Best Available
Copy

Environmental and Experimental Botany, Vol. 22, No. 1, pp. 33 to 38, 1982.
Printed in Great Britain

0098-8472/82/010033-06 $03.00/0
© 1982. Pergamon Press Ltd.

IMPORTANCE OF PLUTONIUM CONTAMINATION ON VEGETATION SURFACES AT ROCKY FLATS, COLORADO

W. J. ARTHUR, III and A. W. ALLDREDGE

U.S. Department of Energy, Environmental Sciences Branch, Idaho National Engineering Laboratory, Idaho Falls, ID 83401 and Department of Fishery and Wildlife Biology, Colorado State University, Fort Collins, CO 80523, U.S.A.

(*Received* 1 *August* 1980; *accepted in revised form* 10 *November* 1980)

ARTHUR W. J., III and ALLDREDGE A. W. *Importance of plutonium contamination on vegetation surfaces at Rocky Flats, Colorado.* ENVIRONMENTAL AND EXPERIMENTAL BOTANY 22, 33–38, 1982.—Vegetation samples collected at Rocky Flats, Colorado were ultrasonically washed to remove attached soil and then analyzed for $^{239,240}Pu$ and ^{238}Pu. Mean plutonium concentrations for washed grass, forb and shrub samples were 1.12, 0.61, 0.03 pCi/g, respectively. This compared to a mean of 28.6 pCi/g for unwashed vegetation samples from an earlier study conducted in the same area at Rocky Flats. The mean plutonium isotopic ratio ($^{239,240}Pu/^{238}Pu$) of 27.2 in vegetation was lower than a ratio of 65.4 in soil, indicating possible differential behavior of the plutonium isotopes in vegetation. Soil attachment to above-ground plant parts ranged from 0.0 to 0.25 g soil per g plant. The potential for plutonium contamination on plant parts due to soil attachment was estimated to range from 0.0 to 206 pCi/g. Plutonium on vegetation was concluded to be the major contributor to total plutonium associated with vegetation.

INTRODUCTION

DURING the 1960's, several square kilometers of grassland east and southeast of the Rocky Flats Nuclear Weapons Plant were accidently contaminated with approximately 11.4 curies (Ci) of plutonium. The majority of contamination resulted from leakage of storage barrels containing plutonium-laden cutting oil, and a small quantity was released from a fire at the plant site.[3] Of the 11.4 Ci released, 3.4 were estimated to be on public and private land, and 8.0 Ci remained on the Rocky Flats site.

In 1975 a study was initiated to estimate plutonium intake by mule deer (*Odocoileus hemionus*) feeding in contaminated areas at Rocky Flats. Part of this study necessitated determination of plutonium concentrations in vegetation. Since surface disturbances and wind were the major factors affecting distribution of plutonium in the Rocky Flats area,[4] vegetation undoubtedly received varying amounts of plutonium contamination due to resuspended soil. Vegetation growing in soils having elevated plutonium concentrations could also assimilate this nuclide. As a means of assessing the relative importance of these two transport pathways, this study was conducted to compare the quantity of plutonium in vegetation to that attached to soil particles on plant surfaces.

STUDY AREA

Rocky Flats Nuclear Weapons Plant is located 12 km northwest of Denver, Colorado. Approximately 1.6 km^2 of land are contained within the security fence and are surrounded by an additional 25 km^2 buffer zone closed to public use. Rocky Flats is located on an alluvial plain with a gravelly topsoil over a layer of stony gravel, sand and clay 3–13 m thick.[6]

ADMIN RECORD

SW-A-005949

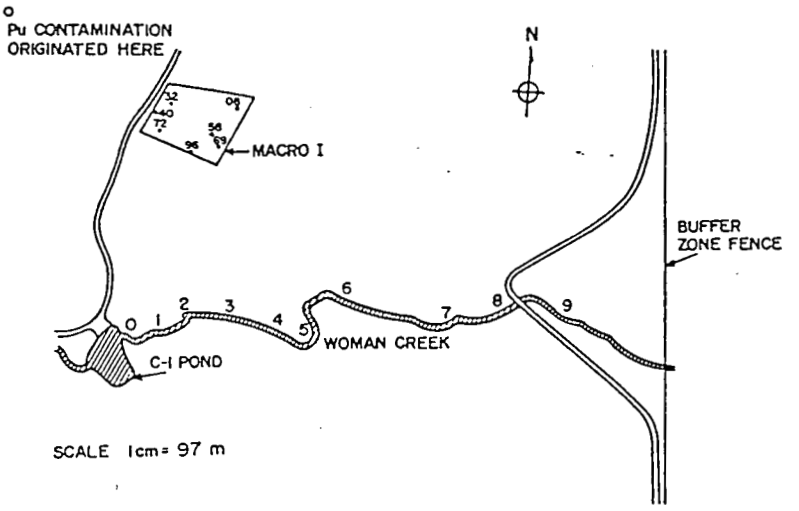

FIG. 1. Location of vegetation sampling points in Macroplot I and along Woman Creek at Rocky Flats, Colorado.

Earlier research studies indicated that greater than 99% of the total plutonium released was contained in soil.[4] Highest levels of plutonium in soil were found adjacent to the former oil barrel storage pad (Fig. 1) and these decreased with distance downwind from the source. A mean plutonium concentration of 835 pCi/g soil has been estimated for an area immediately adjacent to the former storage area and a mean value of 55 pCi/g soil was estimated for an area approximately 1.5 km south.[4]

Two vegetation types dominated the area. A meadow type comprised about 90% of the area and contained a mixture of moist meadows and wet marsh. Slender wheatgrass (*Agropyron trachycaulum*), cheatgrass (*Bromus tectorum*), and blue grass (*Poa compressa*) occurred most commonly in the moist meadow area intermixed with nodding thistle (*Cardus nutans*), great mullein (*Verbascum thapsus*), yarrow (*Achillea lanulosa*), big bluestem (*Andropogon gerardii*), sage (*Artemesia ludoviciana*), blue grama (*Bouteloua gracilis*), golden aster (*Heterotheca villosa*), common dandelion (*Taraxacum officinale*) and salsify (*Tragopogon dubius*). Wet marshy areas were dominated by broadleaved cattail (*Typha latifolia*), sedge (*Carex* sp.) and rush (*Juncus* sp.). Disturbed areas (burns, roads, diggings) supported secondary growth species, primarily cheatgrass, penney cress (*Thlapsi arvense*), knotweed (*Polygonum douglasii*), blue grass, common pigweed (*Chenopodium album*) and sweet clover (*Meliotus* sp.). A streamside vegetation type made up the remaining 10% of the area's plant species composition and was dominated by willow species (*Salix* sp.), lead plant (*Amorpha fruticosa*), plains cottonwood (*Populus sargentii*) and sedge.

METHODS AND MATERIALS

Vegetation sampling at Rocky Flats was conducted in an area immediately adjacent to the plutonium contamination source and an area farther downwind from the source (Fig. 1). Marco I, 0.75 ha in size, was located 125 m southeast of the former oil barrel storage pad. The Marco I grid was designed from a previous research project[4] and contained 100 sampling plots in a 10 × 10 arrangement. The meadow vegetation type dominated this area. The second area sampled was Woman Creek and consisted of 10 sampling plots which ranged from 440 to 825 m from the former barrel storage area. Woman Creek was dominated by streamside vegetation type, and was also downwind from the contamination source.

Seasonal collection dates for vegetation were summer (12 August, 1976), fall (24 November, 1976), winter (8 January, 1977) and spring (12 May, 1977). Grass and forb samples were clip-

ped 2 cm above ground surface and only new stem and leaf growth was clipped from shrubs.

Plutonium accumulation by vegetation within the Macro I area was estimated by compositing samples of cheatgrass, blue grama, and blue grass from plots 8, 32, 58, 72 and 96 during summer (Fig. 1). Samples of each major grass and forb species were also collected seasonally from plot 40 to determine differences in plutonium concentrations between species. In order to estimate whether plutonium concentrations in vegetation changed downwind from the contamination source, composites of grass and forb species were sampled from Macro I plots 40 and 69, and also Woman Creek sampling plot 9. Samples collected from locations 40 and 69 were considered representative of the highest values of plutonium in vegetation and the sample from Woman Creek was indicative of plutonium concentrations characteristic of an area farther downwind from the contamination source (Fig. 1).

Shrubs were sampled from 10 plots established along Woman Creek (Fig. 1). Each season an equal mass of plains cottonwood and sand-bar willow (*Salix interior*) was composited from plots 1, 3, 5, 7 and 9. Single samples of each major shrub species were collected from sampling points 4, 5 and 6 to estimate differences in plutonium concentrations between species.

Previous studies[7, 8] indicated that considerable soil contamination occurred on external plant parts. To remove surface contamination, samples were placed in a 2.8 l. ultrasonic washer filled with distilled water and were cleaned with low frequency sound waves (60 Hz). Washing was conducted for 30 min periods and was continued for 30 min after no soil was visible in the ultrasound pan. The number of washes received by vegetation varied depending on the amount of soil observed in the wash pan. Some vegetation species had to be ultrasonically washed for up to 4 hr before no soil was observed in the wash pan. Washed vegetation samples were oven dried for 72 hr at 80°C and weighed.

For each sample where soil was observed in the wash water, water was filtered through a 1.2 μm filter using a Millipore membrane apparatus. Care was taken to eliminate plant parts from the filtrate. Filters were ashed for 24 hr at 600°C to remove organic materials and weighed. No attempt was made to analyze the filtrate for plutonium.

A ratio of soil to vegetation mass was obtained for each sample where soil was observed during the washing. Analysis of variance procedures were used to test ratios for differences as a function of season and vegetation type. An estimate of the amount of plutonium attached to plant surfaces in the Macro I area was made by relating the amount of soil attached to above ground plant parts to plutonium concentrations reported for the upper 3 cm of soils.[4]

All washed vegetation samples were analyzed for ^{239,240}Pu and ^{238}Pu.[9] The lower detection limit for ^{238}Pu and ^{239,240}Pu was 2.25×10^{-4} pCi/g vegetation. Where both ^{239,240}Pu and ^{238}Pu were detectable, isotopic ratios were calculated. Mean concentrations (pCi/g) of each radionuclide were calculated and analysis of variance (AOV) procedures were used to test for significant differences between plant species, areas and season. Duncan's new multiple range test[2] was used to separate significantly different means ($p \leq 0.05$).

RESULTS

Plutonium concentrations in vegetation

Mean plutonium concentrations in ultrasound washed grass, forb and shrub samples collected at Rocky Flats are presented in Table 1. Forbs from the Macro I area had an overall mean of 0.61 pCi/g and results of analysis of variance indicated a significant difference between seasonal means ($p \leq 0.01$). The mean plutonium concentration for forbs during fall was significantly greater than spring and summer means but not winter ($p \leq 0.05$). In spring and early summer when most forbs are beginning growth, we speculate that less plutonium would accumulate in the vegetation, as compared to late summer and fall when most plants were reaching maturity. In fall, sage, Canada thistle (*Cirsium arvense*), and filarce (*Erodium cicutarium*) had plutonium concentrations greater than 0.45 pCi/g and all species were photosynthesiz-

Table 1. Seasonal and annual $^{238,239,240}Pu$ concentrations (pCi/g) in ultrasonically washed vegetation at Rocky Flats, Colorado

	Spring	Summer	Fall	Winter	Annual
Grass (Macro I)					
$\bar{x} \pm S.D.$*	$0.04_A \pm 0.03$	$0.46_A \pm 0.54$	$2.27_A \pm 1.77$	$1.50_A \pm 0.99$	1.12 ± 1.37
Range	0.01–0.07	0.04–1.17	0.24–4.40	0.25–2.93	0.01–4.40
n	5	5	6	5	21
Forb (Macro I)					
$\bar{x} \pm$ S.D.	$0.16_A \pm 0.27$	$0.28_A \pm 0.29$	$1.67_B \pm 1.04$	$0.95_{AB} \pm 0.90$	0.61 ± 0.81
Range	0.01–0.79	0.02–0.60	0.52–2.54	0.25–2.24	0.01–2.54
n	8	3	3	4	18
Shrub (Woman Creek)					
$\bar{x} \pm$ S.D.	$0.004_A \pm 0.008$	$0.07_A \pm 0.13$	$0.03_A \pm 0.01$	$0.01_A \pm 0.01$	0.03 ± 0.07
Range†	BDL–0.013	BDL–0.26	BDL–0.04	BDL–0.03	BDL–0.26
n	3	4	4	4	15

*Seasonal means are followed by the letters *A* or *B*. Means that do not share the same letters are significantly different ($p \leq 0.05$) for seasons within vegetation types.
†BDL = below detection limit.

ing. In winter, all forbs sampled were dead or dormant yet still contained detectable amounts of plutonium, suggesting that little plutonium is lost until plants decompose.

The mean plutonium concentration in forb samples ($n = 5$) from Woman Creek was 0.14 pCi/g. This high value was caused by the presence of one sage sample (0.66 pCi/g), as compared to other sample values ranging from 0.0 to 0.02 pCi/g. This was probably due to plutonium-contaminated soil particles that may not have been removed during ultrasonic washing. When the sage sample is deleted, a new mean of 0.01 pCi/g is obtained, which is probably a more realistic value for forb species along Woman Creek.

Similar seasonal trends were observed for plutonium concentrations in Macro I grass samples, although considerable variability occurred between samples. No seasonal differences were noted for plutonium concentrations in grasses which had an overall mean ($\bar{x} \pm$ S.D.) of 1.12 ± 1.37 pCi/g. The mean value (0.01 pCi/g) for plutonium concentrations in grasses along Woman Creek was lower than that for Macro I grass species with no plutonium detected in 2 out of 3 grass species.

Shrub species sampled along Woman Creek accumulated the least amount of plutonium with a mean of 0.03 pCi/g. Seasonal differences in mean concentrations for individual shrub species were not significant. Six out of 15 shrub samples had no detectable plutonium.

Plutonium concentration ratios (pCi per g dry vegetation/pCi per g dry soil) were 0.0017 (S.D. = 0.0011) for Macro I (plots 40, 69) and 0.0031 (S.D. = 0.0040) for Woman Creek, respectively. The soil plutonium concentrations used were 835 pCi/g for Macro I[4] and 11.6 pCi/g for Woman Creek. A previous study[4] reported a concentration ratio of 0.0342 for Macro I—greater by a factor of 20 than our value for the same area. The ratio of 0.0342 was calculated assuming a mean plutonium concentration of 28.6 pCi/g for unwashed vegetation, whereas our ratio was based on ultrasound washed vegetation having plutonium concentrations ranging from 0.05 to 2.41 pCi/g.

Based on 33 samples in which both $^{239,240}Pu$ and ^{238}Pu levels were detectable, a mean ($\bar{x} \pm$ S.D.) isotopic ratio of 27.2 ± 15.3 was calculated. This compares to a mean ratio ($^{239}Pu/^{238}Pu$) of 60.6 for unwashed Macro I vegetation.[4] In 1976, an isotopic ratio of 65.4 was obtained for the top 3 cm of soil suggesting that the vegetation isotopic ratio of 60.6 was

*Table 2. Relative mass of soil attached to above-ground vegetation samples collected seasonally at Rocky Flats, Colorado**

	Spring	Fall	Winter	Annual
$\bar{x}$ (g oven dry soil/g oven dry vegetation)	0.005	0.044	0.013	0.018
S.D.	0.003	0.090	0.018	0.048
Range	0.001–0.013	0.002–0.247	0.001–0.061	0.001–0.247
n	9	7	10	26

*No data available for summer season.

mainly due to plutonium in soil attached to the unwashed vegetation and not to plutonium in vegetation.

Soil attachment to plant parts

Ultrasonic washing of vegetation indicated that many species had soil attached to plant parts. This attachment may have occurred due to: (1) wind-blown particles, (2) splash-up during rain and snow or (3) trampling of vegetation. The relative mass of soil on vegetation during each season is presented in Table 2. Variability was high between and within species with an overall concentration ($\bar{x} \pm$S.D.) of 0.018 ± 0.048 g soil per g vegetation. Analysis of variance tests were not significant for ratios between seasons or between forb, grass and shrub categories. Plant species having larger amounts of soil attached were sage, cheatgrass, filaree, snakeweed (*Gutierrezia sarothrae*) and common dandelion. Wash water was filtered only from species where soil was observed in the ultrasound pan. Thus, ratios obtained are not representative of all plants of a particular species and average ratios presented are higher than those of the overall vegetative population.

The mean concentration of 0.018 g soil per g washed vegetation multiplied by the mean plutonium concentration in Macro I soil (835 pCi/g) equals an overall mean value of 15.0 pCi/g for plutonium attached to plant surfaces. This is considerably greater than mean plutonium concentration (0.70 pCi/g estimated for washed vegetation samples, but it is similar to the mean of 28.6 pCi/g obtained from a previous study.[4]

CONCLUSIONS AND DISCUSSION

Mean plutonium concentrations in washed grass, forb and shrub samples were 1.12, 0.61, 0.03 pCi/g respectively. The lower value for shrub samples is probably due to Woman Creek being a greater distance from the source of contamination. Mean plutonium concentrations in vegetation obtained in this study ($\bar{x} = 0.70$ pCi/g) were substantially lower than the plutonium concentration of 28.6 pCi/g obtained for unwashed vegetation samples in 1976.[4] Our plutonium isotopic ratio of 27.2 in vegetation indicated possible differential behavior of the plutonium isotopes in soil and vegetative systems. The 1976 plutonium isotopic ratios for vegetation and soil (60.6 and 65.4, respectively) were quite similar. We hypothesize that the greater plutonium isotopic ratio for vegetation obtained in 1976 was due mainly to plutonium contaminated soil attached to plant parts.

Green and yellow coloration was frequently observed in the water after ultrasound washing of vegetation. Microscopic examination revealed that some of this material was plant epidermal tissue. The possibility exists that ultrasonic washing breaks down plant structure causing release of plutonium that may have been within the plant.

Based on data from ultrasonically washed samples, vegetation uptake of plutonium due to physiological processes at Rocky Flats, Colorado is minimal ($\bar{x} = 0.70$ pCi/g). This low intake of plutonium is probably best attributed to the relatively insoluble state of plutonium oxide.[5] Titanium levels in Rocky Flats' vegetation were also determined as part of another study.[1]

Titanium concentration patterns were similar to those measured for plutonium, indicating possible similarity in mechanisms responsible for behavior of these two biologically inert materials in vegetation. Another plausible explanation is that ultrasonic washing failed to remove all soil and thus both titanium and plutonium measured in vegetation samples was actually associated with soil attached to plant parts.

Soil attachment to above-ground plant parts ranged from 0.0 to 0.247 g soil per g vegetation. We hypothesize that the main factors affecting surface soil contamination on vegetation are wind, proximity of plants to roads, amount of disturbance (trampling, grazing), precipitation and vegetation surface area.

We suggest that future literature discussing plutonium concentrations in vegetation should explain whether an attempt was made to remove surface contamination prior to analysis. This is applicable to any contaminants where a distinction between contamination on and in vegetation is necessary.

Acknowledgements—Research reported herein was funded through U.S. Department of Energy, Contract No. EY-76-S-02-1156 with Colorado State University. Authors thank T. E. Hakonson, C. A. Little and F. W. Whicker for critically reviewing this manuscript and G. S. Hiatt for aid in sample collection.

REFERENCES

1. Arthur W. J. and Alldredge A. W. (1979) Soil ingestion by mule deer in northcentral Colorado. *J. Range Manage.* **32**(1), 67–71.
2. Duncan D. G. (1955) Multiple range and multiple F tests. *Biometrics* **11**, 1–42.
3. Krey P. W. (1976) Remote plutonium contamination and total inventories from Rocky Flats. *Health Phys.* **30**, 209.
4. Little C. A. (1976) *Plutonium in grassland ecosystem.* Ph.D. Thesis, Colorado State University, Fort Collins, Colorado 170 p.
5. Little C. A. and Whicker F. W. (1978) Plutonium distribution in Rocky Flats soil. *Health Phys.* **34**, 451–457.
6. Malde H. E. 1955. Surficial geology of the Louisville Quadrangle, Colorado. *U.S. Geological Survey Bull.* 996-E.
7. Romney E. M., Mork H. M. and Larson K. H. (1970) Persistence of plutonium in soil, plants, and small mammals. *Health Phys.* **19**, 487–491.
8. Romney E. M. and Wallace A. (1976) Plutonium contamination in vegetation in dusty field environments. Pages 287–302 *in* J. G. White and P. B. Dunaway, eds. *Transuranics in natural environments.* NVO-178. Nevada Applied Ecology Group.
9. Wessman R. A., Major W. J. Lee K. D. and Leventhal L. (1971) Commonality in water, soil, air, vegetation and biological sample analysis for plutonium. Pages 73–79 *in* E. B. Fowler, R. W. Henderson and M. F. Milligan, eds. *Proceedings of environmental plutonium symposium held at LASL, August 4–5, 1971,* Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

6/6