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CESIUM-137, PLUTONIUM-239/240, TOTAL URANIUM, AND SCANDIUM IN TREES AND SHRUBS GROWING IN TRANSURANIC WASTE AT AREA B

by

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ABSTRACT

A unique radioecological study was carried out at a Los Alamos National Laboratory (LANL) shallow land burial site called Area B. Area B was the first common transuranic waste burial site for LANL from 1944 to 1948 and had lain fallow for 34 years. During this time secondary succession resulted in invasion of many native trees, shrubs, forbs, and grasses. The purpose of this study was to determine whether any trees or shrubs were rooting directly in waste material, to examine rooting patterns in a shallow land burial site, and to study the distribution patterns of different radionuclides by dissecting vegetative samples into representative compartments.

Scandium, ^{137}Cs, $^{239/240}Pu$, and total uranium were measured in soil, litter, leaf, bark, wood, and root samples from excavated trees and shrubs. Several trees and shrubs were found rooted in transuranic waste material. The radiochemical data were used to calibrate the UPTAKE subroutine of the BIOTRAN model for the Los Alamos environs. The simulation results indicated that higher resolution sampling is needed for ^{137}Cs and $^{239/240}Pu$ to interpret surveillance data and to produce reliable risk assessments.

This study attempted to draw together and interpret the radionuclide data for Area B from several investigations. There is a need to standardize the grid methods and sampling site markers to develop a permanent system where sampling sites can be accurately relocated and tied to engineering drawings. This would allow subsequent studies to use the existing temporal data and hence produce reliable assessments, especially for sites where heterogeneous source terms require high resolution sampling on small grids over long time frames.

I. INTRODUCTION

In 1982, a shallow land burial site known as Area B was renovated in response to a Department of Energy milestone to complete the surface stabilization of all old transuranic (TRU) burial sites by 1987. Before the surface stabilization, a radioecological study was conducted as a collaborative effort between the Environmental Surveillance Group (HSE-8) and the Environmental Science Group (HSE-12). The study examined the rooting patterns of long-lived plant species on the site, aspects of biointrusion of plants into transuranic waste, TRU uptake by plants, and transport of radionuclides from burial trenches.

A. Location and Site Description

Area B is located within the confines of Los Alamos National Laboratory (Fig. 1). The Laboratory is located in Los Alamos County, in north central New Mexico approximately 40 km from Santa Fe, New Mexico.

The Laboratory is situated on the Pajarito Plateau, a dissected, table-like extension on the eastern flank of the Jemez Mountains. The tableland is dissected by a series of eastwardly trending mesas separated by deep, rugged canyons.

The site is found on a narrow east sloping mesa in the northeast portion of the Laboratory land at

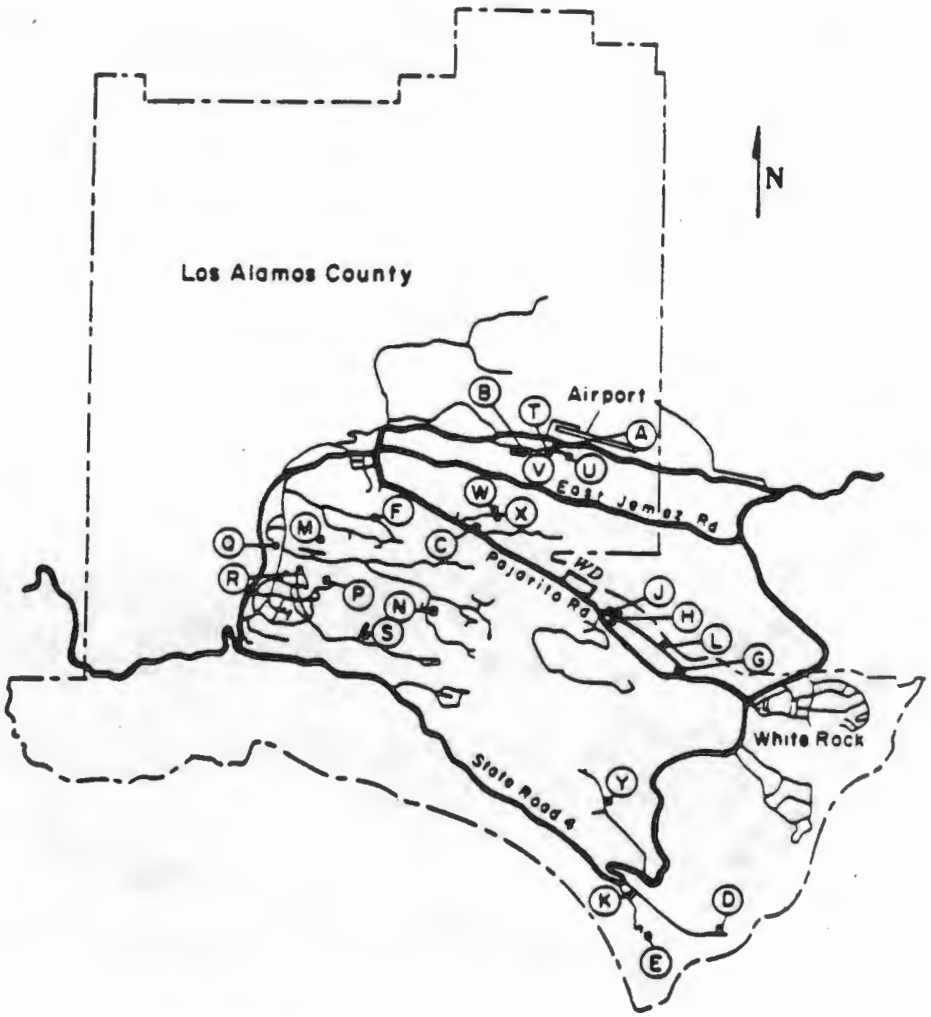

Fig. 1. Map of Los Alamos materials disposal areas.

2190 m elevation, east of the community of Los Alamos, and approximately 120 m west of the entrance to Technical Area 21 (TA-21) (DP Site) (Township and range--SE 1/2 sec 15, T. 19N, R. 6E, and SW 1/2 sec 14, T. 19N, R.6E). The southern fence line is approximately 30 m from the rim of a canyon tributary to Los Alamos Canyon; the northern fence line is bounded by DP Road, an access road to TA-21.

Area B is approximately 15 ha in size. The western two-thirds of the site is paved with asphalt and used by Los Alamos County for storage of privately owned boats, trailers, and cars. The eastern one-third, about 4.4 ha, was not covered with asphalt but through time naturally stabilized with vegetation including trees and shrubs invading from the surrounding mesa top (Fig. 2).

Soils in the area are derived from the Tshirege Member of the Bandelier Tuff, a welded ash flow from Pleistocene volcanism. The burial pits were cut into the Unit 3a of the Tshirege Member. Thickness of the tuff beneath the disposal pits is estimated to exceed 240 m, with the water table at a depth of 360 m below the surface of the mesa.

Topography of the site varies, but there is a pronounced downward slope to the south and east toward the edge of the mesa (Fig. 3). The highest point is 2189 m in the northwest corner of the site with the lowest point at 2184 m.

The mesa top location is within a ponderosa pine (*Pinus ponderosa*) plant community (Tierney and Foxx 1982). Generally the site could be divided into two portions: 1) an area that had little disturbance through time, and 2) an area where subsidence and gopher activity had caused exposure of waste material. The least amount of disturbance was found on the eastern half of the site and was characterized by larger ponderosa pines *(Pinus ponderosa)* (the oldest 27 years old) and Gambel oaks *(Quercus gambelii)*. The understory was primarily native species such as blue grama *(Bouteloua gracilis)* and

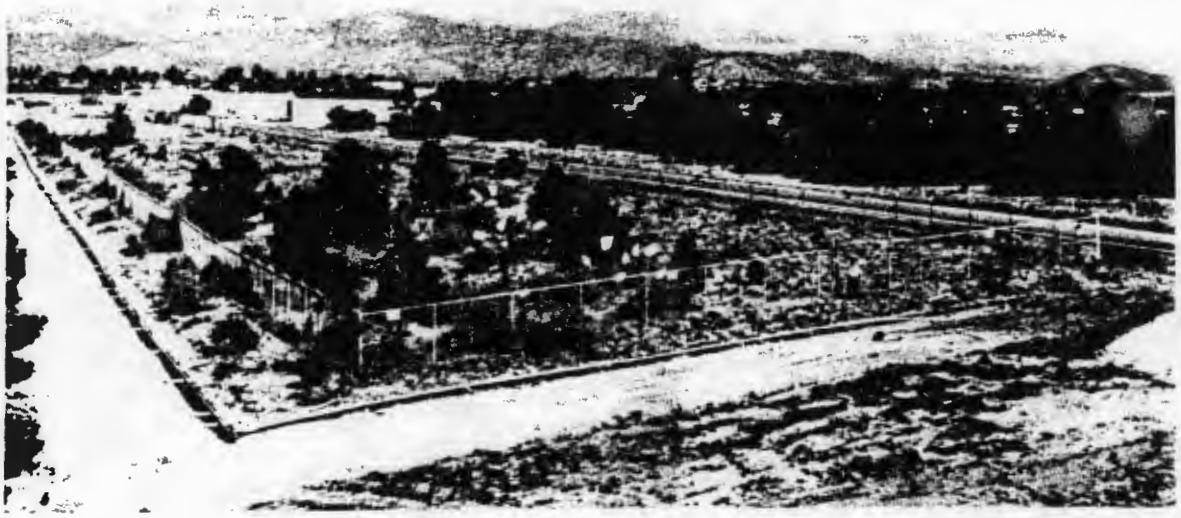

Fig. 2. 1982 Area B photograph looking west from the SE corner before devegetation and renovation.

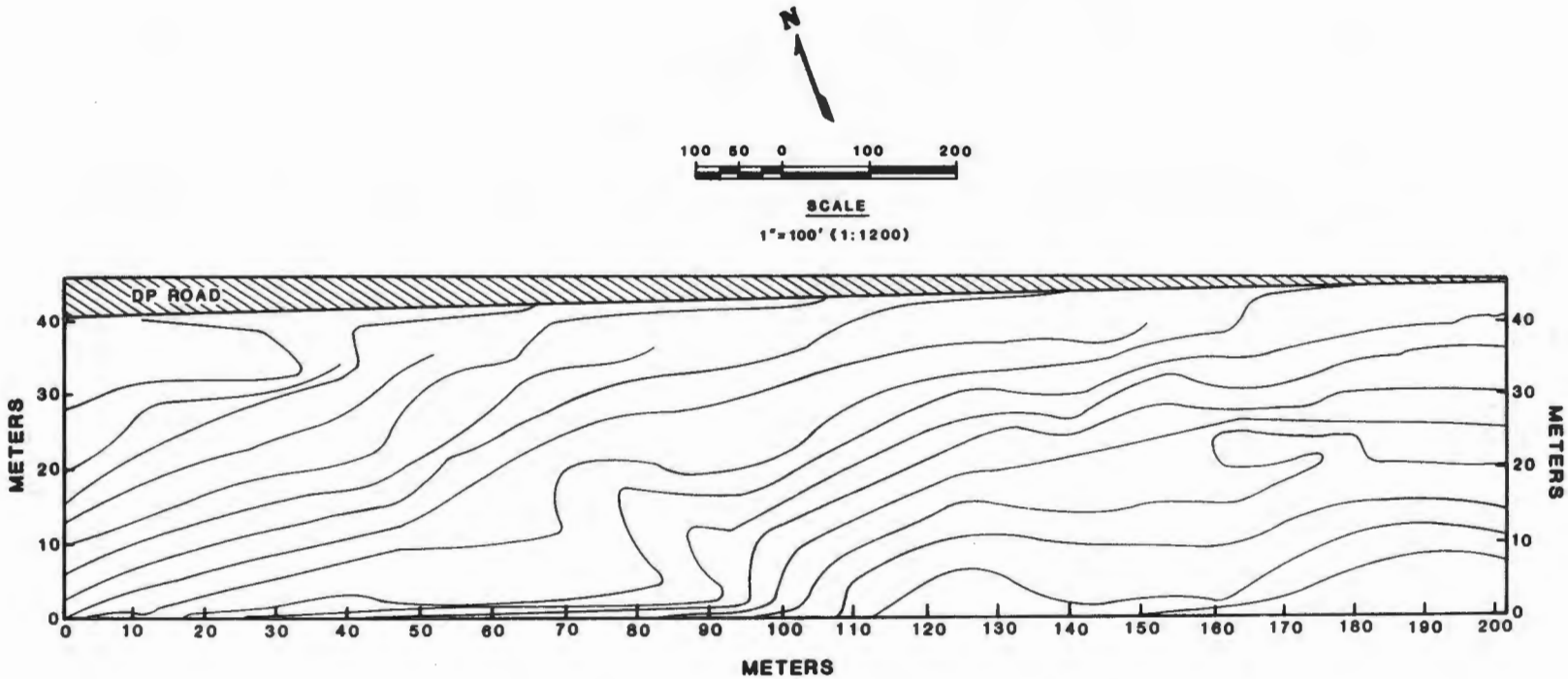

Fig. 3. Area B contour map.

mountain muhly *(Muhlenbergia montana)*. This portion was further stabilized by perennial and annual forbs including wormwood *(Artemisia ludoviciana)*, yucca *(Yucca angustissima)*, pinque *(Hymenoxys richardsonii)*, and fleabane daisy *(Erigeron divergens)*. The western half of the site had numerous areas of subsidence and was characterized by plants most commonly found in disturbed environments such as chamisa (*Chrysothamnus nauseosus*), false terragon *(Artemisia dracunculus)*, sweet clover *(Melilotus albus)*, and cheatgrass *(Bromus tectorum)*.

B. Operational History

Area B was apparently the first shallow land burial site for low-level TRU wastes within the Laboratory confines. Deposition of material into shallow pits began in 1944 and ended in 1948.

Early Laboratory engineering drawings depicted the area as one large pit but it may have contained up to six trenches (Rogers 1977). Depth of the pits was 1-4 m (Rogers 1977). Because of the time and the conditions under which Area B was established, general documentation and records of disposal are scarce. Types of radioactive material disposed in trenches were mainly transuranic waste and fission products from a water boiler reactor (Rogers 1977). Documents indicate about 90% of the waste consisted of contaminated rags, paper, rubber gloves, glassware, and small metal apparatus. Buried waste included large metal equipment, storage batteries, and contaminated toxic wastes. Records indicate material was placed in cardboard boxes. Waste filled the depth and width of the pits without being compacted and alternated with soil layers (as is commonly done now).

After the initial decommissioning in 1948 this waste site remained inactive. Through time, pocket gopher activity and subsidence had exposed wastes including empty acid bottles, wooden crates, metal containers, and other trash-type wastes. Attempts were made to stabilize the subsidence with the use of uncontaminated concrete footings and soil from disassembled buildings. Studies to investigate the distribution of surface and subsurface contamination began in 1966. That study and others ultimately resulted in complete renovation of the site in 1982 by devegetating the area and applying a new surface covering after the area was sampled for this study.

C. Previous Environmental Studies

The first documented study conducted at Area B was a 1966 USGS investigation of the distribution of subsurface moisture and migration of contaminants in the soil and tuff around the perimeter of the site (Purtymun and Kennedy 1966). Thirteen test holes, ranging in depth from 7.6 - 15.2 m were drilled through the soil into the tuff. This study found that the moisture content of the tuff layers was all below saturation. There was no evidence of lateral migration of radionuclides. The presence of a slight increase in moisture content at depths of 3.7 - 7.3 m in five test holes along the north upgradient edge of the site indicated slight lateral moisture movement.

In 1970 a study was made of plutonium and strontium in soils at TA-21 near Area B (Kennedy and Purtymun 1971). The intent was to determine quantities of plutonium deposited from TA-21 stack emissions summarized by an earlier study by Jordan and Black in 1958. Plutonium contours were constructed at 0.005, 0.01, and 0.05 $\mu Ci/m^2$. The estimated amount of ^{239}Pu within the 0.005 $\mu Ci/m^2$ and greater contours (the 0.01 contour included Area B) was 0.26 Ci or 2% of the total release from the stacks through 1969.

During the late 1970s, a series of studies at Area B was conducted as part of the environmental surveillance and radioecological programs at the Laboratory. Above background concentrations of ^{241}Am, ^{238}Pu, ^{239}Pu, ^{137}Cs, ^{233}U, total uranium, and ^{3}H were found in surface soils and vegetation of the site.*

The radiological survey of the eastern unpaved portion of Area B by Trocki in 1977 included a surface phoswich gamma survey (energy levels 12-40 keV), surface soil sampling on a 10 x 15 m grid of the whole area, and surface soil samples where high phoswich readings were obtained (Fig. 4). Trocki found an "apparent pit surface" with surface ^{233}U contamination along with high levels of surface ^{239}Pu

*Unpublished data from L. Trocki, 1977, "Results of a Surface Contamination Survey at Closed Radioactive Waste Burial Area B."

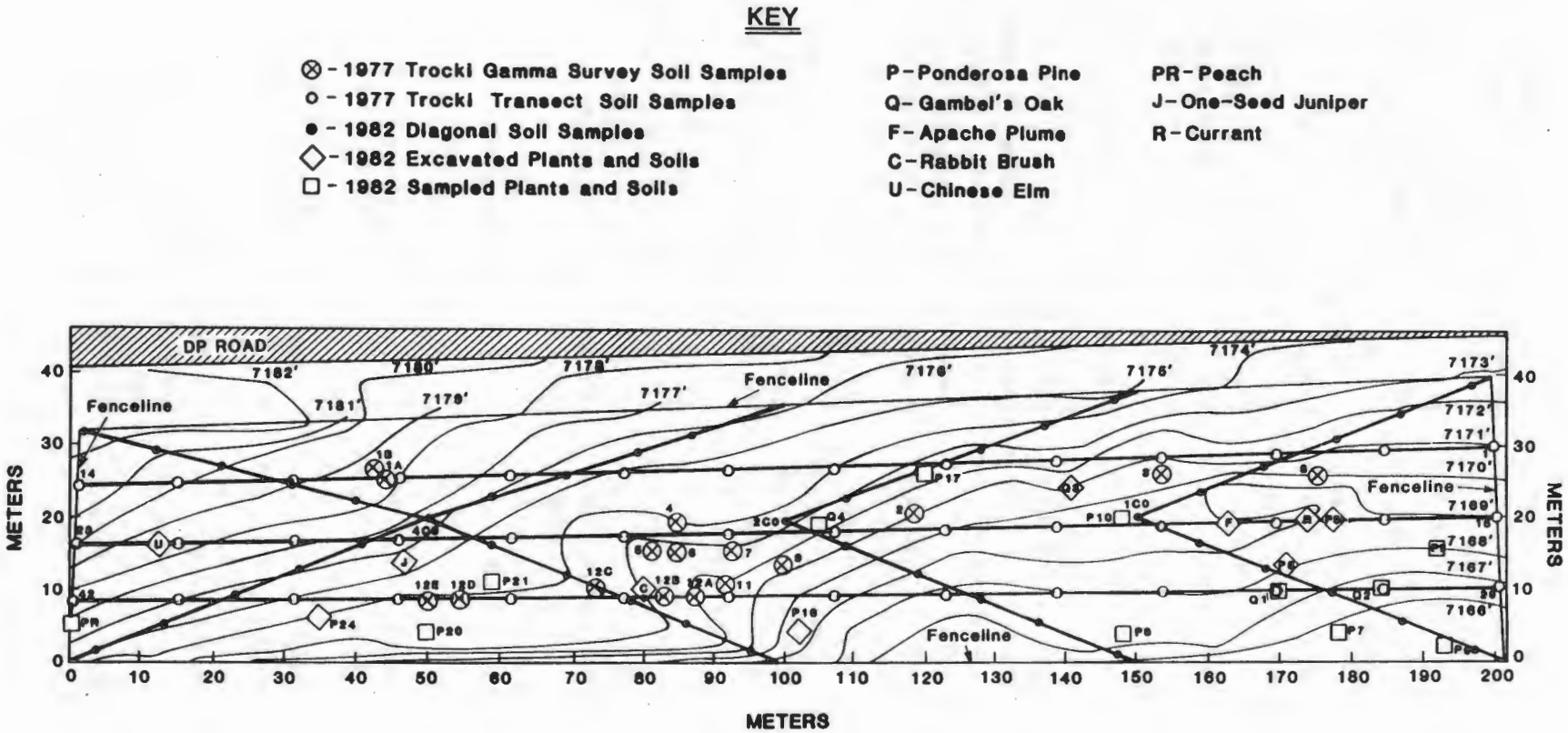

Fig. 4. Area B sampling, excavation sites, and contour maps.

(1370 pCi/g). Waste was exposed at the surface in this area and subsidence was evident. Line transect samples showed elevated tritium levels in the eastern portion of the site that tended to coincide with high gross alpha activity. The inventory of selected sites within Area B showed the soil to have 0.0064 Ci of tritium. Alpha activity mainly from ^{239}Pu was 0.0047 Ci and gross beta due to ^{90}Sr and ^{60}Co was 0.0029. This study found little contamination along the northern fence of the eastern portion where fill from demolished buildings covered the area. It was Trocki's opinion that contamination in the middle and southern portion of the area was most likely due to late 1940s burial practices. Pieces of contaminated equipment were sometimes stored on the surface until a suitable pit location was available. Lack of adequate pit cover probably accounted for the above background gamma activity detected by the phoswich over the "apparent pit surface."

During 1981 a decontamination crew removed most of the ^{233}U and ^{239}Pu in the surface soil near the apparent pit surface area. Soil was placed in drums and disposed at TA-54. After the surface contamination was removed, there remained large areas within Area B with above-background levels of radionuclides along with areas of subsidence and exposed debris.

In 1981-82, to identify the plant species and successional stages on old burial sites, Tierney and Foxx (1982) studied the vegetative composition of several Laboratory disposal sites, including Area B. Area B was found to be in an advanced state of secondary succession resulting in the invasion, through time, of large, long-lived trees as well as shrubs, perennial grasses, and forbs.

In concurrent studies, they examined the literature on rooting ecology of species commonly found on waste disposal sites at the Laboratory and throughout the Southwestern United States (Foxx, Tierney, and Williams 1984). Analysis of data from this extensive literature review indicated that all plant species commonly found on waste burial sites at the Laboratory, with the possible exception of annual grasses, could penetrate trench covers of 1 m or more that did not have a biobarrier. The standard practice at the Laboratory has been to cover the burial sites with 90 cm of top soil and then to revegetate. From this study there was a strong indication that roots of plants growing on burial trenches could be in contact with waste material.

In response to a Department of Energy milestone to complete surface stabilization of old burial sites, Area B was scheduled for renovation in the fall of 1982. Because of the presence of deep-rooted species growing on the site, Area B provided a unique opportunity to verify the likelihood of roots coming in contact with waste material. In addition, the site afforded the possibility of studying the "mining" of radionuclides by mature trees and shrubs that had invaded the waste disposal trenches.

The restabilization of Area B required the removal of all plants. This provided the right set of circumstances to collect and analyze samples from various layers within that ecosystem: soils, litter, roots, stems, and leaves. The objective of the data collection was to look for plants growing in waste, examine the rooting ecology, and sample the associated compartments for radionuclides.

This intense sampling scheme allowed for analyses of the levels of three radionuclides (^{137}Cs, ^{239}Pu, and uranium) and one transition metal (scandium) for each of the environmental layers. This information then became the basis for calibration of the site-specific biological transport model, BIOTRAN. The purpose of this report is to discuss the results of analyses and BIOTRAN simulations and to draw conclusions pertinent for risk assessment and decision making focused on TRU buried waste sites.

D. BIOTRAN Simulations

The four elements, cesium, plutonium, uranium, and scandium, were chosen for analysis in all the soil, litter, and vegetative samples taken at Area B in 1982. The purpose of the simulation study was to investigate the different distribution patterns of these elements at a higher resolution level than had been previously studied.

Early BIOTRAN computer model simulation studies suggested that higher resolution data were needed to understand the short- and long-term distribution of these elements within the internal (wood) and external (leaves, bark, litter) compartments of trees and shrubs. Multiple element analyses would

also give needed comparisons to calibrate the simulation models for elements other than plutonium and uranium at Los Alamos. Multiple element analyses could also give information on the mechanisms of intake and distribution especially for elements where biochemical and physiological mechanisms mediate their distribution in a manner similar to the elemental nutrients.

Figure 5 illustrates the current structure of the computer BIOTRAN model (Gallegos, Garcia, and Sutton 1980; Gallegos and Wenzel 1984; Wenzel and Gallegos 1985). Table I lists the BIOTRAN model components (subroutines) used to develop complex risk assessment and ecological scenarios.

Four BIOTRAN subroutines, CLIMAT, WATFLX, PLTGRO, and UPTAKE were used to develop the Area B waste site scenarios. The input for these simulations was taken from the field data. The purpose for simulating the Area B soil and plant cesium, plutonium, scandium, and uranium concentrations was to calibrate BIOTRAN and to develop a strategy for validating these subroutines by designing appropriate experiments based on the simulation results.

The CLIMAT subroutine is the major driver of a simulation by supplying precipitation, insolation, temperature, and potential evapotranspiration to the other subroutines. Biomass, water, and radionuclides are then transferred within the other subroutines as a function of time by using daily intervals that are summed within a yearly loop. Table I gives a short description of each subroutine.

BIOTRAN is driven stochastically in a Monte Carlo fashion by the CLIMAT subroutine by using random number generators to vary rainfall, snow

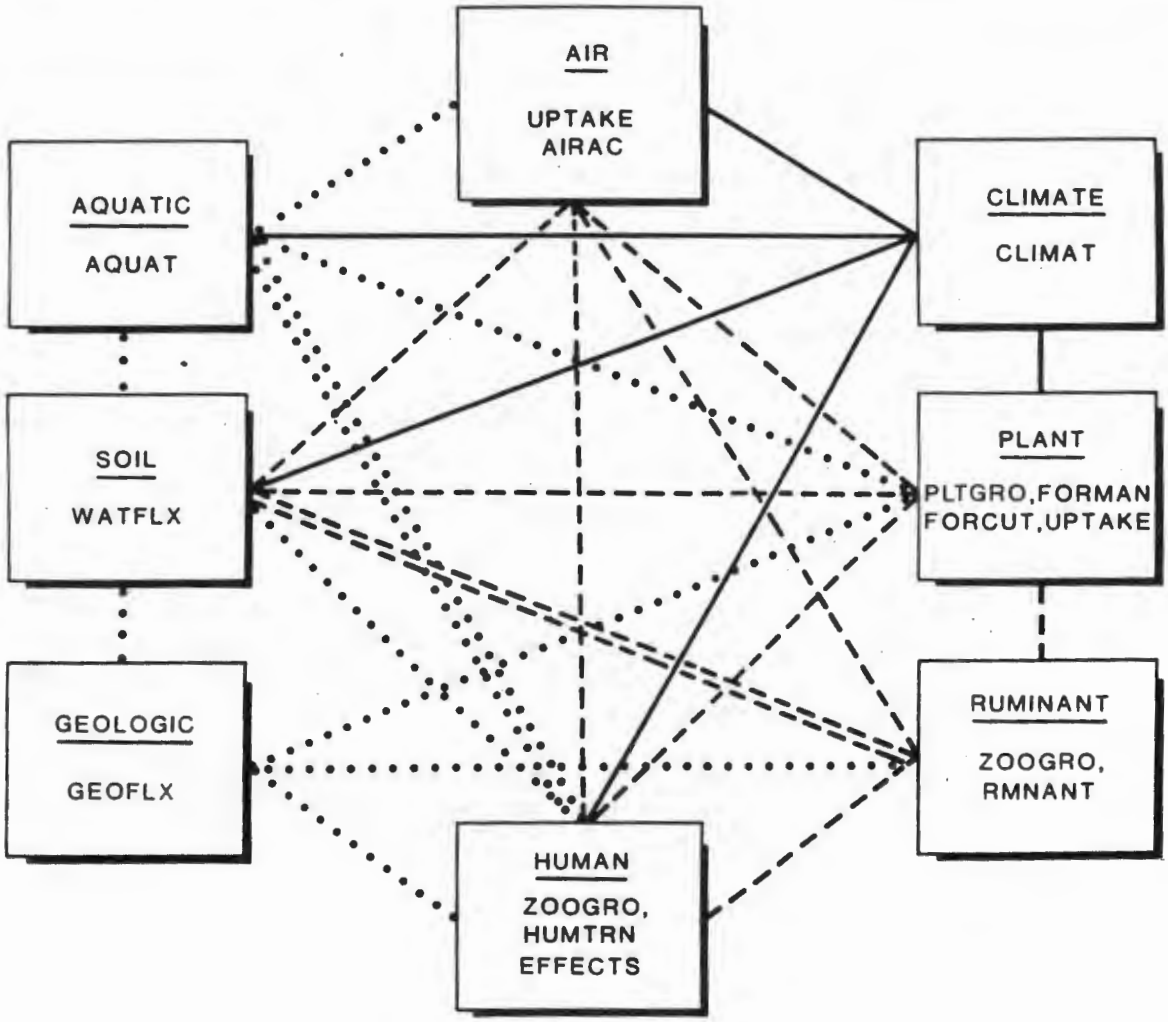

Fig. 5. BIOTRAN structure.

Table I. List of BIOTRAN Submodels

Subroutine Name	Description of Major Processes Modeled
DRIVER	(Under development.) Simulates long term climate patterns using the CLIMAT subroutine. Sets the hydrological cycle and meteorological extremes for climatological changes.
CLIMAT	Simulates daily temperature, solar radiation, precipitation, and potential evapotranspiration.
BIOPLT	Selector for graphical and formatted output.
AIRAC	Sets input for acute and chronic average daily air contaminant concentrations from accidental releases, fallout, or ambient air data.
WATFLX	Simulates soil layer moisture flux and potential evapotranspiration for a particular soil profile and plant type.
PLTGRO	Simulates above and below ground live and dead biomass dynamics for 22 different plant growth strategies (grass, forbs, shrubs, and trees).
FORMAN	Simulates detailed forest stand characteristics and biomass compartments on a daily or yearly basis.
FORCUT	Adjusts biomass for forest fire events and selective cuttings.
AQUAT	(Under development.) Simulates daily limnetic and littoral water characteristics, plankton, plants, and fish.
UPTAKE	Simulates radionuclide transport by wind, rainfall, and root uptake resulting in contaminants in and on plants.
RMNANT	Simulates basic ruminant anatomy, metabolism, and contaminant intake, uptake, and burden based on daily diet regimes of simulated forage and feed.
ZOOGRO	Harvests and stores simulated crops for mammal and human consumption. Manages ruminant grazing.
HUMTRN	Simulates age- and sex-specific human anatomy and physiology; contaminant intake, uptake, and dose based on daily diet regimes of simulated milk, meat, vegetables, and grain. Individual organ burdens and doses are calculated on an age- and sex-specific basis.
EFFECTS	Simulates age- and sex-specific human cancer risk and deaths from the contaminant exposure and body burdens calculated by HUMTRN. Uses 3-D color graphics to visualize population dynamics and risk.

Table I (cont)

Subroutine Name	Description of Major Processes Modeled
GEOFLX	(Under development.) Simulates daily and yearly water and contaminant flow through geologic formations below the soil profile.
HYDRO	(Under development.) Simulates surface and subsurface water and particulate transport for water routing and flow in watersheds.

amounts, and temperature. Precipitation and temperature then determine, for example, the soil moisture input to WATFLX, the growing season for the plants in PLTGRO, and the transport rates of radionuclides in UPTAKE. The temporal effects of the climate and interaction with the biota determine the eventual concentration of radionuclides in the soil, vegetation, and animals in the simulation.

II. FIELD AND LABORATORY METHODS

There were four phases of data collection in Area B: 1) excavation of selected plants, 2) examination of the rooting ecology, 3) sampling of associated soils, litter, and plant material for radionuclide analyses, and 4) determination of leaf surface area.

A. Excavation of Selected Trees and Shrubs

No waste management maps showing the location of individual trenches were available; therefore, trees and shrubs were sampled throughout the site. It was presumed that where there was high-surface contamination or exposed debris such as bottles, piping, or crates, a pit might exist. Therefore, plants selected for excavation were generally contiguous to debris or spots of high-surface contamination. Locations of excavated specimens are shown in Fig. 4.

In the vicinity of each tree or shrub to be sampled, surface soils and litter were collected and placed in plastic bags. Large tarps were placed on the ground surrounding the plant and the specimen was then felled. The tarps protected the aboveground portions from contamination from soil and dust. After felling the specimen, at least 1 kg or more of needles or leaves were collected from the crown at different heights (low, middle, high) and placed in plastic bags for storage. A 0.3 m cross section of the bole was removed and placed in burlap bags for storage.

Once the specimen was cut down, the roots of the plant were uncovered with picks and shovels. During the unearthing process there was continuous monitoring for contamination with a phoswich field portable gamma detector and a hand held air proportional alpha detector (detection limit of about 100 alpha count/60 cm^2). Once the roots were exposed, portions were removed and bagged for radiochemical analysis. Soils were collected along the profile of the excavation and the hole then backfilled with the soil that had been removed.

B. Description and Location of Excavated Trees and Shrubs

Nine native and two domestic plants were excavated. A number of other trees were sampled for surface soils, bole, and leaves but roots were not unearthed. The following is a description of each species sampled and its location within the plot.

1. Ponderosa pine (*Pinus ponderosa*): Ponderosa pine was the most common tree species on Area B and was scattered throughout the site with the largest numbers to the south and west sections. Samples of needles and boles of 24 trees were collected. Four specimens (5, 8, 19, 24) were intensely sampled and roots excavated (Fig. 4).

Pine trees on the site were generally less than 2 m tall. The larger pines were near the southeast corner and the oldest had previously been dated by Tierney and Foxx (1982) to be 27 years old.

Ponderosa pines on the site were medium to tall trees with a narrow, conical or almost flat-topped crown. The crown of one specimen extended to ground level (ponderosa pine 5). Within the plot the trees ranged from 1.2 to 8.8 m tall. The trunks were straight ranging from about 0.6 cm to 30 cm in diameter. Bark was medium to thick (0.4 to 1 cm), dark brown to black in younger trees, and cinnamon red in older trees. Mature trees had irregularly and deeply furrowed bark that was broken into large plates. Needles of this pine species are in bundles of 3, yellowish-green to dark green. In some cases the needles appeared atypically chlorotic.

2. One-Seeded Juniper (*Juniperus monosperma*): Seven one-seeded junipers were found growing on the site. This slow growing tree was generally no more than 3 m tall. The crown was broad, round, and the trunk much branched. Bark was thin, 0-4 mm thick, ash gray to gray, with numerous narrow fissures forming ridges and shredding into scales. Leaves were tiny, scale-like, overlapping, pressing closely to the branchlets. Litter, soils, and bole samples were collected from one specimen. Portions of the root were also sampled.

3. Apache Plume (*Fallugia paradoxa*): One thicket of Apache plume was found growing on the site (Fig. 4). This fast-growing, clump-forming shrub was approximately 1.5 m tall. The shrub has small, densely hairy leaves, white blossoms, and feather-duster shaped fruits. One specimen within the clump was sampled.

4. Gambel Oak (*Quercus gambelii*): Gambel oak was the most common shrub species on the site. Individual plants grew to a height of 4 m. One shrub was excavated but leaves on four oaks were collected for analysis. This thicket-forming shrub generally had a rounded crown and a much-branched base. Bark was thin, light gray to white, becoming rough and scaly with age. Leaves were found to be 8-16 cm long, deeply lobed.

5. Rubber Rabbit Brush, Chamisa (*Chrysothamnus nauseosus*): A few scattered specimens of chamisa were found growing in the western section of the site. This fast-growing plant was generally 1-2 m tall. One specimen was sampled and roots excavated. The bark of chamisa is shreddy and adheres tightly to the branches. Twiglets are covered with matted hairs.

6. Wax Currant (*Ribes cereum*): One wax currant was found growing on the site. It was within 1 ft of ponderosa pine 5 and in the vicinity of contaminated piping. The shrub was approximately 3 ft tall. Leaves and young twigs were covered with a waxy pubescence.

7. Peach (*Prunus persica*): A domestic peach was found growing along the western fence line separating the vegetated and paved sections of Area B. It was probably a remnant from the habitation of the mesa during the early 1940s (Tierney and Foxx 1982). The tree stood approximately 3 m tall. Bark was thick, dark, and reddish brown.

8. Siberian Elm (*Ulmus pumila*): A large Siberian elm (sometimes called Chinese elm) was found growing in the western portion of the site approximately 12 m from the fence line separating the paved and vegetated portions of Area B. This fast growing tree had a deeply furrowed grayish bark.

C. Description of Sampling and Laboratory Preparation Methods

1. Soil Samples. Tierney and Foxx (1982) had previously mapped the site and found the vegetation to be very heterogeneous. The eastern portion of the site was more stabilized than the western. For purposes of sampling, the site was divided into four zones of about 40 x 50 m. Stratified soil samples were collected every 10 m along diagonal transects that traversed each of the four zones (Fig. 4).

Soils were also collected in association with excavated roots. In these instances, soils were collected on the surface beneath the tree, along the trench face, and adjacent to the roots.

Stratified soil samples were collected from 0-2, 2-12, and 12-32 cm layers using stainless steel rings and cylinders of varying sizes and depths. The 0-2 cm soil sample was collected by placing a 30-cm diameter stainless steel ring on the surface of the soil at the sampling point. The soil enclosed by the 30-cm diameter ring was then collected by undercutting the ring with a metal spatula and the sample transferred to a plastic bag. For the second, 2-12 cm soil sample, a 13-cm diameter stainless steel cylinder was used to collect the sample. The core was placed directly on the surface cleared by the 2-cm sample and driven into the ground. The 12-32 cm sample was collected using a 5-cm diameter PVC pipe. All soil samples were frozen until preparation was possible.

In the Laboratory, soils were prepared by air drying and by removing rocks and debris. Soils were then dried at 105°C, ball milled overnight into a homogeneous powder and submitted to radiochemistry.

2. Organic Material. Litter, roots, bole, needles, and leaves were dried, ashed, and submitted for radiochemical analyses.

a. Litter. Litter was collected in a 0.5 m^2 quadrat under the canopy of selected trees and shrubs and frozen until preparation. Litter was separated into needles, deciduous leaves, grass, forbs, and humus-soil fractions. The litter fractions were then dried at 80°C and ashed at 500°C. Leaves, bark, and wood were prepared for radiochemistry using the same methods. The ash was submitted to radiochemistry.

b. Roots. Selected roots were bagged during excavation and frozen until preparation. In the laboratory, the root bark was carefully removed from the root and analyzed separately from the xylem. Using a saw, exposed ends of each root were cut off to decrease the soil contamination on the root wood. In the case of rabbit brush, the bark would not separate from the wood and bark was scraped from the inner wood.

c. Bole. For large pines the bole sample was removed from the stem at approximately 90 cm from the ground. With shrubs and small trees, bole samples were taken within the first 90 cm. Each bole sample was labelled and stored in burlap bags until prepared for analysis. In the laboratory, the bark was separated from the xylem using a knife or wood chisel. The inner and outer bark and the heartwood and sapwood were separated for large boles. The exposed wood ends were carefully cut off using a band saw to lessen cross contamination between bark and wood.

d. Leaves and Needles. Needles were collected from conifers by stripping branches by hand and placing the needles in plastic bags. Needles from larger trees were collected from the lower, middle, and the top of the canopy. From smaller trees a grab sample was taken representative of the whole canopy. In the laboratory, a grab sample of needles was measured to determine surface area and the number of needles in a 1 kg sample count.

Dreicer (1981) found that surface contamination of plant parts by resuspension, deposition, and rain-splash processes make it difficult to determine plant root uptake. If the leaves emerging in the spring could be protected from wind and rain-splash contamination, a truer picture of root uptake might be possible. Therefore, branches of three deciduous species on the site, Gambel's oak, elm, and peach, were placed in ventilated clear plastic bags before leaves began to emerge in the spring (Fig. 6). The remnant leaves were first removed from the shrub or tree in early April. The stems were washed and sprayed with an antistatic spray and then the individual branches placed in a clear plastic bag. Two 30-cm long polypropylene tubes, 0.6 cm in diameter, were inserted at the base and the bag was sealed with yellow marking tape. To prevent sun scorch, a white plastic bag was placed over the transparent bag. The bags remained on the branches until the leaves were fully emerged (for the oak, this was mid-June and for the elm and peach, late May). The branches with plastic bags and emergent leaves were then removed. In the laboratory the branches were removed from the bags, leaves stripped from the branches, and prepared for radiochemistry.

Fig. 6. Ventilated plastic bags placed in Gambel's oaks before leaf emergence.

3. **Leaf Surface Area Measurement.** Leaf surface area measurements were taken for ponderosa pine, oak, elm, and peach. A grab sample of 10 leaves from each sample was taken. Coniferous leaves (needles) were measured in 3 dimensions: length, width, and thickness. Deciduous leaves were measured by use of a planimeter.

D. Radiochemical Methods

Radiochemical analysis of $^{239,240}Pu$, total uranium, ^{137}Cs, and the stable scandium were done by the Radiation Chemistry Group, HSE-9. Cesium-137 (detection limit of 1 pCi/g) was determined by gamma ray spectrometry on ashed vegetative samples and 100 g dry soil samples using a NaI (Tℓ) crystal detector. Total uranium (detection limit of 0.03 μg/g) was measured on 2 g samples using delayed epithermal neutron activation analysis. Plutonium-239 and 240 (detection limit of 0.002 pCi/g) were measured by radiochemical separation of 10 g samples followed by alpha spectrometry. Stable scandium (detection limit about 0.03 μg/g) was measured on 2 g samples, after column separation from the matrix, by thermal neutron activation analysis (Gautier, Gladney, and Perrin 1985).

III. RESULTS

A. Presence of Trees and Shrubs in Waste Material

Ponderosa pine 5 was found during excavation with roots in contact with recognizable waste material. Several other trees and shrubs were rooted in contaminated soil, but field radiation instruments did not recognize the contaminants during the excavation process. The extent of their contamination was apparent only after radiochemical analyses were available. Ponderosa pine 5 was 17 years old, 4.8 m tall, and located in the southeast section of Area B (Fig. 4). Ponderosa pine 5 was found rooting in what may be a trench described by Rogers (1977) as being 90 cm wide and 2 m long. Soils in the area were deep and alpha contaminated debris was exposed at the surface under the tree canopy. The tree was selected for excavation because of the presence of exposed 1.3-cm diameter metal pipe (electrical conduit) having measurable radioactivity (about 2000 alpha counts/minute/60 cm^2). The pipe continued downward beneath the roots. Beneath the roots some copper and electrical wires were uncovered, but had no detectable alpha radioactivity. At about 40 cm deep a mass of rubber gloves was excavated. Radioactivity on the surface of the gloves varied from 0 to 6000 alpha counts/minute. Other gloves in the area had no measurable alpha radioactivity. At 45 cm a large lateral root had come into contact with a rubber glove. The rubber glove and its contents (the glove contained a 6-cm ball of radioactive waste) provided resistance to root growth (Fig. 7). Where the fingers of the glove had not provided resistance, the root had grown between the fingers until the resistance of the rubber had retarded growth. This gave the root the appearance of a hand. Soil and the glove measured 10,000 alpha counts/minute. Excavation was discontinued because of the high radiation levels. In the same layer there were brown Duroglass bottles still filled with liquid, rubber tubing, plaster, and metal tubing that had been painted (Fig. 8). Roots and soils were collected and the hole backfilled. No cardboard or wood materials were found in the excavation site. This was probably due to decay of cardboard and wood and consumption by soil arthropods. There was also indication that some waste material was dumped in the trench without previous packaging. Soils, litter, bole, and needles at three layers (low, middle, and high) in the canopy were sampled on ponderosa pine 5. The canopy extended all the way to the soil surface and had a full and tapered crown. Needles were found to be slightly chlorotic with whitish tips. The bole xylem was knotty and had an irregular appearance. It was also resinous and full of boring insects. Subsurface plutonium was also found later from the radiochemical results in ponderosa 8 and 10, elm, and oak 2 soils.

B. Rooting Ecology

Excavations on Area B were shallow, usually less than 90 cm deep, so it was not possible to make a direct comparison of rooting ecology of excavated species with previous studies done in undisturbed

Fig. 7. Ponderosa pine #5 lateral root growing around a rubber glove.

Fig. 8. Liquid filled Duroglass bottles, painted metal and rubber tubing and plaster found beneath ponderosa pine #5.

soils (Tierney and Foxx 1987). General patterns consistent with previous studies and literature reports were noted.

Three different rooting patterns were noted. The first rooting pattern was observed from specimens that had shallow, spreading, large lateral roots. This rooting type was represented by ponderosa pine and wild currant. Ponderosa pine was found to have a network of large lateral roots that remained in the upper 30 cm of the soil profile and extended some distance from the tree. Although ponderosa pine has been reported to root to 24 m deep, literature and field values indicate an average rooting depth of no more than 2-4 m. Wax currant also had roots in the upper soil surfaces. Roots appeared to extend to depths greater than 30 cm. A few large laterals extended below the soil surface. Roots of this species have been reported to extend to depths of 2.9 m but generally only 1.5 m (Tierney and Foxx 1987). The second type of rooting ecology was found in the Siberian elm and one-seed juniper. The species had extensive fine root development in the upper 30 cm with a few larger laterals extending laterally and downward. Literature values indicate these species may root deeply (Tierney and Foxx 1987, Foxx and Tierney 1984). The third rooting pattern was represented by rubber rabbit brush and Apache plume. These species were found to have one large

taproot that penetrated deeply. Literature values indicate that the taproot may penetrate to depths of 4 m or more (Tierney and Foxx 1987; Foxx and Tierney 1984).

C. Surface Area Measurements

Leaf surface area measurements were done for 9 ponderosa pines, 3 oaks, and 1 elm by measuring a 10-leaf grab sample from each of the leaf samples. The average leaf surface area for these grab samples and the species means are given in Tables II and III. The cm^2/g "dry" ("dry" weight is dry weight in grams minus the ash weight) is given for these samples in the AREABDAT database in Appendix A.

Woodwell (1974) found less variability in leaf concentration values for nutrients when expressed as mg/cm^2 rather than as mg/g of dry leaves. Dreicer's (1981) experiments have shown that surface distribution of particles is a major factor for plant radionuclide concentration. The BIOTRAN UPTAKE model carefully partitions exterior particulate contamination on stems, roots, and leaves from the interior or root uptake concentrations found on the inside of these tissues. Further study is needed to determine this interior/exterior phytodiscrimination. Note from Tables II and III that the leaf surface area varies from plant to plant within species and also between species as well as with height for the same plant.

For the ponderosa pine needles the coefficient of variation (COV) increased as the element concentrations were expressed as ash, dry, and cm^2 indicating that more variability was introduced by expressing the concentration as the dry weight and even more when expressed as a function of area. This was not true for oaks. Some of the COVs for the cm^2 (cesium and plutonium for bagged leaves, and cesium for unbagged leaves) had lower COV for the areas than the "dry" concentrations. This indicated that cesium kinetics appeared different from the other elements and that careful quantification on an area basis would be feasible and perhaps advantageous to delineate differences between surface and internal leaf contamination. For scandium and plutonium bagging the leaves decreased the leaf concentration by a factor of about 5 times (uranium about 2 times); but cesium tended to increase in concentration when leaves were bagged suggesting that nutrients were higher in bagged leaves and that elements that behaved like nutrients (cesium and maybe uranium) could be influenced by the bagged branch microclimate.

D. Radiochemical Results and Database

The data for the excavated and nonexcavated trees and shrubs from Area B were digitized as one file (AREABDAT) shown in Appendix A. Elevation contours and the Trocki (1977) data are also in this database. A description of the data format is given in Appendix A.

The radiochemical data in Appendix A were plotted in Figs. 9-16 as a function of plant type, sample type, and radionuclide to begin the analysis. For plotting purposes, several of the sample types were combined into a new database, called PLOTDAT, given in Appendix B. There are fifteen combined sample types (the AREABDAT sample type numbers are given in parenthesis):

1. unbagged leaves (1), high (7), medium (8), and low (9)
2. bagged leaves (2), high (12), medium (13), and low (14)
3. bole bark (3)
4. bole wood (4), outer wood (15), and inner wood (16)
5. root bark (5)
6. root wood (6)
7. pine litter (18)
8. misc. litter (20)
9. 2 cm soil (21)
10. 10 cm soil (22)
11. 25 cm soil (23), 30 cm soil (26)
12. 45-55 cm soil (29)
13. 80 cm soil (24)
14. 150 cm soil (25), 160 cm soil (27)
15. soil around object or rock (28), no depth soil (30)

Figures 9-16 used DISSPLA color graphics. The figures show the log of concentration as ppb "dry" for scandium and uranium and fCi/g "dry" for plutonium and cesium.

Table II. Mean Leaf Surface Area, Measurements for Ponderosa Pine for a 10 Leaf Grab Sample from Each Leaf Sample

Plant No.	Crown Location	Wet Leaf wt g	Length cm	Width mm	Thickness mm	Area per needle (cm^2)
5	upper 1/3	0.12	14.3	1.57	0.87	0.195
	mid 1/3	0.12	15.0	1.50	0.79	0.178
	lower 1/3	0.11	14.1	1.44	0.77	0.156
8	upper 1/3	0.21	15.8	1.59	0.86	0.216
	mid 1/3	0.09	15.1	1.38	0.77	0.160
	lower 11/3	0.10	13.7	1.42	0.56	0.109
6	entire	0.10	16.9	1.44	0.75	0.183
1	entire	0.10	13.8	1.45	0.76	0.152
17	entire	0.06	14.8	1.44	0.81	0.173
20	entire	0.13	14.2	1.68	0.87	0.208
21	entire	0.09	14.0	1.42	0.76	0.151
24	entire	0.14	15.5	1.67	0.92	0.238
9	entire	0.10	15.2	1.46	0.77	0.171

For plutonium, Figs. 9 and 10 show a maximum concentration range of about eight orders of magnitude from 10^{-2} to 10^{6} units between wood and soil. Both root and bole bark show greater concentrations than their corresponding wood. There is also a distinct decrease in soil concentration with depth, except where buried waste was encountered. The steep soil plutonium concentration gradients shown for oaks are from surface contamination and are similar to the gradient seen for ^{137}Cs in Figs. 11 and 12. Pine needles separated from the litter contained less plutonium than the remaining miscellaneous litter due to soil contamination in the miscellaneous litter fraction. Unbagged leaves had higher concentrations than bagged leaves as expected.

For cesium (Figs. 11 and 12) the concentration ranges were the lowest of the four elements and ranged from 10^{-1} to 10^{4} units. As with the other elements, there was more cesium on the bark than in the wood, but the difference was smaller. There was a steep gradient in the soil concentrations indicating most of the contamination was still on the soil surface. The slope of the gradient is similar to that found for plutonium in oaks. It appears that plutonium and cesium move down the soil profile in a similar fashion as has been described before (Dahlman, Garten, and Hakonson 1980). The major difference observed for cesium is for the bagged and unbagged leaves. Bagged leaves are greater or at least the same concentration as the unbagged leaves. This observation is clearly different than the other three elements, which do not have strong nutrient analogs.

For uranium (Figs. 13 and 14) the concentration range was from 10^{2} to 10^{5} ppb between soil and wood. Root and bole bark were both greater than wood. There was no apparent decrease in uranium soil concentration with depth and only two samples appeared to have deep soil concentrations greater than the surface concentration. As with plutonium, the uranium concentrations in miscellaneous litter and unbagged leaves were higher than pine litter and bagged leaves.

Table III. Mean Leaf Surface Area for Oak and Elm

Species	Plant No.	Crown Location	Mean Wet Leaf Weight (g)	Mean Leaf Area CM^2
Oak	2	entire	0.07	8.38
			0.03	3.88
			0.13	10.03
			0.07	3.92
			0.14	10.41
			0.05	3.28
			0.10	8.47
			0.04	2.91
	3	entire	0.11	10.99
			0.05	3.66
			0.09	10.91
			0.05	4.33
			0.15	13.33
			0.06	5.08
	4	entire	0.11	12.98
			0.05	3.78
			0.08	7.81
			0.04	3.78
			0.23	14.64
			0.08	4.62
Elm	1	entire	0.21	15.16
			0.09	4.92
			0.06	3.25
			0.02	0.77

Scandium concentrations in Figs. 15 and 16 ranged between 10^{-2} and 10^{4} for soil and wood. Bole and root bark were generally greater than their corresponding wood. As for uranium there was no apparent decrease in scandium soil concentration with depth. Litter and leaves showed the same distribution as plutonium and uranium.

Table IV gives the arithmetic means, one standard deviation, and coefficient of variation (COV) for the radionuclide data. Four groups of plants were selected for analysis from the AREABDAT database. These four were ponderosa pine 5, all remaining ponderosa pines (9 trees), peach, and elm, and shrubs--oak, rabbit brush, currant, and Apache plume.

In Table IV and Figs. 9-16, different COVs for soil and vegetation compartments between elements are apparent. The average COV for the scandium and uranium soil results is 0.26 ± 0.20 (n = 20). The average COVs are three times higher for the cesium and plutonium soils (0.92 ± 0.64, n = 20). This indicates that the cesium and plutonium in these soils have a higher variance than the scandium and uranium and are more heterogeneously distributed. The vegetative samples analyzed above and below these soils reflected this heterogeneity also. The average COV for the scandium and uranium vegetative samples was 0.73 ± 0.48 (n = 44) and for cesium and plutonium was 1.07 ± 0.95 (n = 40) indicating that the distribution in the plants followed the distribution

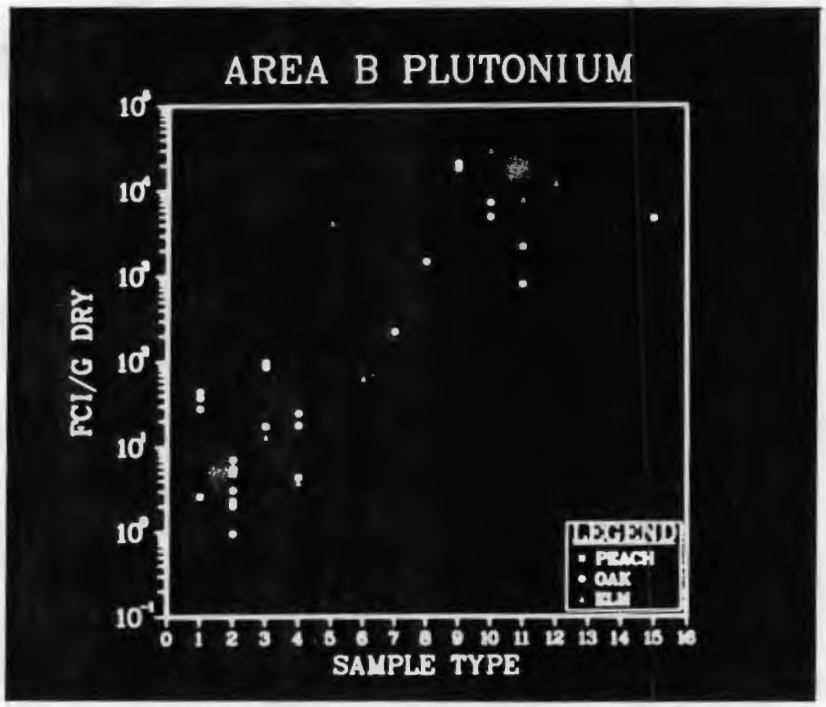

Fig. 9. Plutonium concentrations in soil, litter, and plants. The sample types are listed in the text.

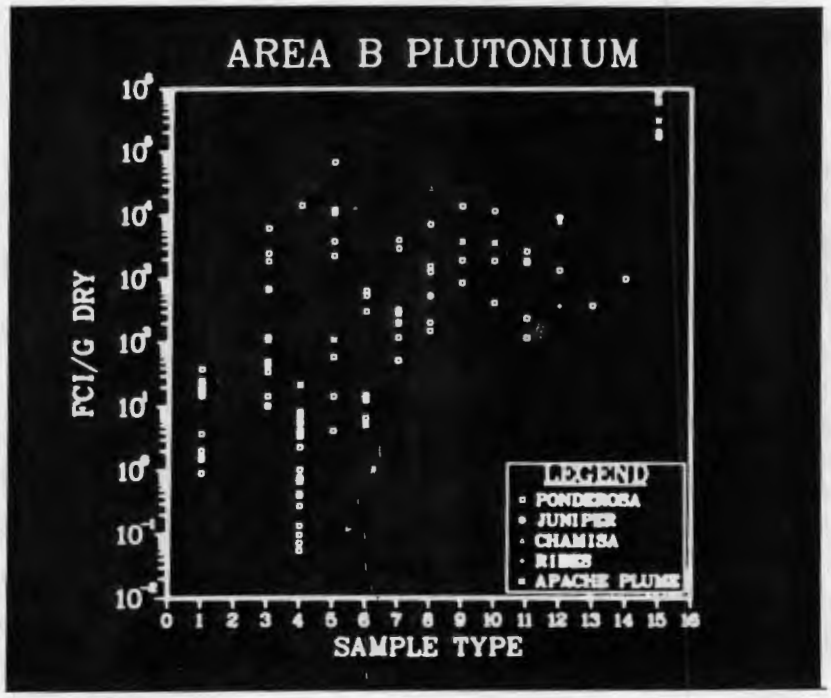

Fig. 10. Plutonium concentrations in soil, litter, and plants.

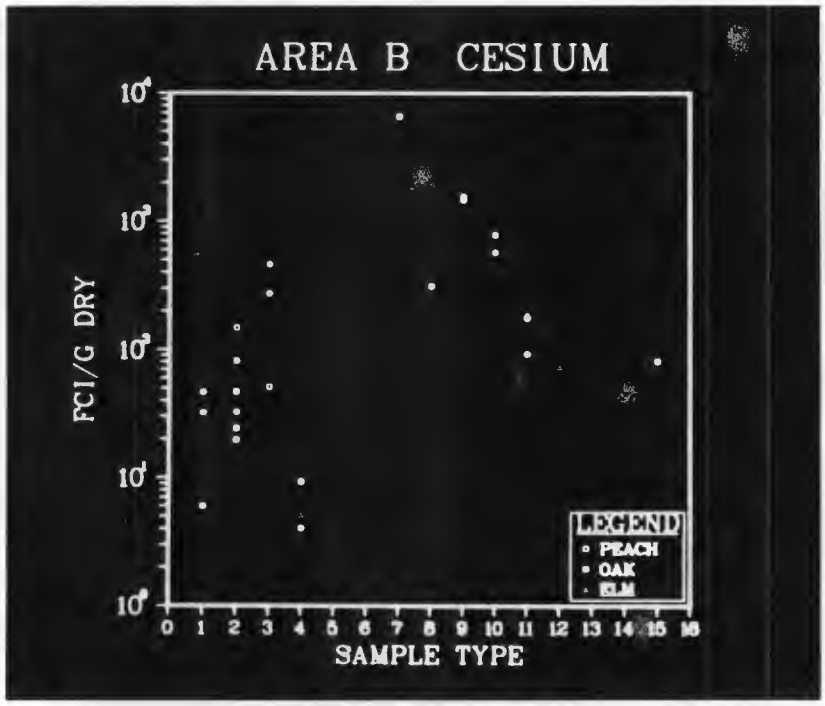

Fig. 11. Cesium concentrations in soil, litter, and plants.

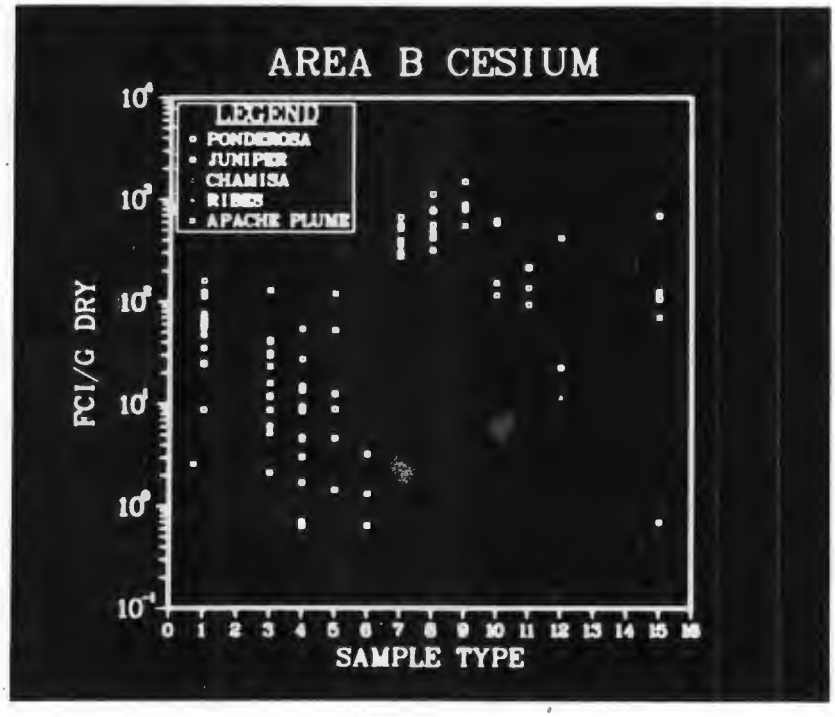

Fig. 12. Cesium concentrations in soil, litter, and plants.

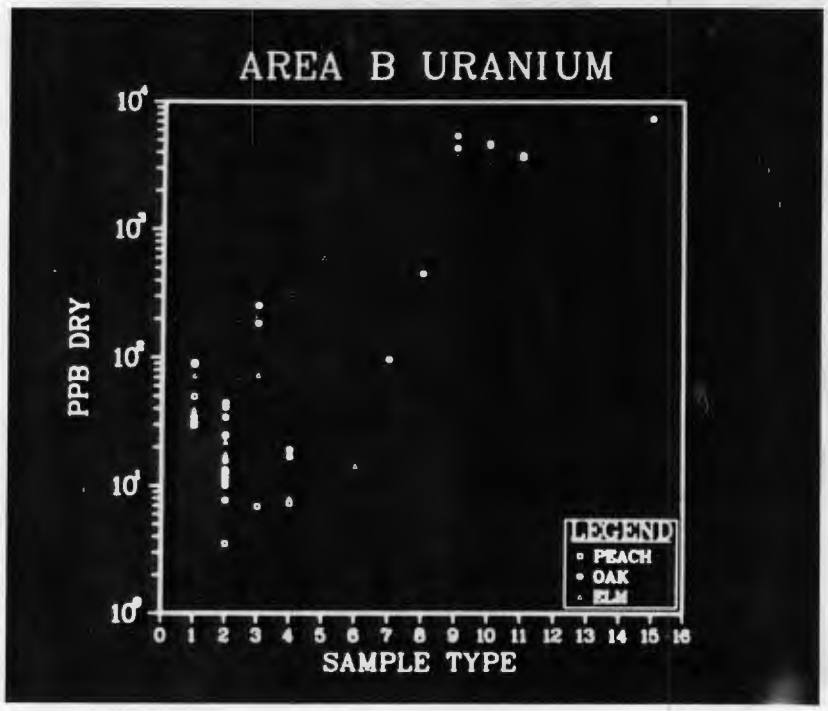

Fig. 13. Uranium concentrations in soil, litter, and plants.

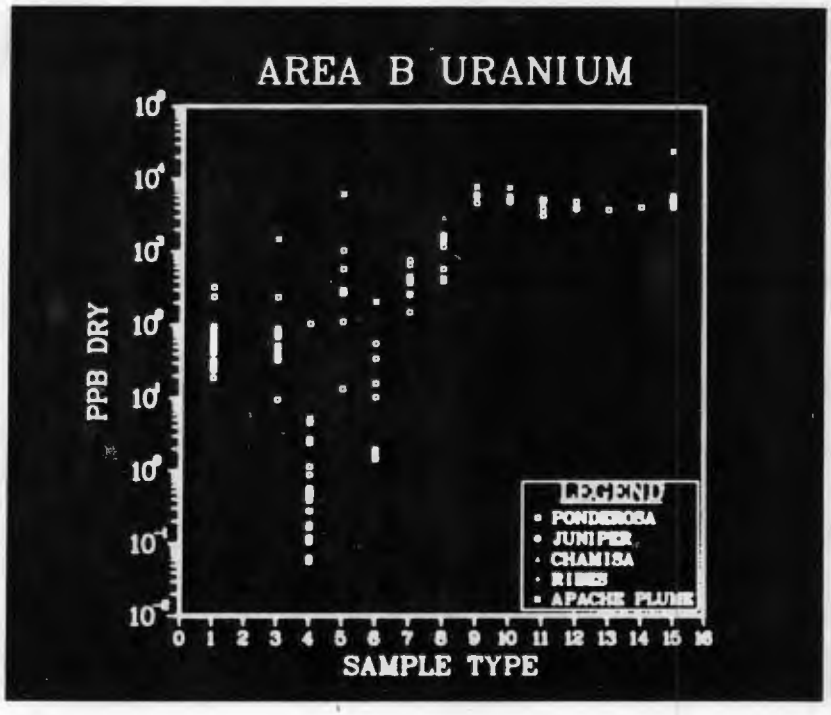

Fig. 14. Uranium concentrations in soil, litter, and plants.

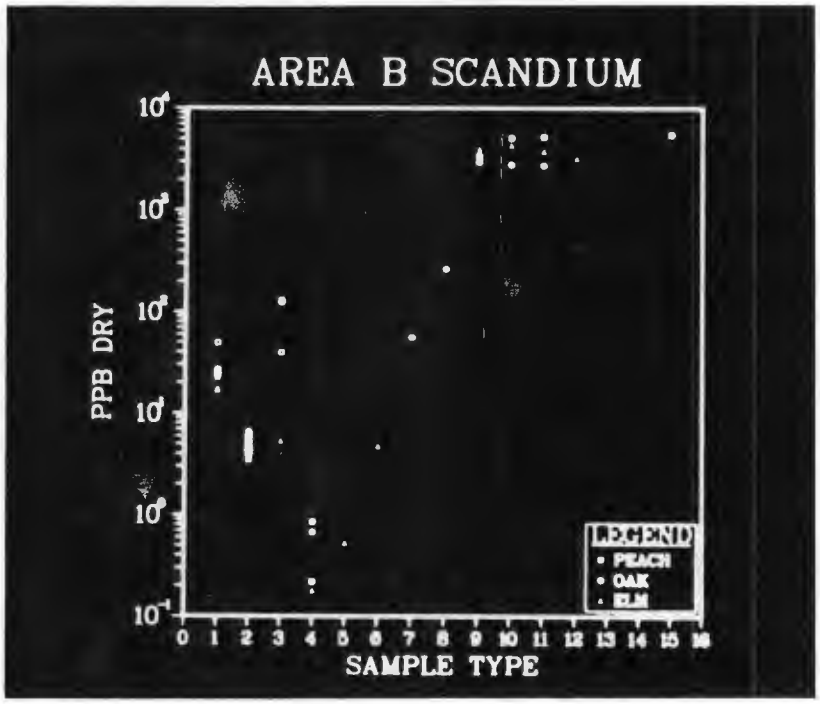

Fig. 15. Scandium concentrations in soil, litter, and plants.

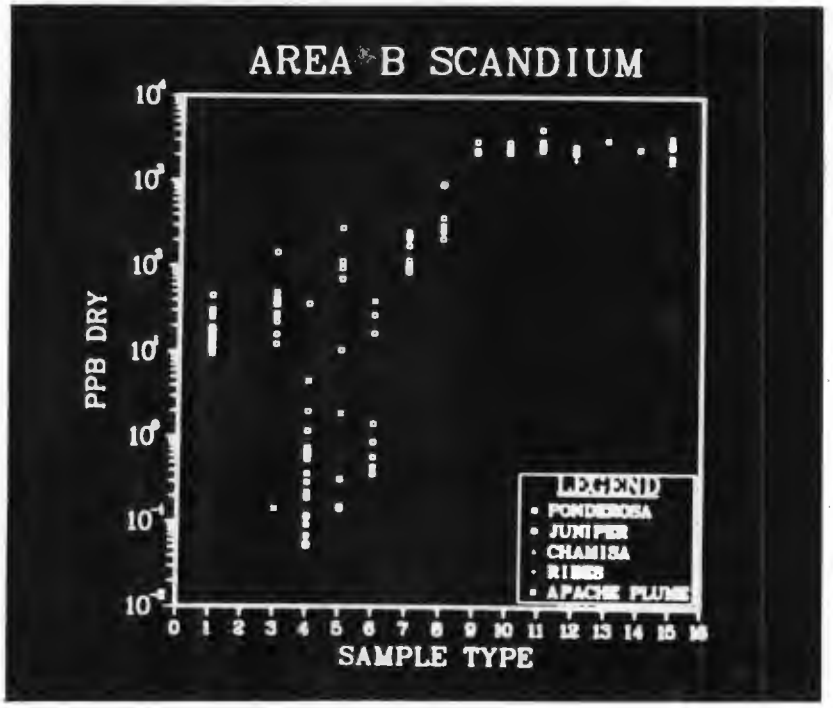

Fig. 16. Scandium concentrations in soil, litter, and plants.

Table IV. Arithmetic Mean, Standard Deviation, and Coefficient of Variation for Radionuclide Results

Sample Type	N	Sc (ppb dry) $\bar{x} \pm 1\sigma$	Sc COV	U (ppb dry) $\bar{x} \pm 1\sigma$	U COV	^{137}Cs (fCi/g dry) $\bar{x} \pm 1\sigma$	^{137}Cs COV	$^{239/240}Pu$ (fCi/g dry) $\bar{x} \pm 1\sigma$	$^{239/240}Pu$ COV
Ponderosa Pine 5 - growing in waste									
Soil around waste debris (>100 cm)	6	2660 ± 434	0.16	5070 ± 581	0.11	168 ± 267	1.6	578000 ± 323000	0.56
Litter - pine needles	2	111 ± 11.3	0.10	430 ± 20.5	0.048	385 ± 28.3	0.074	3570 ± 764	0.21
Litter - misc.	1	262	-	585	-	423	-	7210	-
Unbagged needles	3	10.4 ± .912	0.088	63.3 ± 8.61	0.14	23.1 ± 13.4	0.58	21.6 ± 5.84	0.27
Bole bark	2	9.94 ± 8.86	0.89	70.1 ± 49.4	0.70	28.2 ± 36.8	1.3	10400 ± 5830	0.56
Bole wood	5	0.631 ± .0524	0.083	0.385 ± .195	0.51	8.50 ± 11.21	1.3	5.27 ± 1.59	0.30
Root wood	5	3.78 ± 7.22	1.9	10.1 ± 14.5	1.4	-0.699 ± 1.96	2.8	250 ± 328	1.3
Root bark	4	141 ± 96.0	0.68	314 ± 199	0.63	3.80 ± 6.56	1.7	23900 ± 31000	1.3
Ponderosa Pine - all remaining									
Soil 2 cm	3	2510 ± 382	0.15	5630 ± 793	0.14	1075 ± 389	0.36	5650 ± 7340	1.3
Soil 10 cm	3	2540 ± 366	0.14	5290 ± 497	0.094	289 ± 266	0.92	4720 ± 6180	1.3
Soil 25 cm, 30 cm	4	3070 ± 760	0.25	4430 ± 1030	0.23	110 ± 96.3	0.88	1230 ± 1270	1.0
Soil 45-55, 80 cm	2	2840 ± 368	0.13	4415 ± 827	0.19	186 ± 335	1.8	884 ± 702	0.79
Soil 150-160 cm	1	2450	-	4180	-	-46.9	-	1020	-
Litter - pine needles	7	164 ± 72.2	0.44	426 ± 229	0.54	459 ± 161	0.35	217 ± 102	0.47
Litter - misc.	4	454 ± 322	0.71	867 ± 528	0.61	628 ± 356	0.57	816 ± 741	0.91
Unbagged needles	14	20.0 ± 9.48	0.47	76.7 ± 93.2	1.2	77.9 ± 39.6	0.51	13.0 ± 12.1	0.93
Bole bark	11	43.3 ± 35.6	0.82	74.4 ± 60.7	0.82	30.4 ± 35.7	1.2	557 ± 875	1.6
Bole wood	16	0.391 ± .538	1.4	0.996 ± 1.36	1.4	3.71 ± 6.52	1.8	1.52 ± 2.25	1.5
Root wood	2	14.3 ± 18.1	1.3	36.3 ± 28.7	0.79	-1.62 ± 6.92	4.3	164 ± 211	1.3
Root bark	4	2.80 ± 5.20	1.9	412 ± 445	1.1	15.5 ± 25.4	1.6	997 ± 1940	1.9

Table IV (cont)

Sample Type	N	Sc (ppb dry) $\bar{x} \pm 1_\sigma$	COV	U (ppb dry) $\bar{x} \pm 1_\sigma$	COV	^{137}Cs (fCi/g dry) $\bar{x} \pm 1_\sigma$	COV	$^{239/240}Pu$ (fCi/g dry) $\bar{x} \pm 1_\sigma$	COV
Peach and Elm (Deciduous trees)									
Soil 2 cm	1	4000	-	3730	-	476	-	18100	-
Soil 10 cm	1	4300	-	3240	-	383	-	29500	-
Soil 25 cm, 30 cm	1	3770	-	3720	-	189	-	7850	-
Soil 80 cm	1	3210	-	3800	-	72.7	-	12100	-
Unbagged leaves	4	26.5 ± 15.5	0.58	47.9 ± 18.2	0.38	-34.2	-	2.63	-
Bagged leaves	3	5.18 ± 1.56	0.30	14.4 ± 9.65	0.67	148	-	4.87	-
Bole bark	2	22.5 ± 24.3	1.1	40.0 ± 46.7	1.2	96.0 ± 63.6	0.66	15.3 ± 3.25	0.21
Bole wood	2	0.200 ± 0.0290	0.15	7.68 ± .488	0.064	1.33 ± 5.43	4.1	4.10 ± 0.431	0.11
Root wood	1	4.77	-	14.3	-	-4.27	-	62.1	-
Root bark	1	0.530	-	602	-	83.8	-	4160	-
Oak, Chamisa, Ribes, Fallugia (Shrubs)									
Soil 2 cm	3	2850 ± 613	0.22	5900 ± 1850	0.31	1200 ± 560	0.47	14600 ± 9330	0.64
Soil 10 cm	3	3390 ± 1500	0.44	5660 ± 1750	0.31	664 ± 110	0.17	5320 ± 1850	0.35
Soils 25, 30 cm	3	3510 ± 1500	0.43	4170 ± 748	0.18	163 ± 64.0	0.39	1670 ± 759	0.45
Soils 45-55, 80 cm	2	2260 ± 651	0.29	4020 ± 141	0.035	-13.3 ± 35.4	2.7	4140 ± 5310	1.3
Soil 150, 160 cm	1	2730	-	3770	-	-35.8	-	729	-
Soil no depth, around waste debris area (>100 cm)	2	3630 ± 2690	0.74	15800 ± 11900	0.75	75.5 ± 7.42	0.098	159000 ± 219000	1.4
Litter - pine needles	1	56.5	-	96.8	-	6570	-	255	-
Litter - misc.	2	1100 ± 1190	1.1	1720 ± 1800	1.0	572 ± 366	0.64	14300 ± 18100	1.3
Unbagged leaves	6	23.3 ± 3.67	0.16	62.1 ± 33.3	0.54	28.3 ± 20.6	0.73	36.7 ± 8.15	0.22
Bagged leaves	12	4.86 ± .848	0.17	21.0 ± 12.8	0.61	26.9 ± 31.4	1.2	5.27 ± 3.76	0.71
Bole bark	3	85.4 ± 73.9	0.87	650 ± 746	1.1	252 ± 219	0.87	98.4 ± 12.4	0.13
Bole wood	3	2.05 ± 2.22	1.1	13.8 ± 7.44	0.54	12.0 ± 9.49	0.79	21.6 ± 3.44	0.16
Root wood	1	39.6	-	210	-	-10.4	-	5.21	-
Root bark	1	1.90	-	6250	-	121	-	113	-

in the soils. This observation is important because determination of radionuclide distributions in the environment requires sampling strategies at the proper resolution to determine the distribution. For Area B, scandium and uranium are more uniformly distributed; therefore fewer samples are needed to describe their distribution. Cesium and plutonium require much finer resolution sampling to decrease the COV to the same level as scandium and uranium.

IV. BIOTRAN COMPUTER SIMULATIONS AND CALIBRATIONS

A. Summed Plant Simulations

The Area B data are an opportunity to investigate surface and subsurface transuranic waste transport and uptake over about a 40-year span. Several simulations using the BIOTRAN subroutines WATFLX, PLTGRO, UPTAKE, and FORMAN were made to calibrate the models. The UPTAKE subroutine was expanded to include stems, bole, root, and bark exterior and interior components in addition to leaves and fruits. This allowed both surface and internal radionuclide concentrations to be simulated for leaves, bole, and roots with mass balance between live and dead above and below ground biomass, soil, and litter.

Because serial soil samples were not available over the 40-year history of Area B and the buried radionuclide source term was unknown, the soil radiochemical results from samples taken in 1982 beneath the plants and during excavation were summed by soil layer under four plant types. The soil layer means for the six soil layers are shown in Table V. The soil means were then used as input to the WATFLX subroutine. The BIOTRAN input files called ARBTST1 to ARBTST4 are given in Appendix C. Each element was simulated separately over 40 years for the four plant types: ponderosa pine 5, all remaining pines, oak and elm, and the shrubs. The measured 1982 soil source term was assumed to represent the source term on the first year of the simulation. Other simplifying assumptions were made for the soil source term due to incomplete sampling of all the layers. For ponderosa pine 5, the mean soil concentrations from the remaining ponderosa pines were used as input in the upper profile because only the deep layer was analyzed. In addition, when soil layer results were missing the value from the closest layer above it was used. The radionuclide distribution on and in the four plant types was then simulated using the Table V mean soil source terms as input. The observed and the simulated nuclide concentrations in litter, leaves, bagged leaves, bole bark, bole wood, root wood, and root bark are given in Table VI.

The observed results in Table VI represent the means of the summed tree data for the sampled compartments. The simulated results are the 40-year concentrations in each of the comparable simulated compartments for those summed trees using the Table V soil source term. The underlying distribution for each of the compartments is unknown (probably a log-normal distribution). Since there is insufficient data to determine the distribution in each compartment (litter, bole wood, bole bark, leaves, root wood, and root bark) as a function of the unknown soil layer concentration distribution and root biomass, a simple analysis approach must be made. How well does the simulation match the observed data? What can be learned from the simulations? Why does the simulation match or why does the simulation not match the data? These questions were addressed by Leggett and Williams (1981) using two approaches - one a geometric and the other a statistical reliability index for models. They measured the distance from the y = x line when the observed data are plotted against the simulated data. This gives a measure of how well the model represents the data (but does not indicate which direction). This can be done by analyzing residuals). If the Y_i are the measured observations and X_i are the model predictions, then the geometric method gives the formula

$$k_g = 1 + \left[\frac{1}{n}\sum_{i=1}^{n}\left(\frac{1 - Y_i/X_i}{1 + Y_i/X_i}\right)^2\right]^{1/2}$$

$$= \frac{}{1 - \left[\frac{1}{n}\sum_{i=1}^{n}\left(\frac{1 - Y_i/X_i}{1 + Y_i/X_i}\right)^2\right]^{1/2}}$$

and the statistical method leads to the formula

$$k_s = \exp\left[\frac{1}{n}\sum_{i=1}^{n}\left(\mathrm{Log}\frac{Y_i}{X_i}\right)^2\right]^{1/2}$$

where n is the number of (X_i, Y_i) pairs. Williams and Leggett showed that K_g, based on the geometrical distance of the (X_i, Y_i) pair from the corresponding (x, y) pair on the x = y line, is an intuitive measure of the model's reliability. Models that predict observations well have $k_g \cong k_s$, but where predictions are only within an order of magnitude, k_g/k_s falls below 0.70. The reliability index k_g can then be used as a method of comparing models and therefore determining how well the model fits a set of observations.

Table VII is the summary table for the reliability index analysis for the summed plant simulation and for the individual plant simulation discussed later. The k_g ranged from 2.6 for uranium, 2.7 for scandium, 3.7 for cesium, and 5.3 for plutonium. For the elements where the soil source term is relatively homogeneous, scandium and uranium, the model reliability and the measurement techniques (i.e., the scandium and uranium observation "truly" reflect the distribution in the soil layers) are about a factor of 3.

Table V. Soil Source Terms for Summed Plant Simulation

Plant Type	Layer Size (mm)	Sc (ppb)	U (ppb)	Cs (fCi/g)	Pu (fCi/g)
Ponderosa Pine 5	20	2510	5630	1075	5650
	100	2540	5290	289	4730
	250	3070	4430	110	1230
	500	2840	4415	186	884
	500	2660	5070	168	578000
	500	2450	4180	0.0	1020
All Remaining Ponderosas	20	2510	5630	1075	5650
	100	2540	5290	289	4720
	250	3070	4430	110	1230
	500	2840	4415	186	884
	500	2840	4415	186	884
	500	2450	4180	0.0	1020
Peach and Elm	20	4000	3730	476	18100
	100	4300	3240	383	29500
	250	3770	3720	189	7850
	500	3210	3800	72.7	12100
	500	3210	3800	72.7	12100
	500	3210	3800	72.7	12100
Shrubs	20	2850	5900	1200	14600
	100	3390	5660	664	5320
	250	3510	4170	163	1670
	500	2260	4020	0.0	4140
	500	3630	15800	75.5	159000
	500	2730	3770	0.0	729

Table VI. Simulated and Observed Values for Summed Plants

	Scandium		Uranium		^{137}Cs		$^{239/240}Pu$		Surface layer Litter Adjust ^{137}Cs		Deep layer Adjust $^{239/240}Pu$	
	sim	obs	sim	obs	sim	obs	sim	obs	sim	obs	sim	obs
Ponderosa #5												
Litter	311	161	700	481	135	398	3522	4780	397	398	4276	4780
Leaves	26.1	10.4	122	63.3	60.8	23.1	122	21.6	165	23.1	1813	21.6
Bole bark	18.9	9.94	87.9	70.1	43.7	28.2	89.6	10400	118	28.2	1323	10400
Bole wood	1.14	0.631	5.33	0.385	2.66	8.50	5.38	5.27	7.20	8.50	80.6	5.27
Root wood	5.79	3.78	27.0	10.1	13.4	-0.699	27.1	250	36.4	-0.699	403	250
Root bark	191	141	329	314	18.7	380	1430	23900	32.8	380	17390	23900
Remaining Ponderosa												
Litter	311	275	701	586	135	520	3493	434	519	520	3493	434
Leaves	26.3	20.0	1121	76.7	60.8	77.9	12.3	13.0	213	77.9	12.8	13.0
Bole bark	19.0	43.3	87.0	74.4	43.7	30.4	10.2	557	153	30.4	10.6	557
Bole wood	1.15	391	5.28	0.996	2.67	3.71	0.536	1.52	9.29	3.71	0.560	1.52
Root wood	5.82	14.3	26.8	36.3	13.5	-1.62	2.82	164	46.9	-1.62	2.94	164
Root bark	192	2.80	328	412	18.7	15.5	403	997	39.2	15.5	427	997
Peach and Elm												
Leaves	1.97	26.5	4.55	47.9	-	-	-	-	-	-	-	-
Bagged leaves	2.04	5.18	4.71	14.4	-	-	-	-	-	-	-	-
Bole bark	10.8	22.5	20.8	40.0	6.10	96.0	75.9	15.3	42.9	96.0	13.5	15.3
Bole wood	0.732	0.200	1.70	7.68	0.545	1.33	0.924	4.10	3.78	1.33	0.668	4.10
Root wood	-	-	-	-	-	-	-	-	-	-	-	-
Root bark	-	-	-	-	-	-	-	-	-	-	-	-

Table VI. (cont)

	Scandium		Uranium		^{137}Cs		$^{239/240}Pu$		Surface layer Litter Adjust ^{137}Cs		Deep layer Adjust $^{239/240}Pu$	
	sim	obs	sim	obs	sim	obs	sim	obs	sim	obs	sim	obs
Shrubs												
Litter	824	754	1704	1180	346	2570	21190	9600	2571	2570	21260	9600
Leaves	34.5	23.3	74.5	62.1	16.2	28.3	864	36.7	118	28.3	876	36.7
Bagged leaves	1.05	4.86	5.38	21.0	2.23	26.9	2.52	5.27	14.5	26.9	12.6	5.27
Bole bark	124	85.4	638	650	259	252	300	98.4	1690	252	1516	98.4
Bole wood	0.441	2.05	2.27	13.8	0.930	12.0	1.05	21.6	6.09	12.0	5.43	21.6
Root wood	-	-	-	-	-	-	-	-	-	-	-	-
Root bark	-	-	-	-	-	-	-	-	-	-	-	-

Table VII. Model Reliability Index Summary Table

Element	Geometric k_g	Statistical k_s
Summed Plants		
Scandium	2.69	1.79
Total Uranium	2.59	1.63
^{137}Cs Run 1[a]	3.67	2.01
^{137}Cs Run 2[a]	2.98	1.70
$^{239/240}Pu$ Run 1	5.28	2.76
$^{239/240}Pu$ Run 2	4.81	2.62
Individual Plants		
Scandium	3.51	1.96
Uranium	3.02	1.89
^{137}Cs[a]	4.17	2.13
$^{239/240}Pu$[b]	4.16	2.17

[a]Negative observed values excluded from analysis.
[b]Excluded one outlier.

Because the cesium and plutonium reliability factors are higher, either the model did not correctly predict the observation as well as scandium and uranium or the cesium and plutonium observations were not representative of the "true" soil layer distributions, or a combination of both. We feel that a combination of both nonrepresentative sampling and model improvement is needed to increase the reliability for cesium and plutonium. These observations were tested by repeating the cesium and plutonium simulations using different soil source terms in an effort to decrease the reliability index. The second simulations were done by increasing or decreasing the surface and deep soil layer number 5 by a factor derived from the vegetative samples. The idea was to investigate whether the soil results were accurate for the tree's vegetation results. For cesium, the litter and bole wood observed results were higher than the simulated values indicating that the soil concentration was probably higher than was observed with the samples. To correct this, the surface soil layer input was changed for the four plant types by multiplying the soil layer source term in Table V times the ratio of the observed litter concentration divided by the first simulated litter concentration. The simulated surface soil input was increased by factors of 2.95, 3.85, 9.09, and 7.43 for the ponderosa 5, remaining ponderosas, deciduous trees, and shrubs. This resulted in the movement of the simulated litter concentration closer to the y = x line in the second simulation using the amended surface soil source input. Note that the second simulation resulted in a decrease of the k_g from 3.67 in the first run to a 2.98 in the second run. The conclusion is that observed soil surface concentrations were not representative of what the summed plants actually were exposed to. Amending the deep soil layer #5 in a similar fashion using root, bole wood, and leaf samples for ^{137}Cs did not change the k_g. Similar results were obtained for a second plutonium simulation using a summed factor derived from the internal aboveground samples, bole wood and the underground samples, root wood, and root bark. The factors used to multiply times the fifth layer soil source term in Table V were 16.5, 2.86, 4.44, and 7.53, respectively, for the summed plant types. The results for $^{239,240}Pu$ from run 2 were similar to the amendment found for cesium; the k_g (and k_s) decreased indicating that the observed soil source term was not representative. Extrapolating the results from a point sample to an entire soil layer is probably

a poor assumption and is the reason for lower resolution in the cesium and plutonium simulations.

These findings led to the third set of simulations that were done on individual trees with different source term derivations and simulation assumptions. The purpose for the third set of simulations is to try to find whether the individual tree data had lower variance for each element than the summed data.

B. Individual Tree Simulations

A third set of simulations was done on six individual trees instead of the summed trees to determine whether simulations could be improved. The six trees with the most complete data were chosen for simulation. Each tree was simulated over its lifetime (instead of over the entire 40 years) using the 1982 soil source term shown in Table VIII derived from exponential equations fitted for each tree's soil source term profile. Table IX gives the simulated and observed results for the six trees. Missing soil layer concentrations were calculated for each tree from the equations. The reliability index results for the simulations are shown in Table VII and can be compared with the summed plant simulations. Scandium, uranium, and cesium were similar to those from the summed plant simulations but were higher indicating a lower data resolution for the individual trees. However, the results for plutonium showed a better reliability index for the individual trees indicating that the individual tree method is more appropriate where a highly heterogeneous source term is apparent. Obviously, comparing vegetative samples from a tree growing in contaminated soil with samples taken from that soil will result in a better correspondence. This suggests that for cesium and plutonium multiple samples within each of the soil layers and vegetative compartments instead of single samples could readily give enough resolution to have reliability indices comparable with cesium and uranium.

VI. CONCLUSIONS

Ecosystem modeling of the Oak Ridge, TN, White Oak Creek data has shown the difficulties encountered when plutonium behavior is simulated using environmental data (Garten, Gardner, and Dahlman 1978). They found that the amount of plutonium in the modeled forest was most sensitive to changes in transfers of plutonium from soil to other near soil ecosystem components such as roots and litter. Their model sensitivity was related to the large soil plutonium pool and its central location as the primary source term within the forest model. They concluded that the ability to adequately model plutonium transport in ecosystems is strongly dependent upon higher resolution field data from natural environments and an understanding of the causes of variation in the data.

Garten's observations and our own BIOTRAN simulations suggest that new environmental sampling strategies are needed to gain pertinent data from many compartments simultaneously. The Area B study was an attempt to take many diverse sample types to gain experience in sampling soil, litter, and vegetative compartments to determine the range of concentration for several elements. Earlier studies at White Oak Creek and in Mortandad Canyon at Los Alamos have reported plutonium concentration ratios for mature trees growing in contaminated sediments (Dahlman, Bondietti, and Eyman 1976; Hakonson and Bostic 1976). These studies indicated lower plutonium values for tree leaves than for other exteriorly contaminated tree compartments such as twigs and stems due to their distance from the soil surface source term. They also indicated a strong correlation between cesium and plutonium biota concentrations in both environments even though cesium concentration ratios were higher by at least a factor of 10.

Inspection of the Area B data shows good agreement with the earlier radioecological studies. Work by Olsen (1965) with tulip poplar trees and Klepper et al. (1978) with rabbit brush indicates distinct environmental cesium cycling patterns not found for plutonium. Because cesium is an analog for potassium, it is not surprising that cesium cycling has a large biological component whereas plutonium does not. We found that our models do not account very well for the observed cesium build-up in litter; this is possibly due to the lack of litter decomposition data at Los Alamos to calibrate the litter turnover rate important for mineral cycling mass balance. Other important biologically active constituents in fission and activation products such as strontium, tritium, uranium, manganese, and cobalt could also have strong biologically mediated cycling patterns in the

Table VIII. Individual Tree Simulation Soil Source Terms

Plant Type	Layer Size (mm)	Sc (ppb)	U (ppb)	Cs (fCi/g)	Pu (fCi/g)
Ponderosa 19	20	2240	6000	802	14100
	100	2190	5100	117	11800
	250	4130	5340	138	2740
	500	2744	4782	8.9	120
	500	2744	4782	0.24	2.0
	500	2744	4782	0.0	0.032
Ponderosa 8	20	2350	4720	1520	1975
	100	2920	4910	154	426
	250	2340	3960	0.11	247
	500	2614	4300	0.0	380
	500	2614	4300	0.0	660
	500	2614	4300	0.0	1166
Ponderosa 10	20	2950	6170	903	882
	100	2500	5850	595	1940
	250	3000	5200	219	1810
	500	2807	5726	18.0	1613
	500	2807	5726	0.73	1368
	500	2807	5726	0.0	1160
Elm	20	4000	3730	476	18100
	100	4300	3240	383	29500
	250	3770	3720	189	7850
	500	3798	3615	46	15600
	500	3798	3615	7.1	39790
	500	3798	3615	1.1	101400
Oak 1	20	2970	4346	1480	18700
	100	2790	4570	787	4950
	250	2740	3670	92.5	806
	500	2832	4175	0.78	11.0
	500	2832	4175	0.0	0.044
	500	2832	4175	0.0	0.0
Oak 2	20	3400	5420	1570	21100
	100	5100	4720	574	7320
	250	5240	3810	177	2220
	500	4496	4602	12.0	210
	500	4496	4602	0.0	8.9
	500	4496	4602	0.0	0.0

Table IX. Simulated and Observed Data for Six Individual Trees

	Ponderosa Pine 19 (7 yrs)		Ponderosa Pine 8 (8 yrs)		Ponderosa Pine 10 (20 yrs)		Elm (20 yrs)		Oak 1 (30 yrs)		Oak 2 (30 yrs)	
	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)
Pu												
Unbagged leaves	-	-	1.03	23.0	-	-	-	-	-	-	13.4	32.5
Bagged leaves	-	-	-	-	-	-	-	-	-	-	13.3	1.47
Bole bark	11.7	35.2	2.15	123	3.11	45.3	54.8	13.0	22.7	86.3	-	-
Bole wood	1.24	0.723	0.171	0.340	0.175	0.761	3.15	3.79	0.907	21.0	-	-
Root bark	-	-	-	-	-	-	1196	4160	217	98.0	-	-
Root wood	-	-	0.601	313	-	-	18.8	62.1	-	-	-	-
Litter	105	439	370	273	65.0	156	-	-	-	-	389	577
Cs												
Unbagged leaves	-	-	25.9	109	-	-	-	-	-	-	40.0	16.8
Bagged leaves	-	-	-	-	-	-	-	-	-	-	40.0	1.56
Bole bark	13.1	31.9	29.2	12.1	48.9	5.23	20.8	141	79.7	272	-	-
Bole wood	2.38	-4.42	4.47	-1.49	2.83	3.08	1.21	5.17	3.27	6.17	-	-
Root bark	-	-	-	-	-	-	15.7	83.8	259	460	-	-
Root wood	-	-	16.3	-6.51	-	-	7.42	-4.27	-	-	-	-
Litter	69.0	373	296	555	71.2	467	-	-	-	-	34.0	1434
Sc												
Unbagged leaves	10.2	15.8	9.90	20.1	18.0	45.2	41.1	18.7	18.2	4.32	28.1	24.0
Bagged leaves	-	-	-	-	-	-	41.1	6.00	37.6	128	28.1	4.31
Bole bark	1.74	0.191	1.58	0.0612	1.68	0.0551	1.99	0.179	-	-	-	-
Root bark	-	-	-	-	-	-	264	0.950	-	-	-	-
Root wood	-	-	4.97	1.45	-	-	11.9	4.77	-	-	-	-
Litter	174	145	444	226	219	230	-	-	-	-	65.9	122

Table IX. (cont)

	Ponderosa Pine 19 (7 yrs)		Ponderosa Pine 8 (8 yrs)		Ponderosa Pine 10 (20 yrs)		Elm (20 yrs)		Oak 1 (30 yrs)		Oak 2 (30 yrs)	
	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)	(sim)	(obs)
U												
Unbagged leaves	46.7	49.3	41.8	53.9	85.9	331	88.4	44.1	62.4	32.9	65.7	52.1
Bagged leaves	-	-	-	-	-	-	88.6	19.7	62.4	12.8	138	8.83
Bole bark	46.0	9.57	45.3	80.3	139	33.5	73.7	73.0	129	217	-	-
Bole wood	8.13	0.281	-	-	8.06	0.885	4.29	8.02	5.27	18.0	-	-
Root bark	-	-	-	-	-	-	237	6.02	-	-	-	-
Root wood	-	-	21.5	16.0	-	-	25.67	14.3	-	-	-	-
Litter	436	479	427	902	466	593	-	-	-	-	108	209

southwestern ecosystem similar to that observed here for cesium.

One should also note that this study did not report "concentration ratios" for the elements or "element ratios" commonly found in the literature. Our efforts have been focused on simulating the field data in three dimensions to determine mass balance. Future analyses should be focused on predicting the radionuclide concentration and back calculating to source terms based on the current soil, litter, and vegetative concentrations. This goal is the logical monitoring and surveillance end point required to be able to perform reasonable risk assessments for buried waste sites. To obtain higher resolution simulation with greater confidence, validation studies are needed. Model validation requires high-resolution field data to determine the distribution of these elements in the ecological compartments as a function of time.

Several important concepts useful for surveillance are apparent from the results of this study. All simulations of the Area B data for grouped plants and individual trees were hampered by the high observed data variance and incomplete soil source term sampling. Scandium and uranium soil concentrations were more predictable than cesium and plutonium because their distributions in the soil were more homogeneous. Although some deep and surface uranium contamination was measured at Area B, uranium was localized at the SW corner; therefore, the uranium distribution was more homogeneous than cesium or plutonium. Since Area B was a transuranic waste burial site, trenches with plutonium and also ^{137}Cs (from fission product waste associated with transuranic waste) were the main contaminants. This heterogeneous source term is reflected in the cesium and plutonium data and hence was evident by lower simulation resolution reflected in the geometric and statistical reliability indices. Thorough soil and root source terms need to be developed for such sites as Area B by taking surface and subsurface soil samples in association with individual trees in a manner to reconstruct a three-dimensional soil source term. The tree appears to "see" the integral soil concentration as a direct function of root biomass and root surface area with depth. This concept was clearly illustrated by changing the summed plant simulation soil source terms and simulating the trees and shrubs using soil source terms amended by vegetative concentration factors. The Area B deep soil samples indicate that "hot" spots in the deeper layers are reflected in the above ground biomass compartments.

Each group of investigators at Area B (six separate investigations) used different gridding systems with no record of permanent markers to positively identify where samples were taken. As a result, the data and hence correlation between investigations are probably only reliable to within a few meters. Because many small "hot spots" were seen at Area B, smaller than a few meters, the data could not truly reflect the surface and subsurface radionuclide distribution.

These observations have critical implications for our ability to determine the associated risks on waste sites. We need data from waste sites over long time frames and we need it correlated to the source terms, topography, and biological communities growing there. Systematic stratified sampling of surface and subsurface soils and biomass as well as topographical measurements are needed on the same small grids for the radiochemical results from the major ecological compartments to become interpretable. Resampling is needed on the same grids at specified intervals to ascertain transport in an areal or watershed mass balance mineral cycling context. Radionuclides, their nutrient analogs, and nutrient concentrations also are needed to determine the biological as well as physical transport for these elements.

B. Status of Simulation Capabilities and Surveillance Methods

Major changes in surveillance methods have developed from analysis of the Area B data. The preoperational survey program for new facilities has established two types of sampling strategies, the first a high-resolution radioecological approach on a permanent well-marked site and the second a screening strategy where only soils are sampled at outfalls.

The high resolution sites are ecologically representative 100 m x 100 m sites near the new facilities where samples can be taken on a long-term basis for

temporal comparisons. Detailed quantitative ecological data such as plant density, diversity, cover, age, dbh, and crown cover are measured for biomass calculation by species. Detailed soil hydrology, physics, and biochemistry are also monitored at the site including per cent sand, silt, clay, and per cent H_2O at -1/3, -15, and 0 bar, pH, bulk density, and percent organic matter. Soils are measured for NO_3, NH_4, phosphorus, calcium, potassium, cadmium, lead, and chromium. Nutrient availability is measured using both cation and anion exchange capacities. Vegetative and litter sampling are done on major compartments such as litter, humus, and leaves. Litter and humus are sampled on 0.5 m^2 quadrats and associated soils are taken below the litter samples and leaves above from the associated tree along the permanent 100 m central transect. Small mammal species diversity and density are also measured. Nutrients, heavy metals, and radionuclides are also measured in the plant samples. The high-resolution sampling site represents a "snapshot" of the radioecological status with sufficient data for high-resolution simulations.

The second surveillance strategy is a soil screening sampling method to give an areal overview of the status of nutrients, heavy metals, and radionuclides surrounding the facility and provide preliminary data on the hazard potential. Surface 10-cm-deep soil samples are taken in all drainage areas surrounding the facility. Interpretation and assessment of the risks associated with the screening data rest on the biological and physical interpretation of the high-resolution data from the first strategy. Note that both strategies produce enough data for simulation, assessment, and decision making. Three major sites at Los Alamos have used these two strategies for surveillance sampling, TA-55, TA-53, and TA-18 for new facility preoperational surveys.

The Los Alamos simulation capabilities have grown as new information becomes available. The task is an iterative process where models are continually upgraded to the new data resolution levels. An inherent danger in this process is to overstate the model's predictive capabilities. Another danger is to not use state-of-the-art tools for data analysis, risk assessment, and decision making by restricting model development and utilization. We see model development as a normal process of data analysis and hypothesis testing critical for designing new experiments and decision making. The BIOTRAN models represent the site-specific simulation capabilities at Los Alamos and are constantly being upgraded as new data become available. The simulation results should then be used to assist design of closure experiments and hence decision making. This can be done by upgrading waste management scientific research and engineering development activities to include risk assessment as a major component.

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