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$^{239-240}Pu$ AND ^{241}Am CONTAMINATION OF VEGETATION IN AGED FALLOUT AREAS

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by

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ABSTRACT

Vegetation studies in aged plutonium fallout areas showed variations in the $^{239-240}Pu$ and ^{241}Am contamination levels attributable to differences in the amounts of resuspendable particulate material superficially entrapped upon plant foliage. There was reasonable agreement between the mean activity levels in vegetation and soil samples collected across different activity strata defined by FIDLER survey instrument within each fallout area. The

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ratio of vegetation Pu to soil Pu tended to increase moving out from higher to lower activity strata, which might reflect the increasing proportion of finer particulate material initially deposited in fallout debris at greater distance from ground zero. The Pu/Am ratio was reasonably constant for vegetation samples collected from a given fallout area. This ratio, however, varied among separate test events primarily as the result of differences in the ingrowth of ^{241}Am within the aged source materials. Inventory estimates indicate that standing vegetation contributes a rather insignificant portion of the total contaminant remaining in these aged fallout areas.

1. INTRODUCTION

Several safety tests were conducted between 1954 and 1963 at the Nevada Test Site (NTS) and the Tonopah Test Range (TTR) in the State of Nevada, U.S.A., wherein small amounts of plutonium were dispersed into the surrounding environment by chemical explosives. The Nevada Applied Ecology Group (NAEG) is currently engaged in environmental studies at several of these aged fallout areas, some of which have been exposed to the effects of weathering for more than 20 years. The objectives of the NAEG plutonium program include studies to estimate the total amount and geographical distribution of $^{239-240}Pu$ and the ingrowth product, ^{241}Am, in vegetation and soil of these aged fallout areas. Detailed results from the work underway appear in a series of progress reports [1], [2].

This paper presents, in summary, some pertinent findings concerning $^{239-240}Pu$ and ^{241}Am contamination of the perennial vegetation in these fallout areas. The vegetation involved includes shrub species common to the southern Great Basin and northern Mohave Deserts. Although findings from these desert environments might not be directly applicable to contrasting ecosystems, similar contamination processes probably function merely to a greater or lesser degree wherever plutonium is disseminated into terrestrial environments.

The significance of vegetation in any plutonium contaminated area rests primarily upon its capacity to function as the carrier for plutonium and other transuranics in the food chains leading to grazing animals and man. Two different mechanisms of incorporation are involved in this transport process. First, the contaminant may become superficially entrapped upon vegetation through the processes of resuspension. This is expected to be the most important mechanism in the desert ecosystem where environmental conditions are favorable to wind-driven processes. Earlier studies concerning fission product fallout from nuclear detonations disclosed that superficially contaminated vegetation was a major source of radionuclides to grazing animals [3], [4], [5], [6]. Second, the plutonium disseminated on soil may be taken up through plant roots and translocated to the above ground vegetation. This internal incorporation mechanism, however, should be less important than superficial contamination at the sites under investigation, in view of evidence from earlier studies indicating relatively low uptake of plutonium through plant roots [7], [8], [9], [10], [11], [12], [13], [14]. The NAEG vegetation studies are expected to contribute information on how these mechanisms of incorporation function in the vegetation-carrier transport of plutonium and other transuranic elements from contaminated soil to grazing animals.

2. MATERIALS AND METHODS

Samples of perennial vegetation were collected in conjunction with the sampling of soil for an inventory of plutonium in 10 aged fallout areas at NTS and TTR. The sampling procedure first involved the division of each study area into subregions or strata of contamination using the FIDLER[1] survey instrument. Details of the stratified random sampling methods and the statistical analysis methods used for the NAEG program have been reported by Gilbert and Eberhardt [15] and Gilbert et al. [16]. 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 m^2 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 $^{239-240}Pu$ and ^{241}Am in these large vegetation samples have been reported by Major et al. [17].

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[1] Field Instrument for the Determination of Low-Energy Radiation.

3. RESULTS AND DISCUSSION

Detailed results from vegetation studies conducted in the aged fallout areas have been reported in recent NAEG progress reports [16], [18], [19]. Examples of data have been extracted from these reports and presented herein to illustrate pertinent findings concerning $^{239-240}Pu$ and ^{241}Am contamination of vegetation and soil at this point in time. The "safety-shot" tests at NTS were conducted at Area 5 (GMX) in 1954-55, at Area 11 (Plutonium Valley) in 1956, and at Area 13 (Project 57) in 1957. Tests at TTR (Roller Coaster) were conducted in 1963.

3.1 $^{239-240}Pu$ And ^{241}Am Contamination of Perennial Vegetation

The highest levels of contamination were found in samples of vegetation collected from Area 11 at NTS. Vegetation samples collected from the Roller Coaster test sites at TTR generally contained the lowest levels of contamination.

Inasmuch as the perennial vegetation was sampled at random, it naturally followed that different shrub species would become involved at those sites where species diversity occurred. Table I gives some examples from the two sites having greatest species diversity: NTS Area 5 (GMX), and Area 13 (Project 57). Some plant species were relatively sparse in different areas as indicated by the small n values. The perennial vegetation of the Clean Slate sites at TTR was essentially Atriplex confertifolia (Torr. & Frem.) Wats. The variations in $^{239-240}Pu$ contents between different species were of an order of magnitude that could be attributed to differences in the amounts of resuspendable particulate material superficially entrapped upon 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 $^{239-240}Pu$ and ^{241}Am generally has remained intact in the resuspendable contaminant even though sufficient time has elapsed for some weathering of the original fallout debris.

Of primary importance to an understanding of the environmental plutonium problem is the question concerning the extent to which the contamination of vegetation is attributable to resuspendable material. Earlier studies on fallout from nuclear detonations showed most of the contaminant to be superficially entrapped upon plant foliage [3], [5], [20], [21], [22]. The entrapped debris was of limited size range, and was very difficult to remove by shaking or washing action. Species such as Eurotia lanata (Pursh) Moq. (Table I) generally retained the highest levels of contamination because of their hairy leaf surfaces and long-haired fruiting involucres. The observation mentioned in the preceeding paragraph concerning the good agreement between the Pu/Am ratios among 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 $^{239-240}Pu$ 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 (Pursh) Nutt. foliage at these intervals were 14.4, 33.1, and 44.1 dpm/g ash, and for *Grayia spinosa* (Hook) Moq. 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* (Gray) Payne 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* Ses. & Moq. 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 NAEG resuspension studies on movement of plutonium by wind-driven processes within the fallout pattern 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 [23]. Earlier studies at the Project 57 site in Area 13 showed that contamination was present in resuspended material of respirable particle size during the period of initial cloud passage and also when measured during resuspension studies from D + 3 to D + 28 days and one year after fallout was deposited [24].

A more quantitative indication of the extent of superficial contamination on plant foliage can be observed from the data in Table II concerning the mean $^{239-240}$Pu contents of vegetation and soil samples and the vegetation/soil activity ratio for samples from several fallout areas. Depending upon the location within the activity strata, the

$^{239-240}Pu$ vegetation/soil ratios[2] varied in magnitude from 10^{-2} to 10^{-1}, which was from 100 to 1000 times greater than the amount indicated by discrimination factors previously reported for plant uptake of plutonium through root systems [7], [8], [9], [11], [13], [25], [26]. Some preliminary data from root uptake test using potted soil from each of these fallout areas, indicate discrimination factors from 10^{-4} to 10^{-5} (Romney, et al., unpublished data). Another point to consider is that the vegetation/soil ratios in Table II have been derived from comparison to the top 5 cm layer of soil rather than the full depth of contaminated media in which roots grow. The plutonium contamination decreases markedly at lower depths in the soil profile of these fallout areas [11], [16], [27]; therefore, the actual $^{239-240}Pu$ vegetation/soil activity ratios (i.e., discrimination factors) would be much greater than those listed in Table 2, if calculated on the total root zone basis. The important point of this discussion is that the $^{239-240}Pu$ contents of vegetation samples collected within these aged fallout areas simply are much too high for root uptake. The activity levels on vegetation, therefore, must be attributed large to superficial contamination from resuspendable material.

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

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[2] Commonly referred to as 'discrimination factor,' or 'concentration factor.'

in vegetation and soil samples collected across the different activity strata within each fallout pattern.

Data listed in Table II also show that the $^{239-240}Pu$ vegetation/soil activity ratios 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 top 5 cm surface layer of soil. This can be explained primarily on the basis of the differential particle size distribution which initially occurred within the fallout patterns of these events. The general rule was that the mean fallout particle size decreased with increasing distance downwind from the point of detonation. However, the resuspendable source material that becomes entrapped upon plant foliage represents a limited size range (< 10 μm dia. [23]) of particles which contribute less activity in proportion to the total contamination deposited at points nearer to ground zero. Thus, these vegetation/soil activity ratios are smaller in the higher activity strata near ground zero than they are out in the lower activity strata where finer particulate material accounts for a greater proportion of the total contamination present.

3.2 Biological Significance of ^{241}Am in the Aged Plutonium Fallout Areas

The concern for ^{241}Am in these plutonium fallout areas stems from its ingrowth in the aged source material [27]. Some soil profile samples show a decrease in the Pu/Am ratio with depth, indicating greater vertical movement of ^{241}Am relative to $^{239-240}Pu$ [16], [27]. Findings reported in the literature [13], [14], [28], [29], [30], [31], [32], present strong evidence that americium is much more readily available to plants through roots than is plutonium. As stated by Fowler and Essington [27], "It follows that a possible differential availability to plants of $^{239-240}Pu$ 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 overall average $^{239-240}Pu$ and ^{241}Am ratios for vegetation and soil samples from the aged fallout areas are presented in Table III. 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 [1]. 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.

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 in 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 $^{239-240}Pu$. For example, the mean Pu/Am ratio for wheat straw in one test was 3.79 (n = 18) when grown on potted soil having a mean Pu/Am ratio of 6.73 (n = 12).

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 $^{239-240}Pu$ 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 $^{239-240}Pu$ and ^{241}Am relative to the particle size of source materials will be needed to help explain this apparent anomaly.

3.3 Contribution of Vegetation to the Plutonium Inventory of Aged Fallout Areas

Vegetational biomass at study sites was estimated, using nondestructive dimensional analysis [33], in order to estimate the inventory of $^{239-240}$ for vegetation in the contaminated areas. Data from Area 5-GMX and Area 13 are given in Table IV. It should be understood that these vegetation inventories are only approximate since a) the average biomass data are approximated using dimensional analysis, b) the average biomass data are based on small numbers of 2 x 50 m plots within large areas, c) all species have been lumped together to get an average radionuclide concentration, and d) the formula for S. E. for inventory is only approximate, being based upon a Taylor series expansion. Nevertheless, these results are sufficiently accurate to give a general impression of the relative magnitudes of contamination for vegetation versus that of soil. These comparisons indicate that the standing vegetation contributes a rather insignificant portion of the total amount of $^{239-240}Pu$ present in these aged fallout areas. It would appear, therefore, that the amount of contaminant involved in vegetation-carrier transport through food chains of grazing animals and men would be relatively small compared to the total amount deposited on soil of similar ecosystems.

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

Plant species	Activity strata	n	Vegetation (nCi/g dry tissue): $^{239-240}Pu$ Mean ± S.E.[a]	Vegetation (nCi/g dry tissue): $^{239-240}Pu/^{241}Am$ Ratio ± S.E.[b]
NTS AREA 5 - GMX (1954-55)				
Atriplex confertifolia	5	6	.012 ± .0030	13.4 ± 1.0
	1	27	.011 ± .0030	15.0 ± 1.3
	2	10	.084 ± .031	11.9 ± 0.66
	3	4	.31 ± .073	14.7 ± 1.6
	4	9	.28 ± .047	12.4 ± 0.85
Larrea tridentata	5	6	.0053 ± .0011	13.6 ± 1.4
	1	15	.0040 ± .0016	13.4 ± 1.5
	2	14	.050 ± .010	12.2 ± 0.51
	3	7	.11 ± .036	10.7 ± 1.1
	4	7	.34 ± .095	11.8 ± 0.61
NTS AREA 13 - PROJECT 57 (1957)				
Atriplex canescens	1	2	.0034 ± .0024	17.5 ± 6.1
	3	4	.13 ± .073	9.2 ± 0.58
	4	9	.041 ± .0076	7.1 ± 0.73
	5	3	.12 ± .0052	8.1 ± 0.47
	6	9	.40 ± .11	9.5 ± 0.65
Atriplex confertifolia	1	31	.0047 ± .00056	7.3 ± 0.76
	2	24	.013 ± .0021	10.1 ± 0.62
	3	4	.055 ± .0085	10.2 ± 0.55
	4	5	.073 ± .0027	7.4 ± 1.1
	5	2	.29 ± .016	11.6 ± 0.72
	6	11	.87 ± .24	7.0 ± 0.61
Eurotia lanata	3	4	.18 ± .082	12.9 ± 3.8
	4	3	.19 ± .083	8.0 ± 2.2
	5	5	.37 ± .15	8.8 ± 0.37
	6	11	2.6 ±1.0	8.0 ± 0.26

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

[b] If $R = \overline{Y}/\overline{X}$, then:

$$S.E. = \left\{\left[\sum(y_i^2/x_i) - (\sum y_i)^2/\sum x_i\right] / (n-1)\sum x_i\right\}^{\frac{1}{2}} \qquad [34]$$

TABLE II. $^{239-240}$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		
	n	Mean ± S.E.[a]	n	Mean ± S.E.[a]	n	Ratio ± S.E.[a]	r
NTS AREA 5 - GMX (1954-55)							
5	13	.0083 ± .0016	13	.084 ± .030	13	.13 ± .059	.73
1	47	.0092 ± .0020	41	.059 ± .013	42	.16 ± .035	.53
2	24	.064 ± .014	23	.73 ± .15	24	.075 ± .020	.24
3	13	.26 ± .090	13	4.5 ± 1.2	11	.052 ± .026	-.081
4	17	.31 ± .045	23	7.3 ± 1.6	17	.050 ± .014	.52
NTS AREA 11-D (1956)							
2	10	.18 ± .039	11	.97 ± .19	8	.17 ± .040	.41
3	11	.28 ± .088	12	4.3 ± 1.6	6	.061 ± .069	.35
4	20	.70 ± .14	19	19.1 ± 6.2	15	.060 ± .021	.61
5	10	1.3 ± .23	15	49.9 ± 14.0	5	.027 ± .018	-.60
NTS AREA 13 (1957)							
1	36	.0052 ± .00068	39	.036 ± .0078	35	.15 ± .038	.33
2	25	.013 ± .0020	31	.10 ± .025	24	.24 ± .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
TTR DOUBLE TRACK (1963)							
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

[a] See footnotes, Table I.

TABLE III. Overall Average $^{239-240}Pu$ and ^{241}Am Ratios for Vegetation and Soil in Aged Fallout Areas

Study area	Vegetation (nCi/g ash)			Soil (nCi/g)		
	n	Average Pu/Am Ratio[a] ± S.E.[b]	r	n	Average Pu/Am Ratio[a] ± S.E.[b]	r
NTS AREA 5 - GMX (1954-1955)	98	12.5 ± 0.25	0.98	96	10.2 ± 0.24	0.98
NTS AREA 11-B (1956)	43	8.5 ± 1.0	0.87	51	7.7 ± 0.14	0.99
NTS AREA 11-C (1956)	48	5.2 ± 0.10	0.99	47	6.0 ± 0.08	0.99
NTS AREA 11-D (1956)	54	4.1 ± 0.18	0.96	55	5.8 ± 0.15	0.99
NTS AREA 13 (1957)	137	7.9 ± 0.20	0.99	165	9.4 ± 0.15	0.99
TTR DOUBLE TRACK (1963)	41	15.8 ± 1.40	0.79	33	23.5 ± 0.73	0.99
TTR CLEAN SLATE 1 (1963)	31	16.2 ± 0.52	0.98	45	23.2 ± 1.00	0.99
TTR CLEAN SLATE 2 (1963)	60	11.6 ± 0.64	0.84	59	22.2 ± 0.41	0.99
TTR CLEAN SLATE 3 (1963)	43	17.0 ± 0.93	0.87	60	22.0 ± 0.28	0.99

a $\Sigma y_i / \Sigma x_i$

b See footnote, TABLE I

TABLE IV. Estimated Inventory of $^{239-240}Pu$ for Perennial Vegetation in Aged Fallout Areas

Activity strata	n	Mean ± S.E. (nCi/g dry)	Mean[a] ± S.E.[b] (nCi/m^2)	Estimated Inventory ± S.E. (μ Curies)	Percent inventory by strata	Soil/Veg. inventory ratio
NTS AREA 5 - GMX (1954-1955)						
5	13	.0083 ± .0016	1.8 ± .47	10 ± 2.6	1	2500
1	47	.0092 ± .002	2.1 ± .56	450 ± 120	52	1500
2	24	.0064 ± .014	14 ± 3.9	210 ± 56	24	3000
3	12	.26 ± .090	58 ± 22	93 ± 35	11	4600
4	17	.31 ± .045	69 ± 15	97 ± 21	11	7300
Total	113			860 ± 140	99	AVE. = 2900
NTS AREA 13 (1957)						
1	36	.0052 ± .0007	1.5 ± .27	1300 ± 236	5	1300
2	25	.013 ± .002	3.8 ± .73	9600 ± 1900	36	1600
3	15	.17 ± .055	49 ± 17	4000 ± 1400	15	500
4	18	.077 ± .018	22 ± 5.8	1600 ± 430	6	2400
5	10	.28 ± .077	81 ± 24.3	1700 ± 510	6	1300
6	37	1.2 ± .35	348 ±109	8100 ± 2500	31	2300
Total	141			26400 ± 3500	99	AVE. = 1600

[a] Mean (nCi/m^2) ≃ (mean biomass in g/m^2) x (mean $^{239-240}Pu$ concentration in nCi/g)

[b] $$S.E. \simeq [(\text{mean } g/m^2)^2 \times \text{Var (mean nCi/g)} + (\text{mean nCi/g})^2 \times \text{Var (mean } g/m^2) - \text{Var (mean nCi/g)} \times \text{Var (mean } g/m^2)]^{\frac{1}{2}}$$ [35]

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