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**Measurements of Uranium in
Soils and Small Mammals**

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Measurements of Uranium in Soils and Small Mammals

Felix Rafael Miera, Jr.

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MEASUREMENTS OF URANIUM IN SOILS AND SMALL ANIMALS

by

Felix Rafael Miera, Jr.

ABSTRACT

The objective of this study was to evaluate the bioavailability of uranium to a single species of small mammal, Peromyscus maniculatus rufinus (Merriam), white-footed deer mouse, from two different source terms: a Los Alamos National Laboratory dynamic weapons testing site in north central New Mexico, where an estimated 70 000 kg of uranium have been expended over a 31-yr period; and an inactive uranium mill tailings pile located in west central New Mexico near Grants, which received wastes over a 5-yr period from the milling of 2.7×10^9 kg of uranium ore.

INTRODUCTION

With the increasing use of atomic energy for peaceful and military application the environmental problems associated with radioactive and toxic waste products which may be introduced into the environment must be anticipated. Nuclear power generation using uranium is presently being examined as a viable alternative to fossil fuel power plants. Military applications include the use of depleted uranium ($^{235}\text{U}/^{238}\text{U} < 0.0072$) in conventional weapons tests and as armour-piercing projectiles. The need for appreciable production of uranium to feed these and other industries results in the generation of mill tailings piles which are left exposed to the natural environment. Despite the importance and ubiquitous nature of uranium, its ecological behavior in the environment is not well understood.

The objective of this study was to evaluate the bioavailability of uranium to a single species of small mammal, Peromyscus maniculatus rufinus (Merriam), white footed deer mouse, from two different source terms: a Los Alamos Scientific Laboratory dynamic weapons testing site in north central New Mexico where an estimated 70,000 kg of uranium have been expended over a 31 year period; and, an inactive uranium mill tailings pile located in west central New Mexico near Grants which received wastes over a five year period from the milling of 2.7×10^9 kg of uranium ore.

In this study an attempt is made to determine if uranium concentrations in tissues and organs of Peromyscus can be related to soil uranium concentrations for the respective sites. Uranium concentrations were determined for various soil size fractions and whole, i.e. unfractionated soil samples, to assess uranium availability to Peromyscus through ingestion or respiration of contaminated materials.

LITERATURE REVIEW

Uranium is an ubiquitous constituent of our natural environment. With the introduction of nuclear weapons during World War II an intensive effort was initiated to characterize uranium poisoning and to determine relative toxicities of the various uranium compounds for establishing human health standards in the industry (Yuile, 1973). Decay characteristics for isotopes of uranium with mass numbers 227 through 240 have been reported (Gindler, 1973); of these, only the isotopes ^{235}U and ^{238}U have half-lives sufficiently long (7.1×10^8 and 4.49×10^9 years, respectively) to be found as primordial substances in nature.

Most natural uranium ores contain 0.1 to 2% uranium (U_3O_8) with relative abundances of ^{235}U and ^{238}U of 0.72% and 99.27%, respectively. Milling processes remove about 90% of the total uranium, with the remainder released in the tailings (Tsivoglou and O'Connell, 1964). Depleted uranium, a by-product resulting from the ^{235}U enrichment process, contains about 0.2 wt% ^{235}U and the remainder is composed of ^{238}U . Differences in natural and depleted uranium thus result from a departure of the crustal abundance isotopic ratio: $^{235}\text{U}/^{238}\text{U} < 0.0072$.

Environmental studies in areas of high natural radioactivity (Cannon, 1952; Mericle and Mericle, 1965; Cullen and Franca, 1977) and uranium mill waste investigations (Anderson *et al.*, 1963; Dreesen, 1978), have for the most part been concerned with radiation effects of nuclides other than uranium and other nonradioactive contaminants. The primary processing removes most of the ^{238}U decay chain products that are of major concern for depleted uranium. Principal concern for the isotopes ^{235}U and ^{238}U is their chemical toxicity; the alpha particle emissions from these nuclides have very low penetrability

in air (≈ 3 cm).

The geological distribution of uranium ore bodies has been studied in much detail, and is reported throughout the literature. However, a recent review of the literature (Hanson, 1974) indicates a limited number of indepth studies describing the distribution and fate of uranium contaminants in the environment. The chemical and physical parameters which govern the mobility of uranium are known to be influenced by such parameters as clay content, organic matter content and soil acidity. The natural uranium content of surface waters in the U.S. averages 5×10^{-4} $\mu\text{g U/l}$, although this value is highly variable, and is influenced by factors such as the geological formations in which waters arise and flow through and the size and diversity of their watersheds (Koczy, 1963).

In terrestrial ecosystems studies in uranium-vanadium deposits of the Colorado Plateau showed the following responses of vegetative species to their chemical environment: uranium absorption, anomolous growth habits, and the development of a particular flora or indicator species (Canon, 1952). For plants rooted in uranium bearing rock highest concentrations are usually found in roots and seeds. Uranium concentrations ranged from 2 to 100 ppm, compared to values of generally less than 1 ppm for species from outside this area. Availability of uranium to plants is strongly influenced by soil clay content, organic matter content, soil acidity and depth of the root system. Cannon (1952) reported concentration factors (plant uranium concentration $\div$ soil uranium concentration) for shale derived soils of 0.09 to 0.9 and 0.007 to 0.01 in sandstone soils for a wide variety of plants. Morphological effects in plants from high uranium areas resulting from radiation induced genetic change include double

flowering, leaf splitting and corolla collar variation (Kovalsky et al., 1967).

Investigations of small mammals which permanently inhabit regions of high natural radioactivity have again mostly emphasized radiation effects. Important pathways for potential exposure and biological uptake include: (1) close contact with the soil layer in their burrows and above ground activities; (2) inhalation of resuspended soil particles and gaseous products, as from radon gas; (3) ingestion of contaminated food materials, both vegetation and invertebrates, or soil particles which are attached to the animals pelt and ingested during grooming activities.

In intensive studies of areas with high natural radioactivity in the U.S.S.R. (Maslov et al., 1967) several biological effects were attributed to the increased radiation levels. Morphological and physiological changes were observed in the germinal epithelium of the gonads, tissues of the spleen, bone marrow, liver cornea and other organs. Exposure levels in the study area ranged from 5000-8000 $\mu\text{R/hr}$. The authors concluded that over the several generations of exposure the population as a whole was affected resulting in a decrease in vitality and fecundity of the population.

However, to gain an understanding of the effects attributable solely to uranium, one must rely on the results obtained in controlled laboratory experiments. Based on the results of extensive experimentation, it is generally concluded that the degree of acute uranium poisoning depends on the amount initially absorbed and the consequent level in the bloodstream, irrespective of the route of administration. Critical pathways for the uptake of uranium pertinent to this study are ingestion of uranium contaminated materials and inhalation of respirable size

particles to which uranium may be adsorbed.

In short-term (30 day) feeding experiments with rats, uranium compounds which are relatively insoluble in water, such as U_3O_8 , UO_2 , and UF_4 , were fed in diets containing up to 20% of these substances and were found to be non-toxic and showed little if any interference with growth. Also, no significant histopathological changes were detected. However, 100% mortality resulted from diets which contained 2 to 10% uranium compounds which are highly water soluble, such as UO_2Ac_2 , UCl_4 , UO_2F_2 , $UO_2(NO_3)_2 \cdot 6H_2O$, UO_3 and UO_4 . Diets containing 0.1 to 1.0% of the water soluble compounds resulted in growth depression (Yuile, 1973). Renal effects were also detected in this group, with extent of damage and frequency of occurrence dependent on the compound and dosage.

In oral ingestion experiments with durations of 1 to 2 years, the relative toxicity of uranium compounds was obtained by comparing the dietary percentages required to produce equal body weight depressions: UO_2F_2 , 0.25%; $UO_2(NO_3)_2$, 1%; UF_4 , 20%; UO_2 , at 20% showed no influence on growth (Voegtlin and Hodge, 1953). Rats maintained on a 2% uranyl nitrate diet showed moderately severe initial injury followed by regeneration and progressive atrophic changes in the renal tubules, as well as some degenerative changes in the testes. Uranium analyses of the kidneys and femurs receiving these four uranium compounds showed that only slight increases of uranium were detected for the insoluble compounds with significant increases for the soluble uranium compounds. After cessation of ingestion, uranium in the bone was found to mobilize and be excreted from the body.

Regarding toxicity following inhalation, the solubility of uranium compounds was again a significant factor in producing effects. Particle

size is also an important parameter, with residence time in the lungs generally increasing as particle size decreases. The sequence of events in uranium poisoning following inhalation is similar to other routes of uranium administration and is dependent on maximum levels in the blood stream attained at any given time by solubilization and absorption of uranium.

DESCRIPTION OF STUDY AREAS

The LASL Study Area

The Los Alamos Scientific Laboratory consists of 113 km² in north-central New Mexico on the Pajarito Plateau, which is situated on the eastern slopes of the Jemez Mountains west of the Rio Grande (Fig. 1). The plateau, which has a pronounced elevational gradient in the east-west direction, was formed by successive ash flows from a volcanic area 32 km to the west. The soils of the area have just recently been characterized in detail (Nyhan et al., 1978). The climate is semiarid, with approximately 46 cm of annual precipitation. Nearly 75% of this occurs during spectacular May-October thundershowers and accounts for much of the canyon erosion.

E-F testing site, situated on a mesa top at 2190 m elevation, is located in the western central portion of LASL and has had the most continuous use history for the expenditure of uranium at LASL, beginning in mid-1943. An estimated 65,000-75,000 kg of natural and depleted uranium have been expended at this site in conventional explosives tests over a 30-year period from 1943 to 1973. Reportedly, no cleanup operations have occurred to date due to the lack of known or suspected hazardous conditions. Thus, it is assumed that the uranium inventory is available for uptake and transfer through the food chain, or for surface transport, mainly by storm runoff in the adjacent Potrillo Canyon drainage.

COLORADO

NEW MEXICO

Fig. 1. Location of the Los Alamos Scientific Laboratory in northcentral New Mexico.

The LASL study site exhibits evidences of both uranium's pyrophoric properties and/or resultant explosive properties, in that the overstory vegetation surrounding the site has been burned and is in successional recovery stages. The understory vegetative cover consists mostly of three invader species, Kochia scoparia (Belvedere summer-cypress), Salasola kali (Russian thistle), and Sitanion hystrix (bottlebrush squirreltail). Dominant overstory vegetation found on the outer perimeter of the study site includes Pinus ponderosa (ponderosa pine), Pinus edulis (pinon pine), and Juniperus monosperma (one-seed juniper).

The Uranium Mill Tailings Study Area

The Phillips Petroleum tailings pile is located approximately 40 km north of the southern extension of the San Juan Basin in the Ambrosia Lake mining district in northwestern New Mexico (Fig. 3). Mount Taylor is the prominent extrusion in the area, located approximately 20 km to the southeast of the study area. Major drainage through the Ambrosia Lake area is the southeastern trending Arroyo del Puerto, which has perennial flow resulting from mining and milling processes in the area and is a tributary of San Mateo Creek. Annual precipitation averages about 25 cm.

The tailings for the Phillips Petroleum pile were produced from milling about 2.7×10^9 kg of uranium ore from 1958 to 1963. The estimated volume of tailings was reported in 1977 to be about 1.5×10^6 m³, ranging in thickness from 1 to 10 m (Purtymun et. al., 1977). The uranium was extracted from the ores using a carbonate leaching process, the waste separated from the leach solution and pumped to the pile as a slurry. A series of dikes were built surrounding the pile to contain the tailings. No vegetative species have successfully adapted to the pile, apparently a result of the chemical harshness of the environment. Transport of mill tailings from the pile to adjacent areas is via wind erosion and

■ PHILLIPS PETROLEUM TAILINGS PILE

Fig. 2. Location of the Phillips uranium mill tailings pile.

surface water transport.

The outer periphery of the pile where study areas were located, evidences disturbed habitat with lush growths of invader vegetative species, primarily Sitanion hystrix, Salsola kali, and Kochia scoparia. Native vegetation in the vicinity of the tailings pile is that typically found in a short-grass plains area, dominated by Boutelous gracilis (blue grama) and Hilaria Jamesii (galleta grass).

METHODS

Sample Collection at E-F Site

A polar coordinate sampling scheme was devised for intensive investigation of the vertical and horizontal soil distribution of uranium at this site for a separate study (Miera et al., 1978). Soil cores were collected at the intersection of radii that extended from the detonation point at each 45° azimuth and concentric circles 10, 20, 30, 40, 50, 100, 150, and 200 m from the detonation point (Fig. 3). A polyvinylchloride coring tube (2.5 cm i.d.) with a sharpened end was used to collect two 30-cm deep soil cores spaced 0.5 m apart at each sampling location.

One core from each location was cut into segments corresponding to 0- to 5- and 5- to 10-cm to examine depth distribution patterns. Thirty percent of the duplicate cores collected from the NE, SE, SW, and NW quadrants were randomly selected, and similarly processed, to define the spatial variability of uranium with distance from the detonation point and with depth into the soil profile.

Forty soil samples representing duplicate 0- to 5-cm and 5- to 10-cm depth cores collected from the N, E, S, and W quadrants at distances of 10, 20, 50, 100, 150, and 200 m from the detonation point were processed for soil particle size analysis. These samples were mechanically separated by means of a sonic sifter into six size fractions.

Fig. 3. Soil sampling and small mammal trapping locations at the LASL site.

Modified North American Census of Small Mammals (NACSM) trapping lines (Calhoun, 1948) were permanently marked for collection of Peromyscus during April, 1977. Two trap lines were established at the site parallel to each other and the earthen mounds that bracket the detonation point. Each line was 190 m long and consisted of 20 stations at 10-m intervals. Three snap traps baited with peanut butter were placed within 1 m of each station in positions that appeared most likely to catch small mammals. The traps were prebaited for two nights, followed by three consecutive nights of trapping.

Data recorded on all animals included capture location, species identification, sex, age class, reproductive condition, and weight. The mice were then packaged individually and frozen pending dissection. Measurements of each specimen at dissection included lengths of total body, tail, ear, and hind foot.

Trap line positions were determined by approximated fallout pathways of particulate material from the firing site. Thus, the distance between the parallel trap lines was less than that used for standard NACSM estimates of home ranges and populations. The size of area sampled for rodent populations was determined using the method of Brandt (1962). This method utilizes the average distance between successive points of capture. As snap traps were used for this study, a value of 52.4 m for Peromyscus was also obtained from the data of Brandt, assuming that habitats were not significantly different for these calculations. This value was determined as an average for both sexes during five trapping periods on a grid. Treating the 33 m distance between the parallel trap lines as an overlapping range, the area sampled was determined to be 4.1 hectares.

Sample Collection at the Uranium Mill Tailings Site

Soil and small mammal samples were taken from the north, east, and

south sides, on the outer periphery but adjacent to the tailings pile (Fig. 4). An intensive survey of uranium in the soils at this site could not be undertaken, because the total number of chemical analyses that could be performed was limited. A random sampling pattern was used for the purposes of determining concentration patterns of uranium, spatial variability distribution of uranium to a depth of 10 cm, and distribution of uranium associated with various soil particle size fractions.

Trapping lines similar to those described previously, were permanently marked on each of the three sides of the pile. Four random soil sampling plots were located at stations 2, 8, 10, and 15. Additional soil sampling plots were located at a 10 m distance on each side of and perpendicular to the sampling points located on the trap lines.

At each of the soil sampling points, a 1 m² plot was marked off and all standing vegetation removed to ground level. Two soil samples were collected on the diagonal from the NW and SE corners of the 1 m² plots, in 10 x 10 x 5 cm increments at 0- to 5- and 5- to 10-cm depths. Samples taken from the NW corners of each plot located on the center trap lines were used for analysis of soil particle size distributions.

Two trapping sessions were required to collect sufficient numbers of micr for analyses. For the first session in July 1977 single modified NACSM trapping lines were established parallel to each of the three sides of the pile. Low capture success resulting from inclement weather necessitated a second trapping session in early August 1977. In an effort to increase trapping success, two additional trap lines were established, on each side of and parallel to the original lines. Distance between the parallel lines was limited to 10 m by access and ore haulage roads parallel with the south and east sides of the pile. Three snap traps baited with peanut butter were placed within a 1 m radius of each trapping station.

Fig. 4. Grid locations for small mammal trapping and soil sampling at the perimeter of the Phillips mill tailings pile.

Both trapping sessions consisted of two nights of prebaiting, followed by three consecutive nights of trapping and collections. Similar data was recorded for all individuals as described for E-F Site specimens.

Since most Peromyscus captures were in the second trapping session, the size of area sampled was determined by considering the three lines as a partial grid. The area sampled on each side of the pile was 0.8 hectares, determined by calculating the area of a rectangle with dimensions of 200 m by 40 m.

Sample Preparation

All soil samples collected were oven-dried at 100°C to a constant dry weight ($\approx$ 24 hrs.) and the dry weight recorded. The samples were then passed through a 6-mm screen to remove large pieces of rock, roots, and other debris. With the exception of those samples to be used for particle size analyses, all whole samples were ground to less than 100 mesh in a pulverizer and thoroughly blended to provide a homogenous sample. A 3- to 4-gm aliquot of each blended sample was submitted for uranium analyses, with replicate aliquots submitted for approximately fifteen percent of the total samples.

Soil samples collected for particle size analyses were mechanically separated into six size fractions by means of a sonic sifter to: $<53\text{ }\mu\text{m}$, $53\text{--}105\text{ }\mu\text{m}$, $105\text{--}500\text{ }\mu\text{m}$, $500\text{--}1000\text{ }\mu\text{m}$, $1\text{--}2\text{ mm}$, and $2\text{--}23\text{ mm}$ diameters. Small soil particles were blown from the larger soil separates with an air hose after sifting to minimize contamination of the large particles. The particle diameters may be further described according to US Department of Agriculture Soil Conservation Service standard soil size categories as follows:

<u>Particle Diameter (µm)</u>	<u>Size Fraction</u>
<53	Silt and clay
53-105	Very fine sand
105-500	Medium and fine sand
500-1000	Coarse sand
1000-2000	Very coarse sand
2000-23000	Coarse fragments

All rodents were carefully dissected to minimize cross-contamination of internal organs and tissues by soil particles adhering to the fur. Tissues and organs analyzed separately included the pelt, carcass (skelton and skeletal muscles), lungs, GI (gastrointestinal) contents, liver, and kidneys. To increase sample mass and uranium analytical detection sensitivity, deer micr samples were pooled by pairing according to capture location and sex. Rodent samples were oven-dried at 100°C for 24 hrs, or until a constant dry weight was recorded, and then muffled at 450°C until a white ash was obtained. The entire ashed sample was submitted for analyses.

Analyses of Samples for Uranium

Two analytical techniques were used in the analysis of samples for uranium in this study. The delayed neutron assay technique was used for all samples collected in the Grants area. This technique operates on the principle of a fixed uranium isotopic ratio ($^{235}\text{U}/^{238}\text{U}$) because ^{238}U does not fission with thermal neutrons. As the ratio departs from 0.0072, as with depleted uranium, large errors in determination of total uranium can be made. Thus samples from the LASL area were done by instrumental epithermal neutron activation analysis (IENAA), which is insensitive to ^{235}U (Steinnes, 1971; Gladney et al., 1978).

Delayed neutron measurements were performed by irradiating the sample

for 20 seconds in a thermal neutron flux. The sample was pneumatically transferred to a neutron detector of 27% efficiency and counted for 20 seconds looking at emissions of about 6 short lived decay products. The uranium concentrations were calculated assuming natural isotopic abundance of ^{235}U .

Samples analyzed by the IENAA technique were irradiated with epithermal neutrons for two minutes with the reaction $^{238}\text{U} (n, \gamma) ^{239}\text{U} \xrightarrow{\beta, \gamma} ^{239}\text{Np} \xrightarrow{\beta, \gamma} ^{239}\text{Pu}$. After 2 to 4 days decay, the gamma spectra of the 228 and 278 keV transitions from the decay of ^{239}Np ($t_{1/2} = 2.35$ d) were determined on a large Ge(Li) detector and used for quantitative analysis. Several standard reference soil, plant and tissue materials (International Atomic Energy Agency and National Bureau of Standards) with certified concentrations over the range of expected uranium results were used to standardize all analysis.

RESULTS AND DISCUSSION

Distribution of Uranium in LASL Soils

Highest mean concentrations for the 0- to 5-cm sampling depth at the LASL site were found within 10 m of the detonation point, averaging 4750 and 4910 $\mu\text{g U/g}$ at 0 and 10 m respectively. Uranium concentrations in surface soils decreased exponentially with distance from the detonation point and distances beyond 50 m from the firing point were generally less than 15% of those taken within 10 m (Table I). Surface soil uranium concentrations varied greatly, and presumably were strongly influenced by the variable deposition of uranium debris in the form of fragments from past explosive tests. The Coefficient of Variation $\left[(CV = \text{standard deviation/mean}) \times 100 \right]$ for soils collected 0.5 m apart at individual sampling locations ranged from 18 to 96%, averaging 57% for all distances. For samples collected along individual annuli i.e., sampling locations at the same distance CVs

Table 1. Uranium concentrations in soils collected from two depth increments at LASL.

Distance (m)	<u>0 - 5cm Depth</u>			<u>5 - 10cm Depth</u>		
	Number of Samples	Mean Uranium Concentration ($\mu\text{g/g}$)	C.V. ^a	Number of Samples	Mean Uranium Concentration ($\mu\text{g/g}$)	C.V.
0	2	4750	64	2	3800	33
10	8	4910	100	6	3950	99
20	6	835	68	4	190	66
30	8	1330	84	6	640	66
40	7	710	36	6	470	62
50	8	475	59	7	570	94
100	8	370	91	4	165	50
150	8	730	231	2	40	61
200	7	165	91	2	18	58
All Soils	62	1340		39	1120	

^aCoefficient of Variation

averaged 36 to 100% within 100 m of the detonation point but increased to as high as 230% at 150 m distances.

Uranium concentration isopleths shown in Figure 5 demonstrate the heterogeneity of uranium dispersal resulting from the test shots. Isopleths were obtained by transforming each data point to log values, and then converting the polar coordinate sampling array values to Cartesian coordinate values. The plane surface was generated by a program which interpolated between data points. From the isopleths, areas of highest soil concentrations were detected at the detonation point and 150 m to the west with uranium levels $>3000 \mu\text{g/g}$ soil. Surface soils to the south and northeast of the detonation point also exhibited high uranium concentrations ranging from 300 to $1000 \mu\text{g/g}$ of soil.

Samples taken at the 5- to 10-cm depth within 10 m of the detonation point were only slightly lower than surface values observed for these same sampling locations. For sampling locations at 20 m and beyond, differences between the 0- to 5-cm and 5- to 10-cm depths, mean uranium concentrations ranged from factors of 2 to 18 times less, with one exception at 50 m. However, the large variations associated with observed values (Table 1) again indicate the strong influence of uranium dispersal patterns from explosive test shots. Mean concentrations of 165 and $18 \mu\text{gU/g}$ at 100 and 200 m sampling locations are about 80 and 10 times greater, respectively, than background uranium levels in soils for the area which range from $0.2\text{--}1.2 \mu\text{g/g}$ (Johnson, 1972). These data emphasize uranium's mobility in the soil and also that uranium has penetrated to greater depths than sampled.

To evaluate the potential for redistribution of uranium which might be adsorbed to soil particles by physical or biological means, the particle size composition and uranium content of soil separates were

URANIUM CONCENTRATIONS AT E F SITE

ISOPLETHS: 0-5cm HORIZON

SCALE
0 25 50 100
meters

SOIL SAMPLING LOCATIONS

Fig. 5. Calculated isopleths of uranium concentrations in the 0- to 5-cm soil horizon at the LASL site.

determined. Uranium concentrations in six soil size separates from the 0- to 5-cm and 5- to 10-cm depths are presented in Table 2. Generally, an exponential decrease of uranium concentration with distance was observed for each size fraction, although with appreciable variation noted in values as a function of both distance from the detonation point and depth in the soil profile. Highest concentrations of uranium were found at the 10m distance in the $<53\mu\text{m}$ size fraction for both soil column increments; however, at 20m to 50m distances the highest concentrations were generally associated with the larger particle sizes (1-2 mm). For sampling locations 100- to 200-m from the detonation point the highest concentrations were again associated with the smaller soil particle sizes ($<500\mu\text{m}$).

The fractional distribution of uranium in the soil separates was also determined. The portion of the total uranium in each size fraction was calculated by multiplying the concentration by the soil mass of each fraction. The distribution of uranium for all sampling locations in the six soil size fractions for the 0- to 5- and 5- to 10-cm depths indicated that about 70% of the total uranium and 60% of the soil mass were associated with $<500\mu\text{m}$ soil particles (Fig. 6). For the 0- to 5-cm depth, the percent uranium associated with the $<500\mu\text{m}$ size fractions increased from 50% at the 10 m sampling locations to nearly 90% at the 200 m sampling locations. Soil masses for the respective sampling locations in the $<500\mu\text{m}$ fractions (classified as silt, clay, and sand fractions) were 40 and 70%. The ratio of the total uranium in the $<53\mu\text{m}$ fraction to that in the 2-23 mm fraction for the 0-5- and 5-10-cm depths increased from 1.1 and 2.5 at 10 m to 18 and 21 at the 200 m sampling locations, respectively. These distribution patterns suggest that mobilization and physical transport, i.e., surface water and wind, of uranium associated with smaller soil particles

Table 2. Mean uranium concentrations in soil size fractions at LASL as a function of distance and depth.

Distance (m)	Soil Depth (cm)	Number of Samples	$\bar{x}$ U($\mu\text{g/g}$) in soil separates					
			<53 μm	53-105 μm	105-500 μm	500-1000 μm	1-2mm	>2mm
10	0 - 5	4	7280	2050	3500	2830	2940	2430
	5 - 10	4	5130	2450	2690	1870	1720	1150
20	0 - 5	3	1260	820	1030	595	1700	1290
	5 - 10	3	360	350	450	380	900	145
50	0 - 5	5	795	900	900	530	750	205
	5 - 10	5	425	950	510	420	300	200
100	0 - 5	4	240	430	410	285	720	120
	5 - 10	4	170	205	230	210	160	8
150	0 - 5	2	80	110	130	50	14	17
	5 - 10	2	31	80	55	27	12	33
200	0 - 5	2	170	250	310	115	20	29
	5 - 10	2	29	27	19	20	7	4

Fig. 6. The percent of the total uranium and percent soil mass associated with each size fraction as a function of soil depth and distance from the detonation point at the LASL site. The left-side bar for each size fraction is for the 0- to 5-cm depth and the right-side bar is for the 5- to 10-cm depth.

is occurring with time at this site.

Distribution of Uranium in Soils at the Perimeter of the Phillips Mill Tailings Pile

The highest mean uranium concentrations detected for whole soil samples collected at the perimeter of the Phillips mill tailings pile were about fifty times less than those observed at the detonation point of LASL's E F site; five and four times less than average values at 50 and 100 m, respectively; and within the range of values for samples collected at the 200 m distance. Highest mean uranium values were detected on the east side of the pile averaging 86 ± 28 (1 S.D.) $\mu\text{gU/g}$ soil in the top 5 cm and 103 ± 32 $\mu\text{gU/g}$ in the lower 5 cm of soil. Mean values for both depth increments sampled on each of the three sites are presented in Table 3. Coefficient of variation values of less than 50% were measured for all sample groupings with the exception of the 0-5 cm depth on the north side, compared to CV's which ranged up to four times higher in the LASL soils. For samples taken across the diagonal in each 1 m² plot, the east and south transects generally had CV values of less than 20%. However, samples taken on the north transect reflected high spatial variability, with some samples having almost identical concentrations, and others ranging as high as 100% in variation.

The particle size composition and uranium content of soil separates was determined for the north and east transects as previously described. The results of this exercise are shown in Table 4 and Fig. 7. Over 90% of the soil mass consisted of particles <500 μm in size, with the greatest portion in one size category, the 105 to 500 μm fraction. This trend was observed for both depths sampled and for both transects. The importance of this parameter is that particles less than 500 μm in size are those which are generally associated with wind and water erosion. Again, over 90%

Table 3. Mean uranium concentrations in soils to a 10cm depth collected on the perimeter of the Phillips mill tailings pile.

<u>Transect</u>	<u>0 - 5cm Depth</u>			<u>5 - 10cm Depth</u>		
	<u>Number of Samples</u>	<u>Mean Uranium Concentration ($\mu\text{g/g}$)</u>	<u>C.V.^a</u>	<u>Number of Samples</u>	<u>Mean Uranium Concentration ($\mu\text{g/g}$)</u>	<u>C.V.</u>
North	12	58	68	11	70	48
East	12	86	33	12	105	31
South	8	44	26	8	38	27
All Soils	32	65		31	74	

^aCoefficient of Variation = standard deviation/mean x 100.

Table 4. Mean uranium concentrations in soil size fractions collected on the perimeter of the Phillips mill tailings pile.

			<u>$\bar{x}$ U($\mu\text{g/g}$) in Soil Seperates</u>					
	Soil Depth (cm)	Number of Samples	<u><53μm</u>	<u>53-105μm</u>	<u>105-500μm</u>	<u>500-1000μm</u>	<u>1-2mm</u>	<u>>2mm</u>
North	0 - 5	8	125	82	94	215	82	150
Transect	5 - 10	8	180	120	87	335	95	95
East	0 - 5	8	110	79	56	120	115	N.D. ^a
Transect	5 - 10	6	110	80	68	85	N.D.	N.D.

^aNo data resulting from a lack of soil particles in this size fractions.

Fig. 7. The percent of the total uranium and percent soil mass associated with each size fraction as a function of soil depth at the perimeter of the mill tailings pile. The left-side bar for each size fraction is for the 0- to 5-cm depth and the right-side bar is for the 5- to 10-cm depth.

of the uranium inventory for both transects and depths measured was associated with soil particles less than 500 μm in size and closely paralleled the particle size distributions. These results also indicate that vertical migration of uranium has occurred and that the distribution of uranium to depths sampled was relatively homogenous.

The relatively homogeneous distribution of uranium in these soils is most probably controlled by two mechanisms. First, solubility of uranium at this site resulting from the refining process. As a consequence any precipitation or resulting runoff would distribute the uranium more evenly through the soil matrix. Higher mean concentrations for the lower depth samples on the north and east transects, also supports the hypothesis of a soluble and mobile form of uranium. Second, the uniformity of soil particle sizes in which the uranium is dispersed, as vertical and horizontal mixing of soils is related to particle size composition.

Uranium Concentrations in Tissues of Small Mammals

Trapping efforts for the two study areas yielded twenty and nineteen individuals captured at the LASL and mill tailings study areas, respectively. Mean measurements of selected physical characteristics by sex are listed in Table 5, and are similar for populations of both sites. The means recorded are within the range reported for this species (Hall and Nelson, 1959). The higher mean weight for females from the mill tailings area was attributed to three individuals which were pregnant. Based on the size of area sampled minimal density estimates of Peromyscus for the areas sampled were 5 individuals/hectare at the LASL site and 8 individuals/hectare at the mill tailings study site.

Uranium concentrations detected in tissues and organs of small mammals for the LASL and mill tailings study area are listed in Table 6,

Table 5. Physical Measurements of Adult Small Mammals, ± 1 S.D., Collected LASL and the Phillips mill tailings pile.

<u>LASL E-F Site</u>	<u>Number of Samples</u>	<u>Mean Weight (gm)</u>	<u>Total Length (mm)</u>	<u>Tail Length (mm)</u>	<u>Hind Foot Length (mm)</u>	<u>Ear Height (mm)</u>
Males	11	20.3 $\pm$ 2.2	147 $\pm$ 5	59 $\pm$ 5	18 $\pm$ 2	17 $\pm$ 2
Females	9	19.3 $\pm$ 4.9	144 $\pm$ 9	58 $\pm$ 6	18 $\pm$ 2	17 $\pm$ 3
 <u>Phillips Mill Tailings Pile</u>						
Males	8	18.3 $\pm$ 2.4	138 $\pm$ 6	60 $\pm$ 3	18 $\pm$ 2	16 $\pm$ 2
Females	11	22.8 $\pm$ 5.4	143 $\pm$ 8	60 $\pm$ 2	18 $\pm$ 2	17 $\pm$ 1

with both the mean and medium values reported. For interpretation of these data tissues are grouped as to samples which have the potential for direct exposure to the external environment, i.e. the pelt, GI contents and lungs, and those tissues which have no contact with external uranium sources and included the kidney, liver and carcass (bone and skeletal muscle tissues). Although uranium concentrations were generally higher in all tissue groupings for LASL samples, comparisons of these values with various soil parameters investigated and uranium source terms provided interesting interpretations of the data.

Mean uranium concentrations for composite male plus female values indicates a significant difference in concentrations between tissue types. At both sites GI contents and pelt samples had the highest mean concentrations, all within the range of values reported for soils at each site. The remaining four tissues sampled at the LASL site decreased rapidly in mean concentrations, all ranging from 8 to 16 $\mu\text{gU/g}$ sample compared to values of about 2000 and 500 $\mu\text{gU/g}$ sample for GI and pelt samples, respectively. The low values obtained for lung tissues indicates that inhalation of respirable size particles was not occurring appreciably during the period of time that trapping was conducted. Again the heterogeneity of uranium dispersal in soils as well as the effects of the mobility of Peromyscus are reflected by the range of minimum to maximum values and CV's of 100% to 230% recorded for all tissue groupings.

Tissues from Peromyscus composites sampled in the vicinity of the mill tailings area showed similar results for tissue groupings with GI and pelt samples having the highest mean concentrations of 235 and 155 $\mu\text{gU/g}$ of sample, respectively. Carcass, lung and liver tissues had essentially identical means of about 4 $\mu\text{gU/g}$ (Table 6). One

Table 6. Uranium concentrations ($\mu\text{gU/g}$) in tissues of small mammals collected from two study areas.

<u>LASL</u>	<u>$\bar{x}$</u>	<u>Median</u>	<u>C.V.^a</u>	<u>Max.</u>	<u>Min.</u>	<u>N^b</u>
GI	1985	880	1.22	8100	143	11
Pelt	510	350	0.95	1530	102	11
Kidney	16	19	1.19	60	N.D. ^c	11
Carcass	10	9	1.06	30	N.D.	11
Lung	8.2	24	2.32	62	N.D.	11
Liver	16	19	1.19	60	N.D.	11
 <u>Mill Tailings</u> <u>Pile</u>						
GI	235	170	0.79	590	62	9
Pelt	155	91	0.90	430	40	9
Kidney	47	29	0.91	130	4	9
Carcass	4.6	4.4	0.35	7	1.8	9
Lung	4.5	5.4	0.67	8.4	1.3	9
Liver	4.1	2.2	1.24	17	0.5	9

^aCoefficient of variation = standard deviation/mean

^bNumber of samples

^cBelow detection limit of 0.5 $\mu\text{gU/g}$

significant difference was the higher value detected for kidneys, 47 $\mu\text{gU/g}$, which also supports the hypotheses of a more soluble form of uranium at this site. The kidneys in their physiological function of acting as a filtering medium for body fluids are recognized as the critical organ in reference to uranium (ICRP, 1969). As was observed in the review of the literature for experimental data with mice, oral feedings of water soluble uranium compounds also increased the uranium content of kidneys, whereas this was not found for insoluble uranium compounds. The range of values for all composite tissue groupings at this site was not as great as that observed for LASL samples. This is reflected by CV's of less than 100%, with the exception of liver tissues.

Pooling of samples to enhance analytical sensitivity precluded any meaningful comparisons between sexes at the respective sites. However, inconclusive results obtained for this study indicate differences of uranium concentrations between sexes, to be a topic which warrant further investigation. The value in pooling to increase sample mass is reflected by some minimum values obtained for the LASL site, which even through this exercise were below detection levels of 0.5 $\mu\text{gU/g}$ sample.

Figures 8 and 9 are plots of uranium concentration for samples ranked from the lowest to the highest values detected for tissues which had the largest uranium values. As stated previously, the concentration distributions for these samples is very large, with the distribution positively skewed, i.e. most values occur below the mean. Thus, for these tissues with high uranium concentrations the mean value is biased towards the larger values with the median values (Table 6) probably providing a more realistic estimate.

To test for equality of distribution of uranium concentrations in

Fig. 8. Distribution of uranium concentrations for GI and pelt samples from the LASL site in relation to the mean values. Both distributions are positively skewed.

Fig. 9. Distribution of uranium concentrations for GI, pelt, and kidney samples from the perimeter of the Phillips mill tailings pile in relation to the mean values. All three distributions are positively skewed.

small mammal tissues, a non-parametric method, the Mann-Whitney test, (Snedecor and Cochran, 1967) was used. For this statistical technique both groups to be tested are arranged in a single array and assigned a rank. The smaller sum of ranks is then referred to a table to determine significance. Results of this exercise showed differences between study areas for pelt samples, GI samples and kidney samples ($P \leq 0.05$). For tissue concentrations within the mill tailings study area, GI versus kidney and pelt versus kidney were significantly different, and kidney concentrations were significantly different than carcass, lung and liver tissues ($P \leq 0.05$). For tissues from the LASL site, GI and pelt values were significantly different than the remainder of tissues ($P \leq 0.05$).

Observed concentration ratios (CR) of animal tissue uranium to soil uranium are presented in Table 7 and were calculated using both the mean and median tissue values. The soil value used was that of the 0-5cm horizon. The ratios for the mill tailings samples are generally larger than those obtained for similar tissues at the LASL study site. This is best reflected by compositing all tissues to determine the CR, with the mill tailings ratio being about 4 times larger than the LASL ratio. The mill tailings ratio was slightly greater than 1, with the LASL ratio calculated at 10^{-1} . For individual tissue groupings at the mill tailings site, GI and pelts had the largest ratio, followed by kidney tissues at 10^{-1} , and carcass, lung and liver ratios of 10^{-2} . Use of the median tissue values generally lowered the observed CR's, however, they remained within the same order of magnitude. For LASL study site samples the only CR calculated to be greater than 1 was for GI samples using the mean concentrations. Use of the median GI value decreased the CR by an order of magnitude to 10^{-1} . Pelts also exhibited CR's of 10^{-1} , with the remainder of the tissue groupings having ratios of 10^{-2} and 10^{-3} .

Table 7. Concentration ratios of uranium for the study areas.

	<u>Using Mean</u> $\left(\frac{\text{Tissue U}}{\text{Soil U}^a} \right)$	<u>Using Median</u> $\left(\frac{\text{Tissue U}}{\text{Soil U}^a} \right)$
<u>LASL</u>		
GI	1.48	0.66
Pelt	0.38	0.26
Kidney	1.2×10^{-2}	1.4×10^{-2}
Carcass	7.5×10^{-3}	6.7×10^{-3}
Lung	6.1×10^{-3}	1.8×10^{-2}
Liver	1.2×10^{-2}	1.4×10^{-2}
All tissues	0.32	
<u>Mill Tailings Pile</u>		
GI		2.62
Pelt	2.40	1.40
Kidney	0.72	0.45
Carcass	7.1×10^{-2}	6.8×10^{-2}
Lung	6.9×10^{-2}	8.3×10^{-2}
Liver	6.3×10^{-2}	3.4×10^{-2}
All tissues	1.15	

^aSoil concentrations for the 0-5cm depth were used for purposes of these calculations.

The observed concentration ratio provides an evaluation or estimate of the amount of uranium that moves across physiological barriers under conditions of their passage through the biological system (Comar, 1972), at the time of observation. Pelt, GI and lung values reflect the availability of uranium from the external environment to these consumers. The data indicates ingestion to be the major mechanism of concern for both study areas. Kidney, carcass and liver ratios, are indicators of the metabolic assimilation of uranium. The LASL results indicate that only a small portion of ingested uranium is assimilated, CR's for kidney $U:GI\ U$ being 10^{-3} , and that most of the uranium is excreted from the body. The mill tailings results, however, indicate a larger amount of uranium to be metabolically assimilated, in the order of 10^{-1} , presumably as a result of the more soluble form of uranium at this site.

SUMMARY AND CONCLUSIONS

This study was conducted to evaluate the bioavailability of uranium to Peromyscus maniculatus, the white footed deer mouse, from two separate source terms. One study area was located within the boundaries of the Los Alamos Scientific Laboratory (LASL) in north central New Mexico, at a dynamic weapons testing site where an estimated 70,000 kg of uranium have been expended. The second study area was an inactive uranium mill tailings pile located in west central New Mexico near Grants, generated from the milling of 2.7×10^9 kg of uranium ore. The objective of the study was to determine if the source term and resultant dispersal and distribution patterns in soil, affected the availability of uranium to Peromyscus.

Highest uranium concentrations in soils from the LASL study area were found within 10 m of the shot detonation site, averaging 4750 and 4910 $\mu\text{gU/g}$ soil at 0 and 10 m distances, respectively. Surface soil

uranium concentrations generally decreased exponentially with distance from the detonation point out to the farthest sampling locations of 200 m. Samples taken at 5- to 10-cm below the surface for sampling locations close in to the detonation site were only slightly lower than surface values. For sampling locations 20 m and beyond, results were highly variable, with lower horizon soils ranging from factors of 2 to 18 times lower than surface values. The uranium distribution in six soil size separates for both depth increments gave the following results: at a distance of 10 m, the <53 μ m size fraction had the highest concentrations; at 20 m to 50 m distances, highest concentrations were found for particles 1-2 mm in size; and for sampling locations at distances of 100 m to 200 m, highest concentrations were associated with particles less than 500 μ m in size. High variation in uranium distributions, indicated a strong influence of uranium dispersal patterns from the explosive test shots.

Soils from the perimeter of the uranium mill tailings pile had mean uranium concentrations about 50 times lower than the highest concentrations in LASL soils. Highest mean concentrations were detected on the east side of the pile averaging 86 μ gU/g for the top 5 cm and 103 μ gU/g for the 5- to 10-cm depth. Over 90% of the soil mass and about the same percentage of the uranium inventory at this site were associated with particles less than 500 μ m in size. The relatively homogeneous distribution of uranium in soils of this site were attributed to: the solubility of uranium at this site resulting from the refining process; and, the uniformity of soil particle sizes which optimizes vertical and horizontal soil mixing processes.

Uranium concentrations were generally higher in all small mammal tissue groupings for the LASL site when compared to values obtained

for tissue samples from the mill tailings study area. Concentrations of uranium in tissue samples were found not to be normally distributed about the mean and the distribution of values in tissue samples which had the highest concentrations were positively skewed. Nonparametric statistical testing showed that GI and pelt sample uranium concentrations were significantly different from internal tissue values for both sites. Perhaps one of the more significant findings of this study was that median uranium concentrations in kidneys of mice from the mill tailings study area were 6 times greater and significantly different from values for other internal tissues. This observation was not detected for the LASL study area. Low uranium values detected in lung tissues for both study sites indicated that inhalation of respirable size particle was not occurring appreciably during the sampling period, and that the major route for contamination was via ingestion.

Observed concentration ratios (CR) of animal tissue uranium to soil uranium ($\mu\text{gU/g}$) for all tissue groupings composited were larger for mill tailings samples ($\text{CR} > 1$) than for LASL samples ($\text{CR } 10^{-1}$). The observed concentration ratio was used to provide an estimate of the amount of uranium moving across physiological barriers under conditions of their passage through the biological system. The LASL results indicated that only a small portion of ingested uranium was metabolically assimilated. However, the mill tailings results indicated a larger amount of ingested uranium to be metabolically assimilated and is attributable to a more soluble form of uranium at this site.

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