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Plutonium-239 + 240 and Americium-241 in Soils East of Rocky Flats, Colorado

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ABSTRACT

Soils east of the Rocky Flats (RF) near Golden, CO, were contaminated with Pu-239 + 240 and Am-241 as a result of past waste-storage practices The physicochemical parameters that govern the actinides distribution in the soil are poorly understood Twenty-six soil pits at various distances and directions from a contaminated site at RF were excavated, sampled, and analyzed for actinide activities as well as selected physical, chemical, and mineralogical attributes. Plutonium-239 + 240 and Am-241 activities in the soils ranged from 164 280 Bq/kg to 0.0037 Bq/kg, decreasing with distance from the source More than 90% of the Pu-239 + 240 and Am-241 activities were confined to the upper 12 cm of the soil, regardless of the soil characteristics, or distance and direction from the source Evidence of preferential transport in macropores formed along decayed root channels was observed in four soil pits and had translocated Pu-239 + 240 to a depth of 90 cm This transport mechanism increased by a factor of 30 the level of Pu-239 + 240 activity at this depth Earthworm activity is probably important in the redistribution of actinides in the upper 40 cm of many of the soils investigated Planning of future remedial activities at RF should consider the findings of this contaminant-transport study

Most Pu-239+240 contamination of the soils near the RF was caused by leaking drums of Pu-contaminated oil stored at a former storage site (Krey and Hardy, 1970) Surficial soils in the area east of the former storage site (locally known as the '903 Pad') were contaminated with Pu due to wind dispersal of soil particles during cleanup operations (Krey and Hardy, 1970) Many studies have assessed the spatial distribution of actinides around RF, however, only two studies investigated the vertical distribution and migration of Pu-239+240 in soils at RF In the context of the present study, the term actinides refers only to the most significant transuranic contaminants in the soil of RF (i e , Pu-239+240 and Am-241) Little and Whicker (1978) found that Pu-239+240 activity in soils east of the former storage site increased with decreasing particle size For all depths, sampled in seven increments of 3 cm each, the highest Pu-239+240 activity was associated with submicron size soil particles Two-thirds of the total Pu was found in the top 5 cm of the soils The relationship between Pu-239+240 activity and soil particle-size distribution suggests that Pu-239+240 may be attached to the surface of soil particles Krey et al (1976) found that 90% of total activity of Pu was held in the upper 10 cm of the soil They recommended more detailed studies of soil characteristics and additional measurements of Pu concentrations with depth and time

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The goal of this study was to assess the nature and extent of actinide distributions in the soil environment as well as to provide essential information for risk assessment associated with these radioactive elements in soils east of RF This work supports the Remedial Investigations and Feasibility Studies (RI-FS) at RF The objectives of this study were to (I) assess the distribution of actinides with depth in all major soils, varying the distance and direction from the source of contamination, 25 yr after the initial contaminant release, and (ii) determine the relative importance of selected physicochemical attributes and pedological processes that may govern the fate and transport of actinides

METHODS

Field Sampling

Twenty-six soils were excavated, described, and sampled east of the former storage site at RF (Fig 1) The soil pits were excavated at undisturbed sites, which are characterized by natural short-grass prairie, pasture, and valley side vegetation as defined by Clark et al (1980)

Sampling soil for actinides characterization involves several special considerations (i) potential cross-contamination of subsurface horizons from the more contaminated surface horizons, (ii) collection of sufficient material to obtain representative actinide activities and other soil parameters, and (iii) selection of a realistic sampling design that considers the high cost of actinide analyses and provides sufficient information regarding the vertical distribution of actinides in the soil profile

In light of these considerations, a special sampling method was employed This method involved digging a pit, 3 to 5 m long, 1 m wide, and 1 to 1 3 m deep The vegetation, at the surface of the pit wall selected for sampling, was clipped close to the ground and discarded The surface of the selected wall was then thoroughly scraped with a stainless steel spade to reduce the possibility of cross-contamination Ten soil samples were collected per pit, according to the following depth intervals (in cm) 0- to 3-, 3- to 6-, 6- to 9-, 9- to 12-, 12- to 18-, 18- to 24-, 24- to 36-, 36- to 48-, 48- to 72- and 72- to 96- A bottom-to-top sampling sequence was adopted to reduce further the risk of cross contamination Each soil sample was collected from within a horizontal cavity dug into the pit face at a selected depth An exception to the above sequence was made for near-surface samples (0–12 cm), where the soil was too friable to permit discrete sampling To sample the top section of the profile, the sampling was begun at ground level using a knife and spatula to cut an area ≈25 cm long, 20 cm wide, and 3 cm deep The entire soil mass in this area was collected including roots and partially decomposed organic material Sampling continued in this manner for intervals as deep as 12 cm Additional samples were taken in several pits

Abbreviations RF, Rocky Flats, RI-FS, remedial investigations-feasibility studies CEC, cation-exchange capacity, c/f, coarse fine limit ANOVA, analysis of variance

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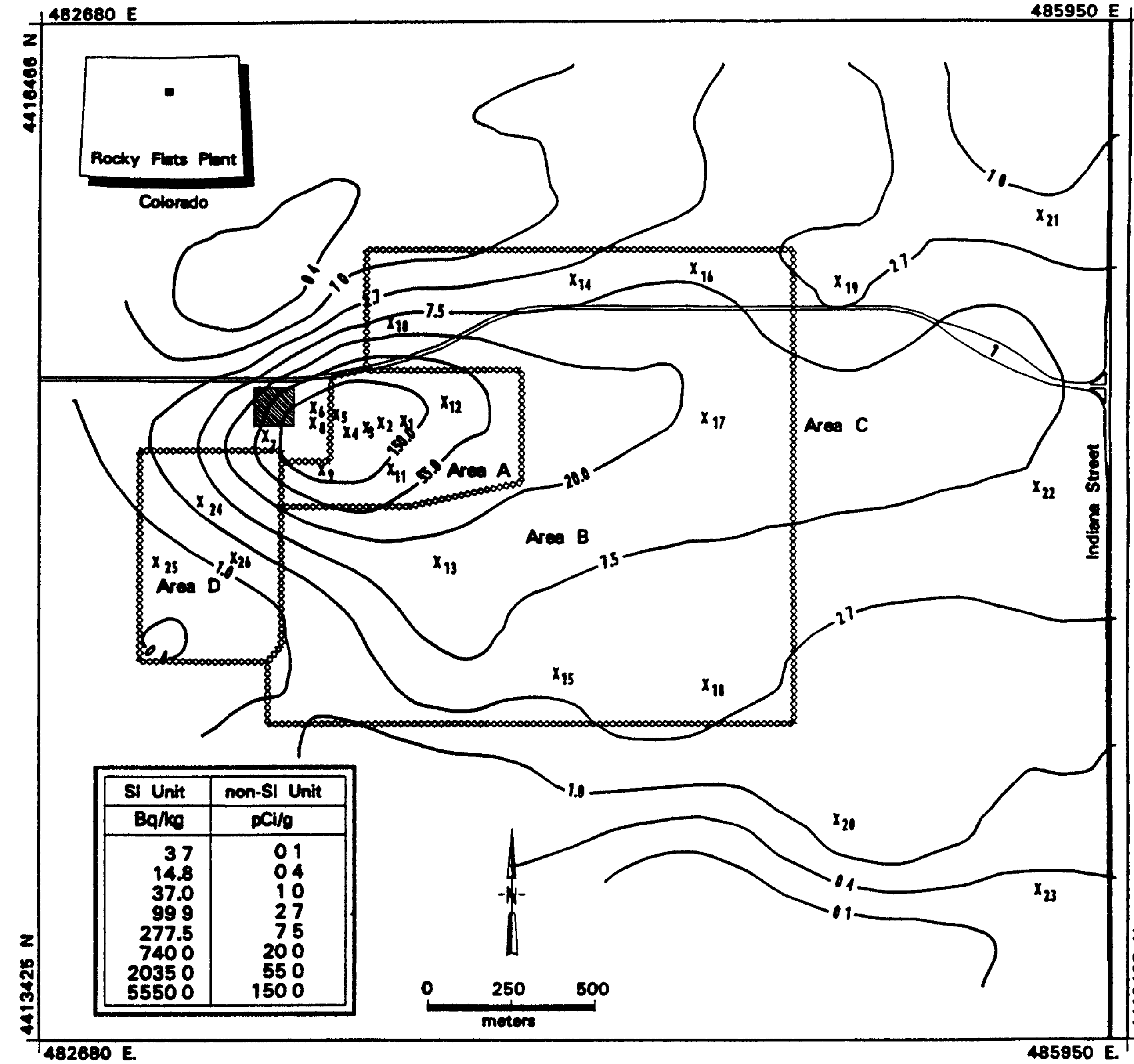

Fig 1 Location of the 26 soil pits denoted as Xn and the spatial distribution of Pu-239 + 240 in soils around RFP (see Litaor, 1993)

where soil exhibited pronounced evidence of macropore flow and biological activities (i e , rodents, ants, and earthworms)

Duplicate soil samples for radiochemical analyses were collected every third pit In the absence of established regulatory guidelines for quality control criterion for actinide activity in soil samples, the common practice of ±35% relative percent difference between duplicate field samples was applied

Sampling for selected physical and chemical parameters was conducted by genetic horizons rather than by the incremental depth procedures The soils were described according to guidelines established by the Soil Survey Staff (1975, 1993), and classified as Cumulic Haplustoll (Pit 1), Aridic Arguistoll (Pits 2, 3, 4, 5, 7, 8, 9, 10, 14, 16, 17, 18, and 23), Pachic Arguistoll (Pits 11, 15, 24, 25, 26), Torrertic Haplustoll (Pit 12), Torrifluventic Haplustoll (Pit 13), Vertic Ustochrept (Pit 19), Typic Ustifluvent (Pit 20), and Aridic Haplustalf (Pits 21 and 22)

Laboratory Analysis

Actinides

The Pu-239+240 and Am-241 activities in the soil samples were measured by alpha spectroscopy at several commercial laboratories The soil samples were digested in HNO_3 and HF using a microwave dissolution procedure (Lamothe et al , 1986, Fischer, 1986), and the Pu oxidation state was adjusted with $NaNO_2$ The solution was then passed through an anion exchange column to separate the Pu from the solution (Talvitie, 1971) The Pu was eluted from the column with HCl–NH_4I solution, was acidified with HNO_3, and the sample was heated to dryness The sample was redissolved and electroplated onto stainless steel discs Upon completion of the electroplating, NH_4OH was added to the solution to prevent redissolution of the deposit

To isolate the Am-241 from the soil matrix the sample was leached with HNO_3 Hydroxides and carbonate-forming elements (e g , Am) were precipitated out of the leachate with NH_4OH and $(NH_4)_2CO_3$ respectively After drying the precipitate was redissolved with nitric acid and passed through an anion-exchange column to remove the nontrivalent actinides Trivalent actinides and lanthanides were coprecipitated with Ca using oxalic acid, Ca carrier, and NH_4OH The precipitate was redissolved with HCl and passed through a column of mixed anion–cation resin to remove some of the Ca and all of the Fe Cesium and the remainder of the Ca were extracted from the solution using double extraction into dibutyl-N,N-diethylcarbamylphosphonate, a back extraction into dilute HNO_3, and then heating to dryness The sample was redissolved in a dilute-acid solution and passed through an anion-exchange column to remove trivalent lanthanides This column was washed with mixture of alcohol, dilute acids, and NH_4SCN for partial separation of Am from Cm The sample was then converted to a sulfate form and heated to dryness Following this, the sample was redissolved and electroplated onto stainless-steel discs Upon completion of the electroplating, NH_4OH was added to the solution to prevent redissolution of the deposit In several cases where electroplating failed for both actinides, the samples were mounted for alpha spectrometric analysis using a neodymium fluoride coprecipitation technique (Hindman, 1983, 1986) Similar percent recoveries for Pu-239+240, Am-241, and Am-243 have been reported for the electroplating and coprecipitation techniques (Sill and Williams, 1981)

Soil Properties

Particle-size distribution was determined by the pipet method (Soil Conservation Service, 1982) Soil bulk density was measured using the clod method described by Blake and Hartge (1986) Specific surface area was estimated on 1-g samples of P_2O_5-dried soil by the free-surface, ethylene glycol monoethyl ether method (Carter et al , 1986) The soil samples were given no chemical pretreatments (Cihacek and Bremner, 1979) Ca-saturated Wyoming bentonite (assumed to be 800 m^2 g^{-1}) was used to calibrate each run Cation-exchange capacity (CEC) was determined by saturation with 0 4 *M* NaOAc–0 1 *M* NaCl, followed by washing with 60% ethanol (v/v), and replacing the index cation by 0 5 *M* $MgNO_3$ (Rhoades, 1982) Organic matter content was determined by dichromate oxidation and titration with $FeNH_4SO_4$ (Nelson and Sommers, 1982) The inorganic C was determined by a modified pressure calcimeter method described by Nelson (1982)

In soil Pits 1 through 5, undisturbed clods from all genetic horizons were air dried, vacuum-impregnated with an epoxy resin, and processed into 50 by 75-mm thin sections ≈ 30 μm thick (Murphy, 1986) Thin-section analysis was performed with a petrographic microscope equipped for viewing in plane-polarized, cross-polarized, and circularly polarized light Concepts and nomenclature followed the International Soil Science Society handbook of Bullock et al (1985) and Murphy et al (1985) The coarse fine limit (c/f) was set at 50 μm The c/f refers to the division between coarse and fine particles used when classifying related distribution patterns

Statistical Analysis

Comparison between groups of soil pits and the 10 sampling intervals was conducted using two-way analysis of variance (ANOVA) (SAS Inst , 1992) Multiple comparisons among the sampling intervals were performed using Bonferroni's method, which controls the samplingwise error rate The samplingwise error rate is defined as the ratio of the number of sample intervals in which at least one error was made to the total number of sample intervals analyzed It is the probability of making at least one error in a test when there are no actual differences between the sampling intervals The mathematical formalism of Bonferroni's method is given by Milliken and Johnson (1992)

RESULTS AND DISCUSSION

Spatial and Vertical Distribution of Actinides

The pits were divided among four regions of expected actinide activity (Fig 1) The four regions were determined using isopleth maps of Pu-239+240 and Am-241 (Litaor, 1993) The length of Areas A, B, and C in the east direction is equal to the range (≈700 m) computed by a semivariogram using 118 surficial soil samples (0 64 cm) between the former storage site and Indiana Street (Litaor, 1993)

The soil immediately around the former storage site was greatly disturbed by past soil removal operations and spreading of fill material on top of the truncated soil Thus, this area was studied separately as a special case (see macrofauna and actinide transport below)

Summary statistics of Pu-239+240 and Am-241 in soils of the four areas are depicted in Table 1 Area A exhibited significantly higher activity of actinides than soils in Areas B, C, and D (Tables 1 and 2) Soils in area B exhibited significantly higher activity of Pu-239+240 than soils in Areas C and D, whereas soils in Areas C and D showed similar actinide activity (Table 2) These results are consistent with the earlier findings of Krey et al (1976) and Litaor (1993) that Pu-239+240 and Am-241 activity decreases with increased distance east of the former storage site

The vertical distribution of actinides observed in Area A suggests that actinides may be transported to greater depths than previously reported for the soils of RF For example, soil Pit 5 exhibited the largest loading of actinides onto the soil surface and the largest increase in Pu-239+240 with depth (Fig 2) This site is located ≈250 m east of the former storage site (see Fig 1) In a site nearby (≈100 m southwest from Pit 5), Little and Whicker (1978) reported a mean activity of 473 Bq/kg for Pu-239+240 at sampling depth of 18 to 21 cm Higher levels of Pu-239+240 were observed at depths of 24 and 36 cm of Pit 5 (1032 and 895 Bq/kg, respectively) In the upper 48 cm, soil Pit 5 is characterized by coarser texture than all other soils studied Soil Pit 5 also exhibited >80% coarse fragments (>2 mm) in the Bt horizon (15–48 cm), and hydraulic conductivity measurements of this horizon ranged between 18 6 cm/h to 21 19 cm/h at a depth of 20 cm (Litaor et al , 1993, unpublished data) These hydrological conditions probably have facilitated the translocation of actinides to greater depth than in finer textured soils at comparable distances from the former storage site

Approximately 90% of the Pu-239+240 activity in the soils under study was observed in the upper 12 cm, followed by a exponential decrease of actinide activity with depth (Table 1, Fig 2) Of the 25 soil pits that were sampled and analyzed, 23 exhibited the distribution

Table 1 Summary statistics of Pu-239 + 240 and Am-241 vertical profiles by areal distribution across the soilscape east of RF

Depth	Pu-239 + 240 Mean	Median	SD	Cumulative	Am-241 Mean	Median	SD	Cumula
	Bq/kg			%	Bq/kg			%
Area A (n = 8)								
0-3	11 655	7770	8658	39 4	1653	1261	1135	38 5
3-6	8 547	8584	5476	28 9	1098	1043	643	25 6
6-9	5 291	3848	4255	17 9	758	540	606	17 7
9-12	2 886	3108	2960	9 8	595	251	677	13 9
12-18	814	240 5	1332	2 7	122	37	173	2 8
18-24	222	107 3	333	0 7	33	11	44	0 8
24-36	111	14 8	296	0 4	22	2 6	51	0 5
36-48	14 8	7 4	22	0 18	2 6	1 5	3 3	0 1
48-72	3 7	1 5	11	0 01	1 1	0 7	1 5	0 06
72-96	3 7	0 7	11	0 01	1 8	0 4	1 8	0 04
Area B (n = 7)								
0-3	407	259	370	61 5	47 4	25 9	58 8	41 8
3-6	177	74	222	26 8	46 2	18 5	58 4	40 8
6-9	29	37	14	4 5	7 4	3 7	4 8	6 5
9-12	26	26	18	3 9	3 7	2 9	2 2	3 3
12-18	7 4	3 7	5 9	1 1	1 8	1 5	1 1	1 9
18-24	5 9	3 7	6 3	0 9	1 5	1 1	1 1	1 6
24-36	4 4	1 5	5 9	0 7	1 1	1 8	0 7	1.2
36-48	1 1	0 4	1 1	0 3	0 7	0 7	0 6	1 0
48-72	0 7	0 4	0 4	0 1	2 2	0 7	0 7	1 0
72-96	1 8	0 4	2 2	0 2	1 1	0 7	0 7	0 9
Area C (n = 5)								
0-3	62 9	44 4	44 4	54 2	2 9	2 2	2 2	21 7
3-6	22 2	11 1	22.2	19 1	5 9	0 7	9 9	38 9
6-9	22.2	11 1	29 6	19 1	3 3	0 3	4 4	21 9
9-12	5 2	7 4	4 1	4 4	1 8	0 2	0 7	11 9
12-18	2 2	1 8	1 8	1 9	0 2	0 07	0 3	1 9
18-24	0 4	0 4	0 3	0 3	0 1	0 07	0 1	0 9
24-36	0 2	0 2	0 1	0 3	0 1	0 07	0 2	0 9
36-48	0.2	0 2	0 1	0 3	0 1	0 07	0 1	0 9
48-72	0 3	0 2	0 3	0 2	0 07	0 04	0 07	0 5
72-96	0 3	0 2	0 2	0 2	0 07	0 04	1 1	0 5
Area D (n = 3)								
0-3	44 4	44 4	22 2	41 5	3 7	4 0	3 7	27 2
3-6	29 6	29 6	14 8	27 6	3 7	5 5	1 8	26 2
6-9	14 8	18 5	7 4	13 8	3 3	3 7	1 1	23 6
9-12	7 4	11 1	3 7	6 9	1 5	1 5	0 4	10 9
12-18	7 4	3 7	11 1	6 9	0 7	0 7	0 4	5 1
18-24	2 2	1 11	2 6	2 1	0 4	0 2	0 4	2 5
24-36	0 7	0 19	0 7	0 7	0 2	0 1	0 2	1 5
36-48	0 4	0 19	0 3	0 3	0 2	0 1	0 7	1 5
48-72	0 1	0 19	0 1	0 1	0 1	0 1	0 1	0 8
72-96	0 07	0 19	0 1	0 1	0 1	0 1	0 1	0 7

described above This pattern was consistent regardless of soil characteristics, level of contamination, or distance and direction from the former storage site

Table 2 Summary statistics of two-way analysis of variance testing the classification of Pu- 239 + 240 activity (log Pu) with area and depth in the 25 soil pits Similar results were observed for Am-241 (not shown)

Source	df	Sum of squares	Mean square	F value	$P > F$
Area	3	838 3	279 4	189 96	0 0001

Bonferroni (Dunn) t tests
Alpha = 0 05 Confidence = 0 95 df = 190 MSE = 1 47
Critical value of T = 2 66

Area comparison	Simultaneous Lower confidence limit	Difference between means	Simultaneous Upper confidence limit
A-B	2 520	3 049	3 578*
A-C	4 123	4 706	5 289*
A-D	3 378	4 070	4 762*
B-C	1 058	1 657	2 256*
B-D	0 315	1 021	1 727*
D-C	−0 110	0 636	1 383

* Significant at the 0 05 probability level

The Effect of Pedogenic Processes on Vertical Distribution of Pu-239 + 240 and Am-241

Several pedogenic features that may control transloca tion and subsequent vertical distribution of actinides were observed in most soil pits under study Abrupt textura and structural boundaries, macroporosity, lateral subsur face discontinuities, burrowing animals, earthworms and ant colonies, were among the more common pedogenic and geologic features observed

Lateral Discontinuities

Lateral discontinuities were located in Pits 2, 3, and 4 (see Fig 1) These discontinuities were clearly identified in the field by their lateral textural and color differences (Fig 3) One soil sample from each horizon on either side of the lateral discontinuity was collected to assess the influence of the different physicochemical properties on actinide activity The BC horizon consisted of 75% sand and 16% clay, with low specific surface area (94 m^2/g), low content of $CaCO_3$ (2 g/kg), low

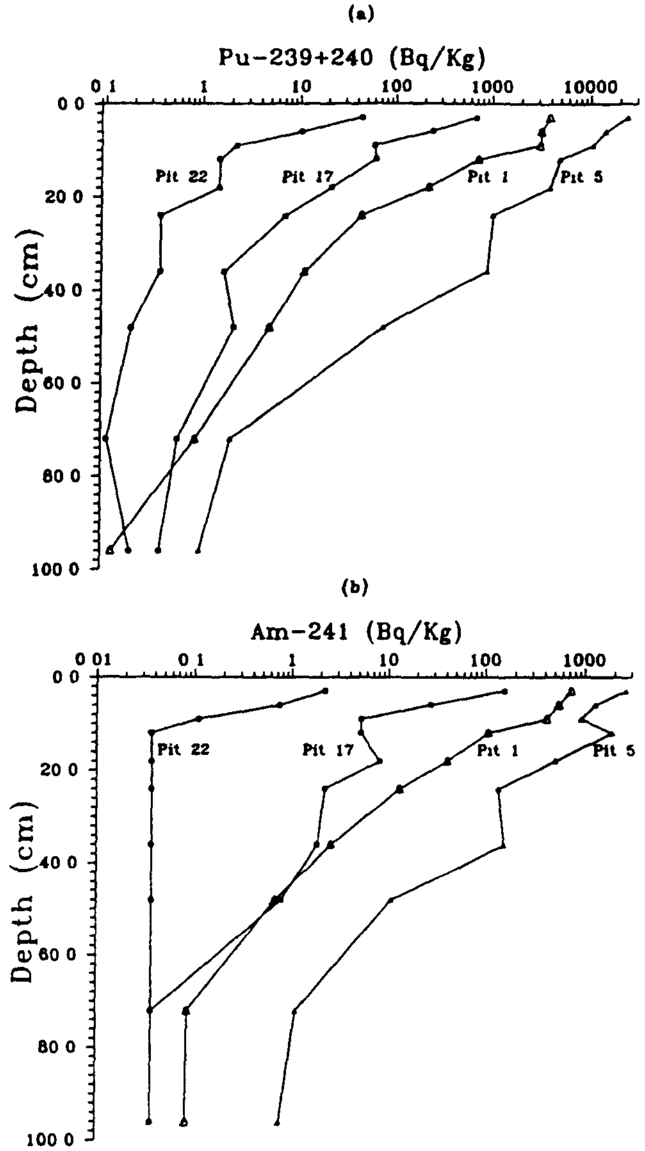

Fig 2 Typical vertical distribution of (*a*) Pu-239 + 240 and (*b*) Am-241 in four soil pits across a west–east transect

CEC (8 8 cmol/kg), small amounts of smectite (5–30%) and trace amounts of clay mica (<5%) (Thompson et al , 1993, unpublished data) By contrast, the BCg horizon consisted of only 16% sand and 40% clay, with moderate specific surface area (168 m^2/g), higher content of $CaCO_3$ (23 g/kg), moderate degree of CEC (29 6 cmol/kg), dominant amounts of smectite (>60%), and small amounts of clay mica (5–30%) The origin of the subsurface lateral discontinuity is not clear It has been hypothesized that the BCg horizon is a truncated layer that was incised and later filled with sandy material (BC horizon) The overlying horizons have developed on top of these two markedly different parent materials

Despite these profound differences in physicochemical and mineralogical attributes between the BC and the BCg horizons, only minor differences were observed in actinide activity Plutonium-239+240 activity in the BC horizon was 3 7 Bq/kg, whereas 1 8 Bq/kg were found in the BCg horizon Americium-241 measured 3 7 Bq/kg in both horizons These relative small differences in actinide activities are probably due to the depth of the lateral discontinuity (>51 cm) That is, most of the actinide activity was found in the top 12 cm of the soil, thus the preferential leaching potential of the sandy BC horizon had little apparent impact on the vertical distribution of actinides

Macroporosity

The distribution and nature of macropores are the most important factors governing water and contaminant transport in soils, according to Gish and Shirmohammadi (1991) Large macropores at various depths (30–100 cm), filled with surface horizon materials, were observed in the field along decayed root channels These filled macropores may serve as prime conduits for actinide translocation with depth To test this hypothesis, samples were taken from the fillings of the two root channels, in soil Pits 3 and 4, and analyzed for actinide activity Two samples that were taken from a decayed root channel at a depth of 90 cm in soil Pit 3 showed significantly higher activity of Pu-239+240 (7 0 Bq/kg and 5 9 Bq/kg) compared with the soil matrix around the macropore filling (0 03 Bq/kg) Similarly, a sample that was collected from the decayed root channel of soil Pit 4 at a depth of 70 to 92 cm showed higher Pu-239+240 activity (8 5 Bq/kg) compared with the surrounding soil matrix at 72 cm (1 5 Bq/kg) and 96 cm (0 7 Bq/kg) These results indicate that preferential flow in macropores is a viable mechanism for mobilizing actinides within the soil profile

It should be noted, however, that the overall actinide activity in the filling of the decayed root channels was very small compared with 8325 Bq/kg and 16 132 Bq/kg of Pu-239+240 observed in the top 9 cm of Pit 3 and 4 respectively Moreover, the large macropore network formed by the root system was not observed at depths >120 cm Hence, the macroporosity network probably does not link the vadose zone to the aquifer below it (≈2 5 m) and probably does not increase the potential for actinide movement to the local groundwater

Burrowing Activity

Evidence of burrowing activity was observed in six soil pits across the study area For example, two krotovinas (filled-in animals burrows or tunnels) were sampled in Pit 18 at 96 cm and Pit 23 at 87 cm (see Fig 1 for pit locations) Horizon BkIV in Pit 22, at a depth of 38 to 55 cm, contained pockets of clay and organic matter These pockets resulted from megafaunal biopedoturbation that translocated surface-horizon material to subsurface horizons Actinide activities in the krotovinas (3.7 Bq/kg) and translocated organic matter (0 18 Bq/kg) were not significantly greater than the soil matrix (ranged between 0 37 and 0 18 Bq/kg) around the krotovinas The sampled krotovinas and organic matter were probably older than industrial activities at RF (>40 yr) If so, they do not reflect current rodent activity, nor the

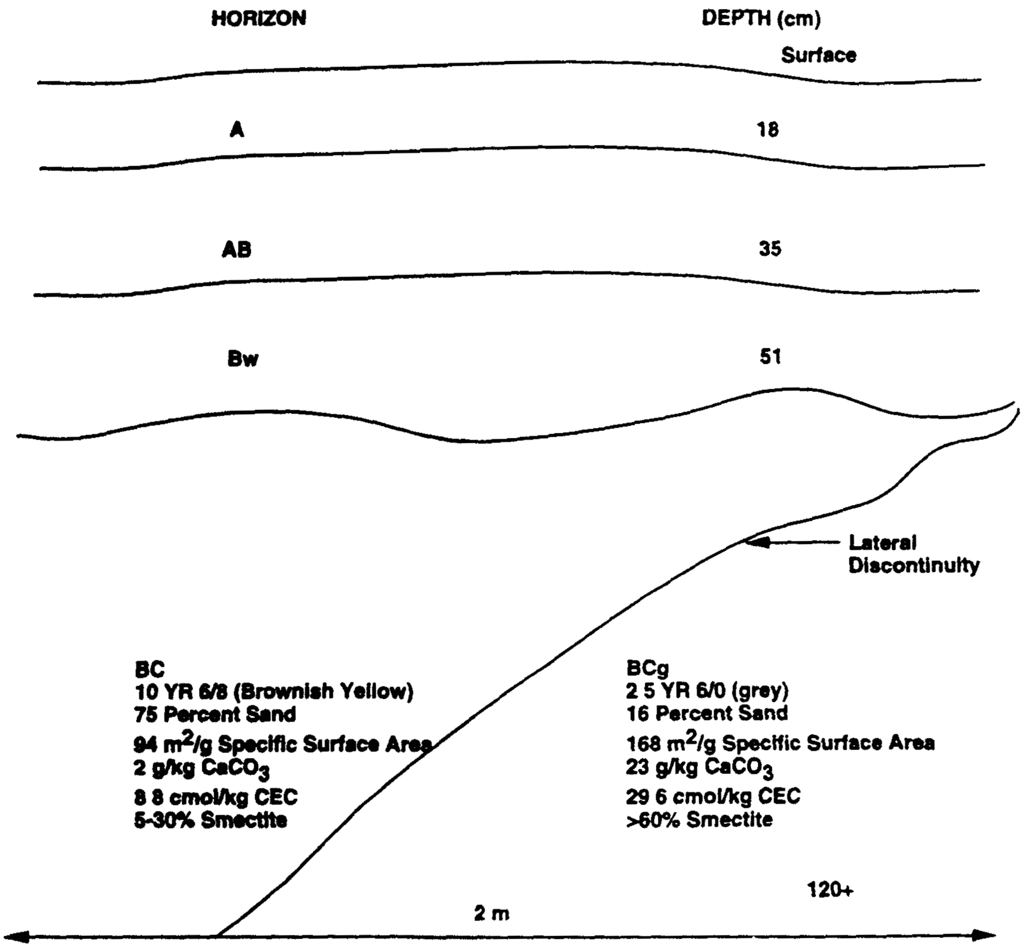

Fig 3 Lateral discontinuity observed at 51 to 120 cm of soil Pit 3

corresponding potential for redistribution of actinides with depth

Macrofauna and Actinide Transport

The area of the former storage site was graded and covered with fill material in 1969 This was followed by the application of a soil sterilant and the site was covered with an asphalt cap The area adjacent to the former storage site was covered with sandy fill material, then revegetated to stabilize the fill material and to reduce wind erosion Three soil pits (6, 7, and 8, see Fig 1) were studied to assess the vertical distribution of actinides in the most heavily contaminated soil in RF The boundary between the fill material and the buried, contaminated soil was clearly identifiable in the field

The fill material is characterized by a low $CaCO_3$ content (<1 g/kg), low CEC (<17 cmol/kg), and high sand content (>80%) The upper 3 cm of the fill material has been enriched with organic C since the material was emplaced in 1969 The organic C increased from ≈0 to 34 g kg^{-1} in 1991 The increase of organic C content suggests that revegetation did stabilize the fill material by adding significant amounts of organic matter It also improved soil aggregation thereby making the soil less susceptible to wind erosion, reduced wind velocity near the surface, and advanced infiltration rate, which in turn decreased the potential for surface flow

The vertical profile of actinides in the soil immediately east of the former storage site showed a unique distribution with depth (Fig 4) The highest actinide activity in the study area was observed in the top 3 cm of Pit 8 This was followed by a significant decrease in the lower section of the fill material (at sampling intervals of 6 and 9 cm), then by an increase in actinide activity at the top of the buried soil (12 cm below the soil surface)

As discussed earlier, the spatial distribution of actinides in the soil east of RF has been attributed to wind dispersal (Krey and Hardy, 1970, Seed et al , 1971, Little and Whicker, 1978) After the placement of the asphalt pad and the fill material around it, however, the level of Pu-239+240 in the air decreased to background level (Seed et al , 1971) Furthermore, airborne Pu resulting from wind erosion in the vicinity of the former storage site, has been minimal since 1970 (Rocky Flats Plant, 1992) Hence, the surprising increase of actinide activity in the top 3 cm of the fill material cannot be explained by wind-transport mechanisms

On the basis of the historical assessment of the site (i e , grading, application of fill material, revegetation, and the low potential and observations of Pu resuspension

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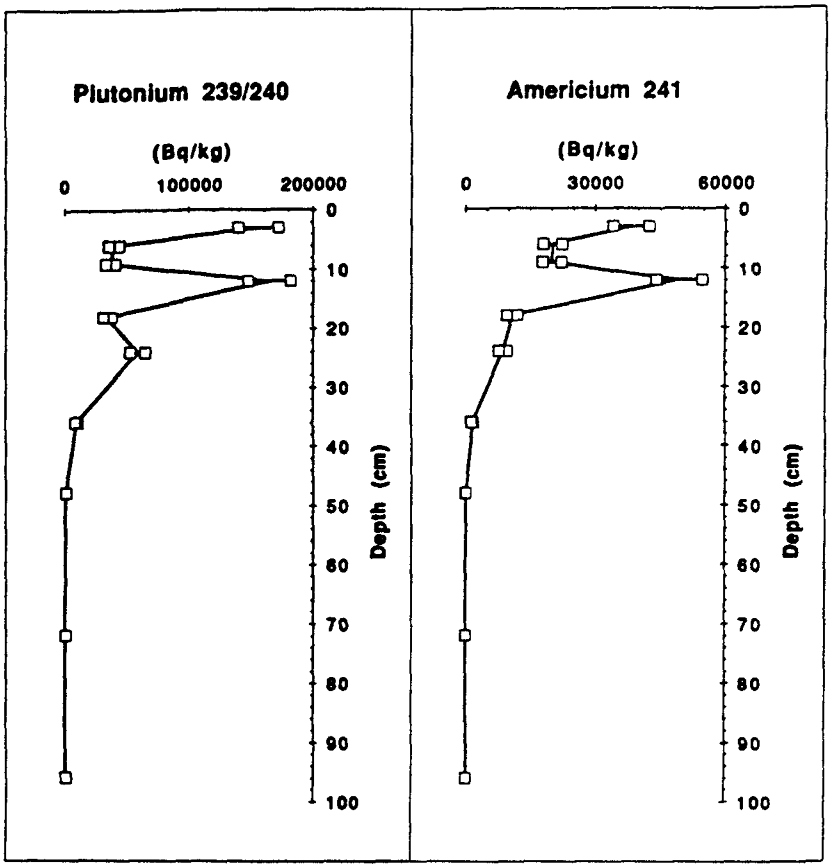

Fig 4 The vertical distribution of actinides in soil (Pit 8) immediately east of the former storage site The negative values correspond to analytical error around the measurements that are close to 0 The analytical error represents the 2σ counting error associated with each analysis

from soils around the former storage site), we speculate that the vertical distribution of actinides in Pit 8 resulted from the upward transport of actinides by earthworms Current literature on the impact of earthworms in similar ecosystems supports this hypothesis Our speculation is based largely on the observed abundance of faunal activity in most of the soils under study, as well as micromorphological analysis Additionally, earthworms and their casts were also collected from soil interstitial waters sampled in Pits 2 and 3 at different depths, using zero-tension samplers There was a strong relationship between the observed earthworms and their castings, and Pu activity in these waters (Litaor et al , 1994, unpublished data)

Evidence of intensive biological activity in grassland soils has been documented by many workers (Baxter and Hole, 1967, Buntley and Papendick, 1960, Jenny, 1980) In general these reports suggest that much of the organic material of A horizons has acquired a granular structure from passage through alimentary tracts of earthworms and other soil fauna Indeed, a weakly granular structure was observed at soil Pits 7 and 8 in the surface horizons composed of the fill material as well as in thin sections made from samples of the fill material

Numerous earthworms and several ant colonies were observed in most of the soils under study Earthworm activity was especially evident in Pits 1, 2, 3, and 4 Micromorphological analyses of A horizon samples from soil Pits 1 through 5 revealed strong evidence of biological activity in the soils For example, ellipsoidal fecal pellets, typically ≈ 80 μm in diam , were observed in the A horizon of soil Pit 1 (Fig 5)

Pasture soils often exhibit numerous features related to earthworm activity in surface and subsurface horizons, including large channels, abundant fecal pellets, channel infillings, and organic intercalations (e g , Thompson et al , 1990) Schmidt et al (1986) showed that earthworms were the major force in vertical transport of cattle dung contaminated with heavy metals The earthworms turned over the experimental soil column from top to bottom, and vice versa, creating layers of contaminated organic matter in their tunnels The study by Schmidt et al (1986) clearly demonstrated the importance of earthworms in reworking contaminated soil material and moving it in all directions

The field and micromorphological analyses, coupled with the above literature review, provided the framework for the following scenario after revegetation of the fill material, earthworms from the buried soil migrated to the new surface to feed on the new seedlings The buried soil horizon provided the earthworms with an additional source of food and shelter from extreme temperature and moisture conditions Hence, the movement of earthworms between the buried soil surface and the newly revegetated surface may have continued indefinitely During this migratory process, actinides from the buried

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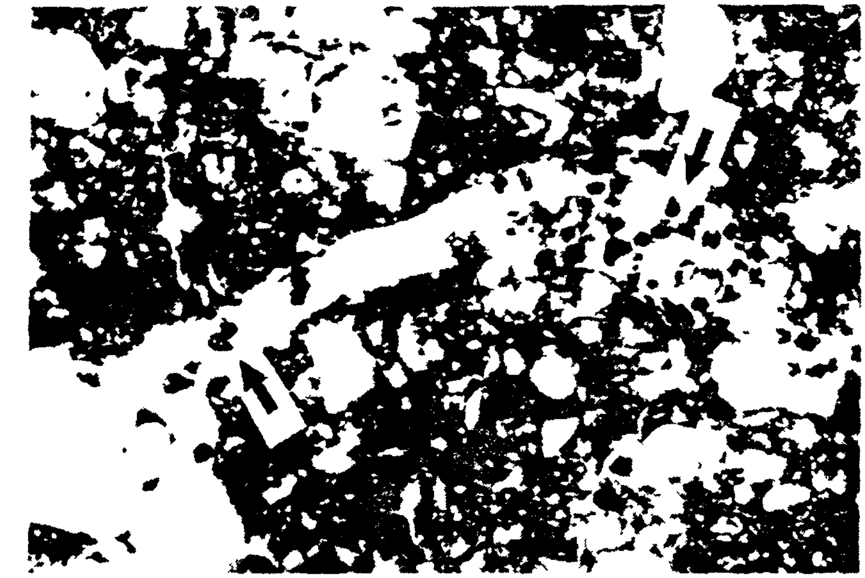

Fig 5 Loose, incomplete infilling of fecal pellets (fp) in a channel in the A horizon of soil Pit 1, frame length 3 5 mm

soil were brought to the top 3 cm of the fill material and were deposited in fecal pellets that probably disintegrated with time to loose continuous infillings of channels and tunnels The relatively low actinide activity in the lower section of the fill material resulted from lack of significant food source for the soil fauna and/or because worms cast preferentially at the surface

CONCLUDING REMARK

The vertical distribution and possible transport mechanisms of actinides in the soils of RF were studied to provide the necessary data for risk assessment More than 90% of the Pu-239+240 and Am-241 activities were observed in the upper 12 cm of the soils Movement of actinides within the soil is probably governed by preferential flow and biological activity such as earthworms If future studies confirm earthworm-induced transport of actinides in the soil environment of RF, the feasibility and cost of soil remediation shall be severely impacted A regulatory requirement by the Colorado Department of Health dictates ameliorative treatment of soils from lands with Pu-239+240 activity >33 3 Bq/kg before construction activity is approved Past cleanup operations have been limited to the upper 10 to 15 cm of the soil (Baker, 1982) Earthworms and other soil fauna, however, may bring Pu-239+240 activity that will exceed the 33 3 Bq/kg guideline from depths >15 cm Hence, future cleanups to greater soil depths may be cost prohibitive because of the greater volume of contaminated soil Before decisions can be made regarding cleanup, more studies are needed to test the proposed earthworm-induced transport mechanism for the migration of actinides

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