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PLUTONIUM CONCENTRATIONS IN ARTHROPODS AT A NUCLEAR FACILITY*

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Abstract—Arthropods were collected for ^{239,240}Pu (^{239}Pu) and ^{238}Pu analysis from three study plots in close proximity to the Rocky Flats nuclear weapons plant and from a site 110 km N–NE of the plant. Mean ^{239}Pu concentrations in arthropods were 265, 16, 0.7 and 0.5 dis/min g^{-1} at the three Rocky Flats study plots and at the control site, respectively. Arthropod ^{239}Pu concentration data were statistically analyzed by season of collection, taxonomic group, and sampling site. Only the collection site differences were significant ($\alpha = 0.01$) and these were correlated with ^{239}Pu concentrations in soil. The mean activity ratio of ^{239}Pu to ^{238}Pu in arthropods was 52, similar to the value of 51 obtained for soil. The mean ratio of ^{239}Pu in arthropods to ^{239}Pu in 0–3 cm soil at Rocky Flats was 9×10^{-3}. Arthropod biomass and Pu concentration data indicated that only about 10^{-8} of the total plutonium inventory is in the arthropod component of the ecosystem. Leafhoppers, grasshoppers and spiders accounted for roughly 80% of the arthropod inventory of ^{239}Pu.

INTRODUCTION

THE Rocky Flats nuclear weapons plant, located near Denver, Colorado, became an area of public concern during the late 1960s due to the discovery of elevated levels of plutonium in the soil surrounding the facility (Ma70). The major source of environmental plutonium is believed to have been leaking drums of plutonium-contaminated cutting oil which were located in a storage area in the southeast corner of the Rocky Flats complex during the period 1958 to 1968 (Kr70). Concern about the contamination, limited information on the environmental behavior of plutonium, and the long half-life and radiotoxicity of the element created justification for a study of its distribution and behavior within the grassland ecosystem surrounding the plant.

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The arthropods were selected as one of five ecosystem compartments to be studied because of their abundance and wide range of behavioral patterns and food habits. The close association of some arthropods with soil, the major plutonium reservoir at present, and the dispersal potential of these animals raised the specific question of biological export of plutonium. The objectives of this study were to ascertain plutonium concentrations in arthropods at selected study sites; estimate total plutonium contained within the arthropod community; and investigate the effects of order, season of collection and sampling location on plutonium concentrations.

MATERIALS AND METHODS

Two study macroplots were chosen for intensive sampling at Rocky Flats; macroplot 1 (MP-1) and macroplot 2 (MP-2). Macroplot 1 was approx. 0.75 hectares (ha) in size and located in the southeast portion of the plant

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property. This area, which was 150 m downwind (with respect to prevailing strong winds) from the formerly contaminated barrel storage area, was chosen because of accessibility, measurable Pu levels and its essentially undisturbed state (Li76). Macroplot 2 was slightly less than 0.75 ha and located 1400 m south of the former barrel storage area, out of the prevailing vector for strong winds. Macroplot H (MP-H), located within 50 m of the original plutonium source, was also sampled but less intensively. This area was not readily accessible since it was inside a security area and entry required special clearance and precautions.

During the latter part of the study, it became evident that MP-2 was slightly contaminated, and therefore not acceptable as a control area. This created need for a fourth macroplot far removed from the Rocky Flats installation, which could be considered completely uncontaminated except by worldwide fallout. Because of vegetative similarity to Rocky Flats the Pawnee National Grasslands located approx. 11 km north of Nunn, Colorado, and 110 km N–NE of Rocky Flats, was chosen as a control sampling site (MP-P).

Macroplots 1 and 2 were sampled seasonally from the fall of 1973 through the fall of 1976. Macroplot H was sampled on only two occasions, both in the summer of 1974. Macroplot P was sampled on three separate dates, all during the fall of 1976. We do not suspect bias among years because the principal contaminating events occurred prior to mid-1969 (Li76) and little plutonium export from the study plots has occurred. Therefore, the total plutonium inventory was presumably stable during the study period. We believe this was also true for fallout plutonium in macroplot P. Any seasonal bias obviously could only be examined with data from macroplots 1 and 2.

Three methods were used in the collection of arthropods; sweep netting, pitfall trapping, and drop-trapping followed by vacuuming. Sweep nets and pitfall traps were used for collection of vegetation dwelling and ground crawling arthropods respectively, to be analyzed for plutonium. These methods were chosen because of the reduced likelihood of acquiring contaminating soil along with the arthropods and the fact that large areas could be sampled in a relatively short period of time. During each sweep net collection, ten 87 m transects at 8 m intervals were swept. The resulting collection area was about 1500 m^2 or 20% of the plot. Three pitfall traps were installed at 50 m intervals along a diagonal transect across MP-1 and MP-2 to sample the ground crawling arthropods over a representative cross section of each plot. A vaporizing insecticide-impregnated plastic strip within the catch container and a powder funnel covering the top of the trap were used to kill the collected arthropods before captives could prey upon each other. These arthropods were collected for plutonium analysis only if no precipitation had occurred during the trapping period, to avoid misleading nuclide concentrations due to washing or leaching. The third collection method employing a drop trap and vacuuming procedure was used to estimate arthropod biomass associated with both vegetation and ground in MP-1 and MP-2. The trap, consisting of a 0.5 m^2 screened aluminium frame, was randomly dropped upon previously undisturbed terrain and the enclosed vegetation and soil surfaces were thoroughly vacuumed to remove arthropods. Sampling, consisting of five trap drops on each macroplot, was carried out bimonthly from fall 1973 through fall 1974, except for the first two months of 1974 when the ground was snow-covered. The drop trap-vacuuming procedure was always carried out during early morning hours. This procedure likely underestimates biomass since mobile individuals may avoid the falling trap and others may be overlooked in the vacuuming procedure.

All arthropod samples were placed in plastic bags and labeled as to collection site and date, taken to the laboratory on the day of collection, and preserved at 0°F. Dead animals were then separated into taxonomic groups, oven dried to constant mass at 52°C, and weighed. The pitfall and sweep net collections were pooled for analytical purposes. Pooling of a large number of oven dried insects was required to obtain a minimum

sample weight (about 1 g) for accurate Pu analysis. When necessary, seasonal collections were pooled within a taxonomic group. Such pooled samples were referred to as "all season". The analysis for ^{239,240}Pu (hereafter referred to as ^{239}Pu) and ^{238}Pu was done by LFE Environmental Analysis Laboratories, Richmond, California, employing an alpha spectrometric technique utilizing ^{236}Pu as a recovery tracer (We71).

RESULTS

Insects collected for plutonium analysis and biomass estimation were predominately leafhoppers and aphids (*Homoptera*); ground beetles, darkling beetles, lady bird beetles and carrion beetles (*Coleoptera*); and grasshoppers and crickets (*Orthoptera*). Other arthropods collected in fewer numbers but containing measurable plutonium activity were spiders (*Aranae*); assassin bugs, plant bugs and boxelder bugs (*Hemiptera*); bees, wasps and ants (*Hymenoptera*); and a representative of the class Crustacea with life habits similar to some ground dwelling insects, the sowbug (*Isopoda*).

Thirty-eight discrete samples of sufficient mass for acceptable ^{239}Pu analysis accuracy were assayed and the results appear in Table 1. All samples contained detectable ^{239}Pu activity, but several samples from macroplots 2 and P did not contain measurable ^{238}Pu activity. Examination of the mean ^{239}Pu activity concentrations by macroplots indicated considerably elevated levels in samples from macroplots 1 and H. The standard errors associated with the mean activity levels are comparatively large, but this was not unexpected because of the diversity of sample types, sampling times, and generally high sampling variability which has been observed for environmental plutonium (Li76). Analytical variability was considered small relative to sampling variability for ^{239}Pu, but not for ^{238}Pu.

Analysis of variance of log-transformed ^{239}Pu data grouped by macroplot consistently revealed a significant difference among three of the four study plots ($\alpha = 0.01$). The only two macroplots that showed no significant difference ($\alpha = 0.05$) were macroplots 2 and P. Macroplot differences were the only statistically significant differences found in comparisons of data categorized by macro-

Table 1. ^{238}Pu and ^{239}Pu concentrations (dis/min g^{-1}) in Rocky Flats arthropods, based upon 1973–1976 collections[a]

			Macroplot							
			1		2		H		P	
Season	Order	Common Names	^{238}Pu	^{239}Pu	^{238}Pu	^{239}Pu	^{238}Pu	^{239}Pu	^{238}Pu	^{239}Pu
Spring	Coleoptera	beetles	0.31	20.4	---[c]	1.55				
	Homoptera	leafhoppers	0.25	13.6	---	1.04				
	Orthoptera	grasshoppers	0.06	4.16	---	0.54				
Summer	Coleoptera	beetles	0.06	7.54	---	0.64	5.42	294		
	Homoptera	leafhoppers	0.15	9.02	---	0.44	9.37	522		
	Orthoptera	grasshoppers	0.07	4.45	0.01	0.26	5.13	272		
Fall	Coleoptera	beetles	0.33	20.3	0.02	0.23			---	0.37
	Homoptera	leafhoppers	0.09	1.3	0.03	0.55			0.01	0.32
	Orthoptera	grasshoppers	0.59	28.0	---	0.52			0.01	0.14
	Isopoda	sowbugs	1.13	61.0	---	0.55				
All Season	Aranae[b]	spiders	0.31	18.9	---	0.81	4.23	256	---	1.76
	Hemiptera	bugs	0.09	5.78	0.02	1.05	1.14	67	---	0.23
	Hymenoptera	bees & ants	0.27	15.3	0.08	0.95	3.25	179	---	0.16
Mean (dis/min g^{-1})			0.29	16.1	0.03	0.70	4.76	265	0.01	0.50
$s_{\bar{x}}$[d]			0.08	4.3	0.01	0.10	1.12	61	0.00	0.26

[a]Twenty-two of 38 ^{239}Pu counting errors (s/count) were less than 5%. Errors ranging from 6–30% were associated with the remaining samples, all of which were <2 dis/min g^{-1}. Twenty of 26 ^{238}Pu counting errors ranged from 5% to 144%.
[b]Subclass, not an order.
[c]— indicates data reported as zero or negative.
[d]$s_{\bar{x}}$ is the standard error of the mean ($s/\sqrt{n}$).

plot, taxonomic group, and season of collection, and no significant interaction was detected. Mean plutonium concentrations in arthropods appear closely related to the mean plutonium levels previously measured (Li76) in 0–3 cm soil representative of each study plot at Rocky Flats (Fig. 1). Concentration ratios* of Rocky Flats arthropods to the 0–3 cm mean soil activity were 1.3×10^{-2}, 8.7×10^{-3} and 5.7×10^{-3} for macroplots H, 1 and 2, respectively. The overall mean concentration ratio and its propagated standard error (Ar72) based on standard errors for both soil and arthropod samples is $9.1 \times 10^{-3} \pm 1.4 \times 10^{-3}$.

The activity ratio of ^{239}Pu to ^{238}Pu was calculated for arthropod samples with detectable levels of ^{238}Pu. These isotopic ratios ranged from 12 to 126 with a mean of 52 and standard deviation of 25, based upon 26 samples. The mean isotopic ratio of 52 in arthropods is very close to the mean of 51 found in MP-1 and MP-2 soils, the compartment containing over 99% of the environmental plutonium at Rocky Flats (Li76).

Arthropod biomass estimates were multiplied by the respective ^{239}Pu concentrations from Table 1 to obtain inventory estimates of ^{239}Pu in the arthropod component of the ecosystem (Table 2). Total arthropod inventory estimates were 1.0 and 0.048 dis/min-m^2 for macroplots 1 and 2, respectively. Leafhoppers, grasshoppers and spiders accounted for 84 and 79%, respectively, of the total arthropod inventories in macroplots 1 and 2. The total ecosystem plutonium inventories were estimated as 1.3×10^8 and 6.3×10^6 dis/min m^{-2} for MP-1 and MP-2, respectively (Li76). The total inventory includes soil, the major plutonium compartment, as well as litter, vegetation, small mammals and arthropods. The fraction of the total inventories in the arthropod compartments are therefore estimated as 7.7×10^{-9} and 7.6×10^{-9} for MP-1 and MP-2, respectively.

*Concentration ratio = (dis/min g^{-1} arthropods) ÷ (dis/min g^{-1} soil).

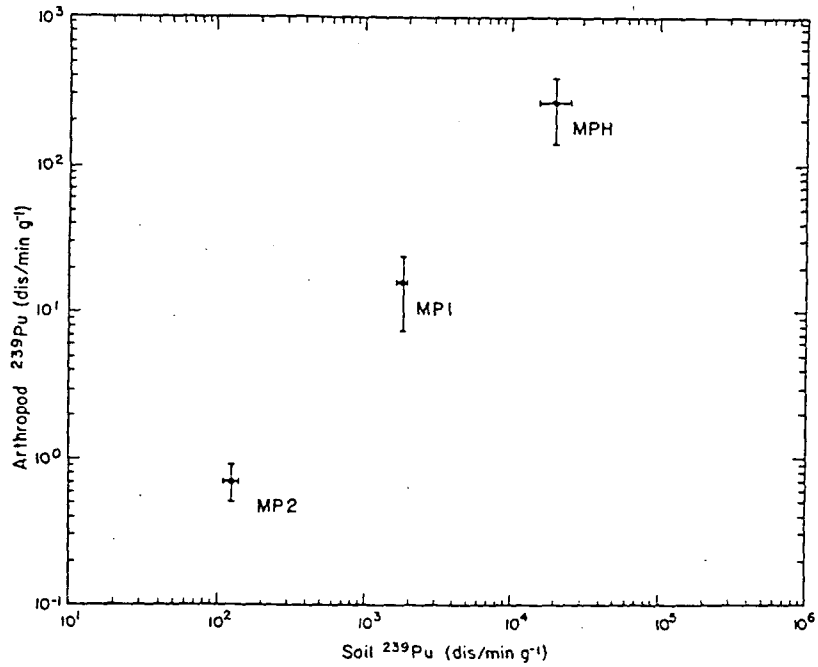

FIG. 1. Concentrations of ^{239}Pu in arthropods vs concentrations in soil at three Rocky Flats study plots. Error bars represent 2 standard errors of means.

Table 2. Biomass and ^{239}Pu inventory estimates for Rocky Flats arthropods

Season	Common names	Macroplot 1 mean biomass (mg/m^2)	Macroplot 1 estimated ^{239}Pu inventory ($dis/min\ m^{-2}$)	Macroplot 2 mean biomass (mg/m^2)	Macroplot 2 estimated ^{239}Pu inventory ($dis/min\ m^{-2}$)
Spring	beetles	3	0.061	4	0.006
	leafhoppers	18	0.245	42	0.044
	grasshoppers	4	0.017	2	0.001
Summer	beetles	4	0.030	8	0.005
	leafhoppers	65	0.586	20	0.009
	grasshoppers	82	0.365	67	0.017
Fall	beetles	12	0.244	15	0.003
	leafhoppers	37	0.048	31	0.017
	grasshoppers	23	0.644	15	0.008
All Season	beetles[a]	6	0.112	9	0.005
	leafhoppers[a]	40	0.293	31	0.023
	grasshoppers[a]	36	0.342	28	0.009
	spiders	12	0.227	8	0.006
	bugs	3	0.017	3	0.003
	bees, ants	2	0.031	2	0.002
	sum of orders	99	1.022	81	0.048

[a]Mean of spring, summer and fall values for biomass and ^{239}Pu inventory.

DISCUSSION

The coefficients of variation within study plots indicated arthropod plutonium concentrations to be more variable than soil but less variable than vegetation from the same sampling areas (Li76). Large sampling variability appears commonly in environmental plutonium studies. Spatial heterogeneity and in some cases, low sample activity contribute to this variability. Other sampling difficulties encountered with arthropods include animal mobility, several possible contamination mechanisms, and difficulty in collecting ground dwelling arthropods without cross contamination by the soil.

Statistical tests on the actual ^{239}Pu data were not valid because the requirements of normality and homogeneity of variance were not met. Bartlett's test for homogeneity of variance showed unequal variances between macroplots. However, after log transformation, the data yielded statistically equal variances ($\alpha = 0.05$). Statistical analyses (Sn67) strongly indicated skewness and kurtosis of the actual data, but this problem was also alleviated by use of log transformations. All sample data were therefore statistically evaluated by analysis of variance on the log transforms of the data.

Data suggested that concentrations of plutonium in soil and in animals were closely correlated. A plot of soil vs arthropod concentrations (Fig. 1), and the similarity in isotopic ratios support this concept. However, since the arthropods were not cleansed of surface activity prior to assay, the relative importance of ingestion and adsorption of dust as plutonium transport mechanisms cannot be determined. Preferential accumulation of ^{238}Pu over ^{239}Pu by arthropods, suggested previously (Li76), was not indicated in this study.

Arthropod biomasses were substantially higher than those of small mammals, emphasizing their relative importance as consumers in grassland ecosystems. While arthropods harbor only about 10^{-8} of the total plutonium inventory in the ecosystem, the data suggest substantially more plutonium in these animals than in mammals, birds or reptiles (Li76). The high degree of mobility of many arthropods, such as bees, grasshoppers and leafhoppers, implies that this group of organisms may effect measurable plutonium dispersal over extended periods of time.

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