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A PRELIMINARY EVALUATION OF THE POTENTIAL FOR PLUTONIUM RELEASE FROM BURIAL GROUNDS AT LOS ALAMOS SCIENTIFIC LABORATORY

by

M. L. Wheeler, W. J. Smith, and A. F. Gallegos

ABSTRACT

In this report an analysis is made of a number of natural phenomena which could result in the release of plutonium from radioactive wastes buried at the Los Alamos Scientific Laboratory (LASL). Background information concerning the history and practice of radioactive waste disposal at LASL is provided. The potential impact of buried radioactive wastes on the environment is addressed through the mechanisms and rates by which the radionuclides can enter the environment. Only mechanisms independent of human activity are considered. They are divided into two classes, acute and chronic. The acute release mechanisms considered are earthquakes, meteorite impacts, and tornadoes. These have been typified by low occurrence probabilities (10^{-6} — 10^{-7}/yr). The chronic mechanisms that have been considered are release through uptake by plant roots, exposure by soil erosion, and transport by soil water. The rates of these processes are low, but may result in radionuclide release over long time periods. The analysis of uptake by plant roots was made using an environmental model currently under development; the model is discussed in some detail.

I. INTRODUCTION AND SUMMARY

Shallow land burial is a common means of disposal for radioactive solid wastes both in this country and abroad. Major burial sites for radioactive wastes are operated by Energy Research and Development Administration (ERDA) - contractors at Savannah River Plant, South Carolina; Oak Ridge National Laboratory, Tennessee; Idaho Nuclear Engineering Laboratory, Idaho; Atlantic Richfield Hanford Company, Washington; and Los Alamos Scientific Laboratory, New Mexico. The hydrogeologic and biologic environment differs widely at the various sites, as do the modes of burial. A determination of the adequacy of the containment provided by the various burial facilities requires site-specific data, and a method for integrating that data. A program has been active at LASL since 1973 with the principal objective of developing a technique for determining the potential impact of waste burial grounds on the environment. The strategy of this program is to develop an evaluation technique using data specific to Los Alamos, but with sufficient generality that the technique may be applied to any waste burial site.

Radionuclides in buried wastes are isolated from the biosphere at the time of burial. They may enter

the environment as a result of migration, or the environment may encroach upon the waste pits through erosion or other mechanisms that breach the burial containment. All processes that result in contact between the wastes and the environment are time dependent, either in a rate or probabilistic sense. The first task in evaluating the potential impact of buried wastes is to identify the mechanisms specific to a given burial site that would result in radionuclide releases to the environment. Second, the rates or frequency of these processes must be determined. Finally, the movement of the released radionuclides through the biosphere must be described in order to arrive at some estimate of the resultant consequences of the release.

Such an evaluation has been performed, on a preliminary basis, for the waste burial grounds at LASL. A review of potential release processes indicates that the frequencies of probabilistic processes are very low (10^{-6} — 10^{-7}/yr). These low occurrence frequencies, combined with the small effects these mechanisms exert on buried wastes, indicate that they can be considered insignificant for radionuclide release. Movement of emplaced radionuclides into the environs by soil moisture involves transport times greatly in excess of the half-life of even ^{239}Pu. Chronic release after soil erosion to the depth of the buried wastes could be expected to occur on a time scale of 50-150 thousand years. The most evident chronic release mechanism on a time scale of a few thousand years has been found to be plutonium uptake by plant roots, with subsequent dispersal to the environs.

An analysis has been made of a release scenario assuming revegetation of waste burial grounds with the natural species of the Los Alamos area. This analysis is based on a preliminary version of an environmental model, and employs estimated input parameters obtained from current literature. Within the uncertainties inherent in the use of a model of this nature, and the uncertainties present in the estimated input data, it appears that plant uptake from LASL burial grounds could, over a period of about 5000 yr, increase plutonium concentrations in the land directly over the waste emplacement by levels that are about the same as those presently observed as a result of global fallout plutonium.

On the basis of these preliminary analyses we conclude that there is no environmental hazard from plutonium in the LASL waste burial grounds which exceeds that already imposed by plutonium from fallout. In addition, the area affected is essentially restricted to the surface of the waste pits. More complete and detailed analyses will be forthcoming as development and validation of the model progresses.

II. LAND BURIAL AT LASL

Wastes containing plutonium and other radionuclides have been buried at LASL since the beginning of the Laboratory operations in the early 1940s. Open pits or trenches have provided the principal disposal facility, but vertical shafts and covered seepage pits have been used for special waste forms. All shafts, pits, and trenches have been excavated in the surface of the Bandelier tuff, the principal rock-type exposed in the Los Alamos area. The tuff (a welded volcanic ash) comprises an upland area referred to as the Pajarito Plateau. The plateau is dissected by numerous canyons trending east-west, all of which drain into the Rio Grande. The bulk of the LASL facilities, including the waste disposal sites, are located on the tops of the resulting finger-like mesas. The locations of the principal radioactive waste disposal sites at LASL are indicated in Fig. 1.

A large variety of waste types have been buried at LASL. The bulk of the material is room-generated trash, such as paper, packing material, protective clothing, broken glassware, obsolete contaminated equipment, etc., which is generally contained in cardboard boxes or wooden crates. A wide variety of disposal operations are performed, ranging from shaft disposal of cylinders containing millicurie quantities of tritium to demolition and burial of entire contaminated buildings. During the early years of the Laboratory, liquid wastes were disposed directly to the ground by discharging them into seepage pits. Since 1952, the sludges resulting from liquid waste treatment have been placed in drums for burial, or mixed with cement and poured into shafts.

Presently, solid wastes are placed in open pits, characteristically about 10 m deep × 15 m wide × 130 m long. The wastes are placed in layers, with each layer covered with the crushed tuff excavated during construction of the pit. Location, physical description, and radionuclide content of the wastes are recorded in log books and on a computerized record keeping system. When a pit is filled to within

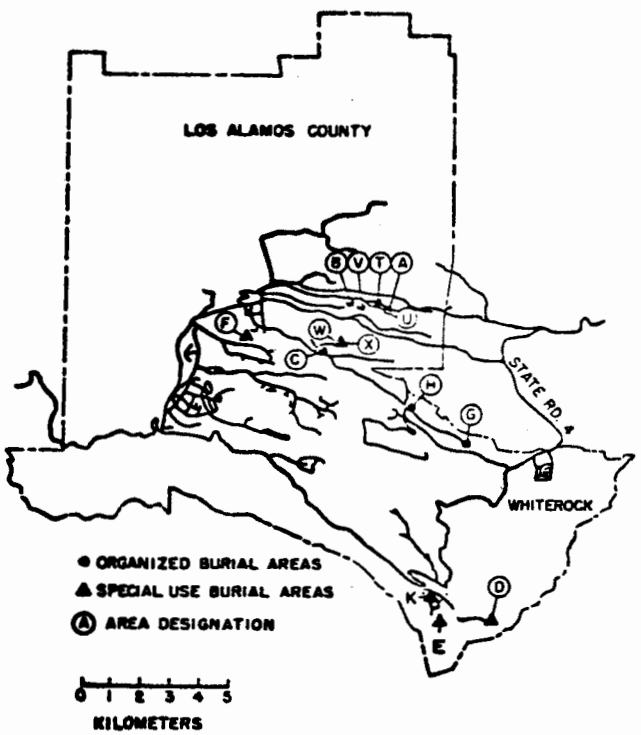


Fig. 1.
LASL waste burial site.

a meter or so of the ground surface, a cover of crushed tuff is overlaid and mounded about 1 m high to facilitate precipitation runoff. The surface is then reseeded with native vegetation for erosion control.

Present practices have evolved from a variety of burial procedures, but historically all practices are similar: wastes are placed in trenches to within a meter or so of the surface and the trench is then backfilled with uncontaminated crushed tuff. A records survey was made of the radionuclide content and composition of solid wastes disposed at LASL since the beginning of the Laboratory. Waste disposal records were highly variable in quality and quite incomplete until the mid 1950s. Detailed records of content and composition were not kept until 1959, and the quality of recording has improved steadily since that time. A summary of the estimated quantities of the major radionuclides present in LASL waste disposal areas is presented in Table I.

III. RELEASE MECHANISMS

Determining the potential impact of buried radioactive wastes on the environment requires definition of the mechanisms and rates by which the radionuclides can enter the environment. The various mechanisms can be divided into two major categories: natural phenomena more or less independent of human activity; and advertant acts by man, such as war, land excavation, sabotage, etc. The processes discussed here all fall into the former category and can be further subdivided into two groups; chronic release processes which occur at a more or less uniform rate when viewed on a time scale of tens to hundreds of years, and acute release processes consisting of single events separated by long periods of nonoccurrence. Chronic release mechanisms include exposure of the wastes by wind or water erosion, subsurface transport of radionuclides by migrating water, and plant or animal transfer of buried material to the surface. Acute release mechanisms of significance in the Los Alamos area include earthquakes, meteorite impacts, and tornadoes. Figure 2 illustrates the interrelationship of the various release mechanisms.

A. Acute Release Mechanisms

The waste material in burial pits or shafts is covered with a minimum of 1 m (usually 1.5 m) of fill material. Natural phenomena that could result in a radionuclide release must breach the cover material, or produce a vertical displacement of at least 1.5 m. The potential for various natural events to expose the wastes in this manner was detailed in a previous progress report.[1] This information is summarized below.

1. Meteorite Impact. A meteorite impacting at a LASL waste disposal site could uncover radioactive material and make it available for distribution to the environment. Analysis of meteorite craters indicates that a crater at least 3 m in diameter would be necessary to penetrate through the burial ground cover and would require a meteorite of at least 0.6 kg.[2] Estimates of the number of craters formed as a function of crater diameter lead to an impact estimate of

TABLE I

ESTIMATE OF RADIONUCLIDE CONTENT OF PRINCIPAL RADIOACTIVE WASTE BURIAL PITS THROUGH 1974

Disposal Area	Pit No.	^{90}Sr		Uranium[d]		^{238}Pu		$^{239,240}Pu$		^{241}Am	
		Ci	nCi/g	Ci	nCi/g	Ci	nCi/g	Ci	nCi/g	Ci	nCi/g
A	1-3							<1[a]			
B	1-5							<1			
C	1							1	0.05	c	
	2							1	0.05	c	
	3							1	0.05	c	
	4			2	0.1			2	0.1	c	
	5			3	0.2			5	0.3	c	
	6			20	1.8			16	0.8	150	6
E	1-6			<1							
G	1	3000	55	6	0.1			74	1	650	11
	2			12	0.2			174	2	352	6
	3			10	0.2			64	1	586	10
	4			6	0.1			60	1	219	4
	5			10	0.2			86	2	315	6
	6			6	0.1	<1		26	0.5	c	
	7							3	0.1	c	
	8					<1		2	0.2	c	
	16			2	0.2					c	
	17							6	0.2	c	
	21			2					0.5	c	
T	1							4[b]	8	c	
	2							2.5	5	c	
	3							3	6	c	
	4							2.5	5		
V	1-3							<0.1		c	

[a]Based on limited quantity of plutonium discarded during pit use.

[b]Estimate of relative amounts to each bed.

[c]Primary source of ^{241}Am is sludge from liquid waste treatment. No sludge was buried in these pits. There are no records to indicate amount present on other waste materials.

[d]Includes isotopes 234, 235, 236, 238.

$1.3 \times 10^{-6}/km^2/yr$ for craters greater than 3 m in diameter.[3] Also, estimates of meteorite fall data as a function of meteorite mass lead to an impact value of $4 \times 10^{-6}/km^2/yr$ for meteorites of mass 0.6 kg or greater.[4] Since the operational area of the Area G waste burial ground is about 0.1 km^2, the calculated probability estimate is $\sim 10^{-7}$ waste-exposing impacts per year.

Further, the consequences of such an impact would be minor, most likely consisting of a small

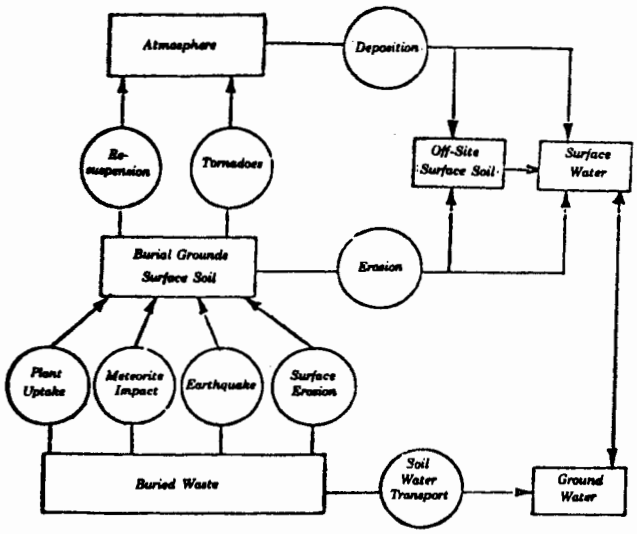


Fig. 2.
Mechanisms for radionuclide release.

area of reduced cover and a smaller area of exposed wastes. Resuspension of radioactive materials could cause low-level air contamination. The effect of erosion on the crater is uncertain; the crater could act to accelerate erosion and exposure of the wastes in the immediate area, but probably would be refilled with eroded soil from surrounding areas, offering little chance for the spread of radioactivity.

2. Tornadoes. Tornadoes or other high velocity wind storms can occur in the Los Alamos area. The principal damage resulting from such storms occurs to above ground structures, as the winds have little excavating capability. Thus the consequences of a tornado would be significant only if waste had already been exposed on the surface. Calculations made on data from an area of one degree of latitude and longitude around Los Alamos indicate the probability of a tornado occurring in the operational portion of Area G is 1.5×10^{-6}/yr.[1] Further, data show that all observed tornadoes have occurred in the eastern, or lower, portion of the region.[5] Investigations by Fujita[6] have shown that both the frequency and intensity of tornadoes decreases very rapidly with increasing elevation. Thus the elevation-corrected probability of tornadoes in Los Alamos would be smaller than the figure given above.

3. Earthquakes. Exposure of buried wastes could occur due to ground displacements by an earthquake. None of the disposal areas at Los Alamos are located on known fault zones, but such zones do exist several km to the west of the Laboratory area. Estimates of earth displacements along the faults indicate that on the order of 130 earthquakes of an average magnitude of 6.7 (Richter) may have occurred in the last million years. This gives an occurrence probability of about 1.3×10^{-4}/yr along the fault zones. However, no faults have been observed in the burial ground areas indicating an occurrence probability of at most 1×10^{-6} earthquakes per year.[1,7]

In addition to the small probability for earthquakes at the burial site, the consequences of an occurrence would be small. The most to be expected would be ground shifting and cracking, with little or no vertical displacement, leaving openings from the surface to the wastes. These cracks would represent a source similar to a meteorite impact, until eroded material fills the crevices.

B. Chronic Release Mechanisms

Chronic release mechanisms occur at a more or less uniform rate for a given environmental setting. This section identifies the nature and rates of various chronic release mechanisms.

1. Erosion. Material on the surface of the earth is continually being transported by wind and water to lower elevations. This process of erosion occurs at a relatively constant rate for a given rock type and geographic area, and is controlled principally by the erodability of the rock or soil and the strength of the erosive forces. The LASL burial areas are located on the tops of mesas, and erosion is continually removing material from both the tops and sides of the mesas. The erosion rate for the mesa tops has been estimated using the age of the volcanic tuff as determined by radiodating, and the estimated original thickness of the ash flow.[1] On this basis, the vertical erosion rate at Area G has been estimated as 2.2 cm/1000 yr.[1] Erosion of the sides of the mesas occurs primarily as successive slumping of blocks of tuff into the canyon. Thus the erosion is characterized by incremental jumps determined by the fracture spacing in the tuff. Determination of the width and age of canyon systems leads to an estimated lateral erosion rate of 10 cm/1000 yr.[1]

Extrapolating forward in time from a geologic history of a million years, these erosional processes will expose the surface of buried waste material in approximately 50 000 yr. During the following 100 000 yr, approximately 2m of waste will be removed, at which time lateral erosion of the mesas will expose the wastes in the trenches closest to the canyon rim. Examination of the present radionuclide content of the various waste disposal pits, as presented in Table I, indicated that ^{239}Pu is the only material of sufficient half-life to be of concern in this time frame. Table II presents a summary of the maximum, minimum, and average ^{239}Pu concentrations in the burial pits which might be anticipated over the next 150 000 yr, based on estimates of present concentrations.

Surface ^{239}Pu concentrations, as estimated for the 50 000 yr period, are above levels currently used as contamination guidelines, and, by present practice, would require some form of area control. However, this stems more from the "as low as practicable" policy than from any known deleterious effects resulting from such contamination. As further work is performed on setting standards for acceptable levels of plutonium concentrations in surface soils, the results will be factored into the present analyses.

2. Water-Related Release Mechanisms. Water from precipitation infiltrates the ground surface, and if present in sufficient quantities may reach the waste material. Dissolution of the radionuclides in the wastes might then occur, followed by movement of the water and radionuclides out of the confines of the disposal pit. This section examines the nature of this process for the LASL waste disposal areas.

All of the waste disposal sites at Los Alamos are located in the surface of the Bandelier tuff. The tuff overlies sediments of the Santa Fe formation, and in some places basaltic rocks occur between the two. The regional water table is located in the sediments at a depth of 200 — 300 m below the surface of the mesas. Figure 3 is a diagram of the hydrology of the Los Alamos area. The mean annual precipitation in Los Alamos over the last 65 years is 460 mm,[8] decreasing significantly with distance from the Jemez Mountains east to the Rio Grande. However, potential evapotranspiration exceeds precipitation everywhere on the Pajarito Plateau, and the majority of the water entering the soil is evapotranspired back to the atmosphere. The soil horizon is generally less than a meter thick. There is a pronounced clay horizon between the soil and the underlying tuff, which effectively restricts downward movement of water into the tuff.

The tuff is highly fractured, with fracture planes spaced one to two meters apart. Within several meters of the surface these fractures are commonly filled with fine-grained soil and *in situ* weathering products. Below about 10 meters the joints are commonly open, and show little or no evidence of weathering. Plant roots have penetrated the filled fractures as the moisture availability is somewhat higher in the finer material. However, there is considerable montmorillonite clay in the fractures, which expands upon wetting. This expansion tends to reduce the permeability and seal the fracture against further downward moisture movement. Fractures intersecting the bottom of the waste pits are commonly open, but are filled with crushed tuff during the pit construction. The water content of the tuff is quite low. Numerous soil sample measurements and *in situ* soil moisture monitoring, with a neutron source and detector probe,[9] indicate that at depths exceeding 5 m the tuff has water content of

TABLE II

CONCENTRATIONS OF ^{239}Pu IN LOS ALAMOS BURIAL GROUNDS

	nCi ^{239}Pu/g Waste			
	Present	50 000 yr	100 000 yr	150 000 yr
Average	1.6	1.4	0.1	0.02
Maximum	8	2	0.4	0.1
Minimum	0.05	0.01	0.002	0.0006

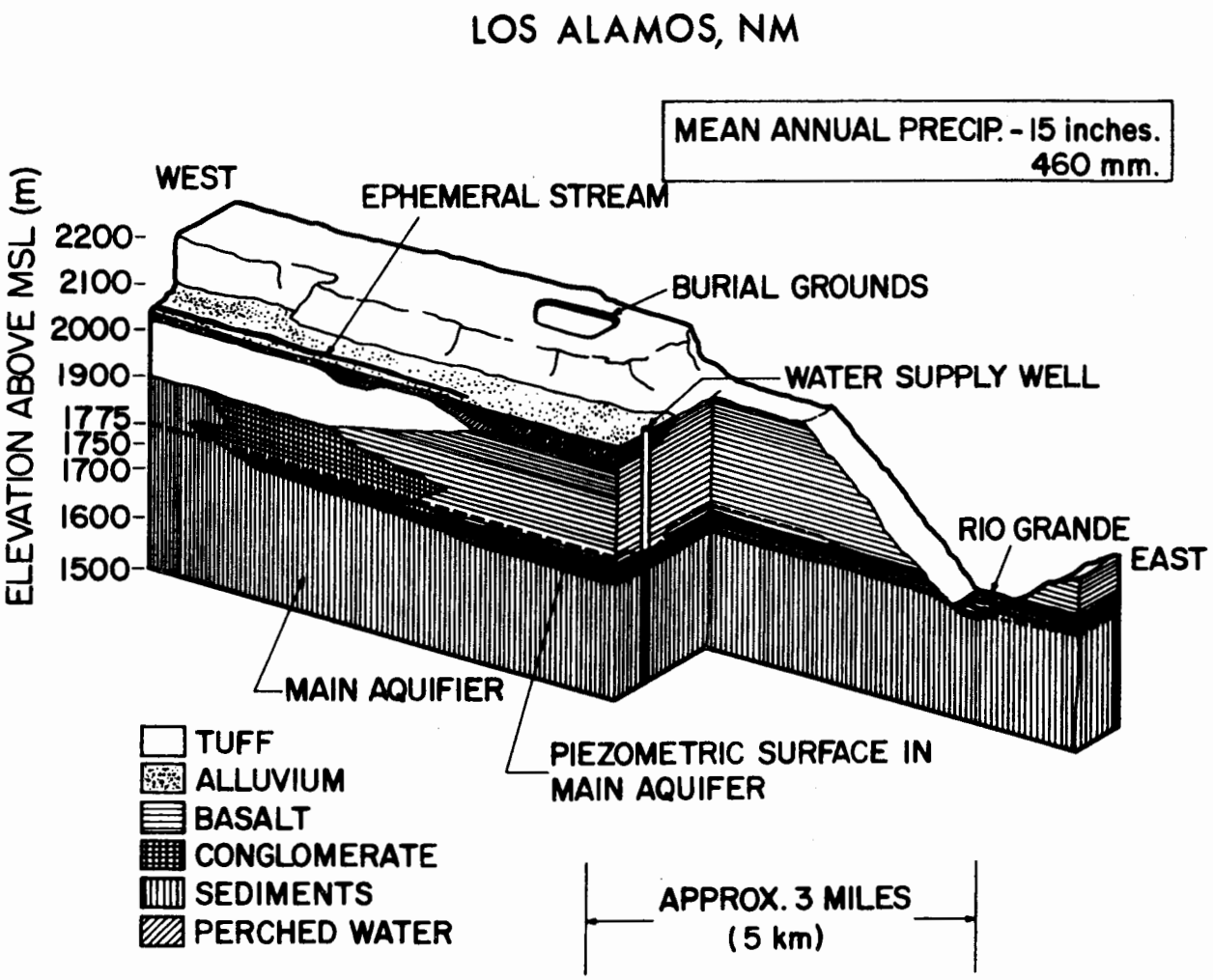


Fig. 3.
Geologic cross section, Los Alamos, NM.

15% of saturation or less. At this low water content, water movement occurs exclusively as unsaturated flow, with much of it being in the vapor phase. The open fractures can conduct water by vapor transport within the fracture, by unsaturated flow along the fracture surface, or by saturated flow if sufficient water is present to fill the fracture.

Some saturated flow may occur in partially filled fractures within a few meters of the surface following heavy precipitation or snowmelt events. However, the absence of weathering in the fractures below about 10 meters indicates this is the maximum depth to which water has penetrated in significant quantities. Flow along the fracture surface is only distinguished from flow in the massive tuff near the surface where flow rates may be driven by saturation or near-saturation conditions. Vapor flux does occur through the open fractures, and is, in part, responsible for migration of tritiated water vapor at the disposal sites.[10] With that exception, all other radionuclides present in the LASL disposal site must be transported in the liquid phase of soil water. Because of low moisture content, saturated flow does not occur in the fracture zones beneath the disposal pits. Thus, the fractures themselves are not pathways for radionuclide migration (other than tritium, as noted). The rate at which water moves through the tuff (the hydraulic conductivity) is also quite low at such low moisture levels. The variation in hydraulic conductivity with water content, as

determined by laboratory measurements, is shown in Fig. 4.[1] For comparison, the hydraulic conductivity of sand and crushed tuff is also present.

When burial pits are excavated, waste material is placed in the pits in layers, and each layer is covered with the excavated tuff. The final cover of all burial pits consists of crushed tuff, to a thickness of from 1 — 5 m. The crushed tuff is frequently devoid of plant material and does not have a significant soil development. Thus, its permeability may be somewhat higher than that of undisturbed areas. Measurements were taken of the water content of fill overlying waste disposal pits, to a depth of 5 m. Soil water contents in fill at depths below 3 m are approximately 10% of saturation and evidence no significant seasonal variation. There is a slight decrease in moisture content with depth, producing a slight downward gradient for water movement. Preliminary calculations indicate this rate is on the order of 1.2 cm/yr[1], but this is a general estimate since the upper several meters are subject to periodic fluctuations in water content, due to the long term variations in precipitation and evaporation.

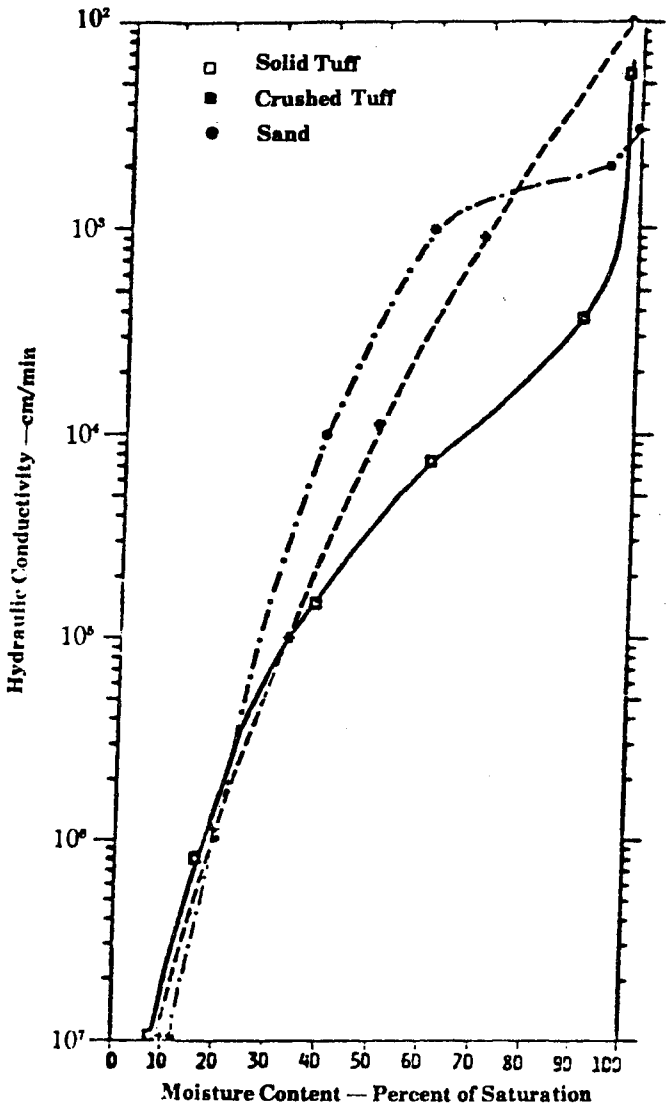


Fig. 4.
Soil conductivity for solid tuff, crushed tuff, and sand.

An alternate approach to estimating the vertical water movement rate uses the observed moisture gradients in the tuff below the disposal pits and the measured hydraulic conductivity of the tuff. Unsaturated moisture flow can be described as[11]

$$q = K(\Theta)\ \partial H/\partial z, \qquad (1)$$

where

q = vertical flow rate,

$K(\Theta)$ = hydraulic conductivity,

Θ = water content of tuff, and

$\partial H/\partial z$ = vertical gradient of driving force.

Under conditions of uniform moisture content, the only significant forces causing moisture movement result from gravity and temperature gradients. Temperature gradients will result in an upward movement while gravity will produce downward movement.[11] In estimating a conservative maximum downward movement rate, the effect of temperature can be ignored. Expressed in units of hydraulic head, the vertical gravitational gradient is unity, and the measured hydraulic conductivity of the tuff at 10% of saturation is approximately 1.5×10^{-4} cm/day.[1] Using Eq. (1),

$$q = K(\Theta)\ \partial H/\partial z$$

$$= 1.5 \times 10^{-4}\ \text{cm}\cdot\text{day}^{-1}\cdot 1$$

$$= 0.05\ \text{cm}\cdot\text{yr}^{-1}.$$

Given the approximations inherent in determining this value, it agrees well with the flux value of 1.2 cm/yr determined from the moisture gradients in the

pit cover material. The higher value is assumed to be a conservative estimate of the water movement rate downward from the disposal pits.

Plutonium, or any other material dissolved in soil water, reaches an equilibrium with regard to the relative quantities absorbed on the soil compared with that in the soil water. This concentration ratio is referred to as the distribution coefficient, K_d. The distribution coefficient is an empirical coefficient, quantitatively describing the effects of a number of processes, including cation exchange, precipitation reactions and physical filtration. These various processes are dependent on the physical and chemical form of the plutonium available for transport, and are thus difficult to extrapolate from one waste to another. Experiments indicate that for plutonium initially in soluble form, a fraction is sorbed less strongly than the remainder.[12] This fraction may move at about the rate of an initial wetting front, but will eventually be sorbed as water movement rate decreases. Thus, when water inputs tend to fluctuate with time, typical of natural conditions, a bimodal distribution of plutonium versus water travel distance may result. The influence on the sorption process of the extremely slow flow rates typical of the Los Alamos area has not been investigated.

The measurements of K_d for the Bandelier tuff have employed either actual waste solutions[13] or a dried plutonium solution which was leached with water. These experiments indicate a value of K_d of 100 to 500, after correcting for the less strongly sorbed fraction. This fraction is sorbed when water movement velocities decrease below wetting front velocities. Field measurements indicate that the moisture content of the tuff beneath the burial pits does not approach wetting front magnitudes at any time. Thus, it is considered appropriate to use the adjusted K_d values.

The sorption of plutonium in the soil solution onto the soil or tuff material retards the rate at which the maximum concentration migrates in the direction of water flow. The relative velocities of the migrating contaminant and the water velocity are expressed as[14]

$$V_i/V_w = 1/(1 + K_d\,\rho/\epsilon), \qquad (2)$$

where $1/(1 + K_d\,\rho/\epsilon)$

V_i = contaminant velocity,

V_w = solution velocity,

ϵ = porosity of medium, and

ρ = bulk density of medium.

For the tuff in the Los Alamos area, $\rho = 1.46$ and $\epsilon = 0.43$ (Ref. 11). Using a K_d value of 200,

$$V_i/V_w = 1.5 \times 10^{-3}.$$

Thus, plutonium in solution will be transported at a rate 1.5×10^{-3} slower than that of migrating soil moisture.

Applying the estimated water movement rate of 1.2 cm/yr, the computed plutonium migration rate is 2×10^{-3} cm/yr. This small value is obviously impossible to validate by actual measurement; however, numerous investigators have applied these principles to movement under saturated or near-saturated conditions, and the principles are valid. In effect, these calculations indicate that plutonium will not migrate from its present location within the waste pits on a time scale which is long compared with the 24 000 yr half-life of plutonium. Thus, transport by moving subsurface waters is not a meaningful release mechanism for plutonium in the LASL waste burial pits.

3. Plant Uptake. Another chronic release process is the uptake of radionuclides by plants whose roots reach sufficient depths to penetrate the buried wastes. This release mechanism is addressed using a technique developed at LASL for simulating the transport of radionuclides through the environment, using as its basis a complex compartmental model.[1,15,16] This computer model is currently applied specifically to north central New Mexico; however, the fundamentals of its structure are, to a great extent, universal. Application to other regions will require only the writing of subroutines to simulate the specific environmental systems. Structural provisions of the model facilitate their incorporation; however, considerable detail on the

climate, biocommunities, and other facets of the region is required for loading and use of the model.

The major compartmentalization of the model is shown in Fig. 5. This environmental simulation model is somewhat unique among its kind in that the design is to model dynamic mass-flow in the biotic and abiotic systems as opposed to those models that rely on static average values for the biomass components. It is primarily a biomass model, using the parameters of the ecosystem that follow the generation and transfer of biomass. The incorporation of radionuclide information, which is the purpose of the effort, is incidental to the functioning of the simulation itself.

In the development of this model only currently available data were used; i.e., no data measurements were made. However, some deficiencies in data available from current literature made it necessary to use several assumed values in lieu of actual measurements.

4. Plant Uptake Simulation. Present policies at LASL require the reseeding of completed burial pits with short-rooted grass species. However, in the absence of permanent maintenance, normal plant succession will occur, eventually producing a stand of mixed piñon-juniper pine with varying understory. Within the time frame of the half-life of ^{239}Pu, artificial controls cannot be assumed, and such revegetation is inevitable. Thus, in simulating plant uptake of radionuclides from the buried waste, a scenario has been used which assumes the undisturbed revegetation of the completed burial grounds by native species.

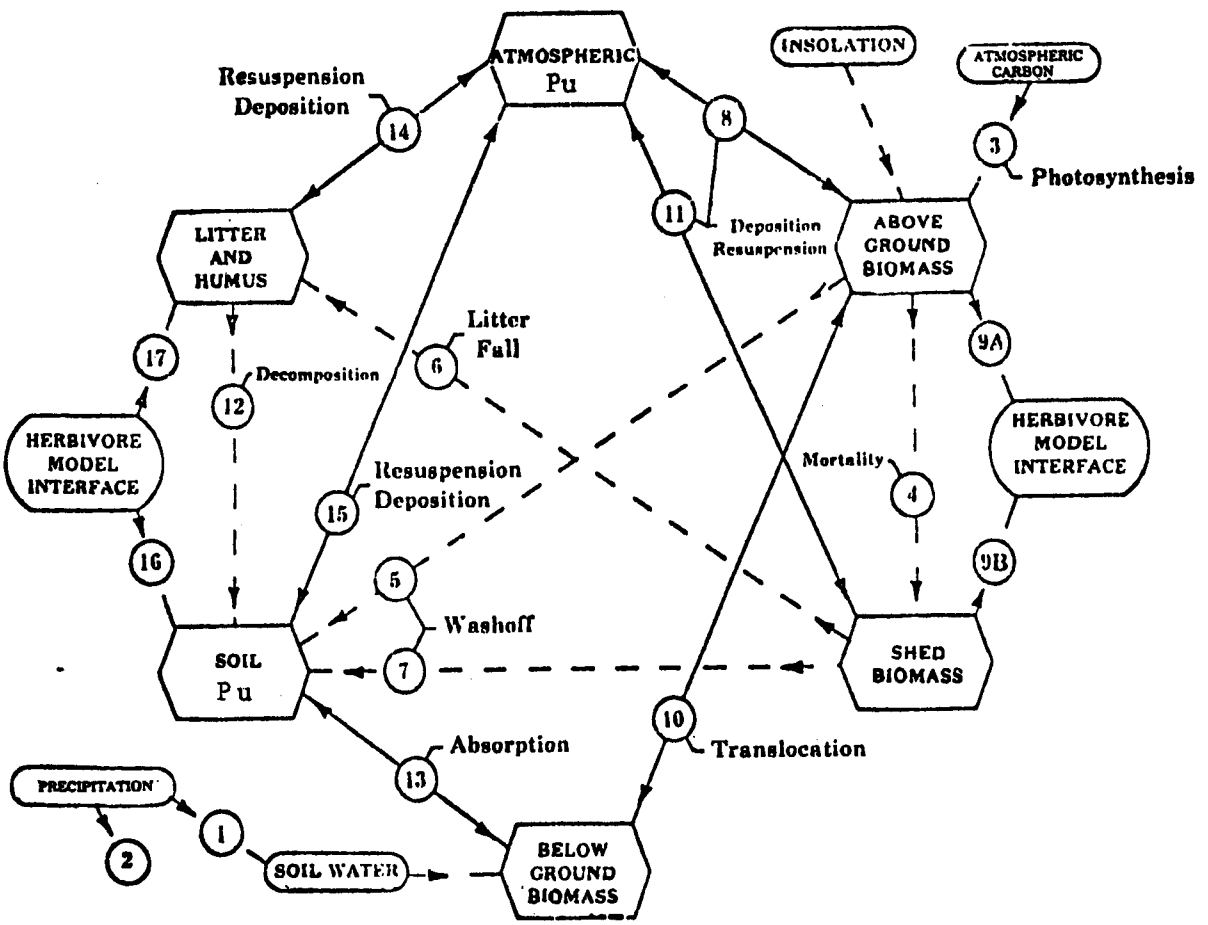


Fig. 5.
Major compartmentalization of the environmental transport model.

The inputs for this simulation are specific to the Los Alamos region, and in particular represent the conditions at an existing waste disposal site, Area G. The climate is simulated stochastically using random selection from a normal distribution based on each parameter's long-term average and variance. The climatic parameters are input to the biological models, which responds to the variations with changes in biomass production and subsequent forest growth or recession. The models for biomass production are iterated on a daily basis within the simulation, and rely on numerous driving and controlling parameters such as soil characteristics, precipitation, solar insolation, water utilization efficiency, temperature, and maintenance requirements of living biomass.

The primary functional design of the model is to simulate the production of biomass and the cycling of plant materials through the soil. As described in program progress reports[1,15,16] this goal has been achieved, and the ability of the model to include trace materials, such as plutonium, as components of the biomass production processes is now being realized.

Data for plutonium wastes buried at LASL (see Table I) indicate a range of average ^{239}Pu concentrations from 0.05 to 8 nCi/g in various waste pits. For this simulation a pit was assumed to have a uniform plutonium concentration of 10 nCi/g ^{239}Pu concentration, covered by a 1.5-m-thick horizon of clean backfilled soil. The only means for radionuclide release considered in this scenario is uptake by plant roots, with transfer to above-ground biomass and subsequent movement to the surface soils via humus decay.

In general, the bulk of a plant's root system is concentrated in the upper 1 m of soil. The root hairs responsible for water and nutrient uptake are found predominantly in the first 15 cm of the soil horizon. Only a very small percentage of tree root biomass is found at depths of greater than 1.5 m.[17,14] An exponential extrapolation of root fraction data for the upper horizons to the lower horizons leads to an estimate, used here, of about 0.2% of the root biomass existing at depths greater than 1.5 m. The estimated root fraction data used in this simulation is given in Table III.

Little data are available for determining the uptake coefficient of plutonium from soil for piñon-juniper. The uptake coefficient, as defined in this model, is the fraction of the radioactivity removed from soil due to respiration of the soil water (mℓ/m^2) by the leaf biomass (g(leaf)). Its units are (mℓ/m^2)$^{-1}$ (g(leaf)/g(soil))$^{-1}$. A value of 4×10^{-9} is used for this parameter; it was determined from data provided by Romney and reported by Martin for uptake by clover over a period of five years.[19] This parameter is of considerable importance and the uncertainty regarding its value for piñon-juniper results in a corresponding uncertainty in the simulation output. An additional problem is the likely variation of the uptake coefficient with root depth in soil. No data have been found to address this problem, thus the same uptake coefficient is used for roots at all depths.

Figure 6 shows the superimposed, above-ground biomass output for three 2000-yr runs simulating the growth of a piñon-juniper forest. Since the burial grounds are assumed to be bare earth at year "0", the forest is seen to grow from zero biomass, developing over a period of 300 to 500 yr to a mature density of 4000 g/m^2, about which value fluctuations occur in response to stochastic variables. The extent of these variations is usually within a factor of 2 of the central value, a realistic range.

Three graphs of the plutonium concentration in the above-ground plant tissue are presented in Fig. 7. This stabilized around a value of about 3×10^{-2} pCi/g for the piñon-juniper forest, using the root distributions and uptake coefficients described above. Again, the effects of stochastic variables are evident. The initial spike, corresponding to output at 10-50 yr, is due to small amounts of plutonium taken up by young plants of very small biomass. The effect of the spike is not significant in terms of the total quantity of plutonium transported to the surface. This fact is indicated by Fig. 8, which graphs the product of the plant mass and plutonium concentration to give the radioactivity contained in plants per square meter of land.

The radioactivity bound in above-ground plant materials (Fig. 8) is seen to rise to an equilibrium value of about 100 pCi/m^2 in 200 to 400 yr. The life span of a tree in the type of forest considered here is about 300 yr,[20] and it appears to take about that length of time for losses to humus to equilibrate with the uptake by roots. Note that the variability evident in this parameter is rather small in contrast to that of the plant biomass (g/m^2) or the concentration in plant biomass (pCi/g), of which it is the product. When suboptimal climatic conditions cause loss of biomass, the concentration of radioactivity increases; when optimal years occur, the biomass may

TABLE III

ESTIMATED PIÑON-JUNIPER ROOT MASS DISTRIBUTION

Depth Interval cm	Fraction of Root Mass	Σ(Fraction)
0 - 25	6.32×10^{-1}	0.632
25 - 51	2.33×10^{-1}	0.865
51 - 76	8.55×10^{-2}	0.950
76 102	3.15×10^{-2}	0.983
102 - 127	1.6×10^{-2}	0.994
127 - 152	4.26×10^{-3}	~0.998
152 - 178	1.57×10^{-3}	
176 - 203	5.76×10^{-4}	
203 - 229	2.12×10^{-4}	
229 - 254	7.80×10^{-5}	
254 - 279	2.87×10^{-5}	
279 - 305	1.06×10^{-5}	
305 - 330	3.88×10^{-6}	~0.002
330 - 356	1.43×10^{-6}	
356 - 381	5.26×10^{-7}	
381 - 406	1.93×10^{-7}	
406 - 431	7.11×10^{-8}	
431 - 457	2.62×10^{-8}	
457 - 482	9.63×10^{-9}	
482 - 508	3.54×10^{-9}	
508 - 787	2.04×10^{-9}	1.00

rapidly increase and the radioactivity will be diluted, but over the long term the total radioactivity bound by the plant remains relatively constant.

Plant detritus serves as the source for release of plant-bound radioactivity to a form mobile in the biosphere. Figure 9 shows the concentration simulated in humus material. For the parameters used in this scenario, a value of 0.6 pCi/m² is found in 200 to 400 yr, again with variability on the order of a factor of 2 or less. This component of the biosystem rapidly degrades into soil materials which are subject to the effects of weather such as resuspension, erosion, and leaching.

Ignoring, for this scenario, the effects of resuspensional and erosional losses on the soil radioactivity, Fig. 10 presents output representing the accumulation of soil radioactivity for the 2000-yr period of this simulation. A value of 0.003 pCi/g in the upper soil horizon results from the set of parameters used in

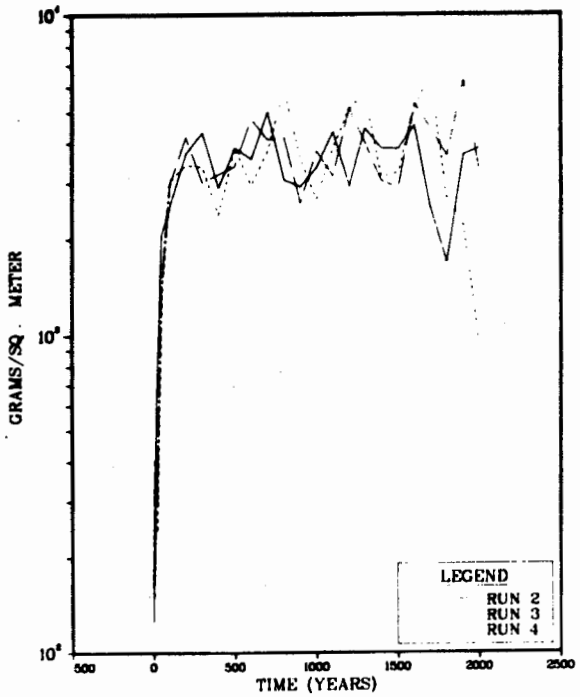


Fig. 6.
Simulated above-ground biomass for piñon-juniper forest, Los Alamos, NM.

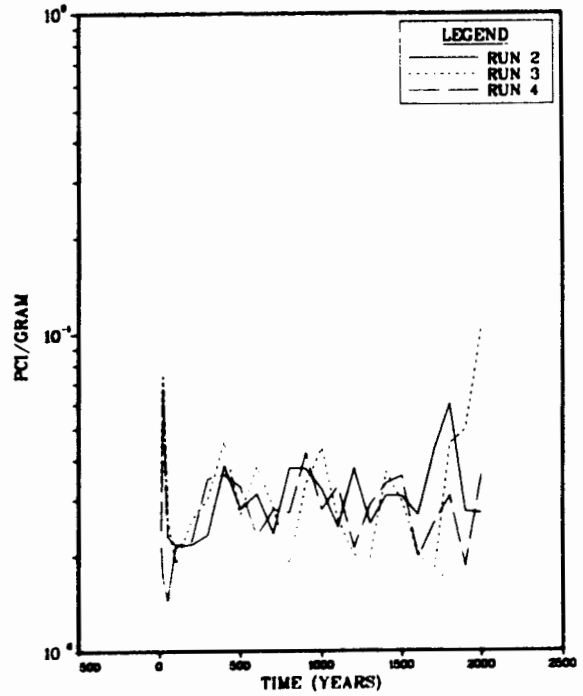


Fig. 7.
Simulated concentration in above-ground biomass for piñon-juniper forest, Los Alamos, NM.

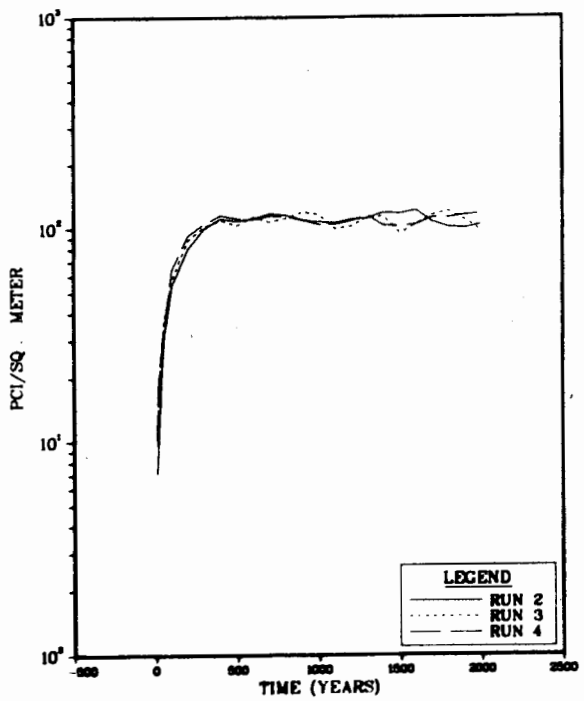


Fig. 8.
Simulated radioactivity in above-ground biomass for piñon-juniper forest, Los Alamos, NM.

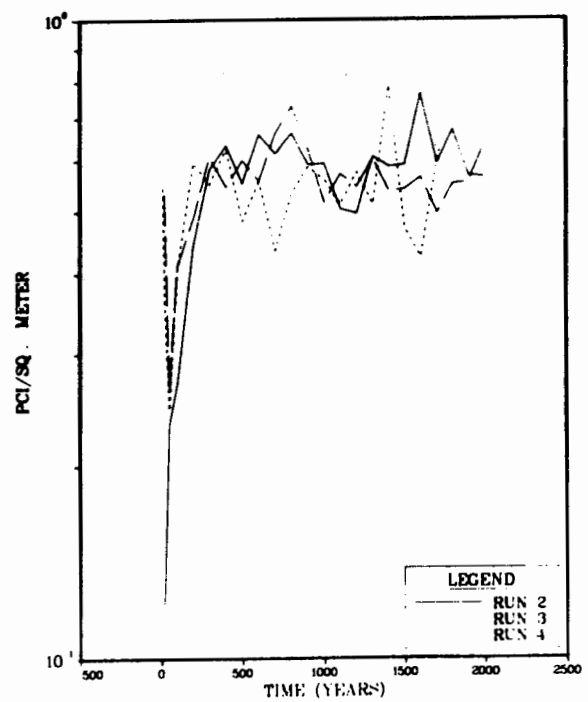


Fig. 9.
Simulated radioactivity in humus for piñon-juniper forest, Los Alamos, NM.

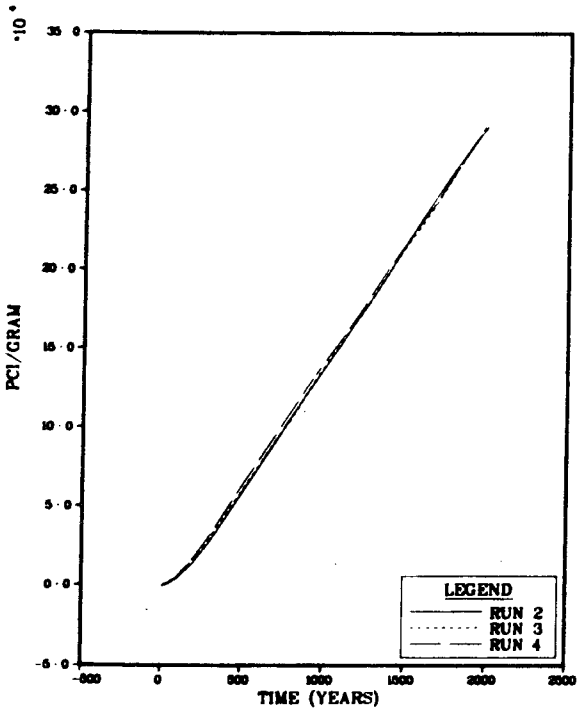


Fig. 10.
Simulated concentration in top 25 cm of soil for piñon-juniper forest, Los Alamos, NM.

the simulation. One significant factor omitted was the loss of the upper layers of soil due to erosion. This can be expected to occur at a rate of several centimeters per thousand years, resulting in the continual removal of the contaminated upper soil layers.

5. Sensitivity and Stability. In order to keep the information derived from these simulations in proper perspective, the response to changes in certain variables and conditions should be examined. For example, since the data for the depth distribution of roots and the root uptake coefficient are scarce, one needs to know the extent to which changes in these parameters affect the output values. Also, it is important to investigate the long-term stability of the model: whether it maintains a uniform stochastic nature or begins to exhibit anomalous characteristics such as a steadily increasing or decreasing magnitude of variability. Many other analyses, especially of the sensitivity to changes in particular parameters, could be made; however, it is felt that a discussion of these few will sufficiently describe the model's abilities and limitations.

The distribution of plant roots with depth in soil is important for defining the contact sustained between the biosphere and the buried waste. A best estimate of the fraction of the root mass extending below 1.5 m (the thickness of clean soil above the buried waste) is made using an exponential extrapolation of data available for the upper soil horizons. This indicates that about 0.2% of the root mass of a mature tree might be found at such depths. In order to determine the sensitivity of the simulation output to error in this important input parameter, tests were conducted using values about an order of magnitude above and below the "best estimate."

Three program runs were made for each of three root fraction values, with results similar to those displayed in the previous section. For ease in display here, however, the three runs were averaged and a single "average run" was plotted for each input value. Figure 11 shows the averaged radioactivity in

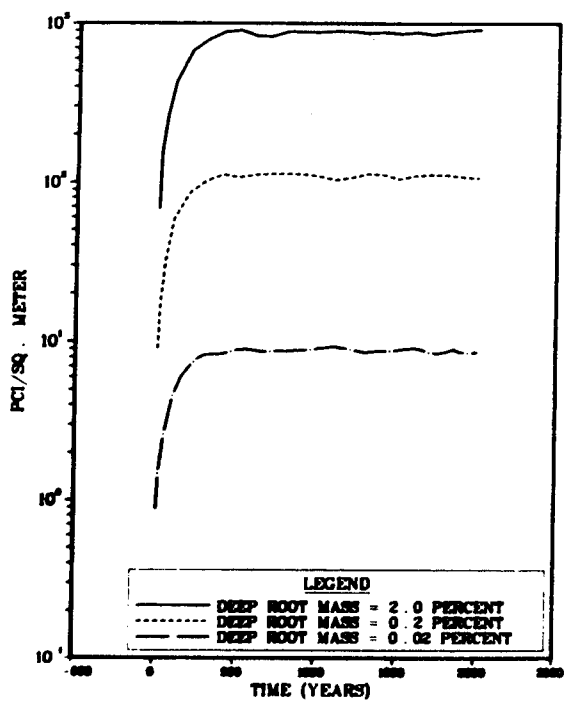


Fig. 11.
Simulated radioactivity in above-ground biomass for differing deep root masses of piñon-juniper forest, Los Alamos, NM.

above-ground biomass outputs obtained for below 1.5-m root fractions of 0.02, 0.2 and 2%. The outputs differ by factors of 10 (ignoring the stochastic variations), indicating that the model responds linearly with variation in root mass distribution. This represents considerable sensitivity to this parameter and provides information allowing simple analytic correction for desired changes in the root distribution input.

Figure 12 provides similar information on variability in the value selected for the root uptake coefficient. The input value currently used is 4.0×10^{-9}, and order of magnitude variations in the input once again produce order of magnitude variations in the output. As expected, the model is very sensitive to the value selected to describe the uptake of plutonium by plant roots from soil and soil water. Again, it is indicated by the linear response that simple analytic corrections can be made for desired changes in this parameter.

The long-term stability of a simulation model such as this is always of concern since the values generated in one iteration affect the results of succeeding iterations. It is necessary to ensure that successive small errors, or perturbations which are insignificant during short runs, do not accumulate and cause unexpected or uncontrolled deviation from proper performance. A run of 5000 yr was conducted in an effort to address this problem.

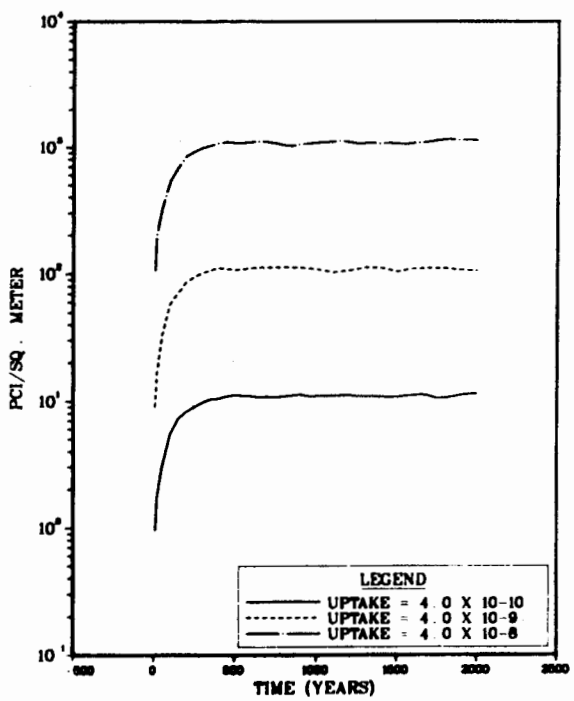
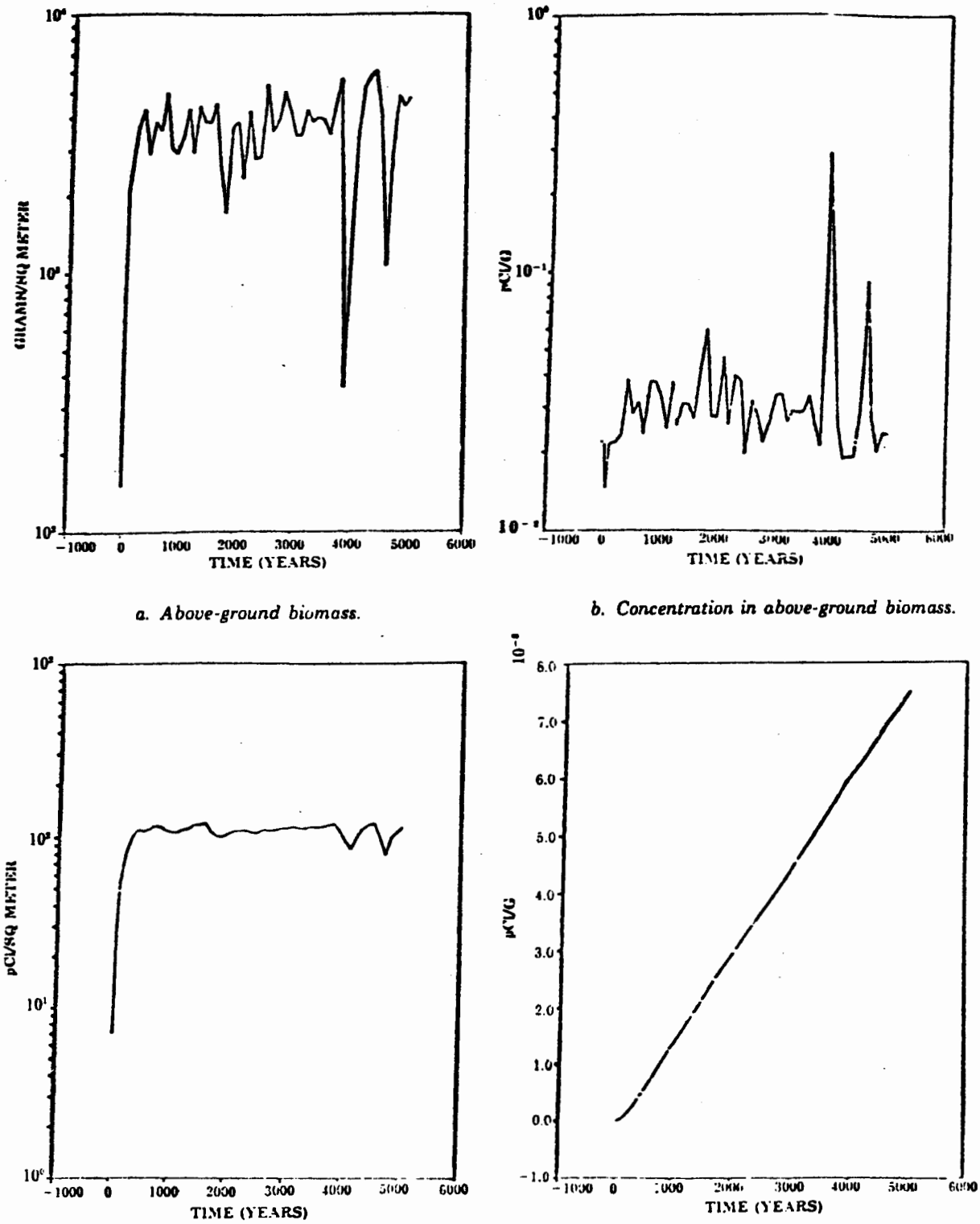


Fig. 12.
Simulated radioactivity in above-ground biomass for differing uptake coefficients for piñon-juniper forest, Los Alamos, NM.

Since equilibrium of the forest simulation generally appears in less than 500 yr, this time frame represents a factor of 10 extension over the necessary time frame for a successful simulation run. Figure 13 is the output from a 5000-yr simulation of a piñon-juniper forest growing over a waste burial ground, using best-estimate input parameters. The drastic recessions of the forest in the later portion of the run are apparent instabilities that deserve close scrutiny. The problem was investigated on the basis that such recessions have been known to occur in nature and that the simulation may actually show acceptable phenomena resulting only from the random nature of the model.

To determine the possible source of the recession centered around 3800 yr, the possible influence of both temperature and precipitation were analyzed. Precipitation of less than 340 mm/yr, or average annual temperatures above 11.5°C, were used to denote suboptimal years for productivity. The percentage of suboptimal years was then determined for the period of recession, (3029-4114), for the normal productivity years in that interval, and for a 100 year period of normal growth. The results, presented in Table IV, indicate that there was a greater than normal percentage of suboptimum years during the recession period. It is concluded that this, rather than any inherent instabilities in the model, accounted for the productivity decline.

6. Model Extensions. Model additions and modifications still under development tend to fall into two categories: those designed to provide more realistic simulation of environmental systems, and those intended to enhance the usefulness of the model. Among the former are developments permitting the simultaneous simulation of numerous highly different plant species. Routines are now being tested which will permit simulation of the process of plant succession which typifies the development of plant communities. Simulation of the wind resuspension of soil particulates is being added. This will provide estimates of soil radioactivity removed by wind erosion, air concentrations resulting from



a. Above-ground biomass.

b. Concentration in above-ground biomass.

c. Activity in above-ground biomass.

d. Concentration in top 25 cm of soil.

Fig. 13.
Five thousand year piñon-juniper forest simulation.

the soil contamination, and plant surficial contamination due to redeposition. The complementary modeling of water erosion is also being developed. The effects of erosion, which may expose the currently buried waste in a period of 50-150 thousand years produces an upper boundary for environmental releases which the action of deep rooted plants cannot exceed.

Two important modifications for increasing the utility of the model are under development. The first is a provision for long-term climatic changes. Because of computer cost and running time limitations, the 5000-yr simulation reported here is the longest run now feasible. With the continual streamlining and generalizing of the code that accompanies further development and revision, it is expected that the maximum simulation period may be somewhat extended. However, the usefulness of such action can be questioned on the basis of whether the environment of today will still be in existence in 5000 yr. The climates of the earth change, and the time scale of drastic changes can well be less than 5000 yr. Effort would perhaps be better spent in investigating the effect of climatic changes on the containment afforded by a burial ground, rather than assuming an indefinite stasis in our global climates. Although modeling of climate changes must be hypothetical, the exercise provides useful insight into potential future problems.

A second major modification is the expansion of the model code to allow simultaneous handling of numerous contiguous land regions of varying environmental characteristics. This modification has been undertaken in order to estimate the effect of burial grounds upon the surrounding, initially uncontaminated, countryside.

7. Significance of Releases. It is interesting to compare the results of this simulation with measurements of plutonium concentrations from worldwide fallout. Measured plutonium concentrations in the soil of northern New Mexico run about 12 fCi/g.[21,22] Figure 13 shows simulated soil concentration to be around 8 fCi/g after 5000 yr. Thus the plutonium concentration in the soil directly over the burial grounds may be expected to about double, over that period of time, due to plant root penetration of the waste. This still leaves the plutonium concentration in the soil within the range of variation now existing on the earth from global fallout.[23]

Simulated concentrations in the biomass of a piñon-juniper forest have an average of about 0.03 pCi/g (Fig. 7). By comparison, fallout plutonium concentrations measured in piñon-juniper trees of the Los Alamos area have shown averages of 0.005 to 0.03 pCi/g.[24] These trees grow in areas where they are subject only to fallout plutonium contamination and represent the response to the plutonium burden already imposed on man's environment. The results of our simulation indicate that the contact sustained between the natural vegetation and the buried waste will tend to about double the plutonium concentrations in the forest biomass directly over the waste emplacement on a time frame of about 5000 yr.

TABLE IV

PERCENT OF SUBOPTIMAL YEARS ATTRIBUTABLE TO TOTAL RAINFALL <340 mm OR AVERAGE TEMPERATURES >11.5°C

Years Analyzed	Percent
Years between 3829 and 4114 which showed decreased productivity	35
Years between 3829 and 4114 which showed normal productivity	21
All years between 3829 and 4114	28
100 consecutive years from a period of normal forest growth	21

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