exposed surface; and the gamma spectroscopy technique, to a depth of several tens of centimeters. Because other FEMP characterization results[4,5] indicate that the contamination is heterogeneously distributed on the scales probed by the field measurement techniques much of the observed data scatter can probably be ascribed to these heterogeneities.

Although the data in Figure 3 exhibit a general linear correlation, the linear regression lines have slopes significantly different from one. This is particularly true for the comparisons of the LRAD and beta scintillator measurements with the gamma spectroscopy measurements. The slope of the regression line for the beta scintillator/LRAD scatter plot is, however, much closer to the expected value of one (Figure 3c). These results are consistent with the uranium contamination being concentrated close to the surface. Because the concentration results from gamma spectroscopy are calculated assuming a uniform uranium distribution as a function of depth, the calculated concentrations are lower than the actual surface concentrations. This results from averaging the uranium concentration over a much larger volume than

the actual volume near the surface in which the uranium is concentrated.

Since a variety of unverified assumptions concerning the distribution of the uranium contamination are inherent in calibrating each of the systems tested, we decided to compare the techniques on a relative basis. One way to do this is to plot and compare the quartiles for the results obtained using each technique. To generate these plots, the population of measurements for each technique is divided into four intervals, each of which contains one-quarter of the total population. The quarter in which each measurement falls is identified to facilitate data comparisons. The quartile plots for both the drum bailing and the incinerator areas are shown in Figures 5a and 5b, respectively. In general, the quartile plots indicate that the techniques give fairly consistent results, particularly for the drum bailing area, where the uranium concentrations are highest. Also, the results are, in general, consistent with the validation data in Figure 2a and 2b. The reasons for the inconsistencies that are observed cannot be determined from the available data but are probably due, at least in part, to inhomogeneities in the actual uranium concentration distribution.

CONCLUSIONS

Although it is premature at this point to reach any final conclusions concerning the characteristics of the various technologies that were tested, several preliminary conclusions can be drawn from the initial field testing experience and the data analysis. The field testing did demonstrate that prototype systems for each of the technologies could be assembled in a configuration suitable for field deployment. Each of the systems was successful in quickly obtaining meaningful field screening data, despite the fact that a variety of operational problems were encountered, as might be expected with initial deployment of first-of-a-kind systems in the field. Unfortunately, possible local inhomogeneities in the uranium concentration distribution have made unambiguous interpretation of the observed differences between the field demonstration results impossible. However, the similarities in the relative concentration profiles, as determined for the different techniques, and the fact that most of the absolute concentration measurements are within an order of magnitude do suggest that all of the methods may be useful for specific field applications. Nevertheless, additional work is needed before definitive statements can be made concerning the practical usefulness of any of the techniques.

At this point, while the techniques may be useful for generating relative concentration contour maps, they will probably have to be used in conjunction with sampling data to establish the absolute values of the contours. Techniques with different position resolutions can be used in a complementary fashion in order to construct contouring data with different resolutions, as the specific needs involved demand. The observed scatter in the low concentration data for the incinerator area indicates that these techniques still require significant improvement if they are to be useful at the nominal target cleanup levels of 35 pCi/g. In general, these initial field tests also indicate that the four techniques show promise for quickly determining concentration contours in the field.

ACKNOWLEDGEMENT

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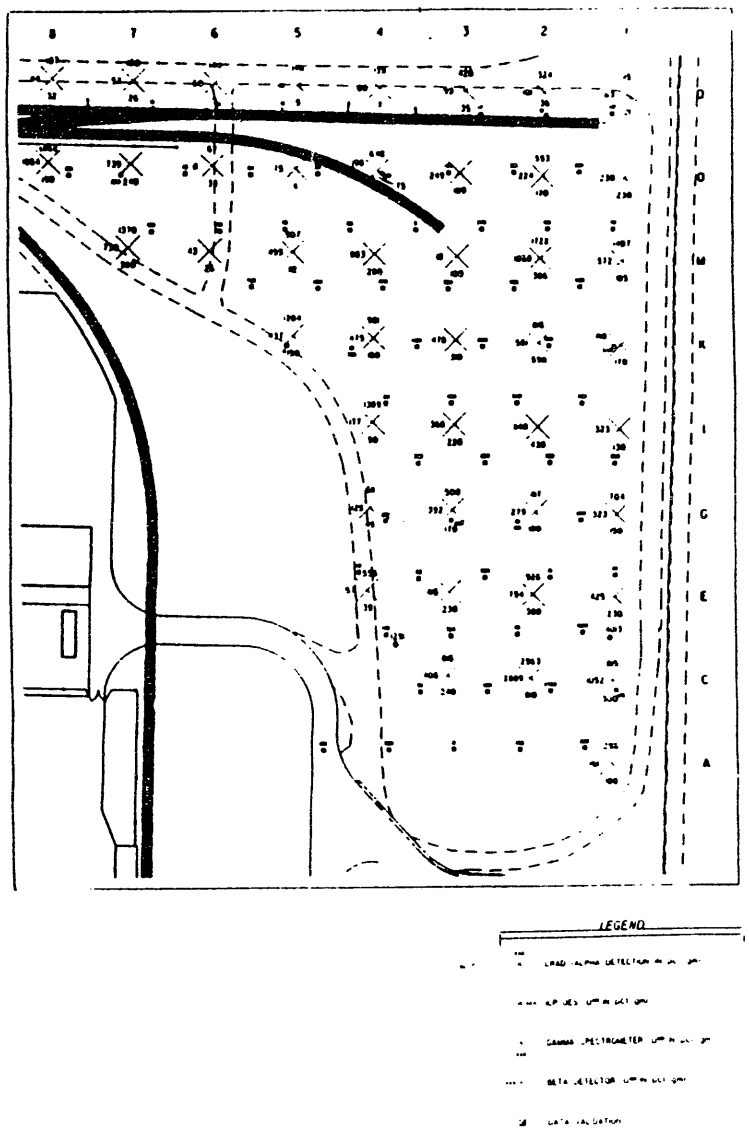
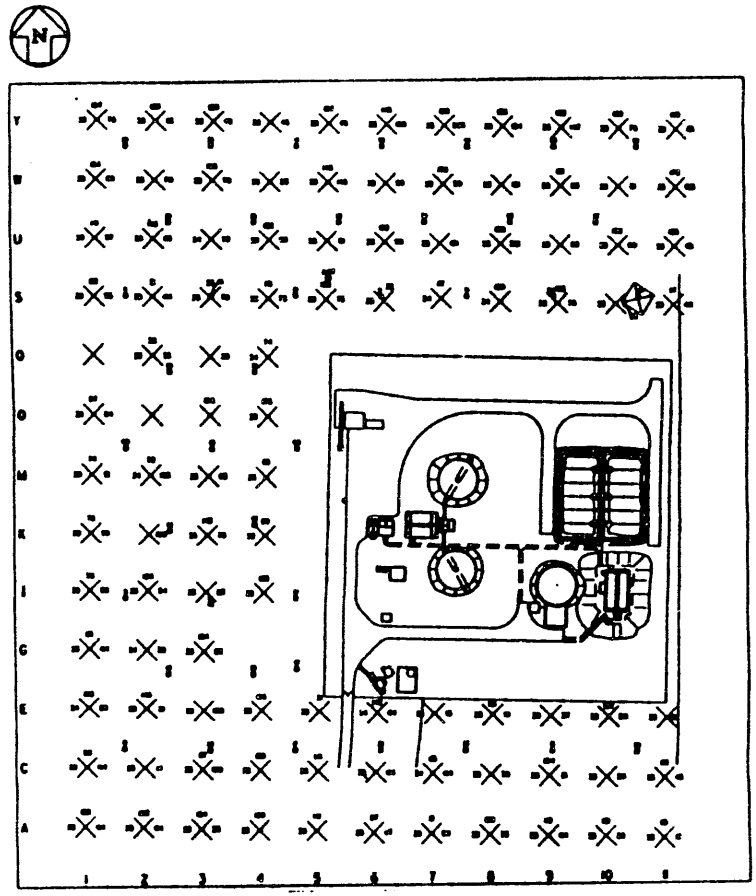
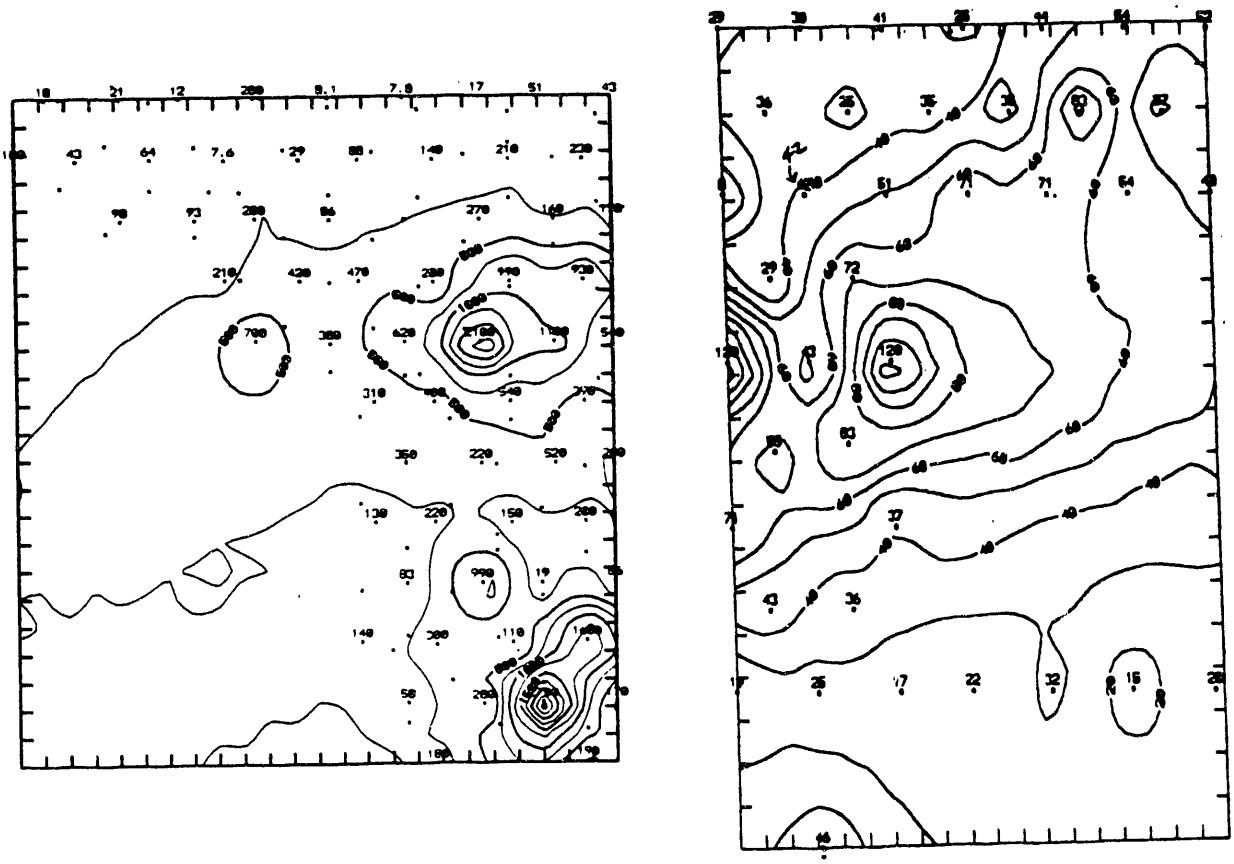
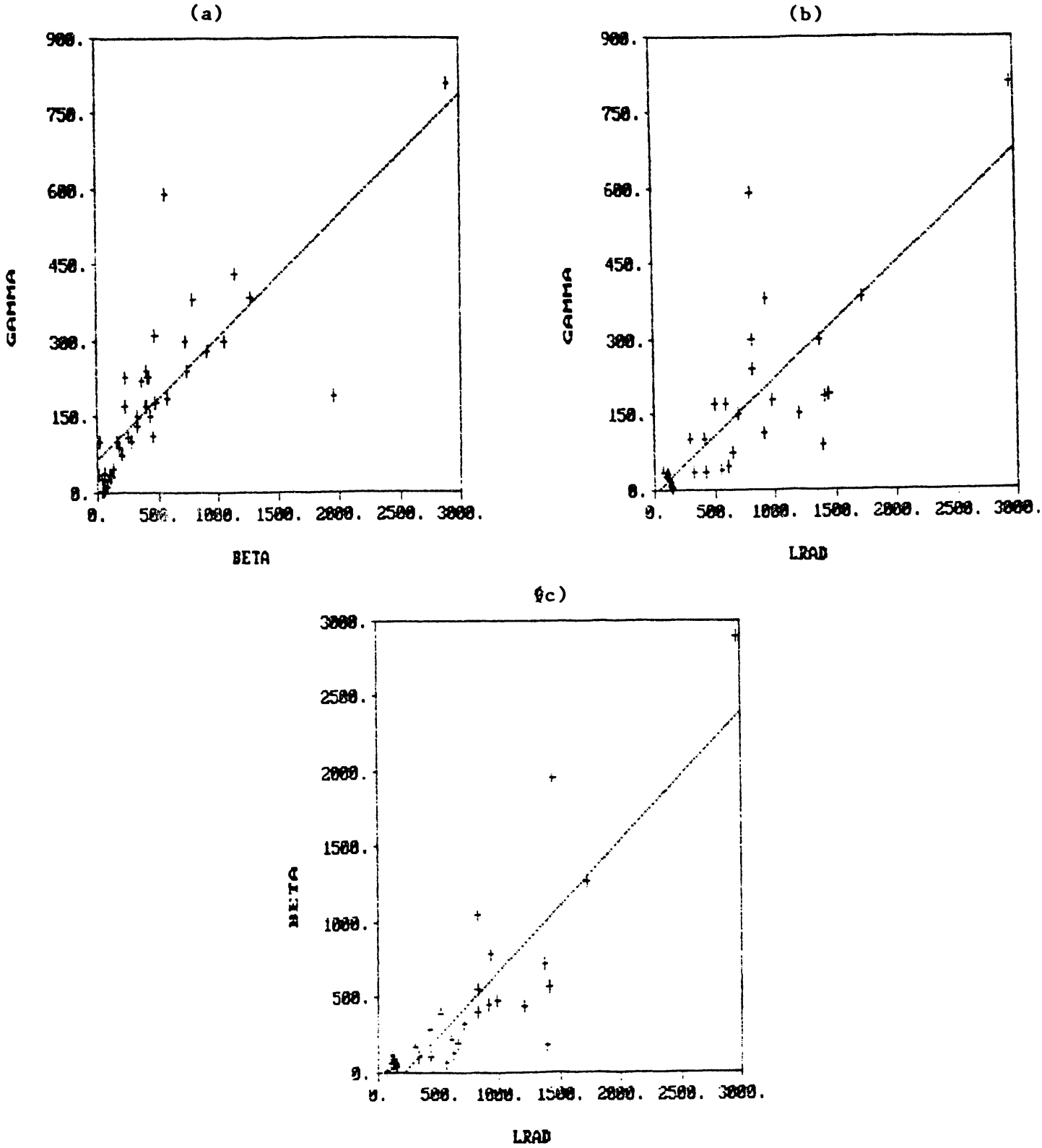




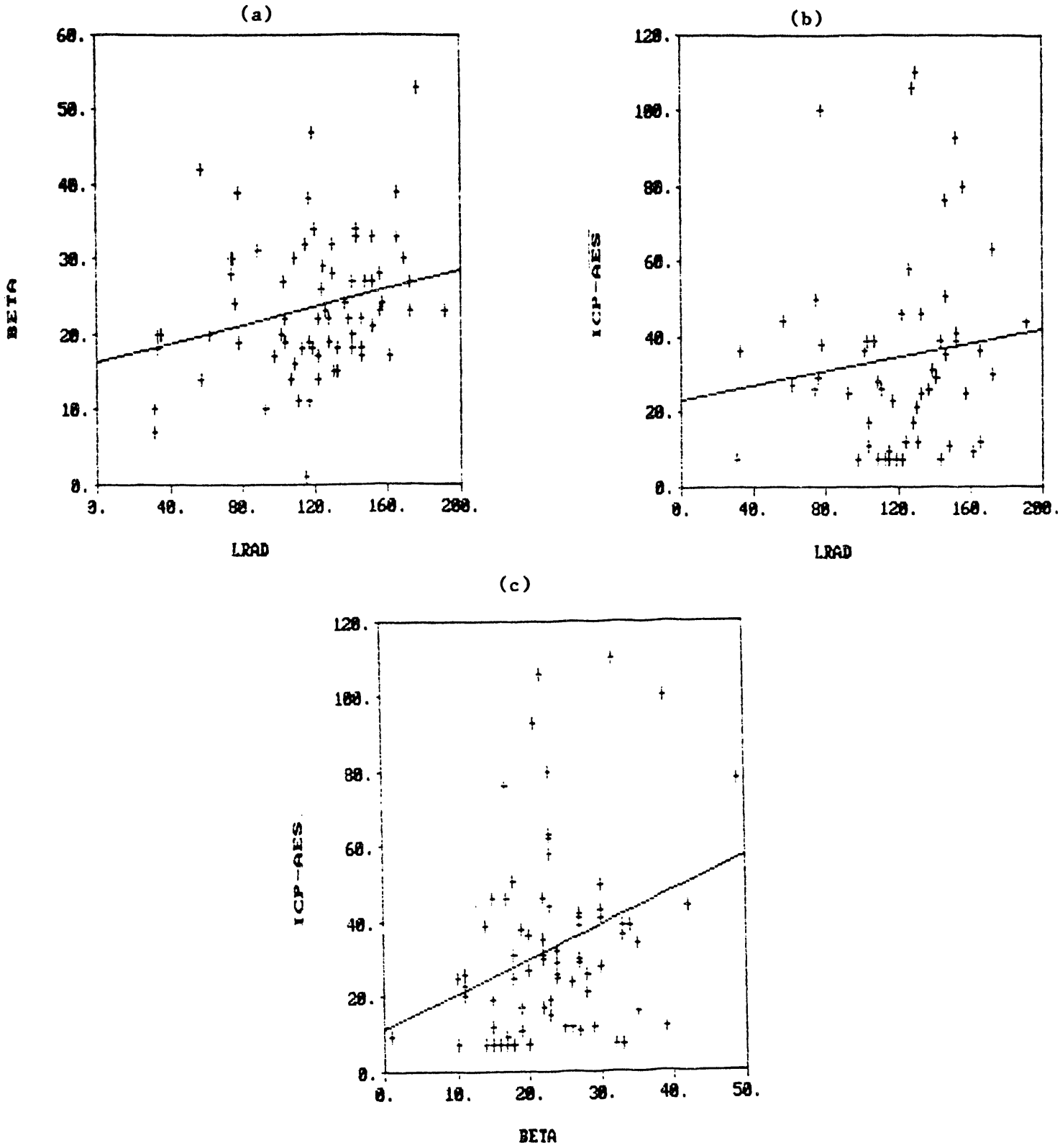
Figures 1a and 1b. Compilation of Field Measurement Data and Validation Data for the Drum Bailing (1a) and Incinerator (1b) Areas



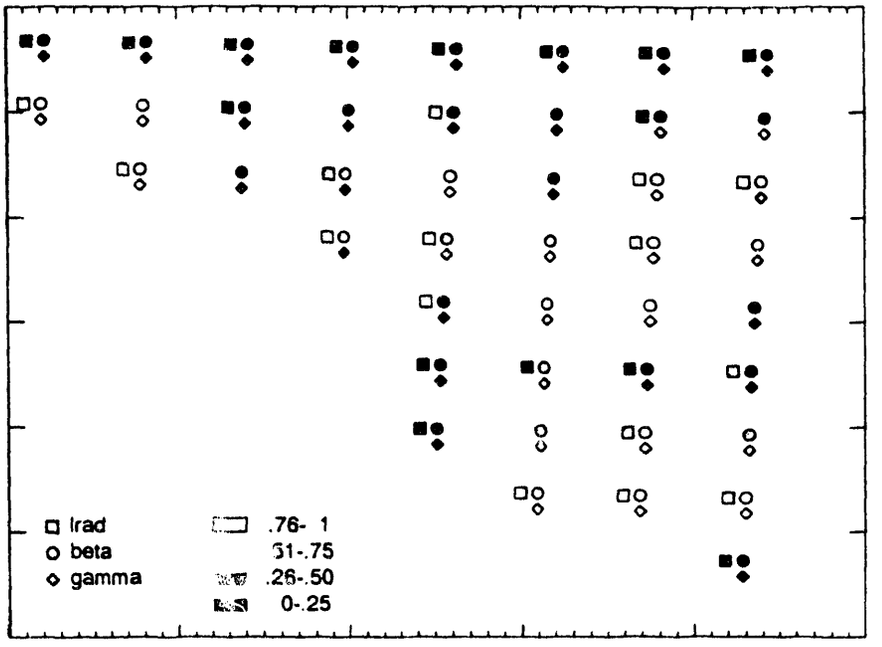
Figures 2a and 2b. Concentration (pCi/g) Isopleth Maps for the Drum Bailing (a) and Incinerator (b) Areas Based on the Validation Data



Figures 3a, 3b, and 3c. Scatter Plots for Uranium Concentration (pCi/g) Data Obtained by Three Techniques from the Drum Bailing Area



Figures 4a, 4b, and 4c. Scatter Plots for Uranium Concentration (pCi/g) Data Obtained by Three Techniques from the Incinerator Area



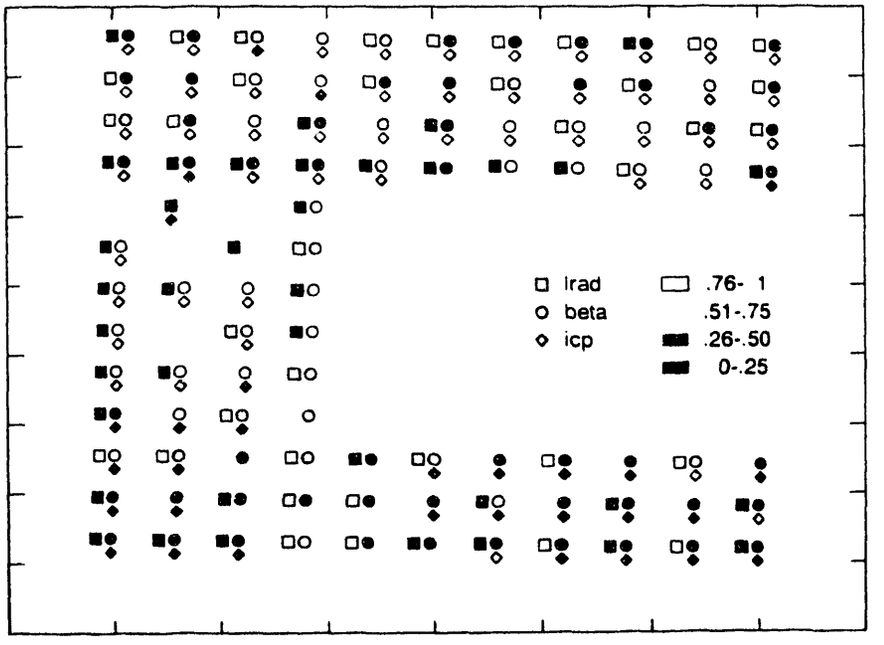


Figure 5a and 5b. Quartile Plots for the Field Data Obtained from the Drum Bailing (a) and Incinerator (b) Areas

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