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Use of Passive Alpha Detectors to Screen for Uranium Contamination in a Field at Fernald, Ohio

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ABSTRACT

This paper reports the results from a field test of newly developed techniques for inexpensive, *in situ* screening of soil for alpha contamination. Passive alpha detectors that are commercially available for the detection indoor airborne alpha activity (i.e., ^{222}Rn) have been modified so they can be applied to the detection of alpha contamination on surfaces or in soils. Results reported here are from an intercomparison involving several different techniques with all measurements being made at the same sites in a field near the formerly used uranium processing facility at Fernald, Ohio, during the summer of 1994. The results for two types of passive alpha detector show that the quality of calibration is improved if soils samples are milled to increase homogeneity within the soil matrices. The correlation between laboratory based radiochemical analyses and quick, field-based screening measurements is acceptable and can be improved if the passive devices are left for longer exposure times in the field. The total cost per measurement for either type of passive alpha detector is probably less than $25 and should provide a cost-effective means for site managers to develop the information needed to find areas with remaining alpha contamination so resources can be allocated efficiently.

BACKGROUND

Among the emerging technologies needed to execute cleanup of U.S. Department of Energy (DOE) sites are new types of alpha contamination monitors. A great need exists for techniques that are easy to implement, fast, and reliable. Often characterization techniques will be implemented by workers in the field who are not highly trained in health physics or chemistry. Also, new techniques are needed that accurately survey soils *in situ* for alpha contamination with very short (same day or next day) turnaround times for the analysis. Examples of DOE sites which could benefit from cheaper screening tools for alpha contaminated soil include: Rocky Flats, Hanford, Oak Ridge, and the Nevada Test Site. Two types of commercially available passive radon detectors have been identified that, with appropriate modification, are suitable for this purpose: alpha track detectors (ATDs) and electret ion chambers (EICs). Each technology is described below.

ATD Technology

ATDs are the basis of a technology that can achieve large numbers of inexpensive surface and near-surface soil measurements that are both accurate and simple to make. Alter and Fleisher[1] described the application of ATDs to the measurement of indoor airborne alpha activity. The underlying principles of the measurement are: (1) that damage tracks in the plastic material are induced by the passage of alpha particles through the material, (2) that the damage tracks can be subsequently enlarged by exposure to sodium hydroxide solution, (3) that the enlarged tracks can be counted with a microscope, and (4) that the density (per unit area) of tracks is proportional to the alpha activity. Plastic ATDs are about 70% efficient in detecting alpha rays while being almost completely insensitive to gamma and beta radiations.

The technology has already been commercialized for the monitoring of environmental radon. Millions of detectors of the ATD type have been sold both to individual homeowners and to federal agencies mandated to screen their facilities for radon. The ATD technology has passed the stringent test criteria, blind testing, and quality assurance procedures set under EPA's Radon Measurement Proficiency Program.

To adapt this technology to other needs, the track-registering plastic element of the radon monitor can be laid directly on or in an alpha-contaminated soil or soil sample. After the plastic is chemically etched and optically scanned, both the surface alpha activity and a spatial map of the contaminant distribution may be determined. Several technical issues are being addressed to improve the performance of the ATD as applied to environmental restoration (ER) and decontamination and decommissioning (D&D) needs. The commercially available application of ATDs to airborne alpha activity measurements produces chips with damage tracks that are randomly distributed. The currently used optical scanning systems have been optimized for randomly distributed tracks. Future applications will yield chips with clusters of tracks due to the presence of macroscopic radioactive particles in some cases. Techniques are being developed to detect and evaluate track clusters. Most available devices for holding the chips during exposure were developed for the indoor environment, which is highly controlled. Future applications will call for ATDs to be left outside and exposed to extremes of temperature, humidity, and wind. Protocols and device holders are being developed to mitigate adverse effects of moisture condensation, abrasion, and ^{222}Rn decay products in soil gas.

Preliminary experiments indicate great potential for ultimate success in the application of ATDs to ER and D&D projects. Pilot measurements have already demonstrated that ATDs can:

- Quantify contamination at 100 dpm/100 cm^2 after an hour's exposure,
- Provide a permanent record of the alpha radiation field,
- Show whether the contamination is homogeneously or inhomogeneously distributed,
- Identify the presence of "hot" microparticles such as PuO_2,
- Be cut to a size and shape to fit into any location, such as inside a crack, and
- Be deployed on interior or outdoor surfaces, on or under the surface of soil, or under water.

The above results have been published[2].

EIC Technology

EICs provide another inexpensive measurement technique that is very easy to implement. Kotrappa and coworkers[3,4] have described the application of EICs to the measurement of indoor alpha activity. The underlying principle of the measurement is that ionizing particles that pass through the air in the sensitive volume of the EIC create electron showers that are attracted to the positively charged face of the electret. neutralizing the charge. The rate at which the charge is neutralized is proportional to the energy deposited in the air. The technology has already been commercialized for the monitoring of environmental radon. Hundreds of thousands of detectors of the EIC type have been sold both to individual homeowners and to governmental agencies (principally school systems) mandated to screen their facilities for radon. The EIC technology has passed the stringent test criteria, blind testing, and quality assurance procedures set under EPA's Radon Measurement Proficiency Program.

To adapt this technology to ER and D&D needs in the field site personnel can use inexpensive (~$750) hand-held voltmeters to record EIC voltages before and after deployment. Before this

technology can be deployed on a wide scale, techniques or protocols must be developed to mitigate the adverse effects of the outdoor environment. Initial field tests have shown that overnight exposures can result in water vapor condensing on the electret which neutralizes the surface charge density. Techniques are also needed to prevent or discriminate ^{222}Rn-induced charge neutralization. Early results suggested that easily measured voltage drops occur within 1 - 2 hours of laboratory exposure to soil containing about 20 pCi g^{-1} of ^{239}Pu.

This paper describes results obtained when ATDs and EICs were used to screen a field near the former Feed Material Production Plant at Fernald, Ohio, for uranium contamination. The results obtained with ATDs and EICs are compared with radiochemical data collected at the same positions. Reasons for discrepancies are also discussed.

EXPERIMENTAL METHODS

Site Description

The grass-covered field where the measurements were made was downwind from a formerly used incinerator. The incinerator had been used to burn general trash that sometimes was contaminated with uranium. All measurements were made within a square area, about 360 m on a side. The staff from the Fernald Environmental Management Project (FEMP) established a set of grid references in the field. These grid references were used to locate measurement positions to an accuracy of about 10 cm. Figure 1 summarizes the uranium contamination as determined by radiochemical analyses of samples from 143 positions within the site. The radiochemical analyses were provided by FEMP staff members who were supporting a field intercomparison of seven methods for measuring uranium contamination in soil (V. Tidwell, Sandia National Laboratory, personal communication).

Measurement Protocols

The measurement positions were marked with wooden stakes by the staff from FEMP. These stakes provided geographic loci that were recorded in field logs. Near each measurement position, grass and other vegetation was scraped away with a small trowel to reveal a generally circular area (radius of about 10 cm) with a reasonably smooth dirt surface.

Before deployment, ATDs were kept in heat-sealed radon-proof Mylar® bags, with four ATDs per bag. After opening a bag and removing the ATDs, the protective polyethylene covering was removed from one side of each ATD and the exposed surface placed onto the soil. The time of placement was recorded in the field log. About twenty-four hours later, the ATDs were retrieved, placed in Mylar® bags that were taped shut, and the retrieval time recorded in the field log. The exposed ATDs in Mylar® bags were returned to Landauer, Inc., where the ATDs were exposed to 6N NaOH for fifteen hours before being optically scanned for radiation damage tracks. The uranium concentration was estimated from the rate at which track density increased per unit time (tracks mm^{-2} h^{-1}).

Before deployment, electrets were kept in small, electrically insulating holders. After removal from the holders, the surface charge remaining on each of two electrets was measured with a hand-held voltmeter. One electret was placed into a holder that contained no shielding material, forming an unshielded EIC. The unshielded EIC was placed onto the soil leaving the electret about 4 cm above the soil. The other electret was placed into an identical holder except that its open face was covered with a gas-permeable piece of filter paper, forming a shielded EIC. The shielded EIC was placed on the soil within 10 cm of the unshielded EIC. The measurement position, electret identification numbers, and the beginning voltages were recorded in the field log. About six or seven hours later, the EICs were recovered and the remaining voltages were measured and recorded. The unshielded EICs respond to alpha activity from both alpha-emitting contaminants in the top layer of soil (a few mm thick) and from radon in the gas in the soil. In contrast, the shielded EICs respond only to alpha activity from radon in the

soil gas. The uranium concentration was estimated from the difference in the time rate of loss of voltage ($V\ h^{-1}$) between the unshielded and shielded EICs.

RESULTS AND DISCUSSION

Field and Laboratory Calibrations

For calibration purposes, the FEMP staff prepared four (4) large boxes containing soils with predetermined uranium concentrations. The soils in these boxes were prepared by mixing, in known ratios, a soil with background uranium concentration (<10 pCi g^{-1}) with a soil having a higher uranium concentration (>200 pCi g^{-1}). The mixing was accomplished by use of a cement mixer. This resulted in a product soil having many very large clumps. While at the site, EICs and ATDs were exposed to these very coarse soils. Figure 2 provides a graphical comparison of the of the results obtained using radiochemical analyses of these soils with those obtained using either ATDs or EICs.

At the conclusion of the entire intercomparison exercise, the FEMP staff retrieved representative samples of the soils in these calibration boxes and sent them to our laboratory. The samples were milled in a ball mill for three (3) hours, at which time they were very fine powders. Portions of each sample were transferred into small plastic dishes for laboratory exposures. Figure 3 provides a graphical comparison of the of the results obtained using radiochemical analyses of these soils with those obtained using either ATDs or EICs in the laboratory. The results shown in Figure 3 were used to determine calibration coefficients used for interpretation of results obtained in the field.

The most noticeable differences between the field and laboratory calibration results are the increased offsets for both ATDs and EICs and the increased deviation from linearity for the EIC results. In the field, both techniques exhibited a higher response for minimal uranium contamination than was the case in the laboratory. There are differences between the experimental protocols that may contribute to the observed differences. First, in the laboratory, ATDs and EICs were exposed for about a week, resulting in much larger instrumental signals and, hence, lower signal-to-noise ratios. Second, milling of the soil samples reduced both physical and compositional heterogeneity. After milling, the surface of the powdered soil was much flatter, yielding a much more uniform coupling of the ATDs and EICs with the soil beneath them. Because the passive alpha detectors are only capable of detecting uranium in the top few mm of soil, more uniform coupling would reduce variation of response among detectors. In the case of the EICs, this would also reduce any variation among EICs in the amount of ^{222}Rn building up within them from decay of ^{226}Ra in the soil.

Intercomparison with Radiochemical Results

At 143 positions within the grass-covered field at FEMP, uranium concentration in the top layer of soil was determined using three different techniques: EICs, ATDs, and radiochemical analyses. The passive alpha detectors were used for *in situ*, screening measurements whereas samples from the top 15 cm of soil were removed and radiochemically analyzed in the laboratory. Figure 4 provides a graphical comparison of the radiochemical results with the EIC and ATD results for the corresponding position within the field.

Compared to the calibration results, the correlation between the screening results and radiochemical laboratory results is markedly reduced. One contributing factor is the short exposure times used for the screening measurements. Based on propagation of errors and binomial counting statistics, the minimum detectable uranium concentration was calculated for a range of exposure times (Figure 5). The minimum detectable concentrations for the screening measurements used here are 48 ± 10 pCi g^{-1} and 32 ± 6 pCi g^{-1} for EICs and ATDs, respectively. The resolution of these two screening techniques is comparable to the mean level of contamination in the study site. Another contributing factor is the heterogeneity in the natural soil matrix compared to milled laboratory samples. In southern Ohio there is a significant fraction of the soil composed of clay. Clay tends to fracture as it dries out. The fracturing results in small channels creating a variable network through which soil gas and the radon it contains may

move to the vicinity of any measurement position. Other causes of small fractures and channels in the soil are the growth and decay of plant roots and the movement of small insects and worms through the soil. The small channels and the varying amounts of soil gas radon introduced into the sensitive volumes of EICs will increase the apparent measurement noise in the response of the EICs. Ongoing work in our laboratory seeks to develop measurement protocols that extend the exposure times for ATDs and EICs possible in humid, temperate sites in the eastern U.S.

CONCLUSIONS

The principal finding of this study is that ATDs and EICs are inexpensive alternative methods for the screening of sites for soil contaminated with uranium or other alpha-emitting isotopes. These techniques can be implemented by general site personnel who do not have specialized training in health physics or chemistry.

The cost for an EIC measurement is less than $20 and may be less than $10. An electret costs $15 and can be used more than once depending on how fast the surface charge is neutralized. Presently, EICs can be read in the field with a $750, hand-held voltmeter. Minimal turn-around time is needed beyond exposure time for each EIC. For this study, a pair of technicians were able to deploy and retrieve about 75 EICs in an eight-hour day, including recording of all needed data. We estimate that labor productivity may be doubled if bar codes and a reader were to be used in these measurements.

The cost of an ATD measurement is also quite low, probably less than $10. If purchased in large quantity (>10,000), radon measurements can be obtained for less than $10 each. For that price, one might obtain: (1) sets of ATDs packaged in heat-sealed Mylar® bags with removable bar-coded labels attached, (2) laboratory processing of the ATDs and disposal of all waste products, and (3) a computerized report of the data from each ATD. For this study, a pair of technicians were able to prepare about 90 measurement positions and deploy four ATDs at each position in an eight-hour day. Within about four hours the following day, the ATDs were retrieved from the 90 positions and prepared for shipment back to the vendor for laboratory analysis. In the future, we believe it will be feasible to set up a small on-site laboratory trailer to process ATDs, decreasing overall turn-around time and obviating the need for off-site shipment of exposed ATDs.

ACKNOWLEDGMENTS

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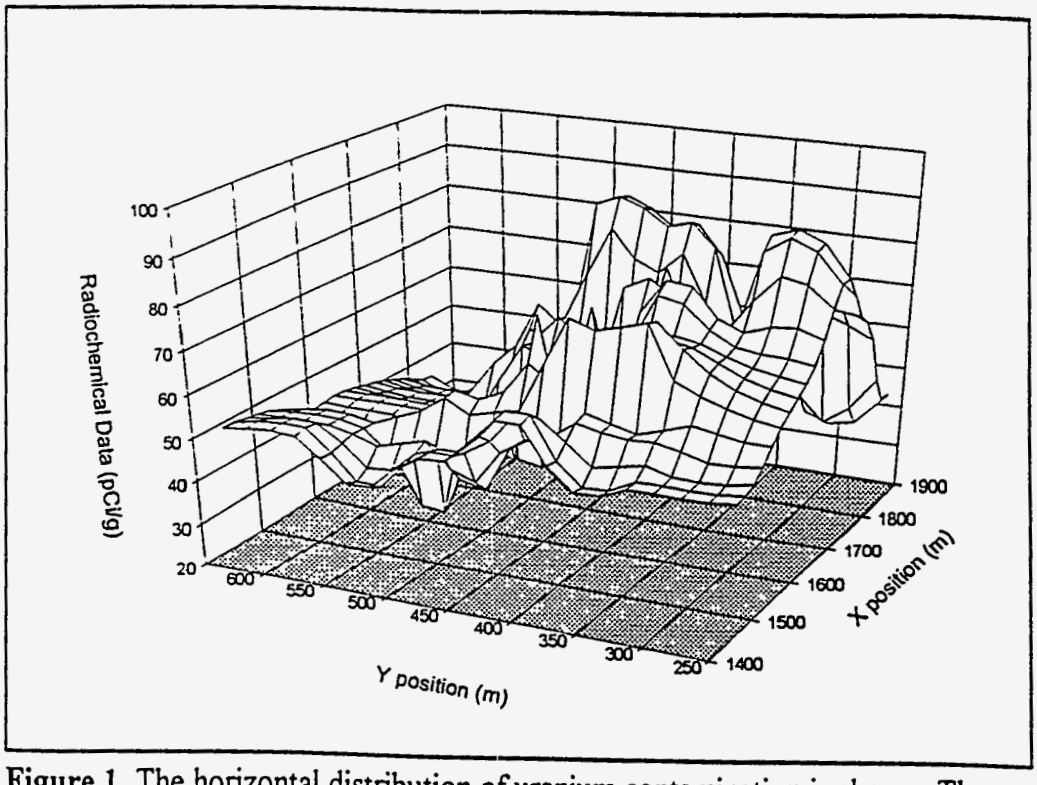

Figure 1. The horizontal distribution of uranium contamination is shown. The uranium concentrations in soil were determined by radiochemical analyses. The mean of 143 data is 53 (± 22) pCi g^{-1}.

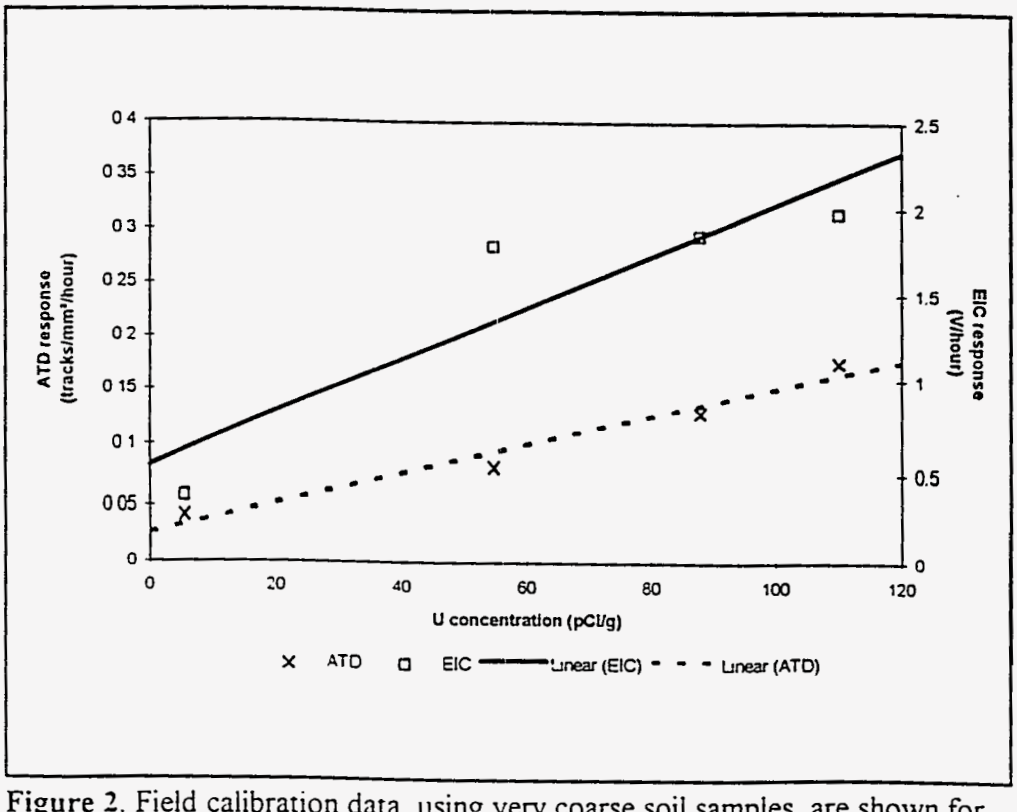

Figure 2. Field calibration data, using very coarse soil samples, are shown for two type of passive alpha detectors.

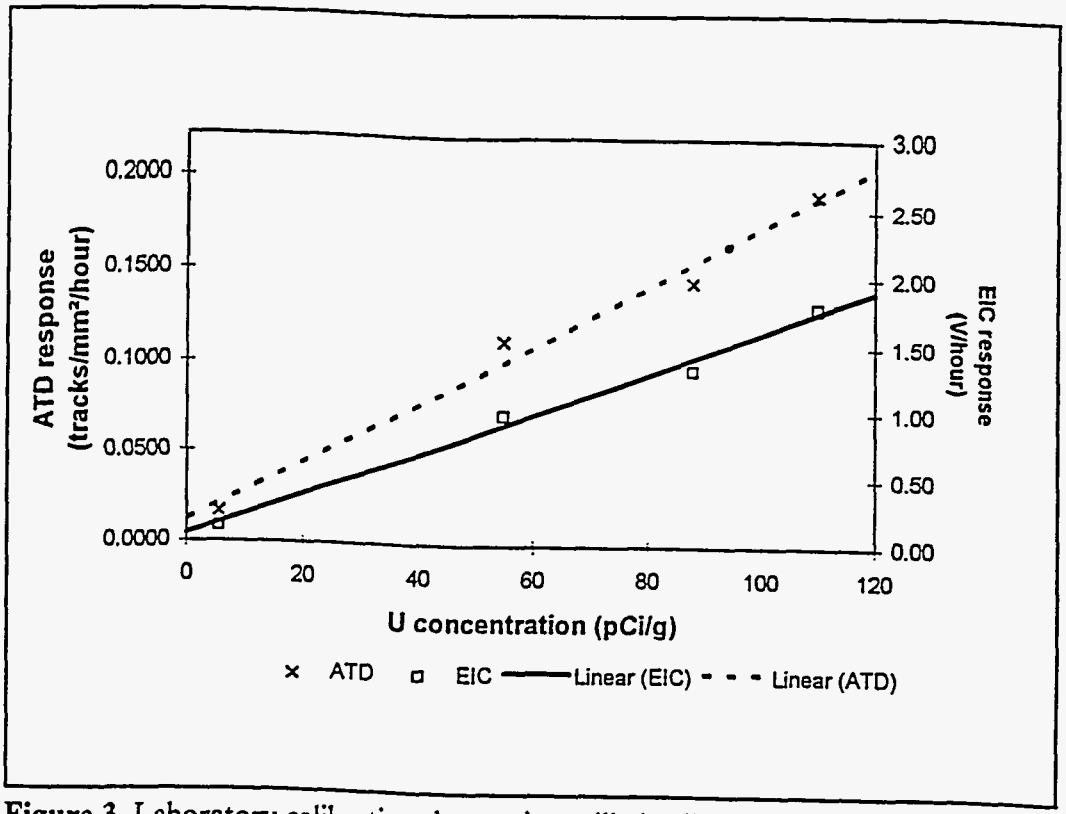

Figure 3. Laboratory calibration data, using milled soil samples, are shown for two type of passive alpha detectors.

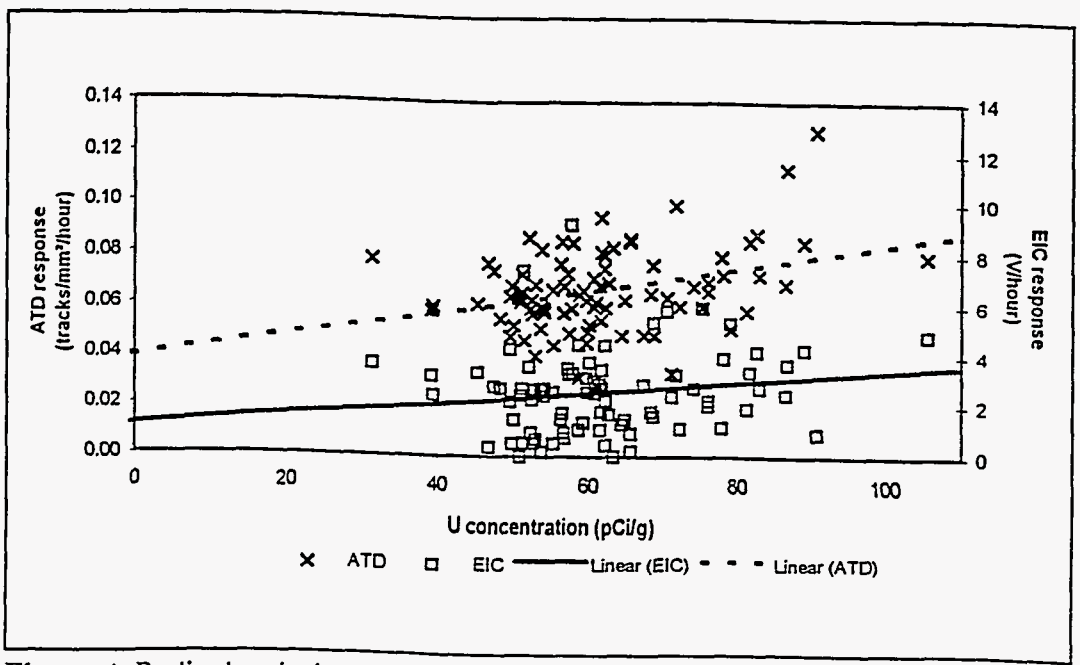

Figure 4. Radiochemical results for uranium contamination at 143 sites are compared with results obtained with two types of passive alpha detectors.

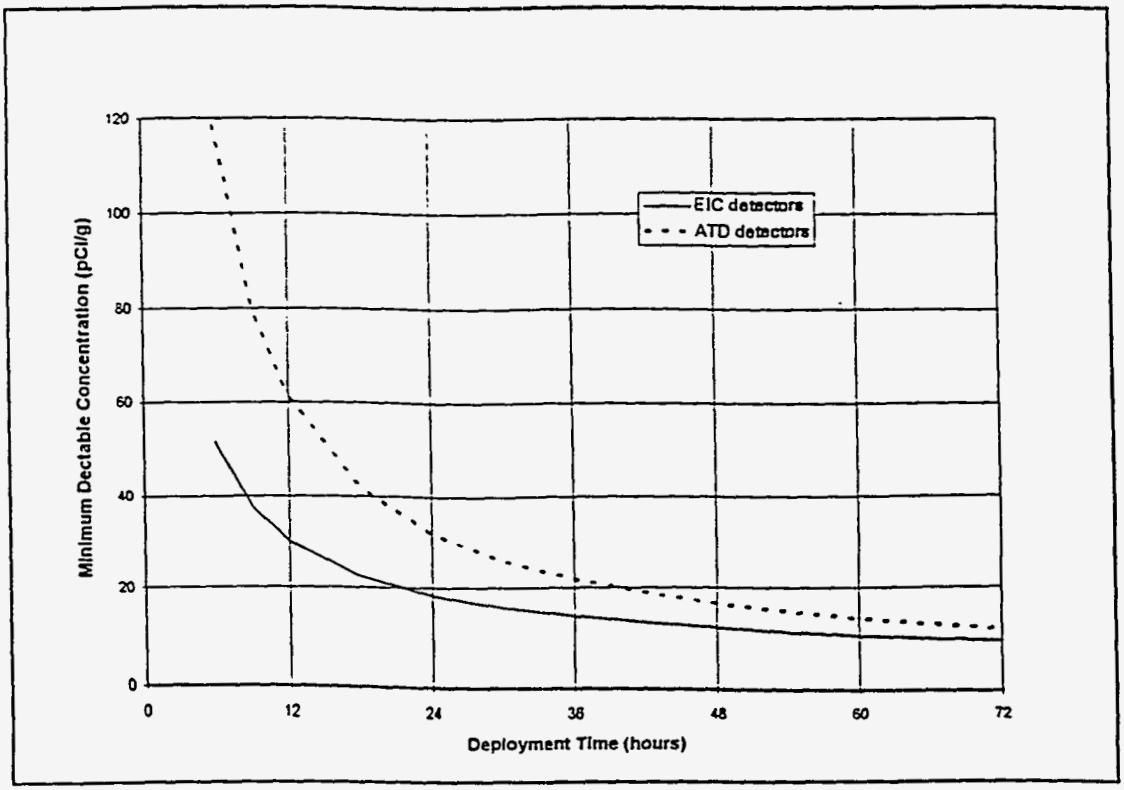

Figure 5. The detectable concentration of U in soil decreases as the time of deployment increases for these passive alpha detectors.

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