# A COMPREHENSIVE APPRAISAL OF $^{241}$AM IN SOILS AROUND ROCKY FLATS, COLORADO

M. Iggy Litaor* and L. Allen†

*Abstract*—Soils east of Rocky Flats Plant (RFETS) near Golden, Colorado, were contaminated with actinides because of accidental release of oils laden with plutonium isotopes. Consequently, these soils were contaminated by $^{241}$Am due to radioactive decay of $^{241}$Pu ($t_{1/2}$ = 14.4 y). A spatial analysis of $^{241}$Am activity in soils east of RFETS was conducted to elucidate the magnitude and the mode of $^{241}$Am dispersion in the soil environment. $^{241}$Am activity of 178 soil samples ranged from 0.037 Bq kg$^{-1}$ to 10,004 Bq kg$^{-1}$ with a mean of 214 Bq kg$^{-1}$, median of 7.03 Bq kg$^{-1}$, standard deviation of 947 Bq kg$^{-1}$, and a coefficient of variation of 4.3. Spatial analysis of $^{241}$Am in soils around RFETS was conducted using indicator kriging, which is a nonparametric technique especially suitable to model a conditional cumulative distribution function (ccdf) of highly skewed environmental data such as $^{241}$Am. The ccdf was used to generate an E-type (mean of the conditional cdf) surface. The resulted surfaces were consistent with the hypothesis that the westerly winds were the dominant mechanism of americium dispersal. The spatial distribution and dispersal mechanisms of $^{241}$Am were similar to those of $^{239+240}$Pu. The ccdf was also used to construct probability of exceedence maps of $^{241}$Am in soils. For the purpose of this report two threshold values for the probability maps were selected: (1) the mean measured background activity of $^{241}$Am (0.4 Bq kg$^{-1}$), and (2) the programmatic preliminary remediation goal for residential occupancy scenario (87.7 Bq kg$^{-1}$). The probability-of-exceedence maps provide estimates of spatial uncertainty associated with each threshold. The E-type maps in conjunction with the probability-of-exceedence maps provide a robust framework for future cleanup options and land use decisions.
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Key words: soil; $^{241}$Am; plutonium; contamination

## INTRODUCTION

ACTINIDE CONTAMINATION of surface soils at Rocky Flats Environmental Technology Site (RFETS), near Golden, Colorado, resulted from leakage of plutonium-contaminated oils from drums stored in an outside storage area. The magnitude and the mode of plutonium dispersion in the soil environment was discussed by Krey and Hardy (1970), Seed et al. (1971), Little et al. (1980) and more recently by Litaor (1995a, b) and Litaor et al. (1995). Weapons grade plutonium processed at RFETS was reported to have isotopic composition of 0.04% $^{238}$Pu, 93.3% $^{239}$Pu, 6% $^{240}$Pu, 0.58% $^{241}$Pu, and 0.04% $^{242}$Pu (Krey and Krajewski 1972; Martell 1975). The initial $^{241}$Am activity in the weapons grade plutonium processed at RFETS did not exceed $10^{-4}$% (Krey et al. 1976). Consequently, nearly all the $^{241}$Am activity in the soil around RFETS resulted from radioactive decay of $^{241}$Pu ($t_{1/2}$ = 14.4 y) to $^{241}$Am.

The physicochemical characteristics of $^{241}$Am in the environment are markedly different than those of $^{239+240}$Pu. Fowler and Essington (1974) ascertained that americium is more soluble than plutonium and may become the radionuclide of prime concern because it has a faster migration rate in soils. Romney et al. (1985) showed that root uptake of $^{241}$Am by various plants was consistently greater than that of plutonium. $^{241}$Am exhibited a higher solubility than did $^{238}$Pu and $^{239+240}$Pu, as observed in rumen contents of cattle grazing on actinide-contaminated desert vegetation (Barth et al. 1985).

The effectiveness of wind transport mechanisms in spreading the actinides across the landscape may vary among different radionuclides. For example, $^{241}$Am was transported in the air across the Hanford site in different particle-sizes and reached maximum concentration at different heights than those of $^{239}$Pu (Sehmel 1978). Hence, the spatial distribution of $^{241}$Am in the soil environment at RFETS may be considerably different than that of plutonium isotopes.

Several studies assessed the spatial distribution and the total inventory of plutonium in soils around RFETS, although no studies have assessed the spatial distribution and inventory of $^{241}$Am in these soils. Human risk analysis was performed only with plutonium data. For example, Johnson (1981) modified the isopleth map of plutonium given by Krey (1976) to correlate plutonium activity in soils with cancer incidence rates in the Greater Denver area. Cancer potency comparison tables suggest that the carcinogenicity of $^{241}$Am is approximately equal to that of $^{239}$Pu for both inhalation and ingestion exposure routes (U.S. EPA 1992). Since $^{241}$Am activity in the soil environs of RFETS will reach its peak by the year 2033 (Krey et al. 1976), a comprehensive understanding of $^{241}$Am spatial distribution is essential for assessing potential human risk associated with surface soils con-

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taminated by $^{241}$Am. Hence, the objectives of the present study are to (1) assess the spatial distribution of $^{241}$Am in soils east of RFETS using robust geostatistical techniques, and (2) provide a measure of uncertainty to the spatial estimation of $^{241}$Am.

## MATERIALS AND METHODS

### Field sampling

The sampling of $^{241}$Am in soils east of RFETS followed the protocol of the Colorado Department of Public Health and Environment (CDPHE) (CDH 1989). This sampling protocol requires the collection of 25 equally-spaced subsamples, to be composited within a 4.05 ha area for americium analysis. The soil at each individual location was sampled with the CDPHE sampler, which was designed to obtain a sample from the upper soil surface 6.4 mm deep, and from an area 5-cm wide by 6-cm long. For this study, the southwest corner of each plot was located by survey and identified with an appropriately marked steel post. The 25 subsamples for the composite sample were located with a hand-held compass and tape measure, using the southwest corner as the starting point. Sampling of the top 6.4 mm of the soil may be difficult, especially in stony soils. The use of this technique was advocated by CDPHE because of the semi-arid conditions in eastern Colorado that increased the potential for wind-resuspension and subsequent inhalation of soil particles containing americium from the top soil.

The rational and density of sampling of $^{241}$Am in soils using the CDPHE sampler was similar to that of $^{239+240}$Pu (Litaor et. al. 1995). In summary, 118 plots within Rocky Flats boundary were sampled in 1991 (Litaor 1995b), whereas 60 offsite samples were taken during the summer of 1992 (Fig. 1). The optimal number of soil samples and the optimal distance between the plots off plantsite were determined using a sampling strategy algorithm for soil sampling suggested by

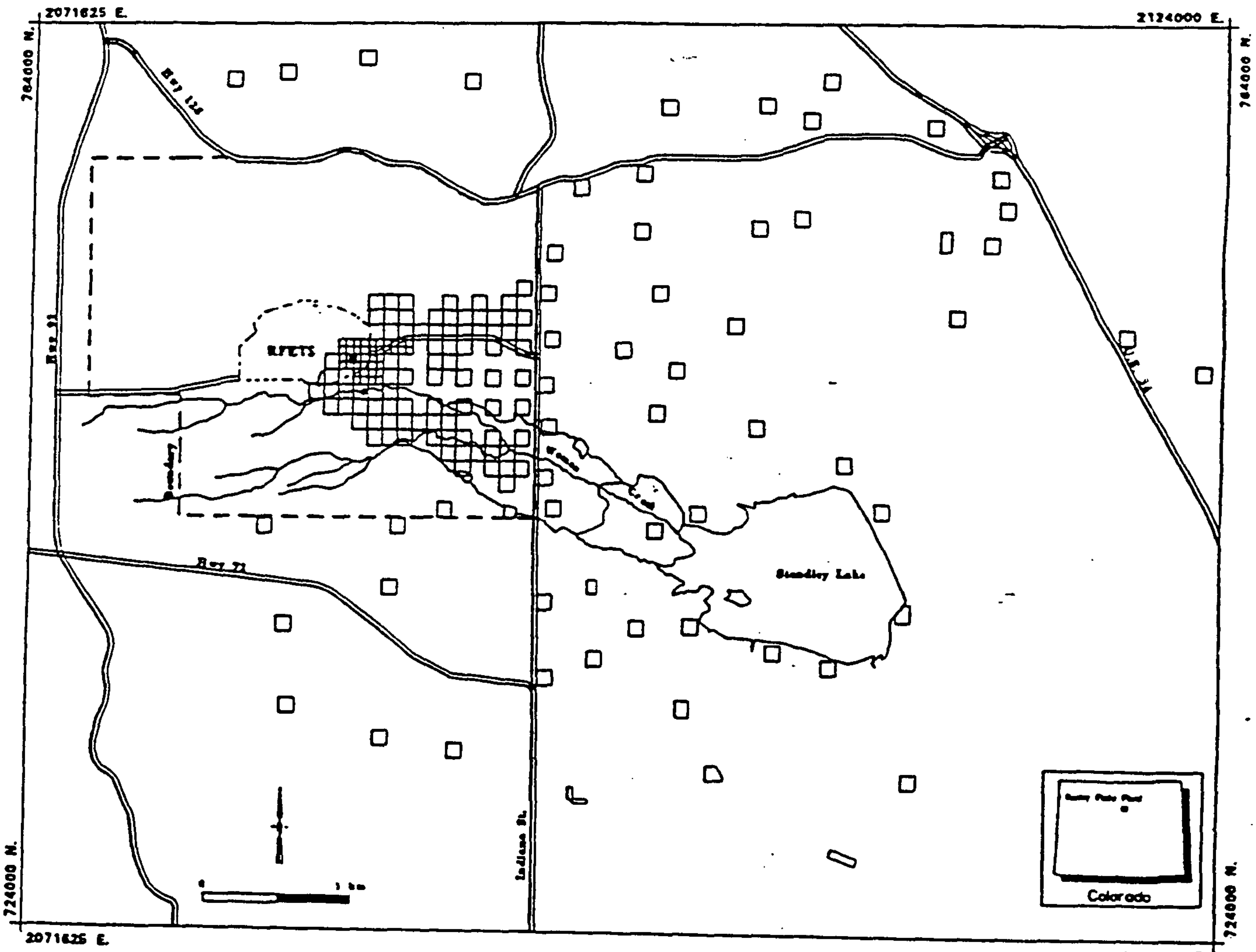


Fig. 1. The locations of soil samples. The former storage site (locally known as the 903 Pad) is depicted as black square. The dashed line represents the fenced boundary of RFETS.

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McBratney et al. (1981). Past soil sampling programs in areas east of RFETS provided sufficient plutonium information (120 soil samples) to compute the optimal sampling design (U.S. DOE 1992). In the absence of similar historical $^{241}$Am data, it was assumed that the optimal sampling design for plutonium will also provide an adequate sampling strategy for $^{241}$Am.

### Laboratory analysis

The $^{241}$Am activity in the soil samples was measured by alpha spectroscopy using various commercial laboratories (see Litaor et al. 1994 for details).

### Geostatistical approach

A determination of the spatial distribution of $^{241}$Am in the soil can be performed using geostatistical techniques such as kriging. Kriging is a generic name for a group of estimation techniques that design to minimize error-variance. Ordinary kriging (OK), which was recently used by Litaor (1995b) to estimate the spatial distribution of $^{239+240}$Pu and $^{241}$Am in soils within RFETS, is sensitive to strongly positively skewed distributions. This sensitivity may result in underestimation of the radionuclide in highly contaminated areas and overestimation in areas of low contamination. The most severe limitation of OK, however, is the difficulty of assessing the reliability and the uncertainty associated with its estimates. There are several geostatistical techniques that model the spatial uncertainty of a given contaminant in the environment rather than produce an "optimal" estimator. These techniques include indicator cokriging, probability, and indicator kriging. Indicator kriging (IK) was chosen for the present study because, as a nonparametric method, it is free from any distributional assumptions and resistant to a highly skewed distribution and outliers. Indicator kriging is also a faster and simpler procedure, compared to more elaborate techniques such as indicator cokriging and probability kriging, but has similar accuracy (Goovaerts 1994).

A detailed example of an IK analysis of $^{239+240}$Pu in soils around Rocky Flats was recently described by Litaor et al. (1995). A complete mathematical treatment and formalism of IK and the theory behind parametric and nonparametric spatial-estimation techniques can be found in Journel (1987), Isaaks and Srivastava (1989), and Deutsch and Journel (1992), among others.

The IK analysis was performed using GSLIB—the geostatistical software library and user's guide (Deutsch and Journel 1992). The original GSLIB software provided for data entry through parameter files entered in a specific format. Minimal internal documentation of parameter values and definitions was provided, and data entry with program parameter files was tedious and prone to execution errors. GSLIB did not provide an interactive display of the modeling results. Hence, a menu-driven windows interface was developed for GSLIB to facilitate the data entry, program execution, and display of data in tabular and graphical formats.

The spatial distribution of $^{241}$Am in the soil environment of RFETS was assessed according to the following steps:

1. A general exploratory data analysis in which univariate statistics was performed and the benefits of data transformation and declustering were assessed;
2. Selection of K cutoffs was performed using the calculated conditional cumulative distribution function (ccdf) of $^{241}$Am. The term conditional cdf means conditional to neighboring data values available for estimating $^{241}$Am activity in unsampled locations. It is customary to select 9 preliminary cutoffs, each representing a 1/10 of the data for variogram analysis. However, spatial data distribution of the ninth cutoff did not yield a meaningful experimental semivariogram. Hence, eight cutoffs were chosen corresponding to the first eight deciles of the ccdf. These 8 cutoffs split the ccdf into 9 intervals from which indicator variogram analysis was performed;
3. Indicator semivariograms were computed for these 8 cutoffs;
4. The accuracy of IK in estimating the ccdf at the 8 cutoffs was examined by a cross validation analysis. This analysis consisted of estimating a ccdf at a datum location where the $^{241}$Am activity was temporarily removed from the data set. This procedure repeated itself at all data locations. The true value at a given cutoff was compared against a computed value at that cutoff;
5. The ordinary IK algorithm was used to generate cumulative indicator functions and to compute the probability estimates for the unsampled area according to certain grid specifications. In general, a 5,500 m. search radius was used with a minimum of 4 and a maximum of 10 data points required to estimate a grid point. The ordinary IK equations system was solved for all the 8 cutoff values. This provided the uncertainty through the 8 selected discrete ccdf values; and
6. The ccdf for any required quantile or probabilities of exceeding a threshold value of interest (e.g., background $^{241}$Am activity) and the E-type estimate (mean of the ccdf) were computed; The ccdf of $^{241}$Am in the soils around RFETS showed large positively skewed distribution (see below), thus, the upper tail of this ccdf was calculated using a hyperbolic model.

## RESULTS AND DISCUSSION

### Exploratory analysis

Americium activity in soils east of RFETS ranged from 10,004 Bq kg$^{-1}$ near the former storage site to 0.037 Bq kg$^{-1}$ at the far southeast corner of the sampling area. The mean activity of $^{241}$Am was 214 Bq kg$^{-1}$, median of 7.28 Bq kg$^{-1}$, with a standard deviation of 942 Bq kg$^{-1}$, a coefficient of skewness of 7.98 and a kurtosis of 74.8. The effect of few outliers on the mean and the variance of the ccdf can be clearly seen in Fig. 2. The positively skewed distribution justified the use of nonparametric spatial estimation techniques such as IK to model the spatial uncertainty of $^{241}$Am in the soil environs.

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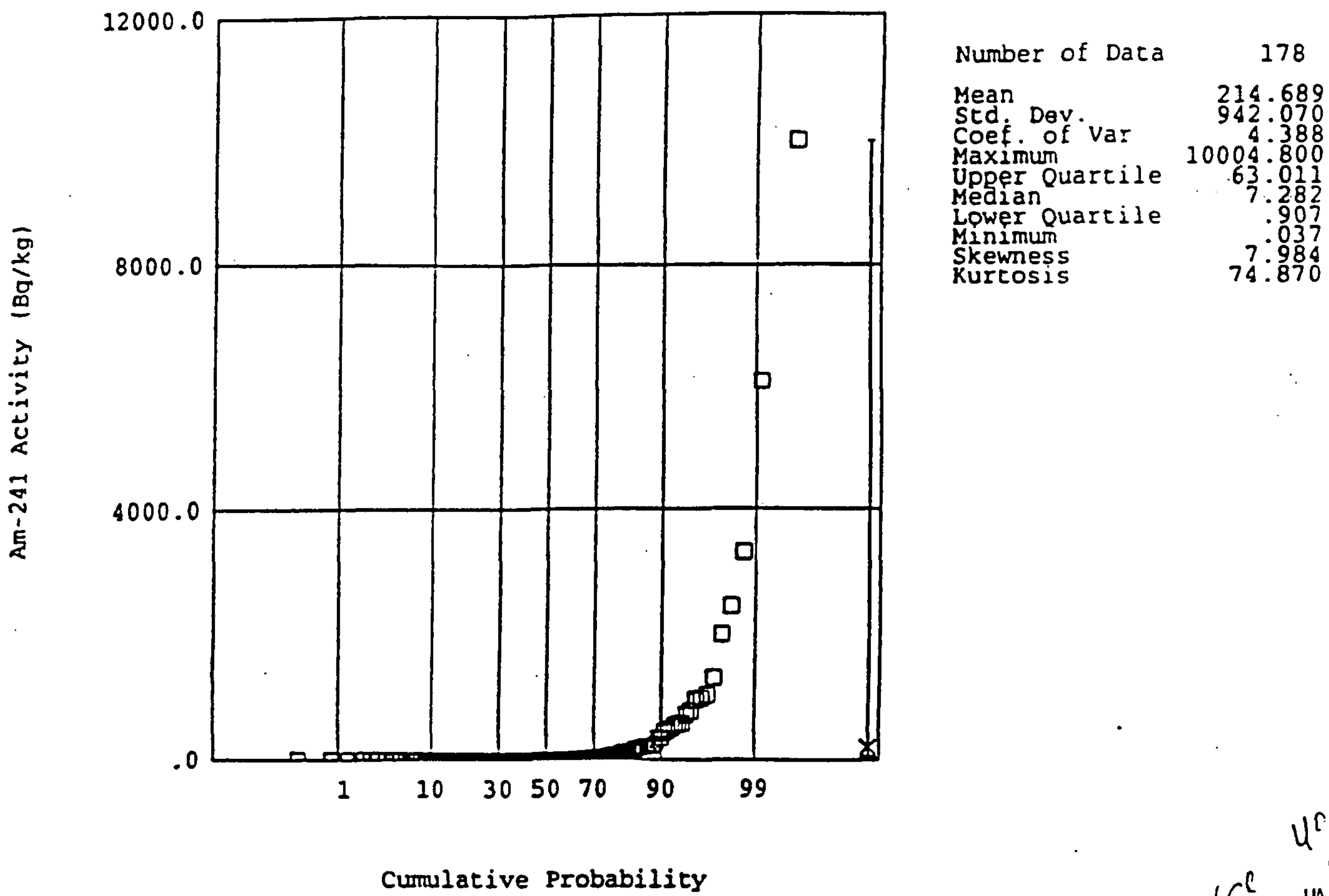


Fig. 2. The cdf of $^{241}$Am activity (Bq kg$^{-1}$) in soils around RFETS.

## Variography

The ccdf depicted in Fig. 2 provided 8 cutoffs from which 8 indicator variograms were modeled. The eight indicator variograms with their models' parameters are summarized in Table 1 and illustrated in Fig. 3. The spatial structure of $^{241}$Am in the first four and the last cutoffs were best described by a power model:

$$\delta(h) = ch^{a} \quad (1)$$

where $\delta(h)$ is the variance, $c$ is a positive slope coefficient, $a$ is the power that bound between $0 > a > 2$, and $h$ is the lag interval. The fifth to seventh cutoffs were best described by a spherical model:

$$\delta(h) = Co + C[1.5(h/Ao) - 1.5(h/Ao)^3] \quad \text{for} \quad h \leq Ao,$$
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where $Co$ is the nugget variance, $C$ is the structural variance, also known as sill, and $Ao$ is the range parameter. The excellent fit of the models to the experimental indicator variograms (Fig. 3) is a clear indication to a strong spatial continuity which is characteristics to contaminant dispersion by wind.

**Table 1.** Indicator variogram models.

| Decile | Cutoff (Bq kg$^{-1}$) | Model | $C_o$[a] | $C$ | $A$ |
|---|---|---|---|---|---|
| 0.10 | 0.37 | Power | 0.001 | 1.0E-8 | 1.66 |
| 0.20 | 0.74 | Power | 0.001 | 1.0E-6 | 1.29 |
| 0.30 | 1.48 | Power | 0.001 | 1.0E-6 | 1.35 |
| 0.40 | 3.33 | Power | 0.001 | 3.0E-4 | 1.00 |
| 0.50 | 7.40 | Spherical | 0.06 | 0.27 | 12,000 |
| 0.60 | 18.1 | Exponential | 0.07 | 0.27 | 3,200 |
| 0.70 | 35.9 | Spherical | 0.03 | 0.28 | 3,000 |
| 0.80 | 107.6 | Power | 0.08 | 6.0E-5 | 1.0 |

[a] Where $C_o$ is the nugget effect, $C$ is the variance component if spherical model, or positive slope if power model, and $A$ is the range in meters if spherical model, or power in the power model.

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Am-241 in soils Around RFETS

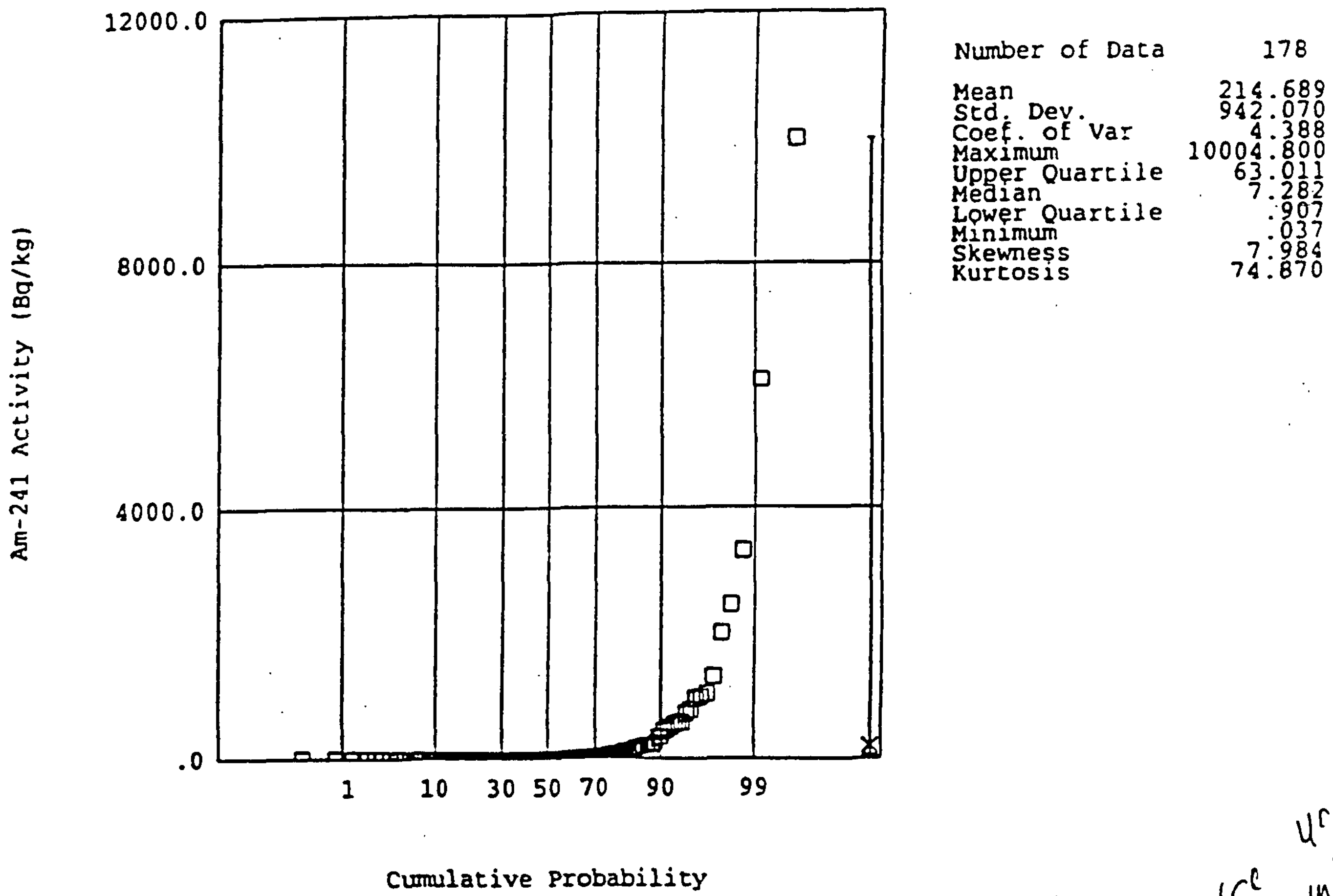


Fig. 2. The cdf of $^{241}Am$ activity (Bq $kg^{-1}$) in soils around RFETS.

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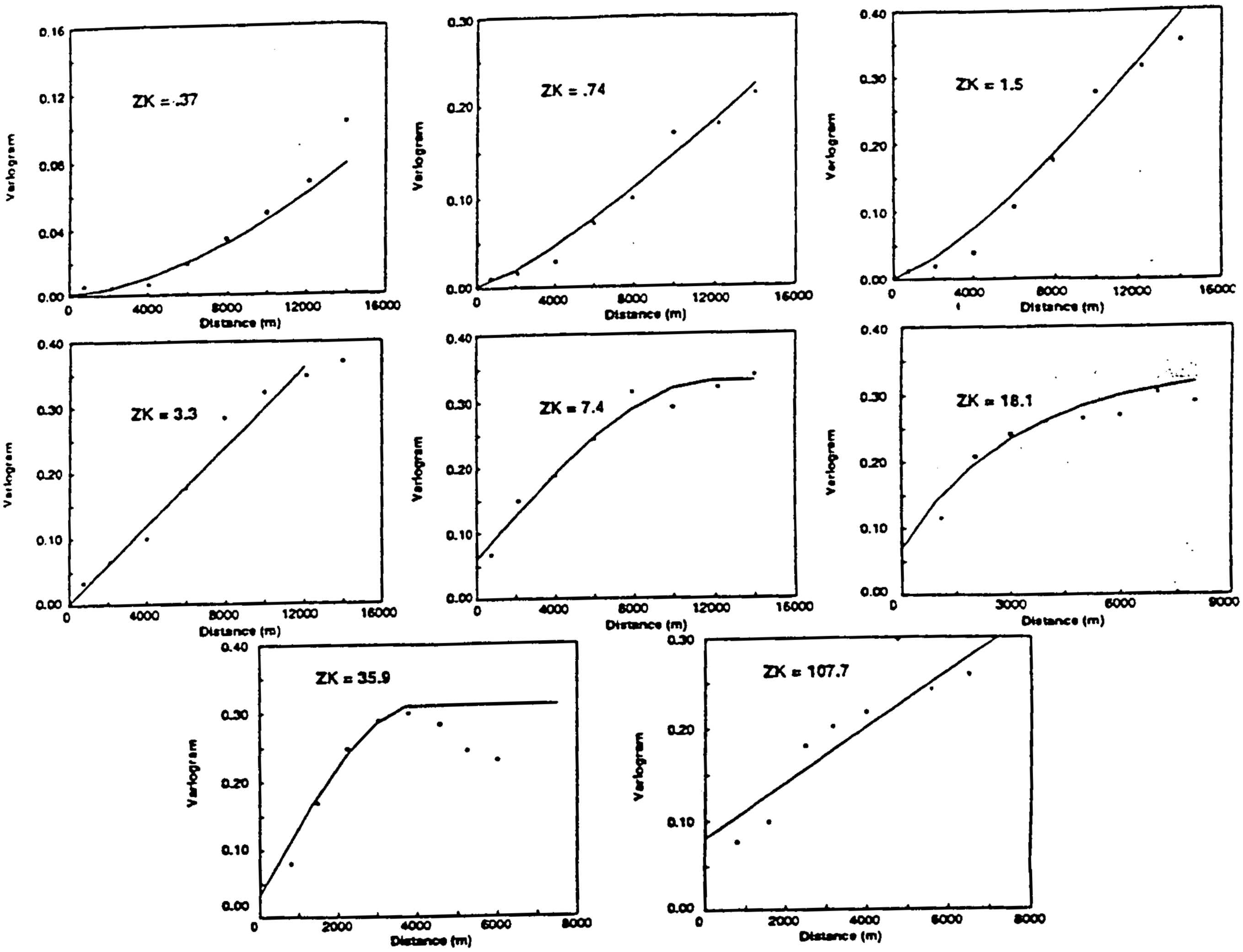


Fig. 3. Indicator variograms and models for the 8 cutoffs.

Table 2. Indicator variogram model validation. The values in the first column represent percentage of the ccdf, the actual and estimate are dimensionless.

| CDF | Cutoff (Bq kg$^{-1}$) | Actual | Estimate |
|---|---|---|---|
| 0.1 | 0.37 | 0.09 | 0.10 |
| 0.2 | 0.74 | 0.197 | 0.215 |
| 0.3 | 1.48 | 0.292 | 0.310 |
| 0.4 | 3.33 | 0.393 | 0.413 |
| 0.5 | 7.4 | 0.500 | 0.483 |
| 0.6 | 18.1 | 0.601 | 0.585 |
| 0.7 | 35.8 | 0.697 | 0.703 |
| 0.8 | 107.6 | 0.798 | 0.703 |

## Cross validation analysis

The indicator variograms model parameters were tested using the cross-validation technique. Indicator values at each cutoff were kriged and the mean of both actuals and estimates computed. Each cutoff represents a point on the underlying ccdf, thus the mean estimated value should be favorably compared with the known mean at that cutoff. For example, if the first cutoff represents the 10% point of the ccdf, the mean of the actual and estimated indicators should be approximately 0.10. Significant deviation from the underlying ccdf would suggest a problem with the modeling strategy. The validation results using isotropic models described in Table 1 and Fig. 3 are summarized in Table 2. The validation results for all data sets indicated that the models adequately represent the underlying cdf. Cross validation analysis conducted on anisotropic models did not improve the estimation results, thus the isotropic indicator variograms were used in the IK analysis.

## E-type estimate of $^{241}$Am in soils east of RFETS

The E-type estimate surfaces of $^{241}$Am, which is the mean value of the cdf at each specified grid point, is depicted in Fig. 4. The E-type estimate E[z'(u)], is considered the closest possible to the true value Z(u) (Goovaerts 1994). The E-type estimates of $^{241}$Am activity showed a clear west-east trend. This trend is characterized by high values near the former storage area with

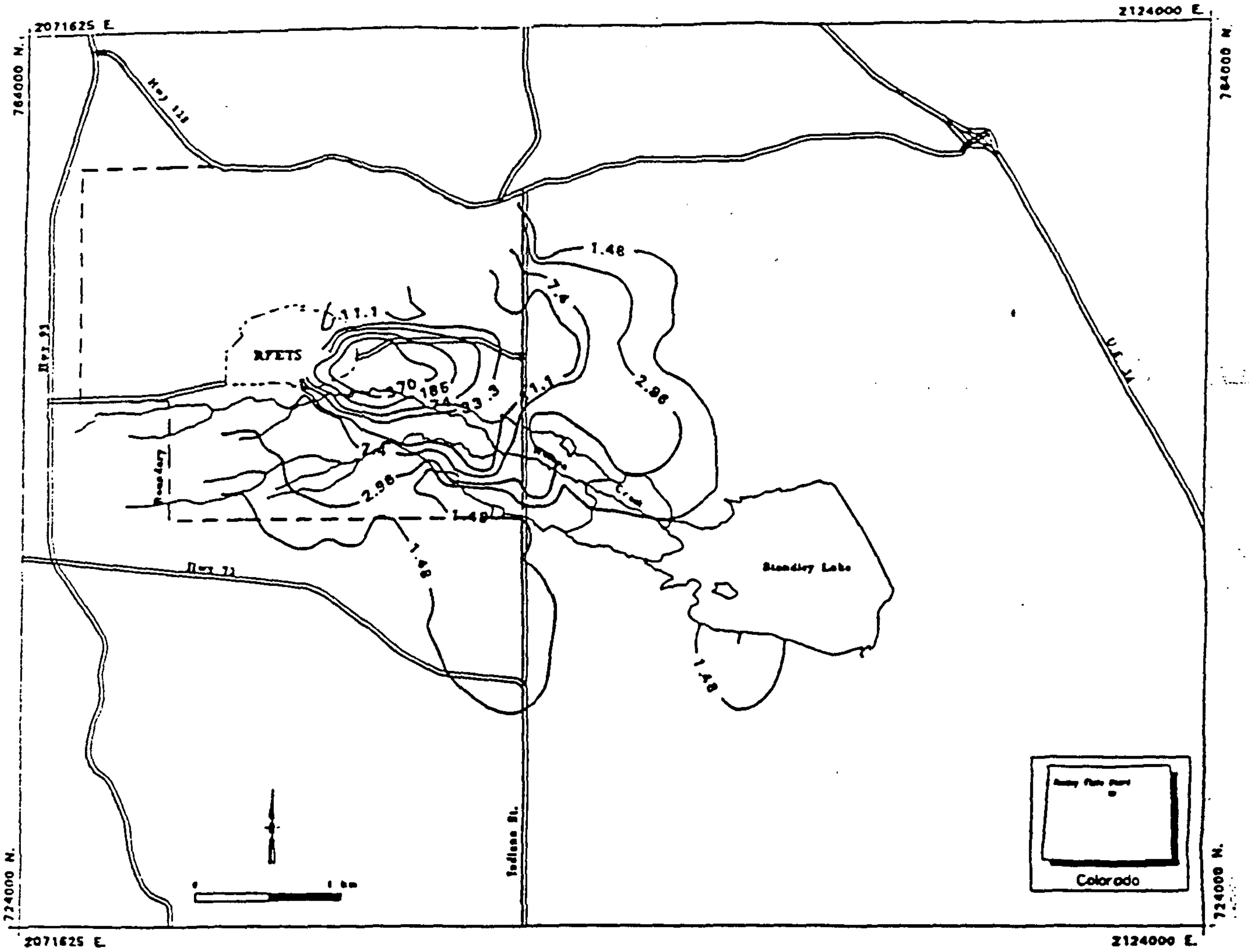


Fig. 4. E-type estimate of $^{241}$Am. The dashed line represents the fenced boundary of RFETS.

a rapid decline towards the eastern plant boundary and the residential areas east of Indiana Street. The $^{241}$Am activity in the soils decreased rapidly in the north and south directions. This pattern reflected wind dispersion consistent with the prevailing winds at RFETS.

The spatial distribution of $^{241}$Am (Fig. 4) is in excellent agreement with the size and *direction* of recently published E-type estimate of $^{239+240}$Pu (Litaor et al. 1995). There was no southeast plume in the observed spatial distribution of $^{239+240}$Pu (Litaor et al. 1995) or $^{241}$Am (Fig. 4). Krey and Hardy (1970) and Krey (1976) constructed plutonium isopleth maps that showed a clear southeast plume from RFETS towards the Denver area. The isopleth map of Krey (1976) was the basis of the cancer incidence assessment for the Denver area (Johnson 1981). Litaor (1995b) questioned the existence of the southeast plume; however, Hardy and Krey (1995) challenged his interpretation and strongly attested for the existence of the plume. Litaor et al. (1995) demonstrated that the southeast plume resulted from the use of extremely small and sparse data sets by Krey and Hardy (1970) and Krey (1976). Assuming that eaolian transport was the dominant process in the dispersion of actinides in the environment, the absence of the southeast plume in the E-type estimates of $^{241}$Am as depicted in Fig. 4 reaffirmed our plutonium studies.

## Probability of exceeding a threshold value

Several maps of conditional probabilities were generated to provide areas of uncertainty around the isopleths of the E-type estimates. To create these maps, two threshold values were selected; the first value represents a background level of $^{241}$Am in the Denver area, and the second is the programmatic preliminary remediation goal (PPRG) value.

## Background level of $^{241}$Am

Fifty soil samples from undisturbed areas along the Front Range of Colorado were collected to assess the background level of $^{241}$Am (U.S. DOE 1995a). The 50 locations ranged from 12 km to 170 km away from RFETS. All locations were upstream and off the main wind trajectories characteristic to RFETS and thus assumed to be unaffected by the site's activities. The background level of $^{241}$Am in soils is assumed to represent the global-fallout of $^{241}$Pu that was originated from atmospheric testing of nuclear weapons. Background activity of $^{241}$Am in Colorado ranged from 1.14 to 0.037 Bq kg$^{-1}$ with a mean of 0.4 Bq kg$^{-1}$ and standard deviation of 0.22 Bq kg$^{-1}$. Based on the locations of the background soils and the tight spread around the center of the distribution (Fig. 5), it was assumed that the arithmetic mean of $^{241}$Am represents a reasonable background threshold value for the Denver area.

The probability map for the background concentrations of $^{241}$Am clearly demonstrates the large uncertainty associated with the E-type estimate isopleths (Fig. 6). Areas within 4 km radius east of the plant's outer boundary (i.e., Indiana Street), exhibited a greater than 80% probability of exceedance the mean global-fallout americium. However, areas, only 7 km east of the outer boundary exhibited a less than 20% probability of exceedance the mean global-fallout americium. Similar pattern was observed for $^{239+240}$Pu (Litaor et al. 1995). Johnson (1981) fitted the censor tracts of cancer incidence in the Denver area within isopleths of plutonium-contaminated soils without taking into consideration the spatial uncertainty around each isopleth. It is conceivable that, had he applied a spatial uncertainty analysis to his epidemiological study, the implied linkage between the cancer incident rates and RFETS' plutonium may have been an artifact of his research design. Hence, the uncertainty depicted in Fig. 6 must be taken into account when attempting to correlate the potential environmental and human health risks from RFETS-derived americium on the Greater Denver area.

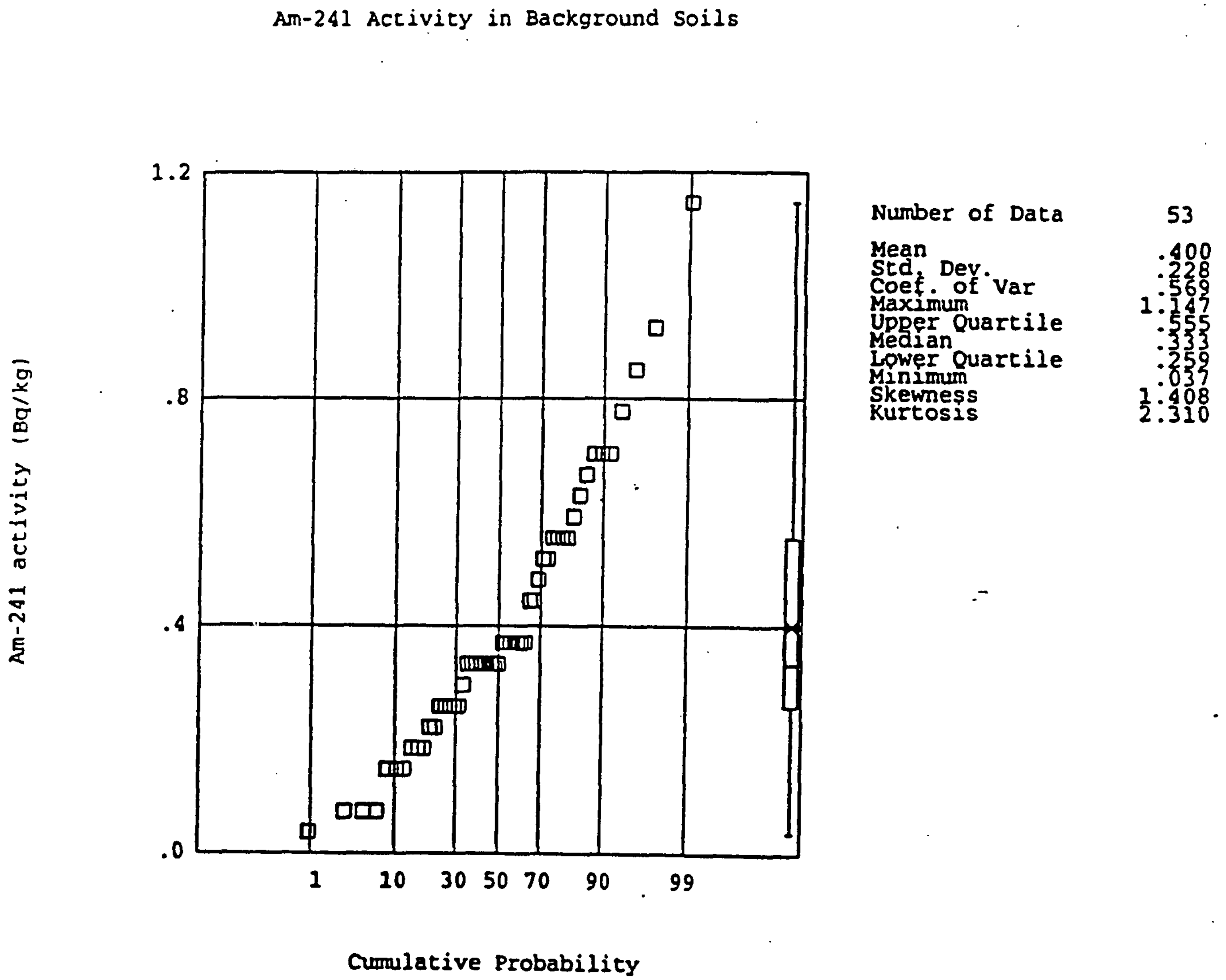


Fig. 5. The cdf of $^{241}$Am activity (Bq kg$^{-1}$) in soils that represent background locations.



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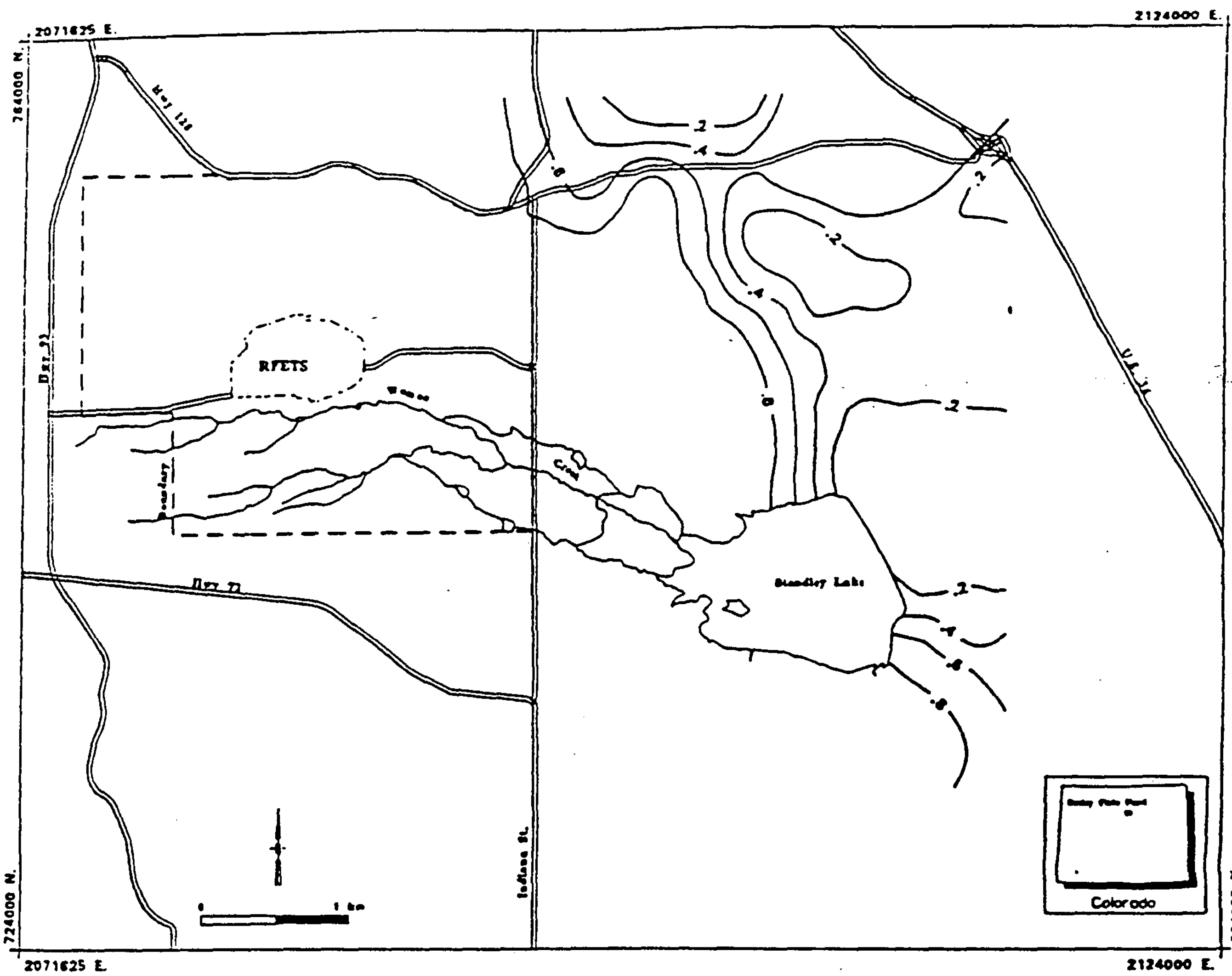


Fig. 6. A contour map of the probabilities that the unknown $^{241}$Am activity exceeds the background concentration of ~~0.37~~ Bq kg$^{-1}$. The dashed line represents the fenced boundary of RFETS.

=> 0.4

## Cleanup options for environmental restoration

Risk-based programmatic preliminary remediation goals (PPRG) were computed for radionuclides in soils in RFETS using an annual radiation dose limit of 0.001 Sv to offsite members of the public (U.S. DOE 1995b). A PPRG of 87.7 Bq kg$^{-1}$ of $^{241}$Am in soil was computed to meet the stringent requirements of a residential scenario. A map showing the probability of exceedance of the computed PPRG for residential scenario at RFETS has been produced (Fig. 7). This map delineates the areas that exceed the PPRG value at a given probability (shown 80 to 20% probability). Any probability of exceedance $[1 - \alpha(u)]$ can be computed using the following relationship:

$$1 - \alpha(u) = \mathrm{Prob}[\mathrm{Am}'(u) > 87.7 \,|\, (n)], \qquad (3)$$

where $u$ is the location of the data, $|(n)$ is the available information, and $\mathrm{Am}'(u)$ is the estimated $^{241}$Am in the unsampled location.

Using the available $^{241}$Am information, a significant portion of the land (358,801 m$^2$ at 80% probability, and 799,539 m$^2$ at 60% probability) east of the former storage site within the buffer zone of RFETS would need to be remediated if meeting the residential scenario requirement is the land use decision. These results also demonstrated the significant increase in the size of the remediated area if the more conservative requirements for the residential scenario (i.e., 40 or 20% probability) is selected.

## $^{241}$Am:$^{239+240}$Pu ratio

The mean $^{241}$Am:$^{239+240}$Pu activity ratio in the 178 soil samples was 0.319 with a standard deviation of 0.531. This ratio did not agree with an earlier work by Litaor (1995b) who sampled 118 soil samples within the plant boundaries and reported a ratio of 0.19 (Table 3). Significantly larger activity ratios were calculated for off plant locations. For example, the *lower quantile* of the $^{241}$Am:$^{239+240}$Pu-activity ratios calculated from 60 soil samples taken from off site locations exhibited similar value to the *mean* activity ratio calculated from samples

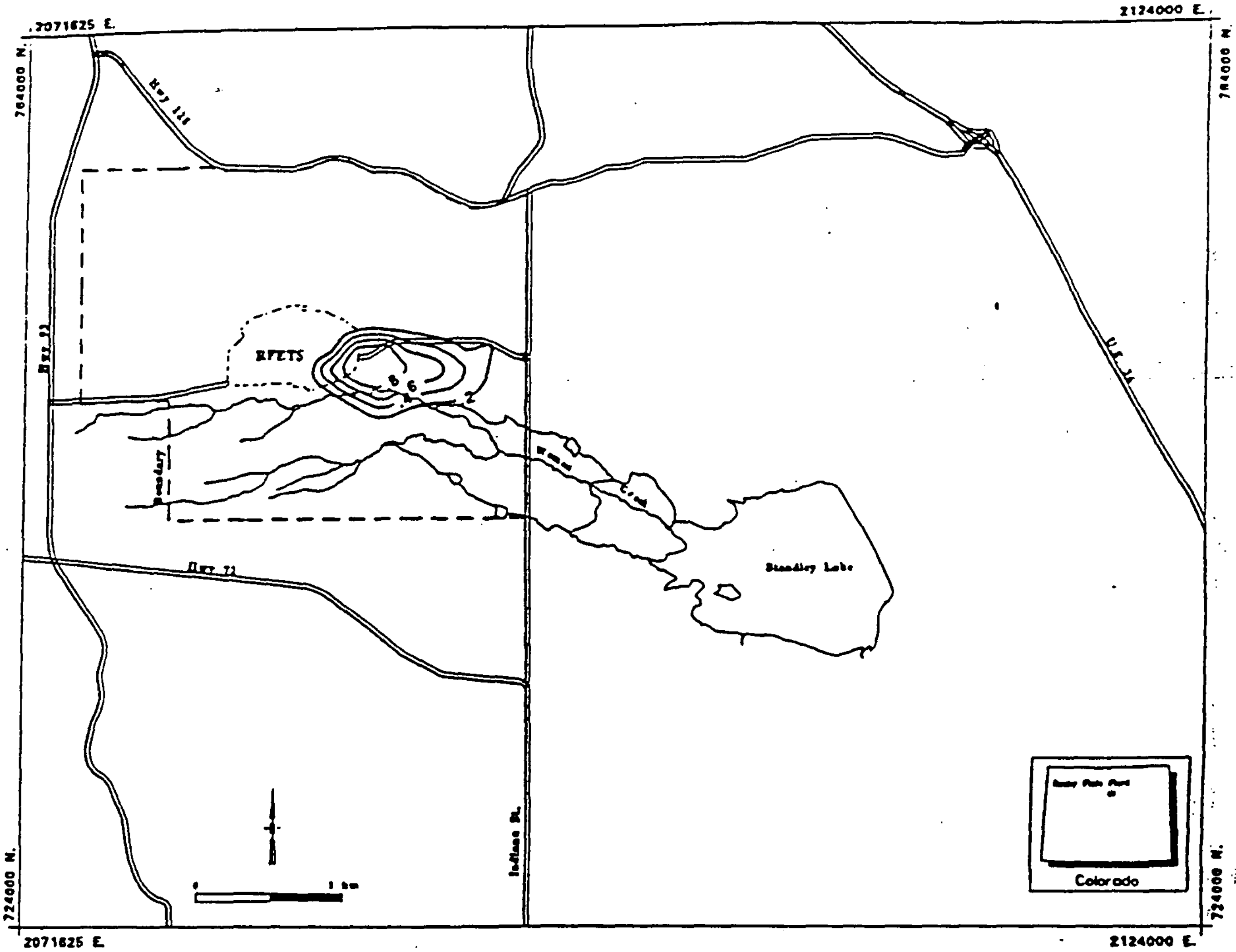


Fig. 7. A contour map of the probabilities that the unknown $^{241}$Am activity exceeds the PPRG of 87.7 Bq kg$^{-1}$.

Table 3. $^{241}$Am:$^{239+240}$Pu ratios observed in off- and on-site samples.

| Statistics | $^{241}$Am:$^{239}$Pu off- and on-site locations | $^{241}$Am:$^{239}$Pu off site samples | $^{241}$Am:$^{239}$Pu on site samples |
|---|---|---|---|
| N | 178 | 60 | 118 |
| Mean | 0.31 | 0.56 | 0.19 |
| Standard deviation | 0.53 | 0.84 | 0.12 |
| Coef. of variance | 1.66 | 1.50 | 0.63 |
| Maximum | 5.76 | 5.76 | 1.12 |
| Upper quantile | 0.29 | 0.54 | 0.20 |
| Median | 0.19 | 0.36 | 0.16 |
| Lower quantile | 0.14 | 0.18 | 0.14 |
| Minimum | 0.00 | 0.00 | 0.02 |
| Skewness | 5.75 | 3.81 | 2.0 |
| Kurtosis | 46.1 | 18.8 | 6.51 |

taken within the plant boundaries (Table 3). From parent-progeny radioactive decay relationships, the history of the site, the isotopic composition inside the stored barrels, the initial amount of plutonium released, and number of years since the initial release, Litaor (1995b) calculated that in 1992 the $^{241}$Am:$^{239\text{-}240}$Pu activity ratio in soils within the plant boundaries should have been 0.17, which agreed well with the 0.19 mean ratio measured in 1991 (see Table 3). The source of the apparent discrepancy between off-site and within plant boundary locations originated from the analytical uncertainty associated with measuring low actinide activities close to the detection limits and possibly preferential eolian transport processes for plutonium and americium over the study area. In general, the analytical errors associated with americium determination were significantly higher than those with $^{239+240}$Pu, regardless of sampling locations (Table 4). The analytical errors for americium and plutonium in samples collected off-site were significantly higher than those collected on-site (Table 4). Errors as high as 400% were recorded for samples taken off-site, with mean error of 82.5% for $^{241}$Am and 53% for $^{239+240}$Pu compared with mean error of 20.7% for $^{241}$Am and 15.6% for $^{239+240}$Pu in soil samples taken within the plant boundaries (Table 4). These large ana-

Table 4. Analytical errors associated with $^{241}$Am and $^{239+240}$Pu determination.

| Statistics | On-site samples Analytical error (%) | | Off-site samples Analytical error (%) | |
|---|---|---|---|---|
| | $^{241}$Am | $^{239}$Pu | $^{241}$Am | $^{239}$Pu |
| N | 118 | 118 | 60 | 60 |
| Mean | 20.7 | 15.6 | 82.5 | 53.1 |
| Standard deviation | 12.4 | 8.5 | 70.3 | 46.4 |
| CV | 0.59 | 0.54 | 0.85 | 0.87 |
| Maximum | 100.5 | 54.9 | 400 | 333.3 |
| Upper quantile | 25.4 | 18.7 | 89.9 | 60.2 |
| Median | 15.8 | 11.9 | 61.5 | 46.6 |
| Lower quantile | 12.8 | 10.7 | 44.7 | 29.3 |
| Minimum | 9.31 | 0.79 | 15.0 | 12.9 |
| Skewness | 2.95 | 1.64 | 2.75 | 4.1 |
| Kurtosis | 14.1 | 3.33 | 8.63 | 22.9 |

lytical errors strongly influence the magnitude of the isotopic ratio, thus caution should be exercised when attempting to use this ratio to ascertain the history of release of actinides in the environment (see Litaor 1995b).

An additional explanation to the observed isotopic ratios in the off-site soil samples may be a selective eolian transport process coupled with multiple source of release. Sehmel (1978) observed that $^{241}$Am was transported on smaller particles than $^{239+240}$Pu, and reached higher heights at the Hanford meteorological tower, thus it may travel larger distances. There is no clear evidence for this phenomenon around Rocky Flat, because the isotopic ratio data gleaned from air monitoring studies are inadequate. Systematic measurements of the isotopic ratio in effluents from the industrial area were performed in 1985 to 1989 (ChemRisk 1992). During these years the isotopic ratios observed in the buildings' effluent varied between 0.13 to 0.31. Illsley (1982) studied airborne matter in ambient air collected during 6 mo in 1978 and 1979 at four locations around Rocky Flats. He found that the mean $^{241}$Am:$^{239+240}$Pu ratio varied between 0.09 to 0.46 with a maximum of 1.7. All the anomalies in the expected isotopic ratio were recorded from an air sampler located immediately north of the industrial area that most likely received its actinide content from industrial effluents rather than resuspension of soil particulates. Indeed, resuspension studies east of the former storage site showed that the isotopic ratio derived from bare soil, grass blades, and soil litter varied between 0.07 to 0.2 (Langer 1984; 1986). On the basis of this limited air sampling information, the observed isotopic ratio in the off-site samples may have been influenced by preferential addition of chronically released $^{241}$Am from the industrial area of Rocky Flats. The magnitude of this addition, however, is probably negligible.

## CONCLUSION

This study has provided a comprehensive appraisal of the extent of $^{241}$Am in the soil environs east of RFETS. The major finding of this work was that the spatial distribution and dispersal mechanisms of $^{241}$Am were similar to those of $^{239+240}$Pu. The area adjacent to the former storage site is the most significantly contaminated with $^{241}$Am in spite of several soil removal operations (Barker 1982). The ccdf was used to generate an E-type estimate (mean of the ccdf) and probability-of-exceedence maps. These probability-of-exceedance maps will provide the background information required for selecting remedial actions and/or corrective measures for cleanup.

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