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PASSIVE FIELD MONITORING OF PLUTONIUM AND AMERICIUM IN SOIL*

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Abstract

Our progress is described in applying passive alpha track detectors (ATDs) and electret ion chambers (EICs) to in-situ field monitoring of soils contaminated with particulate plutonium and americium at a desert site. By varying the exposure times, from a few minutes to about one day, we quantitatively measured alpha activity varying between ~100 pCi/g and ~100 nCi/g. Suggested applications are definition of the boundaries of contamination zones and verification that post-remediated soils are below release limits.

Introduction

At the first Budapest symposium on Environmental Contamination in Central and Eastern Europe, we reported on our preliminary attempts to make passive, in-situ measurements of alpha activity in soils contaminated with plutonium. [1] A "safety shot" conducted in 1955, in an area known as 11B at the Nevada Test Site (NTS), caused the contamination. This communication reports on our efforts to make field measurements on soils with activities ranging over about three orders of magnitude. In particular, we are trying to find niches where the passive, integrating detectors can provide needed information not readily obtainable by other in-situ monitoring techniques.

Methods

The alpha track detector (ATD) and electret ion chamber (EIC) were described previously.[1] When making a surface measurement in the field, the soil surface was prepared by removing extraneous vegetation and rocks and them tamping the soil with the sole of one's boot to make a flat surface.

To deploy an ATD, the protective polyethylene film was peeled from one face and the exposed face placed face down against the soil. A wooden block was used to weight down the ATD. The block's function was to stop the small plastic ATD from shifting or blowing away, prevent baking in the sun, and give protection from rain. The wood also provided thermal mass and insulation for reducing moisture condensation, especially at night when the temperature falls below the dew point. The retrieved ATD was sequentially rinsed with water to remove adhering soil, then air dried, radiation checked, wrapped with polyethylene film, sealed in a Mylar bag and mailed to the manufacturer, Landauer, Inc., for processing. Readings were given as etch tracks per mm^2. The track generation rate is proportional to soil activity.

To deploy an EIC, the lower edge of the chamber portion was firmly pressed into the soil to obtain a seal. An electret was read for initial voltage and then screwed into the top of the seated chamber to initiate exposure. To

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terminate exposure, the electret was unscrewed and its voltage reread with a hand-held voltmeter. The EIC response is expressed as voltage drop per hour. The rate of voltage drop is proportional to soil activity.

The measurement periods depended on the contaminant activity at a particular location; exposures varied between 5 minutes and 3 days. Sets of 3 ATDs and 3 EICs were usually deployed at each location over an area of about $0.5m^2$. On some occasions, "screened" EICs were deployed at the same time. The screen was a closely fitting, circular Tyvek sheet placed on the ground inside the chamber to block alpha radiation coming directly from the soil. The Tyvek, however, is pervious to radon gas passing out of the soil. These "screened" measurements enable one to evaluate and correct for the effects of radon and radon daughter buildup in the EICs.

Field measurements to estimate background effects were also made on the ground adjacent to a car park about 1 km from the location of the safety shot to establish local background levels.

Detector calibration factors were established using soils collected along the contamination fenceline at Area 11B and returned to Oak Ridge National Laboratory for analyses. Processing and analytical procedures are discussed elsewhere. [2]

Results and Discussion

A schematic is shown in Fig. 1 of that portion of Area 11B where we made our measurements. The contours are isopleths of Pu-239 + Am-241 interpolated from in-situ gamma spectroscopy measurements conducted 25 feet above groundlevel;[3] the $^{239}Pu/^{241}Am$ ratio is 6.9. The dotted line is the fenceline of the contamination zone. The solid points 01 to 09 are positions along the fenceline where we made in-situ alpha measurements and collected soil samples. The grid points, E_2, E_3, -- G_4, etc., are surveyed positions.

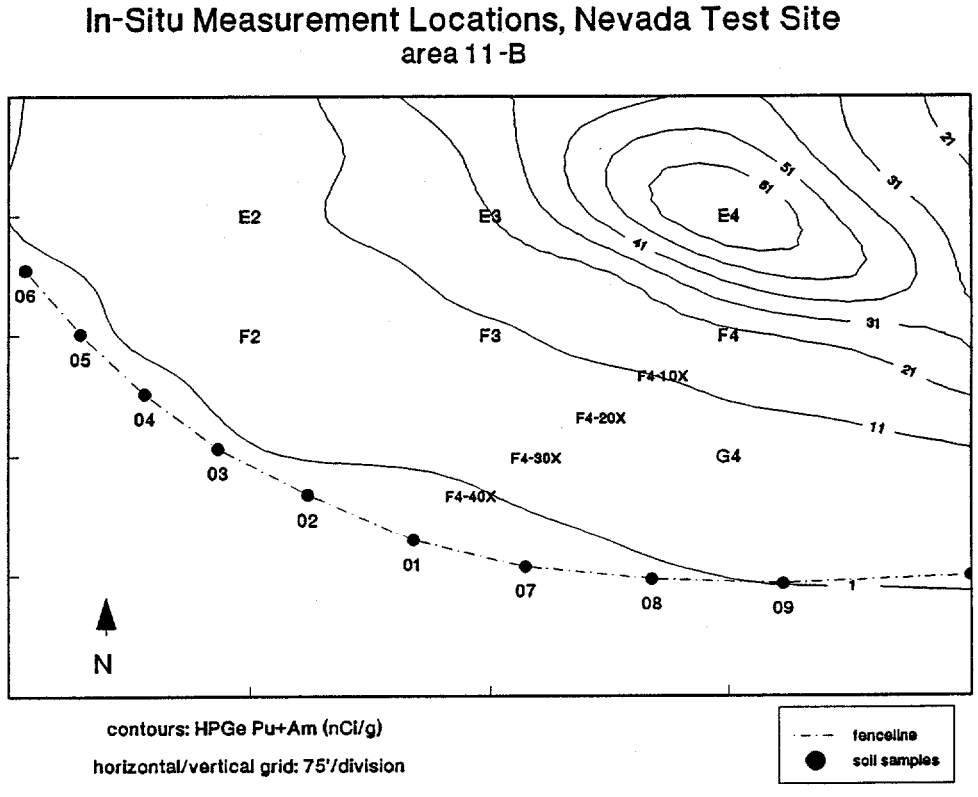

Fig. 1. Schematic of Area 11B at the Nevada Test Site. Contours are surface Pu + Am contamination levels interpolated from in-situ gamma spectroscopy measurements.[3]

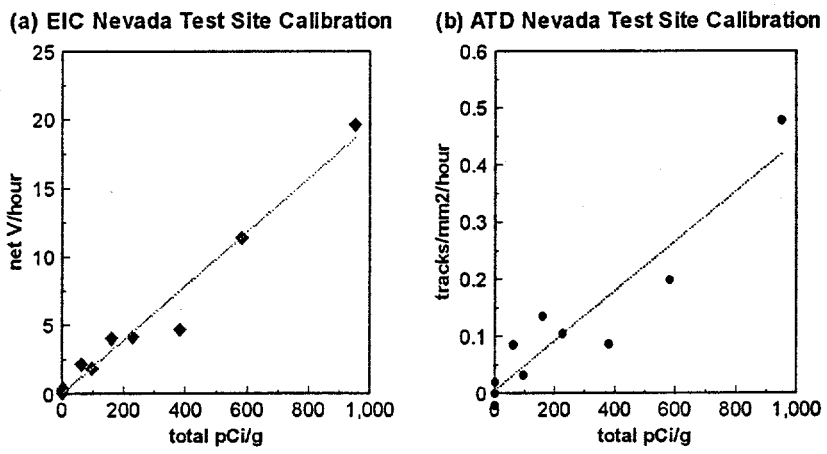

Fig. 2. Laboratory calibration curves obtained on processed NTS soils.

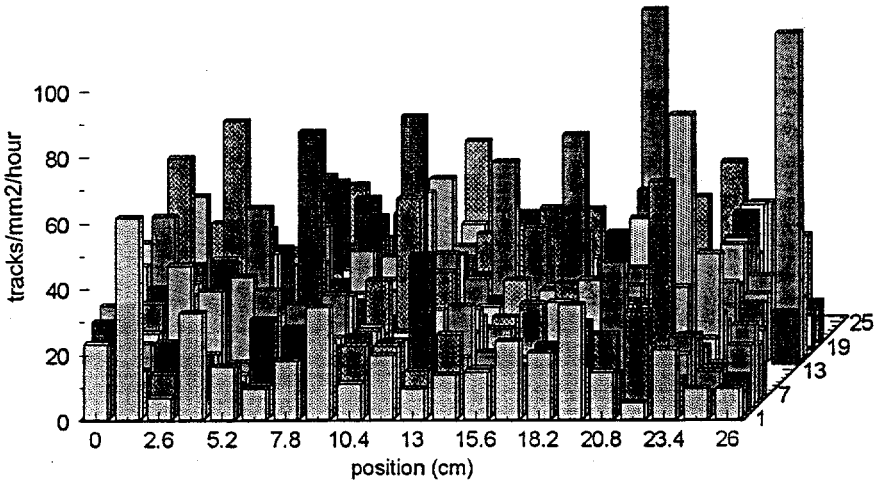

Fig. 3. Spatial inhomogeneity in surface alpha activity encountered at position F4.

The calibration curves for the EIC and ATD devices are shown in Figs. 2a and 2b, respectively.

Total activities were determined by radiochemical analyses for Pu-239, Pu-240, and Am-241. The soils were very inhomogeneous in alpha activity. Even after milling each soil for several hours, and then analyzing ten 0.5 g aliquots, standard deviations ranged from 5% to 29%.

The sub-cm scale inhomogeneity that we encountered during surface measurements is demonstrated in Fig. 3. This particular in-situ measurement was made by placing a 25 cm x 25 cm sheet of ATD face down at position F4 for an hour. Within this small area, alpha activity varies by more than ten-fold.

Because the alpha activity is so variable on a small (sub-cm) scale, one anticipates that in-situ measurements with different types of devices, that are placed side by side at a particular location, will indeed give substantially different results. This prediction is borne out by comparing the in-situ field measurements listed in Table 1.

Table 1. Comparison of surface contamination measurements.

Position	EIC nCi/g	ATD nCi/g	Gamma Spectroscopy nCi/g	Radiochemical nCi/g
E4	60.4 ± 18.9	263 ± 95	92.4	---
F4	26.1 ± 5.7	76.0 ± 21.0	24.6	---
F4-10X	9.3 ± 3.0	24.9 ± 20.5	17.6	---
F4-20X	6.5 ± 3.9	12.± 7.3	8.2	---
F4-30X	1.8 ± 0.4	6.1 ± 5.5	3.5	---
F4-40X	0. 89 ± 0.79	3.6 ± 4.2	1.2	---
fenceline 01	0.16 ± 0.04	0.25 ± 0.10	---	0.08
02	0.34 ± 0.13	0.27 ± 0.31	---	---
03	0.25 ± 0.13	0.34 ± 0.22	---	---
04	0.89 ± 0.21	1.76 ± 1.37	---	0.77
05	0.12 ± 0.05	0,28 ± 0.27	---	0.16
06	0.13 ± 0.07	0.29 ± 0.13	---	---
07	0.14 ± 0.08	0.26 ± 0.14	---	0.38
08	0.38 ± 0.24	0.37 ± 0.16	---	---
09	0.32 ± 0.14	0.44 ± 0.34	---	0.23

In several instances, the variation is close to 100% for in-situ measurements with devices of a particular type that are placed side by side. This variability is again attributable to spatially inhomogeneous contamination. The variability in measurement can be nearly eliminated if repeat measurements are made at the same spot without disturbing the detector's position on the soil. This can be done, for example, by leaving the chamber of the EIC

in place on the soil and making repeat measurements by screwing in and then unscrewing only the electret unit. The readings are then reproducible with a variability of less than 5%.

Intercomparison of results between the different methods (Table 1) indicate that at particular locations, two- or three-fold differences in alpha activity are common within an area of about $0.5m^2$. Again this variability does not surprise us, given the aforementioned spatial inhomogeneity. There is, however, an indication of bias to higher alpha activity measured by the ATD technique. The cause may be linked to much of the alpha contamination being in particulate form; [1,2] the ATD technique is currently optimized to read single tracks rather than clusters of etch tracks deriving from hot particles. We are currently trying to improve our ability to count tracks that appear in clusters.

Conclusions

Two types of passive monitors are useful for making in-situ surface measurement on desert soils contaminated with plutonium and americium. Spatial inhomogeneities on a small scale (sub-cm) can be detected. Exposure times of one day permit one to quantify alpha activities of 100 pCi/g or less. The passive monitors offer an inexpensive means of defining the boundaries of a contamination zone. In a post-cleanup situation, passive monitoring will be a convenient way of verifying that residual soils are as free of alpha activity and hot particles as they need to be.

References:

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[3] Colton, D., "An In Situ Radiological Survey of Area 11 Nuclear Safety Test Locations, Sites A, B, and C, Nevada Test Site, Central Nevada, Conducted May 4 through June 10, 1992," EG&G/EM letter report to U.S. Department of Energy Nevada Operations Office; NRD-93-330 (1993).