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$^{239-240}\text{Pu}$ AND ^{241}Am CONTAMINATION OF VEGETATION
IN AGED PLUTONIUM FALLOUT AREAS¹

E. M. Romney, A. Wallace, R. O. Gilbert,² and J. E. Kinnear

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ABSTRACT

This is an interim progress report of work relating to plutonium in the vegetation of desert environments which has been conducted under the auspices of the Nevada Applied Ecology Group, Environmental Effects Division, USAEC Nevada Operations Office. Information presented herein also appears as part of the NAEG Progress Report, NVO-153, 1975.

Included in this report are data on the $^{239-240}\text{Pu}$ and ^{241}Am contents of vegetation samples collected in conjunction with the NAEG soil sampling program in aged plutonium fallout areas on the Nevada Test Site (NTS) and the Tonopah Test Range (TTR). Radiochemical analyses essentially have been completed on samples collected from the GMX Site in Area 5 and the Project 57 Site in Area 13. About 60% of the vegetation samples collected from the Area 11 Sites at NTS and the Roller Coaster Sites at TTR have undergone radiochemical analysis.

Preliminary data indicate definite variations in the contamination levels from sample to sample collected within a given activity stratum defined by the FIDLER survey instrument. Variations also occur in the contamination levels on different plant species which can be attributed to differences in the amounts of resuspendable particulate material superficially entrapped upon plant foliage. In spite of these variations, there are some indications of reasonable agreement between the mean activity levels of $^{239-240}\text{Pu}$ in vegetation and soil samples

collected across the different activity strata within each fallout area. In addition, there are indications that the ratio of vegetation Pu to soil Pu increases as one proceeds from higher to lower activity strata. A limited particle size range of resuspendable material superficially deposited upon vegetation could account for some of the variations in activity levels and ratios noted when comparing the vegetation and soil data.

Sufficient data have been acquired to indicate a reasonably constant Pu/Am ratio for vegetation samples collected from a given fallout area. This ratio, however, varies among the separate events as the result of differences in the ingrowth of ^{241}Am within the aged fallout source materials. The Pu/Am ratios tend to be lower in vegetation than in soil samples for each event studied, except for the Area 5-GMX Site where multiple tests were conducted. These lower ratios in vegetation samples are presumed to result from a preferential uptake by plant roots of the more biologically available ^{241}Am as compared to $^{239-240}\text{Pu}$ in these aged fallout areas.

The $^{239-240}\text{Pu}$ inventory estimates for Area 13 indicate that the standing vegetation contributes a rather insignificant portion of the total contaminant present in the aged fallout areas.

INTRODUCTION

The previous progress report (Romney et al., 1974) pointed out that the significance of vegetation in plutonium contaminated areas rests primarily upon its capacity to function as a carrier of plutonium in the food chains of man and grazing animals. Earlier studies concerning fission product fallout from above-ground nuclear detonations disclosed that superficially contaminated vegetation

was a major source of radionuclides to grazing animals (Larson et al., 1966; Lindberg et al., 1959; Romney et al., 1963). Preliminary data from the NAEG vegetation studies in Area 13 also indicated that superficial entrapment of contaminants on vegetation might be an important process in the aged plutonium fallout areas on the Nevada Test Site (NTS) and the Tonopah Test Range (TTR). This point is pursued further in this report utilizing the partially completed analytical data from several aged fallout areas.

MATERIALS AND METHODS

Samples of perennial vegetation were collected in conjunction with the sampling of soil for an inventory of plutonium in aged fallout areas at NTS and TTR. The pertinent details on the selection of random sampling sites for soil in activity strata established from intensive FIDLER surveys have been reported by Gilbert and Eberhardt (1974). Briefly, a sample of perennial vegetation was collected in the immediate vicinity of the soil sample, and the two samples from each location were considered to be paired samples. Steps were taken to obtain a random vegetation sample from within a 30 m² circular plot centered upon the soil sampling location. Each shrub was assigned a number and one was chosen for sampling from a list of random numbers. In application, the distribution of shrubs at several locations was such that only one or two plants fell within the sampling plot. For those few locations where no shrubs fell within the plot, the shrub located nearest the soil sampling point was selected.

Samples of foliage varying from 300 to 500 grams of dry tissue were clipped from shrubs into 1-gallon press lid cans and submitted to the analytical laboratory

for radiochemical analysis. Procedures for determining $^{239-240}\text{Pu}$ and ^{241}Am in these large vegetation samples have been reported by Major, et, al., (1974).

RESULTS AND DISCUSSION

It should be noted that some results reported herein are preliminary and subject to change upon further radiochemical analysis and refinement of data. Most of the radiochemical analyses on vegetation samples from Area 5-GMX and Area 13 had been completed for this progress report, but only about 60% of the samples collected from other areas under investigation had been completed.

$^{239-240}\text{Pu}$ Contamination of Vegetation In Aged Fallout Areas

Plots of the $^{239-240}\text{Pu}$ and ^{241}Am data obtained from radiochemical analysis of vegetation samples are presented in the Appendix of this report. The $^{239-240}\text{Pu}$ is plotted against ^{241}Am contents for all plant species in each activity stratum. The least squares weighted regression line through the origin, the average Pu/Am ratio, and the correlation between $^{239-240}\text{Pu}$ and ^{241}Am concentrations were computed for each data set by methods described by Gilbert and Eberhardt (1974). The spread in activity levels, designated on each axis, indicates the extent of variation from sample to sample collected from within the different activity strata which had been defined from readings obtained with FIDLER survey instrument.

Inasmuch as the perennial vegetation was sampled at random, it naturally followed that different plant species would become involved. Table 1 lists the $^{239-240}\text{Pu}$ contents and the $^{239-240}\text{Pu}/^{241}\text{Am}$ ratios in the dry tissue samples of different plant species collected from each of the aged fallout areas. The

Table 1

$^{239-240}\text{Pu}$ Contents and $^{239-240}\text{Pu}/^{241}\text{Am}$ Ratios for Different
Plant Species in Aged Pu-Fallout Areas

Plant species	Activity strata	n	Vegetation (nCi/g dry tissue)	
			$^{239-240}\text{Pu}$ Mean $\pm$ S.E.*	$^{239-240}\text{Pu}/^{241}\text{Am}$ Ratio $\pm$ S.E.**
<u>NTS AREA 5 - GMX (1954-55)</u>				
<u>Atriplex confertifolia</u>	5	6	.012 $\pm$.0030	13.4 $\pm$ 1.0
	1	27	.011 $\pm$.0030	15.0 $\pm$ 1.3
	2	10	.084 $\pm$.031	11.9 $\pm$ 0.66
	3	4	.31 $\pm$.073	14.7 $\pm$ 1.6
	4	9	.28 $\pm$.047	12.4 $\pm$ 0.85
<u>Ephedra nevadensis</u>	5	1	.0066	14.1
	1	4	.017 $\pm$.011	13.1 $\pm$.54
	4	1	.34	15.6
<u>Eurotia lanata</u>	3	1	1.1	13.3
<u>Grayia spinosa</u>	1	1	.011	11.2
<u>Larrea tridentata</u>	5	6	.0053 $\pm$.0011	13.6 $\pm$ 1.4
	1	15	.0040 $\pm$.0016	13.4 $\pm$ 1.5
	2	14	.050 $\pm$.010	12.2 $\pm$ 0.51
	3	7	.11 $\pm$.036	10.7 $\pm$ 1.1
	4	7	.34 $\pm$.095	11.8 $\pm$ 0.61
<u>NTS AREA 11-B (1956)</u>				
<u>Atriplex confertifolia</u>	2	6	.14 $\pm$.049	8.7 $\pm$ 0.52
	3	8	.17 $\pm$.025	7.3 $\pm$ 0.23
	4	12	.64 $\pm$.12	9.8 $\pm$ 3.4
<u>Chrysothamnus vicidiflorus</u>	2	1	.12	7.6
	3	1	.11	7.3
	4	3	.61 $\pm$.36	8.1 $\pm$ 0.52
<u>Hymenoclea salsola</u>	4	2	.21 $\pm$.062	8.0 $\pm$.40
<u>Lycium andersonii</u>	2	3	.096 $\pm$.070	8.9 $\pm$.68
	3	1	.011	11.6
	4	1	.39	7.8
<u>Tetradymia glabrata</u>	3	4	.29 $\pm$.18	7.4 $\pm$.14

Table 1 (continued)

Plant species	Activity strata	n	Vegetation (nCi/g dry tissue)	
			²³⁹⁻²⁴⁰ Pu Mean ± S.E.*	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.**
<u>NTS AREA 11 - C (1956)</u>				
<u>Atriplex canescens</u>	2	1	.11	5.7
	3	3	.34 ± .019	5.1 ± .27
	4	5	.67 ± .16	4.6 ± .48
	5	1	.38	5.7
<u>Atriplex confertifolia</u>	2	6	.15 ± .056	5.2 ± .19
	3	4	.22 ± .084	5.0 ± .22
	4	8	.66 ± .27	5.0 ± .073
<u>Artemisia spinescens</u>	3	1	.79	4.6
	4	1	7.2	5.8
<u>Chrysothamnus vicidiflorus</u>	2	1	.059	5.0
	3	1	.42	5.2
	5	2	1.7 ± .57	4.6 ± .34
<u>Ephedra nevadensis</u>	3	1	.18	5.5
<u>Eurotia lanata</u>	3	1	.59	5.6
<u>Grayia spinosa</u>	2	1	.084	6.1
	5	1	1.9	5.0
<u>Tetradymia glabrata</u>	2	3	.17 ± .026	5.8 ± .40
	3	3	.26 ± .086	5.2 ± .048
	4	3	.90 ± .30	5.5 ± .22
<u>NTS AREA 11-D (1956)</u>				
<u>Atriplex canescens</u>	3	2	.25	4.4 ± .030
	4	1	.43	2.8
	5	1	.90	4.0
<u>Atriplex confertifolia</u>	2	2	.042 ± .016	4.5 ± .052
	3	4	.17 ± .045	3.7 ± .22
	4	13	.60 ± .12	4.1 ± .55
	5	15	1.1 ± .27	3.3 ± .49
<u>Chrysothamnus vicidiflorus</u>	2	1	.095	4.3
	3	1	.13	3.4
	4	1	2.8	4.1
	5	3	1.6 ± .71	3.6 ± .37
<u>Eurotia lanata</u>	2	1	.15	5.4
	3	1	1.1	4.4

Table 1 (continued)

Plant species	Activity strata	n	Vegetation (nCi/g dry tissue)	
			²³⁹⁻²⁴⁰ Pu Mean ± S. E.*	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S. E.**
<u>NTS AREA 11-D (1956) (continued)</u>				
<u>Lycium andersonii</u>	3	2	.13 ± .032	3.9 ± .11
	4	3	.41 ± .082	4.3 ± .38
	5	2	1.2 ± .13	5.1 ± .27
<u>Tetrdymia glabrata</u>	2	7	.23 ± .045	5.8 ± 1.9
	3	2	.32 ± .019	5.0 ± .030
	4	2	.70 ± .17	5.0 ± .35
<u>NTS AREA 11-CD OVERLAP (1956)</u>				
<u>Atriplex confertifolia</u>	6	3	.025 ± .0078	6.4 ± 1.4
<u>Chrysothamnus vicidiflorus</u>	6	1	.019	8.8
<u>Hymenoclea salsola</u>	6	1	.010	5.9
<u>NTS AREA 11 - ABCD (1956)</u>				
<u>Atriplex confertifolia</u>	1	9	.0028 ± .00070	5.9 ± .79
<u>Chrysothamnus vicidiflorus</u>	1	2	.0015 ± .00080	2.9
<u>Eurotia lanata</u>	1	4	.0031 ± .00097	6.0 ± 1.4
<u>Lycium andersonii</u>	1	3	.0053 ± .0052	4.8
<u>Tetradymia glabrata</u>	1	5	.0035 ± .00080	4.9 ± 1.4
<u>NTS AREA 13 (1957)</u>				
<u>Atriplex canescens</u>	1	2	.0034 ± .0024	17.5 ± 6.1
	3	4	.13 ± .073	9.2 ± .58
	4	9	.041 ± .0076	7.1 ± .73
	5	3	.12 ± .0052	8.1 ± .47
	6	9	.40 ± .11	9.5 ± .65
<u>Atriplex confertifolia</u>	1	31	.0047 ± .00056	7.3 ± .76
	2	24	.013 ± .0021	10.1 ± .62
	3	4	.055 ± .0085	10.2 ± .55
	4	5	.073 ± .0027	7.4 ± 1.1
	5	2	.29 ± .016	11.6 ± .72
	6	11	.87 ± .24	7.0 ± .61

Table 1 (continued)

Plant species	Activity strata	n	Vegetation (nCi/g dry tissue)	
			²³⁹⁻²⁴⁰ Pu Mean ± S.E.*	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.**
<u>NTS AREA 13 (1957) (continued)</u>				
<u>Eurotia lanata</u>	3	4	.18 ± .082	12.9 ± 3.8
	4	3	.19 ± .083	8.0 ± 2.2
	5	5	.37 ± .15	8.8 ± .37
	6	11	2.6 ± 1.0	8.0 ± .26
<u>Grayia spinosa</u>	1	1	.00075	28.3
	2	1	.0017	16.9
	4	1	.088	9.1
	6	4	.18 ± .071	2.03 ± 1.4
<u>Lycium andersonii</u>	1	1	.013	41.2
	6	1	.84	8.5
<u>Tetradymia glabrata</u>	6	1	2.3	6.6
<u>TTR DOUBLE TRACK (1963)</u>				
<u>Atriplex canescens</u>	1	1	.0076	40.5
	4	1	.37	28.1
<u>Atriplex confertifolia</u>	1	16	.010 ± .0037	14.3 ± 3.0
	2	9	.072 ± .029	16.8 ± .81
	3	11	.11 ± .036	18.4 ± 1.4
	4	10	.50 ± .18	15.6 ± 3.1
<u>TTR CLEAN SLATE 1 (1963)</u>				
<u>Atriplex confertifolia</u>	1	20	.014 ± .0080	17.5 ± 1.5
	2	12	.027 ± .011	14.1 ± 7.8
	3	9	.012 ± .0033	11.5 ± 1.2
	4	8	.035 ± .011	16.2 ± .77
Mixed grasses	4	3	.021 ± .0063	19.9 ± 2.8
<u>TTR CLEAN SLATE 2 (1963)</u>				
<u>Atriplex confertifolia</u>	1	18	.0066 ± .0013	12.9 ± 1.1
	2	13	.047 ± .0055	13.6 ± 8.1
	3	12	.090 ± .019	12.8 ± 1.0
	4	20	.14 ± .032	10.8 ± 1.3

Table 1 (continued)

<u>Plant species</u>	<u>Activity strata</u>	<u>n</u>	<u>Vegetation (nCi/g dry tissue)</u>	
			<u>$^{239-240}\text{Pu}$ Mean $\pm$ S.E.*</u>	<u>$^{239-240}\text{Pu}/^{241}\text{Am}$ Ratio $\pm$ S.E.**</u>
<u>TTR CLEAN SLATE 3 (1963)</u>				
<u>Atriplex confertifolia</u>	1	10	.013 $\pm$.0042	21.3 $\pm$ 1.3
	2	11	.020 $\pm$.0038	18.6 $\pm$ 1.4
	3	10	.069 $\pm$.015	16.3 $\pm$ 1.4
	4	10	.18 $\pm$.053	16.9 $\pm$ 1.8

* Standard error of mean $\simeq [\text{Var.} / n]^{\frac{1}{2}}$

**If $R = \overline{Y/X}$, then:

$$\text{S. E. (R)} = [(\sum \frac{Y_i^2}{X_i} - \frac{(\sum Y_i)^2}{\sum X_i}) / (n-1) \sum X_i]^{\frac{1}{2}}. \text{ See Snedocor and}$$

Cochran, Statistical Methods, Iowa State University Press, 6th Ed., p. 168. (1967).

data in Table 1 are tabulated by event in order of decreasing time since the event occurred, i. e., Area 5-GMX (1954-55); Area 11-B, C, D (1956); Area 13 (1957); and TTR Sites (1963). The activity strata were labeled in order of increasing activity with numbers 1 through 6, except as follows: (1) a special low activity stratum was designated number 5 at NTS Area 5-GMX; (2) the CD overlap stratum was designated number 6 at Area 11; (3) a zone surrounding the events in Area 11 having FIDLER readings less than 5,000 cpm was designated Area 11-ABCD stratum 1. Some plant species were relatively sparse in different areas as indicated by the small n values. The variations in $^{239-240}\text{Pu}$ contents between different plant species were of an order of magnitude that could be attributed to differences in the amounts of resuspendable particulate material superficially entrapped upon plant foliage. In spite of these variations, there was reasonably good agreement in the Pu/Am ratio of vegetation samples from each event, irrespective of species differences, where the n value was adequate. This indicates that the $^{239-240}\text{Pu}$ and ^{241}Am generally remains intact in the resuspendable particulate material even though sufficient time has elapsed for some weathering of the original fallout debris. The reader's attention is directed to the resuspension studies presented in the previous NAEG progress report (Dunaway and White, 1974) for information concerning the status of resuspendable material in these aged plutonium fallout areas.

The question is raised concerning the extent to which the contamination on vegetation in these aged fallout areas is attributable to resuspendable material. Earlier studies on fallout from above-ground nuclear detonations (Lindberg, 1959) (Romney, 1963) showed most of the contaminant to be superficially entrapped upon plant foliage. The entrapped debris was of limited size range, and it was very difficult to remove by shaking or washing action. The observation noted in the preceeding paragraph, concerning the good agreement between the Pu/Am ratios

for samples of different plant species, indicates a common source of contamination for each event such as the generally intact fallout debris originally deposited within the downwind fallout pattern. Some casual samplings of new foliage from 5 different shrub species were made periodically during the 1973 spring growth season at Area 5 GMX. Data from composites of 10 clippings from each of 5 tagged shrubs indicated a rapid build-up of superficial contamination to a relatively steady level of activity on the new foliage. For example, the $^{239-240}\text{Pu}$ activity was 33.5 dpm/g ash on Eurotia lanata leaf material, within two weeks after new bud stage, and 39.7 and 39.8 dpm/g in samples taken from the same shrubs at 30 and 60-day intervals. Activity levels on Atriplex canescens foliage at these intervals were 14.4, 33.1, and 44.1 dpm/g ash, and for Grayia spinosa they were 2.2, 5.9, and 7.1 dpm/g ash, respectively. The nature of these deciduous shrubs is such that the new leaf flush bursts to maximum within a few days after onset. Ambrosia dumosa broke dormancy for only a short period, and two samples collected one month apart contained activity levels of 39.1 and 43.6 dpm/g ash. Three samples of foliage collected at monthly intervals from an evergreen shrub, Larrea tridentata, contained activity levels of 13.8, 13.2, and 20.9 dpm/g ash. Strong winds occur very frequently in Area 5-GMX during the spring growth season. Results from the intensive resuspension studies on movement of plutonium by wind-driven forces indicate that the ^{239}Pu deposited in this aged fallout area still represents a significant resuspension source. However, the average air concentration of resuspended ^{239}Pu outside of the exclusion area is only a small fraction of the presently accepted maximum permissible concentration for occupational exposure (Anspaugh and Phelps, 1974).

A more quantitative indication of the extent of superficial contamination on

plant foliage can be observed from the data in Table 2 concerning the mean $^{239-240}\text{Pu}$ contents of vegetation and soil samples and the vegetation/soil activity ratio for samples from each of the fallout areas. Depending upon the location within the activity strata, the $^{239-240}\text{Pu}$ vegetation/soil ratios varied in magnitude from 10^{-2} to 10^{-1} , which was from 100 to 1000 times greater than those discrimination factors previously reported for plant uptake of plutonium through root systems (Jacobson and Overstreet, 1948; Rediske, et al., 1955; Wilson and Cline, 1966; Romney, et al., 1970; Wildung and Garland, 1974). Some preliminary data from root uptake tests now in progress, using potted soil from each of the fallout areas, indicate discrimination factors from 10^{-3} to 10^{-4} (Romney, et al., unpublished data). Another factor to consider is that the vegetation/soil ratios in Table 2 have been derived from comparison to the 0-5 cm layer of surface soil rather than the full depth of contaminated media in which the plant roots actually grow. The plutonium contamination decreases markedly at lower depths in the soil profile (Fowler and Essington, 1974; Gilbert and Eberhardt, 1974); therefore, the actual $^{239-240}\text{Pu}$ vegetation / soil activity ratios would be much greater than those listed in Table 2, if calculated on the total root zone basis. The salient point to be gained from this discussion is that the $^{239-240}\text{Pu}$ contents of vegetation samples collected within these aged fallout areas are much too high for root uptake. The activity levels on vegetation, therefore, must be attributed largely to superficial contamination from resuspendable material.

The standard errors of the mean $^{239-240}\text{Pu}$ contents of vegetation and soil samples listed in Table 2 indicate the variation encountered among samples from

Table 2

$^{239-240}\text{Pu}$ Contents of Vegetation and Soil Samples and the Vegetation/Soil
Ratios for Samples From Aged Pu-Fallout Areas

Activity strata	Vegetation (nCi/g dry tissue)		Soil (nCi/g)		Vegetation/Soil Ratio		
	<u>n</u>	<u>Mean $\pm$ S.E.*</u>	<u>n</u>	<u>Mean $\pm$ S.E.*</u>	<u>n</u>	<u>Ratio $\pm$ S.E.*</u>	<u>r</u>
<u>NTS AREA 5 - GMX (1954-55)</u>							
5	13	.0083 $\pm$.0016	13	.084 $\pm$.030	13	.13 $\pm$.059	.73
1	47	.0092 $\pm$.0020	41	.059 $\pm$.013	42	.16 $\pm$.035	.53
2	24	.064 $\pm$.014	23	.73 $\pm$.15	24	.075 $\pm$.020	.24
3	13	.26 $\pm$.090	13	4.5 $\pm$ 1.2	11	.052 $\pm$.026	-.081
4	17	.31 $\pm$.045	23	7.3 $\pm$ 1.6	17	.050 $\pm$.014	.52
<u>NTS AREA 11-B (1956)</u>							
2	10	.13 $\pm$.034	14	.70 $\pm$.38	7	.16 $\pm$.10	.79
3	14	.19 $\pm$.053	14	5.5 $\pm$ 1.4	9	.039 $\pm$.019	-.070
4	18	.57 $\pm$.098	23	32.8 $\pm$ 7.0	13	.013 $\pm$.0036	.23
<u>NTS AREA 11-C (1956)</u>							
2	11	.11 $\pm$.017	13	.83 $\pm$.41	7	.34 $\pm$.29	-.41
3	14	.33 $\pm$.053	10	2.2 $\pm$.65	8	.13 $\pm$.080	.30
4	17	1.1 $\pm$.41	20	25.1 $\pm$ 7.1	11	.065 $\pm$.021	.40
5	4	1.4 $\pm$.41	6	120.0 $\pm$ 52.0	2	.045	
<u>NTS AREA 11-D (1956)</u>							
2	10	.18 $\pm$.039	11	.97 $\pm$.19	8	.17 $\pm$.040	.41
3	11	.28 $\pm$.088	12	4.3 $\pm$ 1.6	6	.061 $\pm$.069	.35
4	20	.70 $\pm$.14	19	19.1 $\pm$ 6.2	15	.060 $\pm$.021	.61
5	10	1.3 $\pm$.23	15	49.9 $\pm$ 14.0	5	.027 $\pm$.018	-.60

*See footnotes, Table 1.

Table 2 (continued)

Activity strata	Vegetation (nCi/g dry tissue)		Soil (nCi/g)		Vegetation/Soil Ratio		
	<u>n</u>	<u>Mean ± S.E.</u>	<u>n</u>	<u>Mean ± S.E.</u>	<u>n</u>	<u>Ratio ± S.E.</u>	<u>r</u>
<u>NTS AREA 11 - CD OVERLAP (1956)</u>							
6	5	.021 ± .0051	5	.36 ± .14	2	.10 ± .082	
<u>NTS AREA 11 - ABCD (1956)</u>							
1	61	.0034 ± .00052	50	.021 ± .0063	26	.097 ± .099	.093
<u>NTS AREA 13 (1957)</u>							
1	36	.0052 ± .00068	39	.036 ± .0078	35	.15 ± .038	.33
2	25	.013 ± .0020	31	.10 ± .025	24	.14 ± .031	.39
3	15	.17 ± .055	15	.40 ± .075	15	.44 ± .12	.39
4	18	.077 ± .018	19	1.1 ± .15	18	.069 ± .025	-.12
5	10	.28 ± .077	20	2.4 ± .43	10	.10 ± .043	.05
6	37	1.2 ± .35	47	14.0 ± 6.4	37	.078 ± .015	.77
<u>TTR DOUBLE TRACK (1963)</u>							
1	17	.010 ± .0035	24	.12 ± .057	14	.094 ± .088	.67
2	9	.072 ± .029	10	5.7 ± 4.0	7	.024 ± .025	.95
3	11	.11 ± .036	10	2.9 ± .97	8	.035 ± .011	.92
4	11	.49 ± .16	9	44.0 ± 15.0	8	.011 ± .020	.11
<u>TTR CLEAN SLATE 1 (1963)</u>							
1	20	.014 ± .0080	21	.36 ± .17	10	.042 ± .035	.33
2	12	.027 ± .011	14	1.5 ± .25	8	.040 ± .047	-.38
3	19	.012 ± .0033	14	2.6 ± 1.0	8	.0036 ± .0018	.03
4	12	.029 ± .0078	10	2.9 ± 9.7	6	.0074 ± .0072	-.26

Table 2 (continued)

Activity strata	Vegetation (nCi/g dry tissue)		Soil (nCi/g)		Vegetation/Soil Ratio		
	<u>n</u>	<u>Mean ± S. E.</u>	<u>n</u>	<u>Mean ± S. E.</u>	<u>n</u>	<u>Ratio ± S. E.</u>	<u>r</u>
<u>TTR CLEAN SLATE 2 (1963)</u>							
1	18	.0066 ± .0013	18	.086 ± .028	12	.092 ± .039	-.099
2	13	.047 ± .0055	12	1.8 ± .74	9	.025 ± .012	.13
3	12	.090 ± .019	14	6.0 ± 2.3	10	.014 ± .0048	.77
4	20	.14 ± .032	22	6.4 ± 1.5	14	.024 ± .010	.16
<u>TTR CLEAN SLATE 3 (1963)</u>							
1	10	.013 ± .0042	28	.24 ± .046	6	.053 ± .022	.34
2	11	.020 ± .0038	12	1.2 ± .33	7	.021 ± .011	-.41
3	10	.068 ± .015	13	4.6 ± 1.3	8	.015 ± .0062	-.24
4	10	.17 ± .055	10	7.9 ± 3.9	6	.039 ± .080	-.29

different activity strata in these aged fallout areas. In spite of the rather large variations, there were some indications of reasonable agreement between the mean activity levels of $^{239-240}\text{Pu}$ in vegetation and soil samples collected across the different activity strata within each fallout pattern. This relationship is illustrated more clearly on the graphs in Figures 1 to 3. The estimated correlations (r values) are listed in Table 2.

Data listed in Table 2 also show that the $^{239-240}\text{Pu}$ vegetation/soil activity ratios were decreased in the sampling strata containing higher levels of soil contamination. In other words, the vegetation/soil activity ratios increased inversely to the level of contamination in the 0-5 cm surface layer of soil. This can be explained primarily on the basis of the differential particle size distribution which occurred within the fallout patterns of these events. The general rule is that mean fallout particle size decreases with increasing distance downwind from the point of detonation. The resuspendable source material entrapped upon plant foliage is of a limited size range that contributes less activity in proportion to the total contamination deposited closer toward ground zero. Thus, these vegetation/soil activity ratios are smaller in the higher activity strata than they are in the lower activity strata where resuspendable material accounts for a greater proportion of the total contamination present.

Table 2 and Figures 1 to 3 give a comparison of the relative $^{239-240}\text{Pu}$ contamination levels of vegetation and soil samples between the different test events. The highest levels of contamination were found in the samples of vegetation collected from Area 11 at NTS. Vegetation samples collected from the Roller Coaster test areas at TTR generally contained the lowest contamination levels.

Figure 1. Mean $^{239-240}\text{Pu}$ contents of vegetation and soil samples from different activity strata of NTS AREA 5-GMX and AREA 13. (Stratum 5 is a special low activity zone.)

Figure 2. Mean $^{239-240}\text{Pu}$ contents of vegetation and soil samples from different activity strata of tests conducted at NTS AREA 11. (11-CD Stratum 6 is an overlap zone of events C and D; 11-ABCD Stratum 1 is a zone surrounding these events having FIDLER readings less than 5,000 cpm.)

Figure 3. Mean $^{239-240}\text{Pu}$ contents of vegetation and soil samples from different activity strata of the Roller Coaster Tests at TTR.

Pu/Am Ratios Indicate Future Biological Significance of ^{241}Am in the Aged Plutonium Fallout Areas

The concern for ^{241}Am in these plutonium fallout areas stems from its in-growth in the aged source material (Fowler and Essington, 1974). Some soil profile samples show a decrease in the Pu/Am ratio with depth, suggesting that greater vertical movement of ^{241}Am relative to $^{239-240}\text{Pu}$ may have occurred (Fowler and Essington, 1974; Gilbert and Eberhardt, 1974). Findings of Cline, 1967; Wallace, et al., 1969; Hale and Wallace, 1970; Price, 1972, 1973; Wallace, 1971, 1972; and Wallace et al., 1971, present strong evidence that americium is much more readily available to plants through roots than is plutonium. As stated by Fowler and Essington (1974), "It follows that a possible differential availability to plants of $^{239-240}\text{Pu}$ and ^{241}Am , coupled with the increase of ^{241}Am with time, may place ^{241}Am in a position of prime importance as a potential hazard."

The $^{239-240}\text{Pu}$ and ^{241}Am contents and ratios for vegetation and soil samples from the aged fallout areas are presented in Table 3. Sufficient data have been acquired to indicate a reasonably constant Pu/Am ratio for the vegetation samples collected within a given fallout area. This ratio, however, varies among the separate events as the result of differences in the in-growth of ^{241}Am in the aged source materials. Data are listed in the order in which the events occurred. It should be noted here that the NTS Area 5-GMX site was used for 22 different tests involving small amounts of plutonium (Dunaway and White, 1974). Each of the other fallout areas under investigation involved a single test event. The Pu/Am ratios in soil are higher at Area 5-GMX than at the Area 11 sites, and this would have to be explained by differences in ^{241}Am content of the original source materials used in these events. The comparative Pu/Am ratios in soil from the Area 11, Area 13, and TTR events appear to indicate normal ^{241}Am in-growth with respect to passing of time since the initial fallout deposition.

Table 3

²³⁹⁻²⁴⁰Pu and ²⁴¹Am Contents and Ratios for Vegetation
and Soil in Aged Pu-Fallout Areas

Vegetation (nCi/g plant ash on paired samples)						Soil (nCi/g)	
Activity strata	n	²³⁹⁻²⁴⁰ Pu Mean ± S.E.*	²⁴¹ Am Mean ± S.E.*	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.*	r	n	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.*
<u>NTS AREA 5 - GMX (1954-55)</u>							
5	12	.082 ± .012	.0060 ± .0009	13.6 ± .83	.94	13	10.9 ± .60
1	38	.080 ± .019	.0059 ± .0013	13.5 ± .83	.95	22	11.0 ± .95
2	20	.64 ± .13	.053 ± .011	12.0 ± .41	.98	23	10.1 ± .70
3	11	2.2 ± .60	.17 ± .041	12.9 ± .89	.97	13	12.9 ± .89
4	17	2.6 ± .33	.21 ± .025	12.3 ± .52	.95	23	9.7 ± .30
Overall	98	.87 ± .14	.070 ± .011	12.5 ± .25	.98	96	10.2 ± .24
<u>NTS AREA 11 - B (1956)</u>							
2	11	.61 ± .19	.070 ± .021	8.7 ± .37	.99	14	6.1 ± .51
3	14	1.4 ± .52	.19 ± .070	7.4 ± .13	.99	14	8.1 ± .19
4	18	3.3 ± .72	.37 ± .075	9.0 ± 2.0	.78	23	7.7 ± .21
Overall	43	2.0 ± .39	.23 ± .043	8.5 ± 1.0	.87	51	7.7 ± .14
<u>NTS AREA 11 - C (1956)</u>							
2	12	1.1 ± .24	.21 ± .043	5.5 ± .17	.99	13	6.2 ± .39
3	14	2.9 ± .51	.54 ± .099	5.3 ± .20	.98	10	6.1 ± .26
4	17	8.2 ± 3.0	1.5 ± .52	5.3 ± .16	.99	20	6.0 ± .13
5	5	8.5 ± 2.8	1.8 ± .67	4.7 ± .25	.99	4	5.9 ± .30
Overall	48	4.9 ± 1.2	.94 ± .21	5.2 ± .10	.99	47	6.0 ± .08
<u>NTS AREA 11 - D (1956)</u>							
2	11	1.0 ± .20	.19 ± .041	5.4 ± .88	.87	11	5.5 ± .22
3	12	1.7 ± .40	.39 ± .083	4.3 ± .17	.99	12	5.6 ± .20
4	20	4.7 ± 1.0	1.1 ± .22	4.2 ± .31	.98	19	5.9 ± .18
5	11	8.3 ± 1.6	2.2 ± .46	3.7 ± .30	.92	13	5.7 ± .30
Overall	54	4.0 ± .62	1.0 ± .16	4.1 ± .18	.96	55	5.8 ± .15

*See footnotes, Table 1

Table 3 (continued)

Vegetation (nCi/g plant ash on paired samples)						Soil (nCi/g)	
Activity strata	n	²³⁹⁻²⁴⁰ Pu Mean ± S.E.	²⁴¹ Am Mean ± S.E.	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.	r	n	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.
<u>NTS AREA 11 CD OVERLAP (1956)</u>							
6	5	.14 ± .024	.022 ± .0051	6.2 ± .55	.95	5	6.2 ± .26
<u>NTS AREA 11 ABCD (1956)</u>							
1	20	.042 ± .0079	.0081 ± .0018	5.2 ± .54	.91	14	7.4 ± .65
<u>NTS AREA 13 (1957)</u>							
1	37	.067 ± .0087	.0072 ± .0011	9.3 ± .99	.63	24	12.6 ± .93
2	25	.18 ± .027	.018 ± .0024	10.1 ± .62	.90	31	9.9 ± 3.5
3	15	2.7 ± .88	.24 ± .073	11.3 ± 1.5	.92	18	9.1 ± .39
4	19	1.0 ± .23	.14 ± .025	7.7 ± .62	.86	21	8.9 ± .18
5	12	6.3 ± 1.6	.67 ± .17	9.4 ± .48	.97	20	8.8 ± .28
6	31	27.3 ± 9.8	2.5 ± .87	7.5 ± .33	.99	49	9.4 ± .20
Overall	137	5.0 ± 1.6	.63 ± .20	7.9 ± .20	.99	165	9.4 ± .15
<u>TTR DOUBLE TRACK (1963)</u>							
1	10	.071 ± .015	.0047 ± .0012	15.0 ± 8.7	.68	7	26.6 ± .85
2	9	.36 ± .13	.021 ± .0075	16.8 ± .81	.99	8	27.2 ± 2.5
3	11	.53 ± .17	.029 ± .0080	18.4 ± 1.4	.97	10	25.8 ± .64
4	11	2.6 ± .82	.17 ± .037	15.2 ± 2.8	.62	8	22.5 ± 1.2
Overall	41	.95 ± .27	.060 ± .015	15.8 ± 1.4	.79	33	23.5 ± .73
<u>TTR CLEAN SLATE 1 (1963)</u>							
1	4	.29 ± .16	.017 ± .0085	17.5 ± 1.5	.99	11	34.9 ± 2.8
2	8	.096 ± .026	.0068 ± .0021	14.1 ± .78	.98	13	29.3 ± 1.5
3	9	.053 ± .016	.0046 ± .0017	11.5 ± 1.2	.95	13	23.9 ± .36
4	10	.19 ± .049	.011 ± .0029	16.8 ± .90	.98	7	24.0 ± .78
Overall	31	.14 ± .029	.0089 ± .0017	16.2 ± .52	.98	44	26.0 ± .76

Table 3 (continued)

Activity strata	Vegetation (nCi/g plant ash on paired samples)					Soil (nCi/g)	
	<u>n</u>	²³⁹⁻²⁴⁰ Pu Mean ± S.E.	²⁴¹ Am Mean ± S.E.	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.	<u>r</u>	<u>n</u>	²³⁹⁻²⁴⁰ Pu/ ²⁴¹ Am Ratio ± S.E.
<u>TTR CLEAN SLATE 2 (1963)</u>							
1	15	.040 ± .0079	.0031 ± .00064	12.9 ± 1.1	.95	13	19.6 ± .68
2	13	.26 ± .032	.019 ± .0022	13.6 ± .81	.84	11	20.1 ± .58
3	12	.44 ± .10	.034 ± .0066	12.8 ± 1.0	.92	13	23.8 ± .80
4	20	.68 ± .13	.062 ± .023	10.8 ± 1.3	.84	22	21.8 ± .67
Overall	60	.38 ± .057	.033 ± .0081	11.6 ± .64	.84	59	22.2 ± .41
<u>TTR CLEAN SLATE 3 (1963)</u>							
1	8	.075 ± .023	.0035 ± .0011	21.3 ± 1.3	.99	22	22.1 ± 1.3
2	10	.10 ± .017	.056 ± .00080	18.6 ± 1.4	.87	13	22.4 ± .42
3	10	.32 ± .067	.020 ± .0038	16.3 ± 1.4	.93	13	21.8 ± .70
4	15	.65 ± .15	.039 ± .0076	16.9 ± 1.8	.80	12	22.0 ± .44
Overall	43	.34 ± .067	.020 ± .0036	17.0 ± .93	.87	60	22.0 ± .28

The mean Pu/Am ratios for soil samples generally are higher than they are for vegetation samples collected from each of the activity strata, except at Area 5-GMX. This is more apparent for the samples from the test areas on the Tonopah Test Range than from Areas 11 or 13 on the Nevada Test Site. We attribute these lower Pu/Am ratios on vegetation samples to preferential root uptake of the more biologically available ^{241}Am from the soil of these aged fallout areas. Preliminary data from plant uptake experiments in progress, using plutonium fallout contaminated soil from the different study areas, indicate a much greater uptake of ^{241}Am through roots than of $^{239-240}\text{Pu}$. For example, the mean Pu/Am ratio for wheat straw in one test was 2.81 when grown on potted soil having a mean Pu/Am ratio of 6.74. The vegetation samples collected from these aged fallout areas, therefore, contain not only the ^{241}Am contributed from the superficial contaminant but also that which has been taken up through plant roots.

We are at a loss to explain the higher Pu/Am ratios for vegetation samples than were found for corresponding soil samples at Area 5-GMX, unless there has been a peculiar partitioning of $^{239-240}\text{Pu}$ relative to ^{241}Am in the resuspendable particulate material compared to that of the total contaminant deposited upon the soil. The situation could be extremely complex as the result of the 22 separate tests conducted at this site. Further studies of the distribution of $^{239-240}\text{Pu}$ and ^{241}Am relative to the particle size of source materials will be needed to help explain this apparent anomaly.

An Estimate of the Contribution of Vegetation to the Plutonium Inventory of Aged Fallout Areas

Measurements of the vegetational biomass in contaminated areas are being made in order to estimate the contribution of $^{239-240}\text{Pu}$ by vegetation to the

Table 4

Estimated Inventory of $^{239-240}\text{Pu}$ for Vegetation in Area 13

Activity strata	n	Mean $\pm$ S. E. * (nCi/gm)	Mean $\pm$ S. E. ** (nCi/m 2)	Estimated Inventory $\pm$ S. E. (Curies)	Percent of Veg. Inv.	Soil Inv. Veg. Inv.
1	36	.0052 $\pm$.00068	1.5 $\pm$.54	.0013 $\pm$.00047	5	1300
2	25	.013 $\pm$.0020	3.8 $\pm$ 1.4	.0096 $\pm$.0036	36	1600
3	15	.17 $\pm$.055	49.0 $\pm$ 23.0	.0040 $\pm$.0019	15	500
4	18	.077 $\pm$.018	22.0 $\pm$ 9.1	.0016 $\pm$.00067	6	2400
5	10	.28 $\pm$.077	81.0 $\pm$ 35.0	.0017 $\pm$.00072	6	1300
6	37	1.2 $\pm$.35	348.0 $\pm$ 150.0	.0081 $\pm$.0036	31	2300
Total	141			.0264 $\pm$.0055	99 AVE.	=1600

* From Table 2

 $^{**}\text{Mean (nCi/m}^2\text{)} = (290 \text{ gm/m}^2) \times (\text{Mean (nCi/gm)}).$

S. E. $\simeq [(290)^2 \text{ Var (Mean nCi/gm)} + 9360 (\text{Mean nCi/gm})^2]^{\frac{1}{2}}$ where 9360 $\simeq$ variance of estimated mean biomass of 290gm/m 2 in Area 13 (data from Table 2 of Romney et al. (1974)).

plutonium inventory of the aged fallout areas. These measurements have been completed for Area 13 and the data were included in an earlier progress report (Romney et al., 1974). The estimated inventory of $^{239-240}\text{Pu}$ for vegetation in Area 13 is given for different activity strata in Table 4. For these estimates, the mean $^{239-240}\text{Pu}$ contents of dry plant tissue were converted to nCi/m^2 using the biomass conversion factor of 290 g/m^2 . These values were then multiplied by the appropriate area factors (see the report of Gilbert and Eberhardt, 1974) in order to estimate the inventory of $^{239-240}\text{Pu}$ for vegetation in each activity strata. The highest contributions of that contained on vegetation were 36, 31 and 15% for activity strata 2, 6 and 3, respectively (column 6 of Table 4). The standard errors are only approximate and probably too small since they are based on regression estimates of biomass. Nevertheless, the results are good enough to give reasonable estimates. Comparisons of the soil and vegetation inventory estimates indicate that vegetation contributes a rather insignificant portion of the total amount of $^{239-240}\text{Pu}$ present in this aged fallout area. It would appear, therefore, that the amounts of contaminant involved in vegetation-transport through the food chains of grazing animals and man would be relatively small in comparison to the total amounts deposited on soil.

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APPENDIX

Plots of $^{239-240}\text{Pu}$ and ^{241}Am Activity Levels in Vegetation Samples Collected from Aged Plutonium Fallout Areas

AREA 5 - GMX STRATA 5

AREA 5 - GMX STRATA 1

AREA 5 - GMX STRATA 4

AREA 11 ABCD STRATA 1

AREA 11 B STRATA 4

AREA 11 C STRATA 2

AREA 11 D STRATA 5

AREA 11 CD OVERLAP STRATA 6

AREA 13 STRATA 3

AREA 13 STRATA 4

AREA 13 STRATA 5

AREA 13 STRATA 6

TTR DOUBLE TRACK STRATA 1

TTR DOUBLE TRACK STRATA 2

TTR DOUBLE TRACK STRATA 3

DOUBLE TRACK STRATA 4

TTR CLEAN SLATE 2 STRATA 1

TTR CLEAN SLATE 2 STRATA 2

TTR CLEAN SLATE 2 STRATA 3

TTR CLEAN SLATE 2 STRATA 4

TTR CLEAN SLATE 3 STRATA 1

TTR CLEAN SLATE 3 STRATA 2

TTR CLEAN SLATE 3 STRATA 3

TTR CLEAN SLATE 3 STRATA 4

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