White et al. 1981

Reprinted from the *Journal of Environmental Quality*
Vol. 10, no. 3, July-September 1981, Copyright © 1981, ASA, CSSA, SSSA
677 South Segoe Road, Madison, WI 53711 USA

ER Record I.D.# 0011994

Factors Affecting Radionuclide Availability to Vegetables Grown at Los Alamos

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ABSTRACT

A field study was conducted in 1977 on ^{238,239}Pu and ^{137}Cs availability to zucchini squash (*Curcurbita melopepo*, hybrid seneca) and green bush beans (*Phaseolus vulgaris*, Landreths stringless) grown under home-garden conditions in an area at Los Alamos National Laboratory used for treated radioactive liquid waste disposal. Radionuclide concentrations were measured as a function of tissue type, height above the soil, fertilization regime, and for the squash, food-cleansing procedures. Analysis of variance procedures were used to analyze the data.

Ratios of the concentration of a radionuclide in oven-dried vegetation to dry soil ranged from 0.0004 to 0.116 for the Pu isotopes, and from 0.051 to 0.255 for ^{137}Cs. Fertilization with cattle manure reduced the Pu concentration ratios by 30% and ^{137}Cs by 50%. Vegetative parts sampled within 20 cm of the ground surface were contaminated about four times as much as those parts growing further from the ground surface. About 65% of the contamination was removed by washing, indicating the presence of surficial contamination. The 50-year radiation dose commitment to humans consuming vegetables from the garden plot would be less than 0.05 mrem and would be due almost entirely to ^{137}Cs.

Additional Index Words: **radiation, fertilizer, soil splash-up.**

White, G.C., T. E. Hakonson, and A. J. Ahlquist. 1981. Factors affecting radionuclide availability to vegetables grown at Los Alamos. J. Environ. Qual. 10:294–299.

The need for information on transuranic element transfer into human food chains has prompted many studies on plutonium (Pu) uptake by agricultural plants. Most studies have been conducted under controlled laboratory or greenhouse conditions using known chemical forms of Pu (Price, 1973; Schreckhise and Cline, 1980; Schulz, 1977). Fewer studies deal with contaminated field sites where Pu transport to vegetation is likely a function of the contaminant source and local environmental factors (Adriano et al., 1980; Hakonson, 1975; Watters et al., 1980; Little and Whicker, 1978). Studies dealing with a wide variety of contaminant sources and ecological settings are required to identify the generic and site-specific attributes of environmental Pu behavior.

This paper presents the results of a field study conducted in 1977 on Pu and ^{137}Cs availability to zucchini squash (*Curcurbita melopepo*, hybrid seneca) and green beans (*Phaseolus vulgaris*, Landreths stringless) grown in Mortandad Canyon, an area at Los Alamos used for treated radioactive liquid waste disposal. The study was designed to simulate a home vegetable garden. Concentrations of radionuclides were determined in the two vegetable crops as a function of tissue, height above the ground surface, fertilization with steer manure, and food preparation treatment. The radiation doses to humans from eating the vegetable crop were calculated to assess the potential hazard from agricultural use of the area.

MATERIALS AND METHODS

Industrial liquid wastes generated by the Los Alamos National Laboratory are collected at a central location, treated to reduce the levels of radioactive contaminants to below Federal guidelines for drinking water, and then released to the local environs (USDOE, 1979).

Mortandad Canyon has been used for disposal of this liquid waste since 1963. The quantities of ^{238}Pu, ^{239,240}Pu (denoted as ^{239}Pu in this paper), and ^{137}Cs released to the canyon from 1963 through 1977 were 0.051, 0.039, and 0.85 Ci, respectively (USDOE, 1979). Prior to 1968 the isotopic composition of Pu in the waste was predominately ^{239}Pu. Subsequently, more ^{238}Pu than ^{239}Pu was processed so that the ^{238}Pu inventory is now greater than the ^{239}Pu inventory.

A 9- by 13-m plot was established in Mortandad Canyon about 2,500 m downstream from the liquid effluent outfall on an alluvial floodplain formed from storm runoff. The plot was fenced and commercial garden preparation (6%N–12%P–6%K) was applied just prior to cultivation of the soil. The soil is an unnamed member of the Typic Ustorthents group. One-half of the plot received an additional amendment of cattle manure (hereafter referred to as the fertilization treatment) applied at the rate of 5 kg/m^2 (dry weight). A garden rototiller was used to cultivate the plot to a 15-cm depth. The plot was seeded with zucchini squash and green beans.

The term "plant" as used in this paper refers to the above-ground vegetative parts. The term "fruit" refers to the edible reproductive parts. Plant roots were not sampled during this study. Green beans were sampled at maturity by pooling beans from four randomly selected plants within each plot. A total of 13 samples of bean fruit was collected. Green bean plants were also collected and bagged separately.

Squash fruits were collected at maturity from individual plants. Squash plants were sampled as a function of height above the ground surface with a 1-m^2 template divided into four equal quadrants; the heights from which samples were taken were 0–20 cm and >20 cm above the ground surface. Bean plants were not collected as a function of height above the ground.

To evaluate the effect of washing on radionuclide concentrations, squash plants and fruit from two of the quadrants in a particular height strata were pooled together for a wash treatment, while the samples from the remaining two quadrants were analyzed for radionuclide content without the wash treatment. All squash fruit and plant samples designated for the wash treatment were thoroughly washed with water in an attempt to remove surficially attached soil particles. Although the wash treatment appeared to be effective in removing coarse particles, it may not have effectively removed the smaller sized particles. All bean plant and fruit samples were washed before the radionuclide analyses.

Soil (minus roots) was collected for chemical, physical, and radionuclide analyses from the rooting zone of individual plant samples. Thus, the depth to which soil samples were taken varied depending on the depth of root penetration by the two vegetable species.

Samples were dried at 100°C and then ashed at 400°C. Samples were analyzed for ^{137}Cs on 10- by 20-cm NaI(Tl) detector coupled to a multichannel analyzer. The samples were then subjected to wet chemical techniques followed by alpha spectroscopy to quantify the Pu content (USDOE, 1979). Most of the chemical and physical analyses on garden-plot soils were conducted by Colorado State University's Soil Testing Laboratory in Fort Collins, Colo (Soltanpour et al., 1979). Soil particle size analyses were performed in our laboratory using mechanical sonic sieving methods (Nyhan et al., 1976).

[1] Research funded under Contract No. W-7405-ENG-36 between the U.S. Department of Energy and the Los Alamos National Laboratory, Los Alamos, NM 87545. Received 23 June 1980.

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STATISTICAL ANALYSIS

Analysis of variance (ANOVA) procedures involving nested factors were used to analyze the vegetable crop data. Calculations were made with the multivariate ANOVA (MANOVA) procedure (Cohen and Burns, 1977) of SPSS (Nie et al., 1975). Because nested factors are random effects, e.g., the ANOVA model is a mixture of fixed and random effects, the F ratios are not constructed with a common error term in the denominators. Rather, the F ratios utilize different error terms depending on the nesting structure. The complexity of the linear models used necessitates the following complete descriptions.

Bean data were analyzed as a three-factor experiment (Winer 1971:361-363, 514-539) with plants nested within fertilization plots. The linear model utilized was

$$y_{ijk} = \mu + \alpha_i + \beta_{j(i)} + \gamma_k + \alpha\gamma_{ik} + \beta\gamma_{j(i)k} + e_{ijk}, \qquad [1]$$

where

y_{ijk} is the radionuclide concentration for the ijk^{th} cell;
μ is the overall mean;
α_i with $i = 1, 2$ is the fertilizer effect;
$\beta_{j(i)}$ with $j = 1, \ldots, 13$ is the plant effect nested within fertilizer plots;
γ_k with $k = 1, 2$ is the tissue effect, e.g., stem and leaves or fruit; and
e_{ijk} is the error term for the ijk^{th} cell.

Squash plant data were analyzed as a four-factor design (Winer 1971:539-559) with plants nested within fertilization plots. The linear model utilized was

$$y_{ijkl} = \mu + \alpha_i + \beta_{j(i)} + \gamma_k + \delta_l + \alpha\gamma_{ik} + \alpha\delta_{il} + \gamma\delta_{kl} + \alpha\gamma\delta_{ikl} + \beta\gamma_{j(i)k} + \beta\delta_{j(i)l} + \beta\gamma\delta_{j(i)kl} + e_{ijkl}, \qquad [2]$$

where

y_{ijkl} is the radionuclide concentration for the $ijkl^{th}$ cell,
α_i with $i = 1, 2$ is the effect either with or without fertilizer,
$\beta_{j(i)}$ with $j = 1, \ldots, 8$ is the plant effect nested within fertilizer plots,
γ_k with $k = 1, 2$ is either the lower or upper portions of plant (height),
δ_l with $l = 1, 2$ is either the washed or unwashed tissue effect, and
e_{ijkl} is the error term for the $ijkl^{th}$ cell.

Squash fruit were also analyzed with a four-factor hierarchial design. The linear model utilized was

$$y_{ijkl} = \mu + \alpha_i + \beta_{j(i)} + \gamma_{k(j)} + \delta_l + \alpha\delta_{il} + \beta\delta_{j(i)l} + \gamma\delta_{k(j)l} + e_{ijkl}$$

where

y_{ijkl} is the radionuclide concentration for the $ijkl^{th}$ cell,
α_i with $i = 1, 2$ is the effect either with or without fertilizer,
$\beta_{j(i)}$ with $j = 1, \ldots, 8$ is the plant effect nested within fertilizer plots,
$\gamma_{k(j)}$ with $k = 1, \ldots, 3$ is the fruit effect nested within plants,
δ_l with $l = 1, 2$ is the fruit peel or meat effect, and
e_{ijkl} is the error term for the $ijkl^{th}$ cell.

Each of the radionuclides was analyzed separately for each of the above designs, and was also analyzed for MANOVA for each of the above designs where the observed concentration is a vector of concentration:

$$y = [^{238}\text{Pu}, ^{239}\text{Pu}, ^{137}\text{Cs}].$$

The multivariate tests give overall tests of the various hypotheses for each of the models above, and provide a means of evaluating the three radionuclides simultaneously rather than trying to interpret them separately. Significance levels of multivariate tests utilized Pillai's method (Morrison, 1976).

Radionuclide concentrations were analyzed both untransformed and with logarithmic transformations. Conclusions from the two analyses were identical for the effects of interest, and so only the analyses for untransformed data are presented.

RESULTS AND DISCUSSION

Study plot soils receiving the fertilization treatment differed significantly ($p < 0.01$, *t-test*) in chemical composition from those receiving no fertilizer treatment (Table 1). Of the parameters measured, only Ca, Cu, and pH remained unchanged in soil after addition of the fertilizer.

Table 1—Chemical and physical characteristics of soils from Mortandad Canyon garden plot in 1977.

Constituent	Soil treatment: Fertilization‡	Soil treatment: None	Significance
	mean (SD)†		
Ca, meq/liter	4.9 (2.2)	3.6 (0.91)	NS
Mg, meq/liter	1.7 (0.66)	0.93 (0.22)	<0.01
Na, meq/liter	3.9 (1.3)	1.1 (0.12)	<0.01
K, meq/liter	5.3 (0.91)	0.27 (0.05)	<0.01
Sodium absorption ratio	2.2 (0.59)	0.75 (0.15)	<0.01
pH	8.1 (0.20)	7.9 (0.10)	NS
Conductivity, mmhos/cm	1.6 (0.49)	0.60 (0.10)	<0.01
Organic matter, %	1.4 (0.29)	0.60 (0.07)	<0.01
P, ppm	52 (6.1)	16 (4.9)	<0.01
K, ppm	557 (71)	66 (4.9)	<0.01
NO_3-N, ppm	16 (7.1)	7.3 (3.7)	<0.01
Zn, ppm	2.6 (0.22)	1.9 (0.22)	<0.01
Fe, ppm	15 (0.93)	12 (1.9)	<0.01
Cu, ppm	2.2 (0.27)	2.1 (0.20)	NS
Mn, ppm	4.2 (0.39)	2.8 (0.59)	<0.01

† means based on sample size of six.
‡ Fertilized with cow manure.

Table 2—Radionuclide concentrations in oven-dry soil and soil size fractions from Mortandad garden plot.

	<43 μm	53-100 μm	100-500 μm	0.5-0.1 mm	1.0-2.0 mm	2.0-23 mm
	mean (SD)†					
% Soil weight	9.0 (5.0)	14 (5.0)	22 (4.0)	19 (4.0)	21 (4.0)	16 (6.1)
pCi ^{137}Cs/g	217 (43)	196 (47)	125 (36)	69 (35)	54 (30)	114 (67)
% ^{137}Cs in fraction	17	23	23	11	10	16
pCi ^{238}Pu/g	14 (2.4)	17 (3.4)	10 (6.1)	8.1 (5.6)	5.4 (3.9)	6.8 (4.1)
% ^{238}Pu in fraction	12	22	32	14	10	10
pCi ^{239,240}Pu/g	2.6 (0.52)	3.7 (3.2)	2.8 (0.87)	1.7 (0.83)	0.95 (0.46)	1.3 (0.55)
% ^{239,240}Pu/g in fraction	11	25	30	15	10	10

† Mean and SD based on sample size of 26.

Table 3—Effect of soil fertilization on concentrations of Pu and ^{137}Cs in plants (above-ground biomass minus fruits) and fruits of bush beans (mean ± one SD of 13 determinations).

Radionuclide	Unfertilized	Fertilized†
	pCi/g oven-dry tissue	
	Plant	
^{238}Pu	0.92 ± 0.41	0.52 ± 0.23
^{239}Pu	0.14 ± 0.06	0.08 ± 0.04
^{137}Cs	17.3 ± 3.7	8.0 ± 3.7
	Fruit	
^{238}Pu	0.26 ± 0.60	0.31 ± 0.72
^{239}Pu	0.04 ± 0.09	0.05 ± 0.11
^{137}Cs	5.9 ± 4.1	2.8 ± 1.2

† Fertilized with cow manure.

Table 5—Effects of soil fertilization, washing, and height of material sampled on concentrations of Pu and ^{137}Cs in the above-ground biomass of squash plants (minus fruits) (mean ± one SD of eight determinations).

	Unfertilized		Fertilized	
Radionuclide	0–20 cm	20 cm	0–20 cm	20 cm
	pCi/g oven-dry tissue			
	Washed			
^{238}Pu	1.34 ± 0.40	0.34 ± 0.25	0.81 ± 0.31	0.17 ± 0.09
^{239}Pu	0.19 ± 0.04	0.05 ± 0.04	0.12 ± 0.05	0.02 ± 0.01
^{137}Cs	24.9 ± 7.5	1.9 ± 1.4	10.5 ± 5.4	4.5 ± 3.3
	Unwashed			
^{238}Pu	3.3 ± 1.4	0.63 ± 0.50	2.26 ± 0.76	0.44 ± 0.22
^{239}Pu	0.56 ± 0.25	0.10 ± 0.88	0.36 ± 0.13	0.07 ± 0.04
^{137}Cs	66 ± 36	22 ± 11	33.5 ± 7.7	10.5 ± 4.0

Radionuclide concentrations in soils (Table 2) did not differ significantly ($p > 0.05$) between treatments (fertilizer vs. no fertilizer) and, overall, averaged 85 pCi ^{137}Cs/g dry weight, 10 pCi ^{238}Pu/g, and 1.9 pCi ^{239}Pu/g. The soil was sandy with over 90% of the soil mass consisting of particles >53 μm. In addition, over 80% of the radionuclide inventories were present in sand size fractions. Relatively little of the radionuclide inventory was present in the silt-clay (<53 μm) fraction; these particles contained only about 10% of the radionuclide inventory (Table 2), even though the radionuclide concentrations in this fraction were relatively high (Table 3).

Treatment Effects

The fertilization treatment had no significant effect ($p > 0.26$, Table 4) on Pu concentrations in beans (Table 3), although it decreased ^{137}Cs concentrations. Bean fruits had lower concentrations ($p < 0.001$, Tables 3 and 4) of all three radionuclides than bean plants did.

The differences for tissue effects (e.g., fruits vs. plants) are explainable due to the differential times of the material being available for contamination by soil from wind or water movement, and to the fruits usually having lower radionuclide concentrations (Schulz, 1977; Schreckhise and Cline, 1980). The leaves and stems had been exposed to raindrop splash-up and/or wind erosion for the full growing season, while the fruits appeared late in the growing season and were soon harvested. The potential for the fruits to become contaminated by physical processes was much less than for the remainder of the plant.

The differences between the effect of fertilization (Table 4) for Pu ($p = 0.26$) and ^{137}Cs ($p < 0.001$) is attributed to the increase of K, a biological analog of Cs. The increased K concentrations in the soil (8.4 times greater in fertilized soil, Table 2) decreased the uptake of ^{137}Cs by 55% (Table 3). The significant ($p = 0.004$, Table 4) fertilization × tissue effect for ^{137}Cs further indicates the importance of biological uptake for this radionuclide.

The three main effects (fertilizer treatment, washing, and height above the ground; Table 5) were each significant (Table 6) for all three radionuclides. The fact that washing significantly reduced radionuclide concentrations by al 65% (Table 5) indicates that much of the measured radioactive materials must have been associated with the plant surface. Likewise, because the lower portions of the plant had about four times higher concentrations than the upper portions (Table 5), surface contamination of the plant by soil is further supported. The lower radionuclide concentrations ($p = 0.066$, Tables 5 and 6) on the fertilized plot (35% for Pu, 50% for Cs) is attributed to the increase in organic matter, which stabilizes the soil and decreases mobility from wind- and water-driven processes. This hypothesis is further substantiated by the significant interaction term ($p = 0.055$, Table 6) for fertilization × height. Fertilization

Table 4—Analysis of variance table for the beans.

Source	Radionuclide	Sum of squares	Degrees of freedom	F	Significance	Multivariate significance
Fertilization	^{238}Pu	0.397	1	1.306	0.264	
	^{239}Pu	0.009	1	1.329	0.260	<0.001
	^{137}Cs	503.332	1	47.367	<0.001	
Plants within fertilization	^{238}Pu	7.304	24			
	^{239}Pu	0.165	24			
	^{137}Cs	255.023	24			
Tissue	^{238}Pu	2.459	1	9.980	0.004	
	^{239}Pu	0.054	1	9.847	0.004	<0.001
	^{137}Cs	904.890	1	71.932	<0.001	
Fertilization × tissue	^{238}Pu	0.621	1	2.521	0.125	
	^{239}Pu	0.014	1	2.558	0.123	0.014
	^{137}Cs	125.800	1	10.000	0.004	
Tissue × plants within fertilization	^{238}Pu	5.914	24			
	^{239}Pu	0.132	24			
	^{137}Cs	301.917	24			

Table 6—Analysis of variance for squash plants.

Source	Radionuclide	Sum of squares	Degrees of freedom	F	Significance	Multivariate significance
Fertilization	^{238}Pu	3.677	1	5.813	0.030	
	^{239}Pu	0.110	1	6.970	0.019	0.066
	^{137}Cs	3064.038	1	9.783	0.007	
Plants with fertilization	^{238}Pu	8.855	14			
	^{239}Pu	0.220	14			
	^{137}Cs	4384.757	14			
Washing	^{238}Pu	15.688	1	37.832	<0.001	
	^{239}Pu	0.471	1	38.993	<0.001	<0.001
	^{137}Cs	8075.943	1	34.273	<0.001	
Fertilization × washing	^{238}Pu	0.247	1	0.597	0.453	
	^{239}Pu	0.017	1	1.399	0.257	0.254
	^{137}Cs	1014.343	1	4.305	0.057	
Washing × plants within fertilization	^{238}Pu	5.806	14			
	^{239}Pu	0.169	14			
	^{137}Cs	3298.900	14			
Height	^{238}Pu	37.305	1	157.885	<0.001	
	^{239}Pu	0.966	1	111.601	<0.001	<0.001
	^{137}Cs	9097.583	1	74.877	<0.001	
Fertilization × height	^{238}Pu	1.434	1	6.069	0.027	
	^{239}Pu	0.043	1	4.989	0.042	0.055
	^{137}Cs	1406.906	1	11.579	0.004	
Height × plants within fertilization	^{238}Pu	3.308	14			
	^{239}Pu	0.121	14			
	^{137}Cs	1701.010	14			
Washing × height	^{238}Pu	8.042	1	23.311	<0.001	
	^{239}Pu	0.254	1	24.250	<0.001	0.006
	^{137}Cs	1403.345	1	12.605	0.003	
Fertilization × washing × height	^{238}Pu	0.219	1	0.633	0.439	
	^{239}Pu	0.016	1	1.482	0.244	0.286
	^{137}Cs	12.311	1	0.111	0.744	
Washing × height × plants within fertilization	^{238}Pu	4.830	14			
	^{239}Pu	0.149	14			
	^{137}Cs	1558.684	14			

reduced the accumulation of contaminants more on the upper parts of the plant than it did on the lower portions (Table 5).

The above explanation of the decrease in splash-up due to fertilization would also suggest that the fertilization × washing interaction should have been significant (Table 6). More material should have been washed off of the unfertilized plants than the fertilized plants because there was more material present to wash. The ^{137}Cs term was marginally significant (p = 0.057), but neither of the Pu terms were significant (Table 6). We believe this interaction term was not significantly rejected because the amount of additional material to be washed off was small in relation to detection limits, and the variation in the data obscured the interaction.

Table 7—Effect of soil fertilization on concentration of Pu and ^{137}Cs in washed fruits of squash. Three fruits were taken from each of eight plants in each fertilization plot (mean ± SD of 24 determinations).

Radionuclide	Unfertilized	Fertilized
	pCi/g oven-dry tissue	
	Peel	
^{238}Pu	0.024 ± 0.012	0.017 ± 0.031
^{239}Pu	0.006 ± 0.003	0.005 ± 0.012
^{137}Cs	9.5 ± 7.1	4.6 ± 2.3
	Meat	
^{238}Pu	0.006 ± 0.017	0.002 ± 0.001
^{239}Pu	0.006 ± 0.022	0.002 ± 0.004
^{137}Cs	6.2 ± 4.8	4.1 ± 1.8

Cesium-137 concentrations in plant material from the fertilized plot were even further reduced (Table 5) than were Pu concentrations. The greater decrease for ^{137}Cs from fertilization can be attributed to the increase in K and, hence, discrimination against ^{137}Cs by the plant, illustrating the greater biological activity of ^{137}Cs.

The reduction of radionuclide concentration in squash fruit due to fertilization (Table 7) was not significant (p >0.174, Table 8) for Pu. We attribute the lack of a fertilization effect for Pu to the short time that the squash fruits were in the garden and thus subject to surface contamination, and to the low levels of activity observed in the fruits with the effect thus obscured by the variation due to fallout background. As the fruits reached the sizes where they are normally utilized for food, they were harvested from the garden and stored until analyses. They were washed before being prepared for analyses, and thus, as illustrated in Table 5, some of the surficial contamination was removed. In addition, the greater biological activity of ^{137}Cs increased the fertilization effect, and thus a significant difference was found for ^{137}Cs but not for Pu.

For ^{238}Pu, the washing did not remove all the activity on the peel (Table 7), with the ^{238}Pu concentration in meat being only 20% of the concentration in peel. We attribute the difference to surficial contamination, although biologically active Pu could have been preferentially deposited in the peel of the fruit rather than the meat. The difference in ^{239}Pu concentration between the peel and the meat of the fruits was not significant (p = 0.48, Table 8), which suggests that there was little con-

Table 8—Analysis of variance for squash fruit.

Source	Radionuclide	Sum of squares	Degrees of freedom	F	Significance	Multivariate significance
Fertilization	^{238}Pu	8.0×10^{-4}	1	2.053	0.174	
	^{239}Pu	1.2×10^{-4}	1	0.736	0.405	0.238
	^{137}Cs	2.9×10^{2}	1	4.245	0.058	
Plants within fertilization	^{238}Pu	5.4×10^{-3}	14			
	^{239}Pu	2.2×10^{-3}	14			
	^{137}Cs	9.6×10^{2}	14			
Fruit within plants	^{238}Pu	6.0×10^{-3}	16			
	^{239}Pu	2.6×10^{-3}	16			
	^{137}Cs	2.0×10^{2}	16			
Peeling	^{238}Pu	6.5×10^{-3}	1	19.770	<0.001	
	^{239}Pu	6.0×10^{-5}	1	0.527	0.480	0.006
	^{137}Cs	8.7×10^{1}	1	10.174	0.007	
Peeling × fertilization	^{238}Pu	4.0×10^{-5}	1	0.136	0.715	
	^{239}Pu	9.0×10^{-5}	1	0.505	0.483	0.261
	^{137}Cs	4.4×10^{1}	1	3.385	0.075	
Peeling × plants within fertilization	^{238}Pu	4.6×10^{-3}	14			
	^{239}Pu	2.2×10^{-3}	14			
	^{137}Cs	1.2×10^{2}	14			
Peeling × fruits within plants	^{238}Pu	5.4×10^{-3}	16			
	^{239}Pu	2.7×10^{-3}	16			
	^{137}Cs	1.8×10^{2}	16			

tamination by Pu, and that any difference in concentration was obscured by variation due to the fallout background. Concentration of ^{239}Pu was only 20% of that for ^{238}Pu in the soil (Table 2).

In contrast, both fertilization and peeling the fruit produced significant differences (Table 8) for ^{137}Cs. The decrease in ^{137}Cs concentrations due to fertilization (Table 7) can be attributed to the increase of K in the fertilized plot, although the effect was only marginally significant ($p = 0.058$, Table 8). The greater concentration of ^{137}Cs in the peel vs. the meat (Table 7) may be explained by surficial contamination that was not removed by wash treatment. The higher concentrations of ^{137}Cs allowed smaller differences to be detected than for Pu.

Concentration Ratios

The dimensionless ratio of concentration in plant tissue to concentration in soil was calculated for the five plant tissues analyzed in this study (Table 9). Combined cell means were used for each tissue, thus the concentration ratio varied for fertilized vs. unfertilized plots, washed vs. unwashed, and lower vs. upper strata. These concentration ratios were calculated using the soil data in Table 2 and tissue data from Tables 3, 5, and 7. All of the statistical results discussed for Tables 4, 6, and 8 apply to the concentration ratios, as the only difference in the data is the constant factor, soil concentration.

In Table 9, we see that the largest concentration ratios for Pu were in squash plants, followed by bean plants. We would expect these results, as squash plants are better collectors of soil than bean plants because of their large flat leaves close to the ground surface.

The ^{137}Cs concentration ratios were always greater than those for Pu (Table 9), substantiating the fact that ^{137}Cs is biologically more active than Pu. Because greenhouse studies have shown that biological uptake is <1% of the total Pu content of vegetation (ERDA, 1976), we can use the concentration ratios in Table 9 to partition the amount of ^{137}Cs splashed up on the plant

Table 9—Concentration ratios (pCi/g dry weight vegetable ÷ pCi/g dry weight soil) for various plant tissues from the Mortandad garden plot.

Tissue	^{238}Pu	^{239}Pu	^{137}Cs
Bean plant	7.2×10^{-2}	5.8×10^{-2}	1.5×10^{-1}
Bean fruit	2.9×10^{-2}	2.4×10^{-2}	5.1×10^{-2}
Squash plant	1.2×10^{-1}	9.7×10^{-2}	2.5×10^{-1}
Squash peel	2.0×10^{-3}	2.6×10^{-3}	8.3×10^{-2}
Squash meat	4.0×10^{-4}	2.1×10^{-3}	6.1×10^{-2}

as opposed to the amount taken up biologically. The squash plant data are taken as an example to illustrate the calculations. The average concentration ratio for Pu was about 0.11, while the concentration ratio for ^{137}Cs was 0.25. Thus if no biological uptake were present for ^{137}Cs, we would expect a concentration ratio of 0.11 as for Pu. Instead, the concentration ratio of 0.25 suggests that 43% of the ^{137}Cs on the squash plant was due to splash-up, while 60% was due to biological uptake. Similar calculations for bean plants, bean fruits, squash peel, and squash meat gave the percent of ^{137}Cs splashed onto the tissue as 43, 52, 28, and 2, respectively. These values illustrate that about 45% of the ^{137}Cs on the unwashed plant material was due to splash-up. The squash fruits were washed as in normal food preparation, so the amount of ^{137}Cs remaining on the peel surface was lower than for the unwashed plants. The 2% figure for squash meat illustrates the obvious—nothing was splashed into squash meat.

Radiation Doses

Radiation doses to humans consuming vegetables from the garden plot were calculated for all vegetable combinations. Doses from zucchini squash (meat only) and for green beans from the unfertilized plot are presented in Table 10. Unfertilized plot data were chosen because contamination concentrations were higher than in the fertilized plot. Thus, calculated doses represent the upper portion of the range of calculated doses. Doses were dominated by the ^{137}Cs contamination. The

Table 10—Estimated radiation doses to humans from ingestion of garden vegetables grown in the unfertilized garden plot contaminated with $^{238,239}Pu$ and ^{137}Cs.

Vegetable	Annual ingestion†	1st-year dose‡			50-year dose commitment‡		
		Total body	Bone	Liver	Total body	Bone	Liver
	kg	mrem					
Zucchini squash	0.23	2.5×10^{-3}	2.5×10^{-3}	4.2×10^{-3}	4.0×10^{-3}	4.7×10^{-3}	6.3×10^{-3}
Green beans	0.68	1.6×10^{-2}	1.6×10^{-2}	2.8×10^{-2}	2.7×10^{-2}	4.2×10^{-2}	4.4×10^{-2}

† Ingestion rates taken from USDA, 1978.
‡ Radiation Dose Factors (mrem/μCi intake) taken from Corley et al., 1977; concentrations of ^{137}Cs and $^{238,239}Pu$ in vegetables taken from Tables 3 and 7. Calculated dose includes contributions from both ^{137}Cs and $^{238,239}Pu$.

only place where $^{238,239}Pu$ made a significant contribution (38%) was in the 50-year dose commitment to bone from green bean ingestion. As can be seen from the data, doses are extremely low and represent no hazard. The largest total body dose in the table (0.027 mrem) is equivalent to the dose received from cosmic radiation in 7 min from flying at typical jet aircraft altitude of 9,144 m (30,000 feet). This dose represents 0.23% of the estimated 138 mrem Los Alamos residents received from natural radiation in 1979 (Environmental Surveillance Group, 1979).

SUMMARY AND CONCLUSIONS

The results of this study demonstrate that a major source of contamination of green beans and zucchini squash by ^{137}Cs and $^{238,239}Pu$ was due to soil on the plant surfaces. The lower portions (0–20 cm) of zucchini and squash plants were contaminated about four times the higher portions (>20 cm). About 65% of the contamination on zucchini squash plants could be washed off with normal food preparation techniques. Fertilization of the garden plot with cattle manure reduced contamination by about 30% for Pu and even greater (about 50%) for ^{137}Cs. The reduction due to fertilization is attributed to a stabilization of the soil surface, and, in the case of ^{137}Cs, reduction in ^{137}Cs uptake in the plant was due to increased K concentrations.

Fifty-year radiation doses to humans consuming vegetables from the garden plot would be <0.05 mrem and would be due almost entirely to ^{137}Cs.

ACKNOWLEDGMENTS

The authors wish to thank E. M. Karlen, J. L. Martinez, M. Dreicer, K. V. Bostick, E. Montoya, T. Jacques, and C. Burns for their contributions to the research presented in this paper.

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