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TRANSURANIC SOLID WASTE MANAGEMENT PROGRAMS
JULY-DECEMBER 1975

ABSTRACT

Progress is reported for three transuranic solid waste management programs funded at the Los Alamos Scientific Laboratory (LASL) by the Energy Research and Development Administration (ERDA) Division of Fuel Cycle and Production (NFCP).

Under the Transuranic Waste Research and Development Program, continued studies have shown the potential attractiveness of fiber drums as an acceptable substitute for the current mild steel storage containers. Various fire retardants have been evaluated, with one indicating significant ability to inhibit fire propagation. Continued radiolysis studies, under laboratory and field conditions, continue to reaffirm earlier LASL results indicating no significant hazard from radiolytic reactions, assuming no change in current allowable loadings. Care must be exercised to differentiate between radiolytic and chemical reactions. Other efforts have identified a modification of chemical processing to reduce the amounts of plutonium requiring retrievable storage. Studies are also in progress to enhance the sensitivity of the LASL MEGAS assay system.

The Transuranic-Contaminated Solid Waste Treatment Development Facility building was 72% complete as of December 31, 1975, which is in accord with the existing schedule. Procurement of process components is also on schedule. Certain modifications to the facility have been made, and various pre-facility experiments on waste container handling and processing have been completed.

The program for the Evaluation of Transuranic-Contaminated Radioactive Waste Disposal Areas continued development of the various computer modules for simulation of radionuclide transport within the biosphere. In addition, program staff contributed to an ERDA document on radioactive waste management through the preparation of a report on burial of radioactive waste at ERDA-contractor and commercial sites.

I. TRANSURANIC WASTE RESEARCH AND DEVELOPMENT PROGRAM (G. Maestas, R. Martin, R. Nance, A. Zerwekh)

A. Corrosion Studies

Corrosion testing of mild steel coupons from waste drums, of USS-100 Fe-Cr alloy coupons, and of mild steel coupons coated with asphalt, Bitumastic, enamel, WD-40, and hot-dip galvanize, was continued in air containing 50 and 100% relative humidity at ambient air temperature. Samples were taken from coupons exposed for periods of up to 24 months to determine depth and extent of corrosion by electron microprobe examination.

The first cell for interim storage of ^{239}Pu-contaminated waste at the LASL disposal area was closed. This cell is at a depth of from 2 to 8 m below ground surface and contains approximately 165 m^3 of air-filled void space. The temperature in the cell remains essentially constant on a daily basis. For the fall months it was

13.9°C; for the winter it has been 11°C. The relative humidity observed for both seasons is 85 to 87%. This is excessive for mild steel drums, and an effort has been made to reduce it. On several days when the ambient relative humidity was low (10-20%), outside air was drawn through the cell at a rate that exchanged the cell atmosphere every 20 min. The relative humidity in the cell was reduced to approximately 30% in 8 h or less and remained there, or lower, as long as the pumping continued. When the air flow was stopped, the internal moisture returned to the 70 to 80% range in a few hours and in 24 h was back to approximately 85%. The cell temperature and humidity were compared with those found in 0.1- and 0.6-m-diam holes in the area. At the same depth as the bottom of the storage cell (8 m), the temperature and relative humidity were the same. Thus, moisture obviously permeates the surrounding soil fairly rapidly, and air must be moved through the cell almost constantly to maintain low relative humidity. For the LASL storage area, this might be accomplished economically with a wind-driven blower.

As droplets of water have been found to condense on the surface of sensors and the connecting cord in the interim storage cell, a corrosion-inhibiting oil, Texaco Compound L, is being sprayed on all mild steel drums going into interim storage. A shaft will be installed in interim storage cell No. 2 so that drums on all sides of the shaft, six layers deep, can be examined periodically. If the corrosion protection is adequate, or if coated fiber drums can be substituted for metal containers, it may be possible to tolerate high humidity in storage.

B. Storage Container Studies

The continuing increase in dollar costs and difficulty in procuring mild and stainless steel containers and 2.29-mm polyethylene liners have prompted a search for acceptable substitutes. The ultimate goal is to use a safety-qualified, but less expensive, fiber drum for burial and retrievable storage of solid waste. However, it then becomes necessary that the fire hazard be addressed, since combustibles are included in retrievable storage. Although the probability of combustion within a container of combustibles appears rather low, the possibility exists for pyrophoric materials that could explode and rupture a container. The contents would thus be exposed to more oxygen and possible ignition.

A test was performed to establish additional baseline data on the containment capability of mild steel drums. A 50-g piece of cheesecloth was soaked in 16 M HNO_3 for 10 min, air-dried, and placed among the simulated transuranic waste contents in a 115-ℓ mild steel drum. Attempts to ignite the contents with a nichrome element at 800°C caused a small pressure spike 1.5 min after energizing, but there was no explosion and no increase in temperature at that point. The test was terminated after about an hour. Later examination of the contents showed the nitrate-impregnated rag to be completely blackened and the rags and paper in contact with the heated element were charred. The balance of the contents were neither burned nor charred; the drum was not breached and no visible smoke escaped during the test.

Two fire retardant coatings were tested and found unsatisfactory in that they allowed fiber drums to burn and flame to propagate from one drum to another. The tests were made using Kraft fiber drums (from Continental Can Co.) with fire retardant impregnated into each of the 14 layers making up the drum.

An electrical heat source of about 850°C was placed among contents simulating combustible radioactive waste in a covered container. The heat source was kept active for 1 h with no flame occurring. The test

drum was then covered with a metal drum for another 2 h. The contents continued to smoulder and burned through the drum wall. Removal of the metal covering was followed by ignition of the contents and treated fiber. Thus, although the drum burned more slowly than untreated fiber, the retardant was unable to prevent fire propagation.

A third fire retardant coating, Thermo-Lag 226, appears to be a satisfactory fire retardant, based on preliminary tests of fiber treated with the coating. Simulated waste was ignited inside a 12-ℓ uncovered fiber container and the walls remined intact. Subsequent efforts to ignite the container with a propane torch were unsuccessful due to the insulating char formed when the coating was heated previously. Thermo-Lag is a subliming, heat blocking, proprietary substance which absorbs heat by its endothermic decomposition. A char layer is slowly formed underneath the coating, and heat is dissipated by the flow of sublimate gases from the coating and through the char. Heat is also dissipated from the char layer by radiation, blocking additional heat from the protected substrate.

Handling tests were performed with various models of fiber containers. Two-hundred-and-ten-liter drums were filled with about 230 kg of dirt and moved about with a special drum handler on a fork lift. The drums maintained their integrity through severe handling and abuse and remained intact after drops from 1.2 m. The bottom disk, which appears to be the weakest part of the drum, gave way when drops were made from 2.5 m.

Radioactive sludge handling procedures were duplicated using water-filled drums. Although water subjects the container to greater stresses during handling than sludge, it provided "worst case" data. Three tests were performed, each with groups of nine containers. A 0.13-mm polyethylene bag was placed inside each drum and filled with water to within 15 cm of the top. Loading procedures were duplicated at the sludge generating plant, and the drums were transported to the waste disposal area and unloaded. Small water leaks were observed from 2 of the 18 containers after unloading; however, neither sludge nor solid waste would have escaped through the breaks. Fiber containers have subsequently been adopted for continued use in sludge burial.

Representatives of the drum manufacturer visited LASL to discuss waste container needs. Results of fiber drum tests were reviewed and specifications were defined for more sample drums. Although fiber containers currently being used for sludge disposal appear to be adequate, it is felt that the margin of safety can be substantially increased, at a relatively small cost, by strengthening the bottom disk and its bonding with the wall. New drums for fire tests will also be strengthened mechanically, in addition to being coated with Thermo-Lag on the outside.

C. Radiolysis Studies

Radiolysis effects studies continued with the measuring of gas generated by radiolytic attack on 52.5 mg of various waste matrices, each contaminated with 62 mg of ^{238}Pu, contained in selected experimental cylinders. Each of these Type 304 stainless steel cylinders has a volume of 300 cm^3 and is fitted with a pressure gage and a miniature brass shutoff valve. The rates of gas production were monitored (see Fig. 1) and samples were tested for explosivity threshold. Comparision of the curve representing water-soaked cellulosics with that for dry cellulosics shows a significantly increased rate of gas formation when water is present. Clearly, there is radiolysis of the water in addition to the radiolytic degradation of the waste. Comparison of the mol percentages of hydrogen for gas mixtures from wet and dry cellulosics (Table I) further illustrates this point.

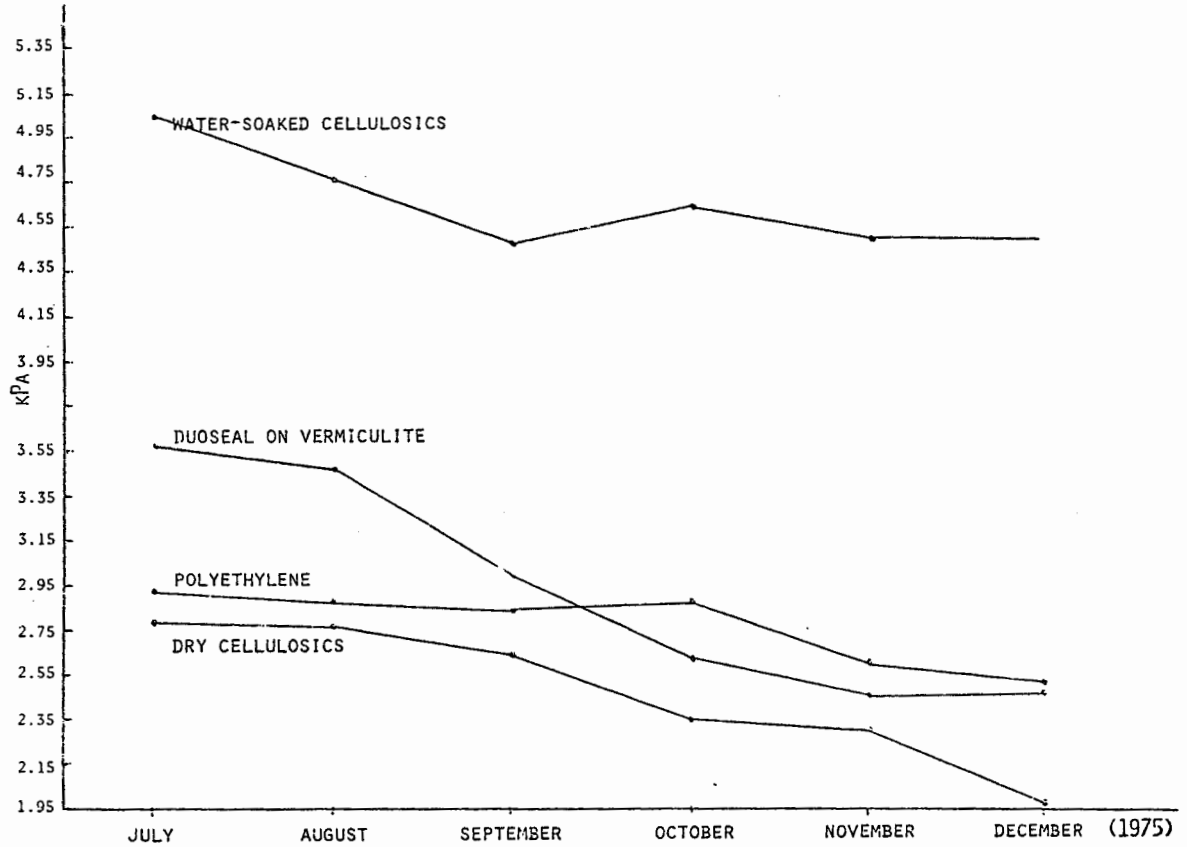


Fig. 1. Gas pressurization, average kPa/d, computed monthly.

Continuous gas formation in contaminated hydrogenous materials is a matter of concern in interim storage facilities. Although the limits of explosivity for hydrogen in air are known, no information was found describing its explosive properties in oxygen-deficient air, with or without other gases present. Therefore, a number of explosivity tests (Table II) have been performed on gas mixtures generated in the laboratory, and one explosimeter test was made on the gas mixture from drum No. 232 after it had been stored for 238 days.

Eight drums of ^{238}Pu-contaminated hydrogenous waste in interim storage have now been instrumented for monitoring temperatures each month and withdrawing gas samples at 1-, 3-, 6-, and 12-month intervals, and semiannually thereafter.

TABLE I

COMPOSITION OF GAS SAMPLES FROM VARIOUS WASTE MATRICES CONTAMINATED WITH ^{238}PU

Cylinder No.	Waste Matrix	Gas Composition (mol %) H_2	CH_4	O_2	CO	CO_2	N_2
35	Water-soaked cellulosics	85	<0.1	<0.1	4	7.1	4
33	Duoseal on vermiculite	89	2	<0.1	0.8	0.5	8
29	Polyethylene	98	0.5	<0.1	0.4	1.2	0.3
1	Dry cellulosics	48	1.8	0.2	14	35	1.3

TABLE II

EXPLOSIMETER TESTS ON VARIOUS GAS MIXTURES

Gas Composition (mol %)						Test			
H_2	CH_4	O_2	CO_2	CO	N_2	No.	Gas Source	Matrix	Explosible
12	0.3	16	9	2	60	2	Cylinder 26	Cellulosics	Yes
17.5	0.5	13.4	14	3	51	3	Cylinder 26	Cellulosics	Yes
16	0.6	13.2	14	8	47	10	Cylinder 27	Cellulosics	Yes
31.9	0.1	12.2	7	3.1	45.1	14	Cylinder 35	Water-soaked cellulosics	Yes
21.7	0.8	11.7	14.6	6.7	43.9	7	Cylinder 1	Cellulosics	Yes
42.8	1	11.6	0.3	0.2	43.6	12	Cylinder 33	Duoseal on vermiculite	No
25.8	0.8	11.1	15.8	4.1	41.5	8	Cylinder 24	Cellulosics, plastics, rubbers	Yes
47.2	0.3	10.9	0.6	0.3	40.2	13	Cylinder 29	Polyethylene	Yes
22.1	0.8	10.6	15	4.6	38.1	15	Cylinder 24	Cellulosics	Yes
40.5	0.1	10.6	6	3	39	4	Cylinder 35	Water-soaked cellulosics	Yes
48.7	0.2	10.4	0.5	0.4	39.1	19	Cylinder 29	Polyethylene	Yes
26	1	10	19	8	36	9	Cylinder 22	Cellulosics	No
35.5	0.1	9.9	8.3	3.9	36.3	21	Cylinder 35	Water-soaked cellulosics	Yes
49.8	1	9.8	0.3	0.4	37.7	20	Cylinder 34	Duoseal on vermiculite	No
49.2	1.1	9.8	0.4	0.5	38.6	11	Cylinder 34	Duoseal on vermiculite	No
51.9	1.2	9.7	0.4	0.4	35.9	17	Cylinder 33	Duoseal on vermiculite	No
27.1	0.8	8.8	21.3	7.9	33.5	18	Cylinder 26	Cellulosics	No
57.1	1.2	8	0.4	0.5	32	5	Cylinder 33	Duoseal on vermiculite	No
36.4	0.1	7.9	7.6	3.3	29.2	16	Cylinder 35	Water-soaked cellulosics	No
63.4	0.3	7.2	0.8	0.4	27.5	6	Cylinder 29	Polyethylene	No
9	0.5	6	17	6	61	1	Drum 232	Hydrogenous solid trash	No

Some of these samples will be tested for explosivity. Four of the instrumented drums have been in storage for 1 yr. They have not pressurized because their seals are not gas tight. It has been shown that gases diffuse through the drum seals and walls of the concrete casks that surround the drums.

Matrices typical of Mound Laboratory wastes were individually contaminated with ^{238}Pu at levels shown in Table III, mixed together, and placed in 300 cm^3 stainless steel experimental cylinders. After 257 days at ambient temperature (20°C), the cylinders have not begun to pressurize. The experiment will be continued to 1 yr, at the request of Mound Laboratory and ERDA/ALO. The gas contents will then be sampled, and the contaminated solid waste removed and examined to determine the extent of radiolytic attack.

A meeting was held in November at Rockwell International at Rocky Flats, attended by representatives from ERDA's Albuquerque Area Office and Rocky Flats Area Office, Mound Laboratories, and LASL. The purpose was to determine the possible causes for unexpectedly rapid pressurization in 210-ℓ steel drums containing alpha activity greater than the allowable limit of 14.6 Ci. Additional concurrent experiments conducted on Mound contaminated waste have reinforced the former conclusion that 200 g of weapons grade ^{239}Pu on hydrogenous waste (4×10^5 nCi/g) does not generate excessive gas pressures in 20-yr retrievable storage.

The relatively rapid pressurization of the RF drums, together with the significant percentage of NO_X found in the gas mixtures, strongly suggested a chemical reaction was the major contributor to gas

TABLE III

CONTAMINATION LEVELS OF SIMULATED MOUND LABORATORY WASTE

Wt% of Contents of Drum	Matrices	Cyl #30 Total ^{238}Pu 22.8 mg	Cyl #31 Total ^{238}Pu 12.1 mg	Cyl #32 Total ^{238}Pu 3.6 mg
		Contamination Levels, nCi/g		
		Strong	Medium	Weak
19	Plastics 45% Polyethylene 45% Polypropylene 10% Polyvinylchloride	15×10^6	7.5×10^6	2×10^6
56	Gloves, Drybox Neoprene/Hypalon/Pb	4×10^6	2×10^6	4×10^6
25	Cellulosics Rags and Paper	2×10^6	1.5×10^6	1×10^6

After 257 days at ambient temperature (20°C), laboratory test cylinders of these mixtures have not pressurized.

formation. A thorough review of the contents of the drums brought agreement that the pressurization problems were indeed caused by one or more chemical reactions. One of the waste mixtures thought to have been responsible for chemical gas generation consisted of ion exchange resin, plutonium, americium, nitric acid, thoicyanate, and other unspecified impurities both in particulate and dissolved forms. The following experiment was performed to determine if the mixture volume could be reduced, the gas generation potential eliminated, and the plutonium and americium left recoverable by combustion at less than 600°C.

A 21.8-g sample of AG 50W X 8 styrene sulfonic acid ion exchange resin, 100-200 mesh, hydrogen form, 50 to 56% H_2O was heated in a covered petri dish to constant weight according to the following schedule.

Temperature (C°)	Wt. Loss (%)	Description of Residue
120	51.4	Dark Brown
200	8.3	Black
300	13.2	Black
400	18.8	Black
500	7.9	Gray ash (1.2 mg)

The volume reduction was significantly large, and the oxide residue should be incapable of gas formation. The combustion was complete at 500°C, avoiding the formation of high temperature plutonium oxide, and a tar residue was not observed.

D. Handling of Waste from Plutonium Processing Areas

1. Nonretrievable Waste

Assay of Room Waste

The MEGAS (Multi-Energy Gamma Assay System) has been in use for about 18 months for evaluating room-generated waste. During this report period, procedures have been developed for improved fission product identification. The MEGAS was moved from an area of highly variable background to one of generally lower and more stable background. This move improved the assay of the majority of those boxes that were in the category of less than 10 nCi/g of waste. A summary of room waste boxes from the plutonium processing areas is presented in Table IV.

Dumpster Lockup Program

The procedures instituted for locking dumpsters situated in the inner exclusion area of the plutonium processing area were described in the Jan-June 1975 semiannual report. In general, the program has been successful in improving the segregation, bundling, and proper marking of <10 nCi/g waste that is too dense or too large to be placed in room trash boxes for

TABLE IV

ROOM WASTE BOXES FOR JULY-DECEMBER, 1975

	<1	1-10	10-100	100-5000
July	142	7	3	3
August	188	14	0	0
September	139	7	1	0
October	168	5	0	1
November	186	16	0	6
December	229	13	3	0
	1052	62	7	10

In summary, 98.5% of the boxes contained <10 nCi/g of waste.

MEGAS assay. There has also been better control of cold waste and segregation of potentially hot waste from the cold dumpster.

Reduction of Waste Volumes

Plutonium recovery is governed by technical, safety, and economic considerations. In the past, difficulties have required the use of a 0.4 wt% plutonium discard limit for the residues from incinerator ash processing; recovery personnel have succeeded in lowering the actual discard level to about 0.2 wt% for these residues. By contrast, residues from the more easily recoverable LASL materials can be discarded below 0.05 wt% plutonium. In 1975, discards to retrievable storage from Hanford ash recovery operations were a significant fraction of all of the plutonium placed in retrievable storage.

Investigations have begun to attempt to improve the percentage of plutonium recoverable from the dissolution of incinerator ashes. Table V summarizes some of the early survey dissolution experiments with ash heel lot LS-4429. The most efficient plutonium dissolution procedure appears to be the two-step process of 1) aqueous hydrofluorination (HF) and 2) nitric acid-aluminum nitrate [HNO_3-$Al(NO_3)_3$] dissolution of the presumed plutonium fluoride.

The results for ignition of samples from ash heel lots LS-4429 and LS-4625 are given in Table VI. Nitric acid (HNO_3) dissolutions conducted on ignited samples produced light-colored solutions, contrasting strongly with the very dark brown solutions typical of most HNO_3 incinerator ash solutions. Apparently, the action of HNO_3 on the organics in the ash lots produces very dark brown soluble organics that obscure subsequent anion exchange operations. Most of the original organics are left in the ash as an acid-insoluble gummy material that coats process equipment and slows filtration. The most recent dissolution experiments with ash heel lot LS-4625 are shown in Table VII. Lower ratios of concentrated hydrogen fluoride (HF)/g ash will be correlated with plutonium recovery in future experiments. Experiments with as-received Hanford heel lots will be started to determine the extent of plutonium dissolutions using aqueous HF followed by HNO_3 and HNO_3-$Al(NO_3)_3$.

2. Retrievable Waste

The largest volume of waste going to retrievable storage from plutonium processing originates in the metal fabrication area. Sorting of glovebox-generated waste at the source has proven to be quite efficient in that much of the waste can be sent directly to retrievable storage, rather than to Recovery for sorting and processing. This also minimizes use of packaging materials and prevents an increase in the Pu-contamination level of some materials because of mixing. A summary of the retrievable waste accumulated by the plutonium operations group is shown in Tables VIII and IX.

E. Investigation of Heat Accumulation in Stored Nitrated Cheesecloth

Cheesecloth is used routinely as cleaning rags in gloveboxes. Experiments performed in 1973 showed that fires and explosions may occur when extensively nitrated cheesecloth is heated beyond a critical point that is dependent on the degree of nitration. Although these experiments used electrical resistance heating as the source of heat, the objective of the present

TABLE V

PRELIMINARY SURVEY EXPERIMENTS WITH ASH HEEL LOT LS-4429

Run No.	Type	mℓ Reagent / Original g ash	Reaction Time (hrs)	mg Pu in Solution x 100[a] / mg Pu in Original Sample	
2	10 M HNO_3-0.09 M CaF	5	1.75	51	
3	10 M HNO_3-0.09 M CaF-0.1 M SO_4	5	2	40	
4	16 M HNO_3-0.09 M CaF_2	5	2	37	
5	7 M HNO_3-0.09 M CaF_2	5	2.3	55	
6	10 M HNO_3	5	2.2	28	
7	10 M HNO_3-0.09 M CaF_2 with -120 Mesh Ash	5	1.9	63	
8A	15 M NaOH	5	3.6	<1	30
8B[b]	8 M HNO_3-0.09 M CaF_2	5	1.7	30	
9	10 M HNO_3-0.09 M CaF_2-0.1 M Ce^{+4}	5	1.8	51	
11	7 M HNO_3-0.09 M CaF_2	5	1.8	57	
12A	Concentrated Aqueous HF followed by diluted HF	5	4 each	3	114
12B[b]	16 M HNO_3;HNO_3-$Al(NO_3)_3$	7	7	111	
13A	Concentrated HCl	10	3	53	87
13B[b]	Concentrated HBr	5	3	34	
14A	Concentrated HCl	10	4.8	43	89
14B[b]	10 M HNO_3-0.09 M CaF_2	10	1.8	46	
15A	Concentrated Aqueous HF	5	5	2	113
15B[b]	16 M HNO_3;HNO_3-$Al(NO_3)_3$	15	6	111	
16A	Concentrated Aqueous HF	5	24[c]	2	116
16B[b]	16 M HNO_3;HNO_3-$Al(NO_3)_3$	15	7	114	
17	Diluted NaOH/6 M HNO_3-0.5 M CaF_2	3/5	2	35	

[a] These values may reflect a small, but noticeable, ^{238}Pu contamination.

[b] In each case, the residue from equilibrium A was the feed for the same number equilibration B.

[c] This equilibrium was conducted at room temperatures; all others were at elevated temperatures.

TABLE VI

EFFECT OF IGNITION ON ASH HEEL LOTS

Lot	LS-4429	LS-4625
Original color	Dull grey-black	Dull black
Ignition loss @ 700°C, wt%	11	35
Ignited product color	Red-brown	Red-brown
Color of HNO_3 solutions		
Original material	Dark brown	Black-brown
Ignited material	Pale green/tan	Pale green/tan

TABLE VII

SUMMARY OF LS-4625 ASH HEEL RECOVERY EXPERIMENTS USING CONCENTRATED AQUEOUS HF EQUILIBRATIONS FOLLOWED BY HNO_3-$AL(NO_3)_3$ DISSOLUTIONS

LS-4625 Run No.	ml conc HF / g ignited ash[a]	% Original Pu: HF Filtrate	% Original Pu: HNO_3-$Al(NO_3)_3$ Filtrate	% Original Pu: Dissolution Residue	g dissolutions residue x 100 / g initial sample	Wt% Pu in dissolution residue
4[b]	8.3	1	95.7	3.3	23 wt%[c]	0.3
11	5	0.5	90	9.5	35 wt%	0.6
12	2.5	0.5	93.6	5.9	29 wt%	0.4
13	10	0.8	90	9.2	28 wt%	0.7
14	5	1	96	3	32 wt%	0.2

[a] Ignited ash was 65 wt% of the original dried leached ash heel received from the CMB-11 Recovery Section.

[b] This run was held close to 100°C for 5.5 h of equilibration. All other runs were conducted for 18 h at room temperature.

[c] Based on the weight of ignited residue; all other values in this column are based on dried residue weights.

TABLE VIII

DRUMS OF ^{239}PU CONTAMINATED WASTE TO 20-YR RETRIEVABLE STORAGE FOR THE PERIOD OF JULY-DECEMBER 1975[a]

	LASL Residues		Off-Site Residues		
	Combustible	Non-Combustible	Combustible	Non-Combustible	Process Solids
No. of 115-ℓ barrels[b]	46	22	0	3	4
No. of 210-ℓ barrels	56	65	14	11	6
Total ^{239}Pu, g	124	371	97	199	823
Total wt, kg	1921	3106	353	519	360
g ^{239}Pu/kg ^{239}Pu residues	0.06	0.12	0.27	0.38	2.29

[b] Effective 10/31/75, 115-ℓ barrels were discontinued as outer containers for all ^{239}Pu retrievable waste. Only 210-ℓ DOT 17C barrels, which had passed the Rocky Flats Plant quality control testing, could be used.

TABLE IX

DRUMS OF ^{238}PU WASTE SENT TO 20-YR RETRIEVABLE STORAGE[a]

Number of 115-ℓ drums	34
Total ^{238}Pu, g	277.8
Total wt, kg	806.8
g ^{238}Pu/kg residues	0.34

[a] These data are from the Plutonium Processing Group only and not for all of LASL.

work is to identify those areas where chemical reactions, such as cellulose nitration, may be a source of heat and possibly fire.

The experiments conducted during this period are summarized in Table X. There appeared to be no significant heat buildup with dried nitrated cheesecloth in run 10-3. For wet nitration reactions, the time that elapsed between the start of storage, the time the reaction started, and the maximum temperature of reaction appeared to have a strong negative correlation with the ambient temperature of storage. This indicates that temporary storage at summer temperatures could precipitate a strongly exothermic reaction between HNO_3 and cellulose. A recent ERDA Safety Memo discussed possible similar reactions in HEPA filters sent to retrievable storage in Kentucky. The possibility of nitration heat triggering a rapid decomposition of adjacent dry nitrated cheesecloth was first investigated in run 16-5. Further work will determine if a fire can actually be produced by

TABLE X

HEAT ACCUMULATION IN CHEESECLOTH NITRATION REACTIONS

Run[a] No.	Time to Reach Max Temp, hr	Wet Nitrated Cheesecloth: Wt Before HNO_3 Added, g	g HNO_3 / g Cheesecloth	Temp, °C Max	Temp, °C Ambient	Dry Nitrated Cheesecloth[b]: Wt Before Nitration, g	Wt Gain Nitration, wt%	Temp, °C Max	Temp, °C Ambient	Storage Conditions
10-3	(No Temp increase)	----------	----------	---	-------	120	1	31°	31°	Stoppered Dewar in 31°C Water Bath
10-4	95	100	2	65	31	----------	----------	---	------	Same as 10-3
16-1	92	100	2.8	48	10-14	----------	----------	---	------	18-ℓ S.S. drum stored 0.6 m below grade
16-2	204	100	2.8	17	5	----------	----------	---	------	Centered in vermiculite filled 210-ℓ drum
16-3	7	100	2.6	92	29-32	----------	----------	---	------	18-ℓ S.S. drum stored in heated outer drum simulating summer temps
16-5	12.5	100	2.6	100-110	30±3°C	100	20-25	140	30±3°C	Same as 16-3

[a] The prefix number refers to the molarity of the HNO_3 used in the equipment.

[b] The cheesecloth was immersed for 4 h in HNO_3 and then "wrung out" and dried overnight in an open hood.

such a set of circumstances.

F. Loss of Hydrogen from Storage Containers and Simulated Burial

Recent work has confirmed that the consumption of hydrogen (H_2) by the MSA (Mine Safety Appliance) catalytic monitoring head was a large factor in preliminary determinations of H_2 loss from the 200-ℓ, 2.3-mm (90-mil) polyethylene drum liner and the 0.3-mm (2-mil) PVC plastic bag. Table XI compares correct H_2 loss by diffusion with the original continuous run MSA values. The last semiannual report characterized the loss of H_2 from an earth (tuff) covered pit constructed in solid tuff. These data were correct, since the loss of H_2 by diffusion was so rapid that its consumption by the MSA catalytic head was judged to be negligible.

G. Assay Instrumentation

The automated, computerized, box counter has been moved from the CMB-11 counting room into a separate area that has a 30% lower nominal background rate and is isolated from the large (factor of 10) fluctuations in background experienced when barrels of high-level, plutonium-bearing material are moved into the counting room. The lower background has resulted in a 200% improvement in amplifier stability.

The computer analysis codes for improving the reliability of the NaI (Tℓ) system, incorporating automatic operator diagnostics, and permitting the inclusion of a Ge detector in the system have been updated. The changes will be introduced without disrupting the routine assay schedule at DP-Site.

TABLE XI

DETERMINATION OF H_2 LOSS FROM WASTE CONTAINER MATERIALS
- COMPARISON OF SPOT SAMPLING WITH CONTINUOUS MSA MONITORING -

a. Determination of H_2 loss by diffusion at 22-28°C from a 200-ℓ 2.3-mm (90-mil) polyethylene drum liner at atmospheric pressure.

Time, minutes / Type of sampling	% H_2 concentration present at 1000 min after original injection of 2-3.5 vol% H_2					
	2000	3000	4000	6000	8000	10 000
Mass spec values, spot sampling[a]	98%	93%	88%	--	--	--
MSA values, spot sampling[a]	97%	93%	89%	78%	73%	70%
MSA values, continuous sampling, composite of runs 1 and 4[b]	75%	60%	43%	--	--	--

b. Determination of H_2 loss by diffusion at 22-28°C from a 0.3-mm (12-mil) heat-sealed PVC plastic bag at atmospheric pressure.

Time, minutes / Type of sampling	% H_2 concentration present at 1000 min after original injection of 2-3.5 vol% H_2		
	2000	3000	4000
Mass spec values, spot sampling[a]	93%	89%	86%[c]
MSA values, spot sampling[a]	92%	87%	84%[c]
MSA values, continuous sampling, composite of runs 1 and 4[b]	58%	--	--

[a] These values reflect the loss of H_2 by diffusion.

[b] These values reflect the sum of diffusion loss and loss by consumption of H_2 due to the continuously operating MSA catalytic head.

[c] Extrapolated values.

A large-area, planar, intrinsic germanium detector (1000 mm^2 by 12 mm thick) was purchased to supplement the NaI detector. The high resolution of this detector (568 eV at 6.4 keV, and 720 eV at 122 keV) will be used to identify the isotopic content of boxes having activity greater than the 10-nCi/g limit. The low energy efficiency of the Ge detector has been measured to be consistent with the expected behavior: E(14.4 keV) = 87.4%, E(84 keV) = 99.6%. Counting rates in the detector positioned 43 cm from an 88-μg plutonium source (predominately ^{239}Pu) are approximately 1/s for the 13.6-keV line and 1.5/s for the 17.2 keV line. A 1000-s counting time was used to acquire sufficient statistics from this low-level source. The transmissions for a "typical" trash box at energies of 13.6, 17.2, and 59.5 keV were measured to be 0.48, 0.68, and 0.87. However, these values could vary considerably with the matrix density of individual boxes.

Preliminary work began on investigating the 4-π neutron barrel counter located at DP-Site as a possible low-level waste measuring instrument for high density materials where gamma- and x-ray signatures are not possible. The addition of a 10-cm lead shield between the sample and the neutron counters could make the device suitable for measurement of low levels of plutonium in the presence of hot radioactive wastes generated in spent fuel reprocessing. The neutron assay is based on coincidence counting from spontaneous fissions (^{240}Pu or ^{242}Pu) or from the total neutron counting rate associated with spontaneous fission and (α,n) reactions.

For the spontaneous fission coincidence measurements, counting rates are low--4.1 X 10^2 and 7.7 X 10^2 fissions/s/g for ^{240}Pu and ^{242}Pu. A knowledge of computing backgrounds is essential. Backgrounds produced primarily by cosmic-ray showers in a simulated lead or iron liner were measured. The results, expressed as equivalent grams of plutonium (6% ^{240}Pu) per 100 kg of material, were 2.86 ±0.04, and 0.53 ±0.02 for lead and iron at 2200-m altitude. Thus, measurements with a sensitivity of about 1 g of plutonium in a 210-ℓ drum should be possible. At sea level, where the cosmic-ray background is a factor of 4 lower than at Los Alamos, the technique should be more sensitive. The addition of cosmic-ray veto counters may also lower this troublesome background. As measurement of total neutron flux is affected by the chemical composition of the sample, as well as by the isotopic composition and backgrounds, the errors with this type of measurement will be biased on the high side. Hence, the total neutron flux measurement may be valuable as a go/no-go indicator for disposal.

The manual for the box counter system, "Computerized Low-Level Waste Assay System Operation Manual," LA-6202-M, has been published and is available for circulation.

II. TRANSURANIC-CONTAMINATED SOLID WASTE TREATMENT DEVELOPMENT PROGRAM (L. C. Borduin, W. E. Draper, R. A. Koenig, A. S. Neuls, C. L. Warner)

The objective of this program is the development and demonstration of alternative production-scale volume reduction processes for TRU-contaminated solid waste. Achievement of this goal is currently focused in three major efforts: (1) construction of the new Treatment Development Facility (TDF); (2) final design and procurement of the initial waste treatment process (controlled-air incineration); and (3) experimental operation of pre-facility study equipment. Beneficial occupancy of the TDF is expected for mid-April 1976. Final assembly of the initial process is scheduled for completion July 1976. Experimental studies continue to produce valuable data requisite for specification of process train components.

A. Facility Development

Construction of the TDF, which will house the waste reduction systems, was 72% complete as of 31 December 1975. Progress remained essentially on schedule for this reporting period.

The single major design change following construction contract release was the substitution of concentric runs of plastic (PVC) pipe, instead of the originally proposed Duriron pipe, for the facility acid waste collection system. This modification will provide positive leak detection and prevent possible contamination of the building subsoil. In addition, all drain traps, piping junctions, and bends have been housed in pits with removable steel covers to permit periodic inspection of drain-line integrity. A process trench to house pipe runs associated with on-going development studies was included as part of this contract revision. The covered trench provides a safe location, as well as accessibility, to facilitate modifications normally required during development efforts.

Construction activity on the building exterior is essentially complete. Remaining tasks include completing ventilation ducting, ceiling panel installation, boiler and water-conditioning system assembly, finishing plumbing and electrical fixture installation, and finishing interior painting and carpeting.

Leveling and fencing the utility yard southwest of the building is presently being considered for completion with remaining contingency funds. This will serve to provide a weather barrier for extended operation and testing of the auxiliary generator, cooling towers, and HEPA filter, as well as define and visually screen a storage area for large process equipment and piping.

B. Process Design

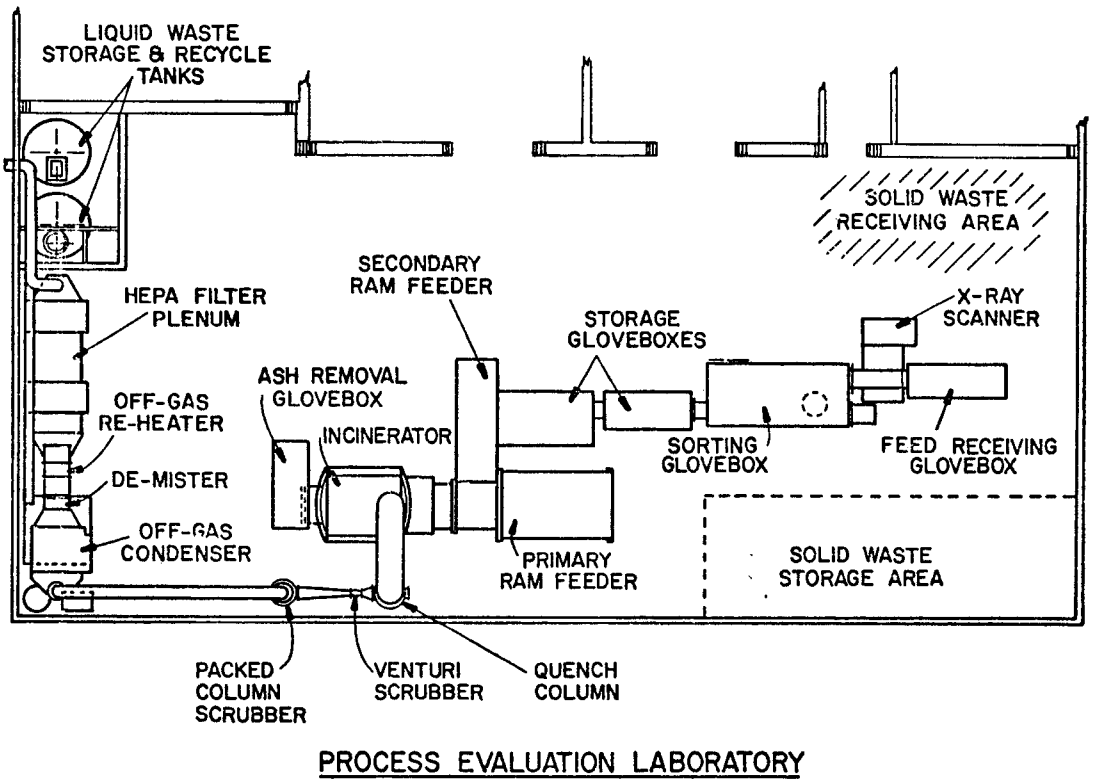
The initial TDF solid waste treatment process draws heavily on conventional incineration and air pollution technology. The conceptual design for the incinerator and off-gas system was included in LA-6100-PR[1] and remains essentially unchanged. The system is designed around a controlled-air incinerator, with a high-energy aqueous scrub system coupled with HEPA filters to provide off-gas cleanup. Feed preparation is performed in a series of stainless steel gloveboxes designed to meet LASL specifications for plutonium service.

Process design activity at the end of this reporting period focused on finalizing the configuration of the off-gas condenser/reheater/HEPA-filter enclosure and support module; the last of the major component design efforts. The secondary ram feeder, a mechanism for transferring wastes from the storage gloveboxes to the incinerator ram feeder, has been designed. Contamination-control enclosures for the ram feeders and the combustion-gas supply system are in process. Exterior dimensions of all major process components and enclosures have been fixed to permit equipment layout. After reviewing several possibilities, the equipment arrangement shown in Fig. 2 was selected. The process and instrumentation diagram was reviewed and finalized to permit specification of individual piping and instrument components.

Purchase requisitions for all major vendor-supplied components have been issued. Items include the off-gas condenser, induced draft process exhaust fan, scrub-solution receiver tanks, recycle-stream heat exchanger, and scrub-solution pumps. Off-gas system components, including the quench chamber, venturi scrubber, and packed-column scrubber, were received from the John Zink Company. Process instrumentation specifications are 75% complete; procurement is initiated as the individual components are identified.

C. Pre-Facility Studies

Pre-facility studies were conducted in a plywood sorting glovebox mockup to assess suitability of selected materials handling



PROCESS EVALUATION LABORATORY

Fig. 2. Planned equipment layout, controlled-air incinerator system.

devices. The proposed method for cutting up tramp metal proved to be inadequate, and the glass bottle crusher was difficult to operate in the glovebox enclosure. Resultantly, a small power hack saw was purchased and modified for glovebox installation, the glass crusher was modified to a more compact form, and a shard receiver compartment was included for safer handling of crushed glass. The bulk materials compactor tested satisfactorily.

The incinerator was operated to establish shutdown procedures in the event of utility failure. A dual-purpose experiment was devised to determine combustion characteristics in the event of fuel gas pressure loss and whether burner shutdown would be a suitable measure in the event of water pressure failure. Immediately after introducing a new charge, both fuel gas burners were shut down. Burning continued at a steadily diminishing rate until temperatures in both chambers reached subignition levels. No visible stack emissions were observed, and exit temperatures after 1 h were below 540°C--the upper temperature limit for the off-gas system. The emergency quench water system has thus been deemed adequate and, should water pressure failure occur, burner shutdown with all other systems continuing in operation is an acceptable system shutdown procedure.

The Hastelloy quench weir, which provides initial contact between hot combustion gases and quench solutions, was tested to determine liquid distribution, flow rates, and associated pressure drops. Observation and data indicate that the weir performs adequately. Inspection of the other scrubbing equipment showed all to be in compliance with design specifications and in good repair. Testing of the complete off-gas system is awaiting installation in the TDF high bay.

Vendor liquid-filter designs for particulate removal from the scrub solution recycle have been reviewed, and a unit has been ordered from Commercial Filters, Inc., for development study. Test objectives include determining filter

cartridge changeout requirements, porosity requirements, pressure drop data, and mechanical reliability.

D. Miscellaneous

A "mini-189" proposal outlining study programs for processing LWR fuel-cycle waste was forwarded to the Savannah River Operations Office for review. Controlled-air incineration technology is promising as a volume reduction method for low- and intermediate-level fuel reprocessing wastes. Should such a study be funded, study objectives for irradiated and non-irradiated transuranic wastes appear entirely compatible, with the possible exception of the feed preparation train.

III. EVALUATION OF TRANSURANIC-CONTAMINATED RADIOACTIVE WASTE DISPOSAL AREAS

(A. F. Gallegos, L. J. Johnson, W. J. Smith, M. L. Wheeler, W. J. Whitty)

In August 1975, LASL was requested by the Nuclear Fuel Cycle and Production Division of ERDA to prepare a section on radioactive waste burial for the Technical Alternatives Document on the Management of Radioactive Waste from the Light-Water Reactor Fuel Cycle. Project personnel from the evaluation of waste disposal areas program were selected to prepare the waste burial section.

Radioactive waste in all categories except high-level waste from fuel reprocessing is presently buried at six commercial burial sites in the United States and five major ERDA facilities. These facilities are expected to continue operation for the foreseeable future. Our task was to describe past and present practices at these sites and to identify technology which exists or which could be developed to improve the containment at these burial sites. The purpose of this effort was to provide a large background of experience on which to base suggested modifications.

The objective of shallow earth burial is the isolation of undesirable waste materials from man and his environment. The extent to which a burial ground provides this isolation is thus a direct measure of its acceptability. Radionuclide containment is affected by various means which fall into three general categories--waste form, engineered containment measures within or adjacent to the burial trench, and the natural system surrounding the burial site. Examination of present practices at both ERDA and commercial sites indicated that primary reliance is generally placed on the natural system for containment, and instances of radionuclide migration into man's environment frequently resulted from the inadequacy of this system to provide complete containment.

The information accumulated for this report has brought to light many areas in which engineering or waste form modification could be used to augment the natural containment mechanisms. The completed alternatives document, incorporating the section on land burial, was published as ERDA 76-43, in May 1976.

A. Abiotic and Biotic Transport of Transuranics

Efforts have continued to develop and refine previously described modules[1,2] involving biological transport of plutonium and other transuranics (TRU). New submodules have been developed and tested as integral parts of the general model for risk analysis.[2] This report is limited to a discussion of abiotic, plant, and herbivore modules. In addition, progress in the development of interregional modeling aspects is described, including present output capabilities and applications.

1. Abiotic Module

The portions of the module which have been revised and extended include soil-water aspect, root distribution, transuranic root uptake, solar radiation aspect, resuspension of transuranics, and new data base considerations. Soil-water availability was considered previously to be based on the top 250 mm of soil without regard

to shallower or deeper soils in any given region.[1] Also, the total root biomass had been considered exclusively at this depth for purposes of plant growth. Soil water now takes into account the soil depth which has been added to the data base for a given region. A soil-water factor,

$$SF = 0.632/(1 - \exp - 0.00450SD), \quad (1)$$

where SF = soil Factor and SD = soil depth, mm, SD $\geq$ 2 mm, is calculated to adjust the total amount of water removed via evapotranspiration;[1] the relationship was approximated from evapotranspiration studies of conifers.[3] The factor varies from 79 at 2-mm soil depths to 0.632 at greater depths; it equals 1 at 250 mm in accordance with Gilbert.[1] Hence, soils deeper than 250 mm can enhance soil water availability by as much as 60%, and shallower soils can deplete it to about 1% of that available at this depth.

Root biomass distributions for varying soil horizons can be calculated in accordance with Eq. (1). The module now considers 33 soil horizons that consist of a top 20-mm horizon and 32 250-mm horizons. This makes it possible to test different activity configurations for transuranic soil activities including those at LASL waste disposal sites. The model also provides for root distribution from card input.

The uptake coefficient for transuranics by roots has been modified to correspond with the evapotranspiration scheme used in the module, and with a partition coefficient between $^{238}PuO_2$ microspheres ($\sim$150-μm diam) and water derived from an experiment by Adams.[4] The partition coefficient K_d determined in this manner ($\sim 1 \times 10^{-11}$ at pH 8.2) was tuned to 4×10^{-9} to be in agreement with a root uptake coefficient derived by Martin[6] from Romney,[7] and modified for use in the previous scheme.[1] Unfortunately, experiments at other pH values were not found in the literature to support the tuned K_d; however, the estimated value seems justified on the basis of pH variability around root hairs and mycorrhizae, and the possible presence of organic chelators in the soil. Different uptake coefficients can be included in the model for different soil horizons by card input. The solar radiation function used in the model for plant growth[1] has been modified to saturate its effect on growth at 450 Langleys (LY/day) in accordance with IBP studies,[8] which brings this model more in agreement with the results of Gilbert[1] and photosynthetic light efficiencies. The change was made possible through formulation of a new water utilization scheme which is described later in this report.

The resuspension of particulates containing transuranics was modified to account for weathering effects and adjustment for ground cover by plants. The previous scheme assumed a constant resuspension factor of 1.2×10^{-8}/day from the top 2 cm of soil similar to the model of Martin.[6] The new scheme adjusts the resuspension rate for average annual plant crown cover (yearly running average for woody plants) by the following relationship.

$$RS = 2.5 \times 10^{-8} \times \exp(-0.046CCV), \quad (2)$$

where RS = resuspension factor, day^{-1}, and CCV = crown cover fraction. The resuspension factor ranges between 2.5×10^{-8} to 2.51×10^{-10}/day between bare ground and total crown cover of 100%. The value at about 16% ground cover corresponds to the 1.2×10^{-8}/day used previously. Resuspension has been further modified to respond to a weathering effect that has a half-life of 70 yr as described by Langham;[9] however, above-ground produced humus not weathered until it has become part of the soil matrix is considered resuspendable in the model. The weathered activity is transported to the top 250-mm soil horizon.

Our earlier plant model accounted for deposition variations on plant surfaces, assuming a leaf area index,

LAI = 1,

$$\frac{M^2 \text{ leaf surface}}{M^2 \text{ soil surface}}$$

with precipitation events as outlined by Wilson.[10] This scheme has been modified to allow for small precipitation events by using the following ingrowth factor.

$$IF = 1 - \exp(-0.6518P),$$

where IF = ingrowth factor and P = precipitation in mm water equivalent. The optimum size for a precipitation event to produce a maximum effect occurs at about 8 mm; thereafter, larger precipitation events do not affect the amount of deposition as a result of atmospheric scrubbing, nor increase the amount of particulates weathered from a plant surface. The same factor is used to increase the weathering of particulates from the surface to the soil horizon below 2 cm by a factor of 10 above that occurring with a half-life of 70 yr. Resuspension is not considered during precipitation events, which occur about 13% of the time in this model, for the climatic pattern of the study area.[1]

The data base for operation of the model on an interregional basis is also included within the abiotic module, as reported earlier.[2] This includes regional environmental characteristics,[1] and additional information pertaining to agricultural activities. Most of this information has now been recorded in the proper format for incorporation into the regional model and is awaiting completion of other segments of the interregional computer code.

2. Plant Module

Root uptake of transuranics by plants was treated in an earlier model version[1,2] to be a constant (1.1×10^{-5}/g) for a given fine-root biomass, without regard to transpiration variations throughout the year. In addition, only one soil horizon (top 250 mm) was considered for uptake and one uptake coefficient. The new scheme utilizes the following equation for a given calendar date.

$$RU_i = \sum_{j=2}^{32} FR_j \cdot U_j \cdot SA_j \cdot E_i \cdot N_i, \quad (4)$$

where i = day number 1, 2, ...365;

j = soil horizon (250 mm) 2,3,4,...n;

RU_i = root activity uptake, activity/m^2 on i^{th} day;

FR_j = fraction of roots in j^{th} soil horizon;

U_j = partition coefficient in j^{th} horizon;

SA_j = soil-specific activity, activity/g on j^{th} horizon;

E_i = evapotranspiration rate, mℓ/m^2 on i^{th} day;

N_i = needle biomass, g/m^2 on i^{th} day.

Since the LAI is proportional to leaf biomass, transuranic uptake is directly related to the total transpiration stream of a given plant as described for conifers by Running et al.[3] Leaf biomass is calculated by the following relationship.

$$N = 0.928\ B \exp(-0.00311B) + 0.082B, \quad (5)$$

where N = leaf biomass, g/m^2; and B = aboveground biomass, g/m^2. Our previous modeling scheme assumed that the leaf fraction for all woody plants was equal to the second term of Eq. (5), where the total aboveground biomass was assumed as the leaf biomass as in Gilbert.[1] Our new scheme shows a gradual transition from an herbaceous to a woody plant depending on above-ground biomass accumulation (see Fig. 3). Most of the change occurs between 1-2000 g/m^2 of

the latter which is in the range of some vegetables and many woody shrubs; above 2000 g/m^2, the leaf biomass is once again approximated by the second term of the equation as before.

In the last progress report[2] mention was made of a number of uncertainties involved in the use of single water utilization efficiency (mℓ of water required per gram of photosynthate synthesized) throughout the year because the diversity of moisture-temperature interactions would certainly cause some variations.[12] Also, it was not appropriate to use the phenological water utilization scheme of Gilbert[5] for plants other than native grasses and related vegetable plants. A new strategy was to develop a dynamic scheme to account for moisture-temperature interaction effects on this parameter throughout the year. The following general relationship was developed.

$$WUT_j = WUTM \times EOT_j^{-1} \times ESW_j^{-1}, \qquad (6)$$

where j = day number, 1,2,3,....365;

WUT_j = water utilization efficiency on j^{th} day mℓ/g;

$WUTM$ = maximum water utilization efficiency, mℓ/g;

EOT_j = effect of temperature on photosynthesis on j^{th} day, $0 \leq EOT_j \geq 1$, fraction; and

ESW_j = effect of soil water on photosynthesis on j^{th} day, $0 \leq ESW_j \leq 1$, fraction.

The effect of temperature and soil water on photosynthesis was defined for warm- and cool-season plants in an earlier report.[1] The maximum efficiency for grasses and herbaceous plants was set equal to the maximum of Gilbert[5] (350 mℓ/g photosynthate). The value for evergreen trees and shrubs was set to 750 mℓ/g photosynthate, the min-

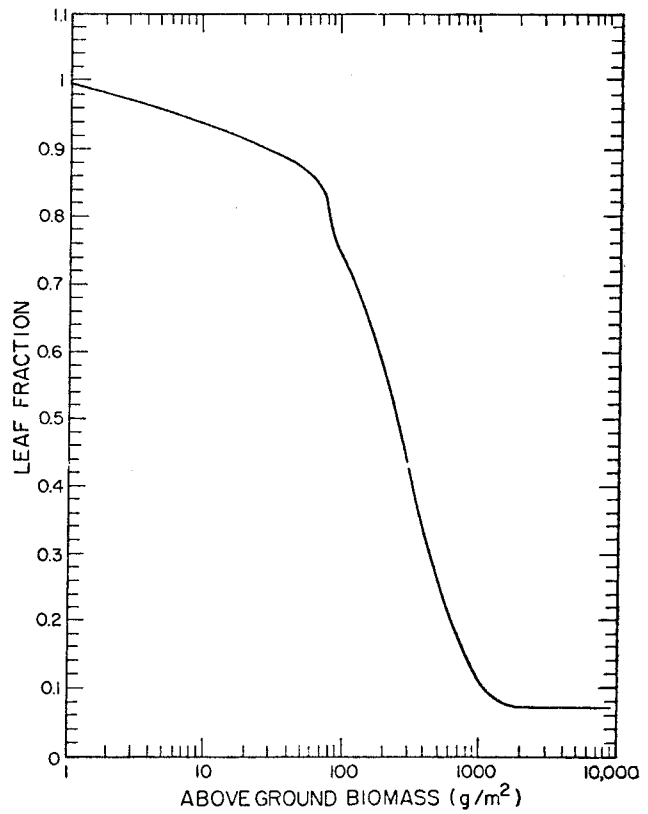


Fig. 3. Plant leaf biomass.

imum efficiency for grasses using the phenological equation derived by Gilbert and previously used in this model. Hence, any combination of temperature-soil moisture conditions which reduce efficiencies, such as cold dry periods, is represented in the model down to 1% of the maximum efficiency.

In addition to leaf biomass, living biomass in stems and roots requires photosynthate for respiratory needs. The growth rate of a plant is highly dependent on its maintenance loading through time. Respiration of roots, stems, and leaves is still considered separately for grasses. Maintenance requirements of conifers on the basis of sapwood thickness, as considered in the last report,[2] are related to water storage capacity and utilization.[13] After experimentation with several strategies for modeling maintenance requirements, an approach was selected which allows tuning of plant growth by adjusting maintenance loads of various plants. The model was tuned by embodying the maintenance load

into the overall photosynthesis equation[1] to obtain a new relationship.

$$PS_j = N_j \cdot EOT_j \cdot ESW_j \cdot PSM_j - (N_j + f \cdot N_j) \cdot R_j, \qquad (7)$$

where

PS_j = net biomass production g/m^2 on j^{th} day;

N_j = leaf biomass, g/m^2 on j^{th} day;

PSM_j = maximum photosynthetic rate possible, g biomass photosynthate/g leaf biomass on j^{th} day;[1]

R_j = respiratory fraction on j^{th} day;[2]

and

f = maintenance factor.

The maintenance factor has been tuned to a maximum of 6 for pinon-juniper, 3 for ponderosa, about 2 for Douglas fir, and approaches 1 for some deciduous trees. Tuning this factor for plants other than ponderosa pine and pinon-juniper stands is still in progress.

The exchange of transuranics between above- and below-plant biomasses has been modified to depend on daily photosynthate status rather than on an assumed respiration rate as considered previously.[1] This new scheme allows exchange through phloem flow in response to photosynthate requirements by the given plant, assuming exchange with xylem elements. Since the specific activity of transuranics is separate for root and above-ground biomass in the model, a phloem flow of equal amount in either direction may in fact result in a net flow of transuranics in one direction, depending on the relative transuranic specific activity of these compartments. The model thus allows transuranics entering the root compartment to translocate to above-ground parts by the following relationship.

$$TR_j = \frac{|PS_j|}{B_j} \cdot \left[1.0 - \frac{|PS_j|}{B_j}\right] \cdot \frac{B_j}{(B_j+R_j)} \cdot RA_j, (TB_j, RB_j); \qquad (8)$$

where

TR_j = activity transferred from below-ground to above-ground biomass, activity/m^2 on j^{th} day;

B_j = above-ground biomass g/m^2 on j^{th} day;

R_j = below-ground biomass g/m^2 on j^{th} day;

RA_j = activity of below-ground biomass, activity/m^2;

BA_j = activity of above-ground biomass, activity/m^2, on j^{th} day;

TB_j = activity transferred from above-ground to below-ground biomass, activity/m^2, on j^{th} day; and

RB_j = activity of above-ground biomass, activity/m^2 on j^{th} day.

The value of TB_j can be calculated by substituting RB_j in the equation and R_j for B_j in the previous factor of the equation. Thus variable amounts of phloem are exchanged between compartments in response to both positive and negative values of net biomass production. Transuranic activity associated with movement of water in the xylem vessels is assumed to equilibrate with phloem elements along their course and to be a portion of the upward movement of transuranics (TR_j) in the model as noted earlier. The second term on the right side of Eq. (8) becomes important for juvenile stages of plants and, in particular, perennials where above-ground biomass is comparable to $|PS_j|$ during the off-season; it does not permit excessively high specific activities during this period. The third term of Eq. (8) is important for perennial grasses as a mediator representing slowing and stopping movement of transuranics

to crowns during the off-season. It also allows for storage of transuranics in roots by the same mechanism.

Crown cover determinations for herbaceous and woody plants are based on a specific plant type net annual productivity (see Fig. 4) by the following relationship.

$$CV = 1 - \exp(-0.00154PN),$$

where CV = fraction crown cover and PN = net annual production g/m^2. The equation is used directly for calculating crown cover for grasses and other herbaceous plants; however, above about 2000 g/m^2 for woody plants in general, crown cover is calculated from a net yearly production running average rather than from current values which may be quite small or even negative during years with adverse growing conditions. Also, the crown diameter for above-ground biomass estimates exceeding 2000 g/m^2 must be adjusted for the shape of the plant and is more accurately determined by relating the stem diameter and the crown area rather than by Eq. (9). This topic will be considered in more detail later in this report.

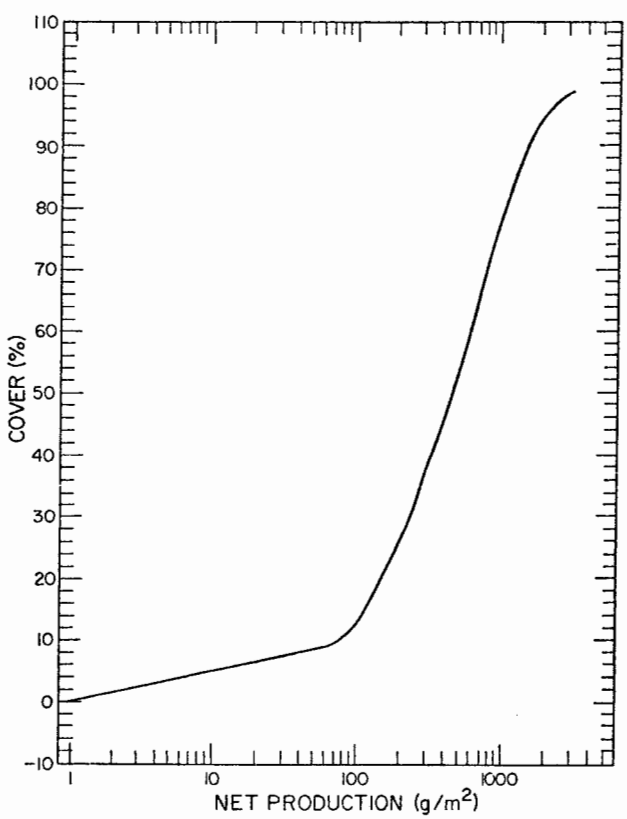


Fig. 4. Percentage crown cover.

Several compartments in addition to those already outlined[1] were added to the plant module to account for below-ground behavior of transuranics more effectively. These include below-ground dead, litter, and humus compartments as companions to their above-ground produced counterparts. The addition of these compartments makes it possible to trace transuranic cycling and redeposition in various soil horizons as described earlier.

Plant Submodules

The plant submodules are categorized within the plant module based on growth strategy differences for native plants, or on agricultural use. They are divided into forage, agricultural crops, and tree and shrub types. The forage submodule includes the following plants, of both warm- and cool-season types: annual grasses, perennial grasses, and irrigated natural pasture grasses. The basic growth strategies for these plants remain unchanged[1,2] with the exception of those changes previously mentioned.

The agricultural crops submodule includes corn and sorghum, warm and cool season annual vegetables and grains, fruit trees, and alfalfa.

The growth strategies vary considerably, depending on type of crop. Corn and sorghum are modeled to be twice as efficient in water utilization under irrigation as other vegetables and grains;[2] alfalfa was modeled as a cool-season perennial plant. Fruit trees are treated under the trees and shrubs designation. In all cases involving agricultural crops, planting schedules, irrigation schedules, and amount of water applied are taken from the data base included in the abiotic module. Most of the latter has been computer coded for storage on an interregional basis.

model thus sets a quasi-equilibrium biomass for leaves based on the potential of the environment to support the maintenance requirements of the plants in question. It is important to note that production for forests during a given year may be quite small or even negative under adverse climatic conditions.

The maintenance requirements of plants were considered in an earlier section of this report. For tuning purposes, the respiring biomass of woody plants has been modeled as multiples of the leaf biomass at any given time. It would appear from the outcome of growth simulations in this model that the advantage gained by plants that can survive under adverse climatic conditions, such as drought and frigid weather, is partly offset by larger maintenance requirements relative to leaf biomass. A good example is the difference in modeled maintenance requirements between pinon-juniper associations and those of Ponderosa pine, as noted previously.

In addition to differences in growth strategies between trees and shrubs and other plant types, the former also assume a specific structure, and a given above-ground biomass larger than 2000 g/m^2 will assume a distribution of plants based on stem diameter. For above-ground biomasses larger than 4000 g/m^2, the "forest" may acquire economic importance other than firewood use and be subject to timber management schemes such as those followed by the U. S. Forest Service. Since minerals including transuranics are stored in woody tissue for ultimate recycling via humus, it is important to model the fate of transuranics in wood which are transported concomitantly with wood for either fuel or home-building activities. In the event of fire, most of the minerals in wood would deposit at the source as resuspendable particulates, and a small fraction (ca..10%) would be transported via air circulation for deposition on land surfaces and in the respiratory tract of organisms. Pollen release is also a potential vector of transuranics to organisms via resuspension and inhalation.

The beginnings of a forest structure module based on a log-normal distribution of forest volume was presented in a previous report.[2] The model gave encouraging results for undistributed and unmanaged forest stands, although not in a dynamic sense; it was, however, fairly representative of young stands (ca...100-2000 g/m^2). What is required is a module which can respond to catastrophic events such as fire, adverse climate, and timber management schemes such as those of Larson.[17] There are two modules now under testing which attempt to meet these objectives for tracking transuranics, a forest structure submodule and a forest management submodule. The former includes the dynamics of diameter class evolution in 2-in. intervals, while the latter includes the modeling of events other than climatic events which would alter the diameter class structure of the forest stand in question. Only a brief description of these modules will be presented.

The forest structure submodule has been developed as a hybrid between a log-normal distribution generator for the 0-2-in. diam class and a compartmental model for other class intervals: 2-4, 4-6, 6-8, 8-10...n-n+2, in. diameters (see Fig. 5). The log-normal distribution equation[2] has been modified to include an additional term.[18]

$$f(M) = \frac{1}{\sqrt{2\pi}\ \ell n\sigma} \exp\left\{-\left[\ell\ \frac{(M-M_o)\,(M_\infty-M_o)}{\dfrac{(M_\infty-M)\ \bar{M}}{\sqrt{2}\ \ell n\sigma}}\right]^2\right\}, \tag{13}$$

where, as before,[2]

M = DBH of tree category, inches;

M_o = minimum DBH observable, inches;

M_∞ = maximum DBH observable, inches;

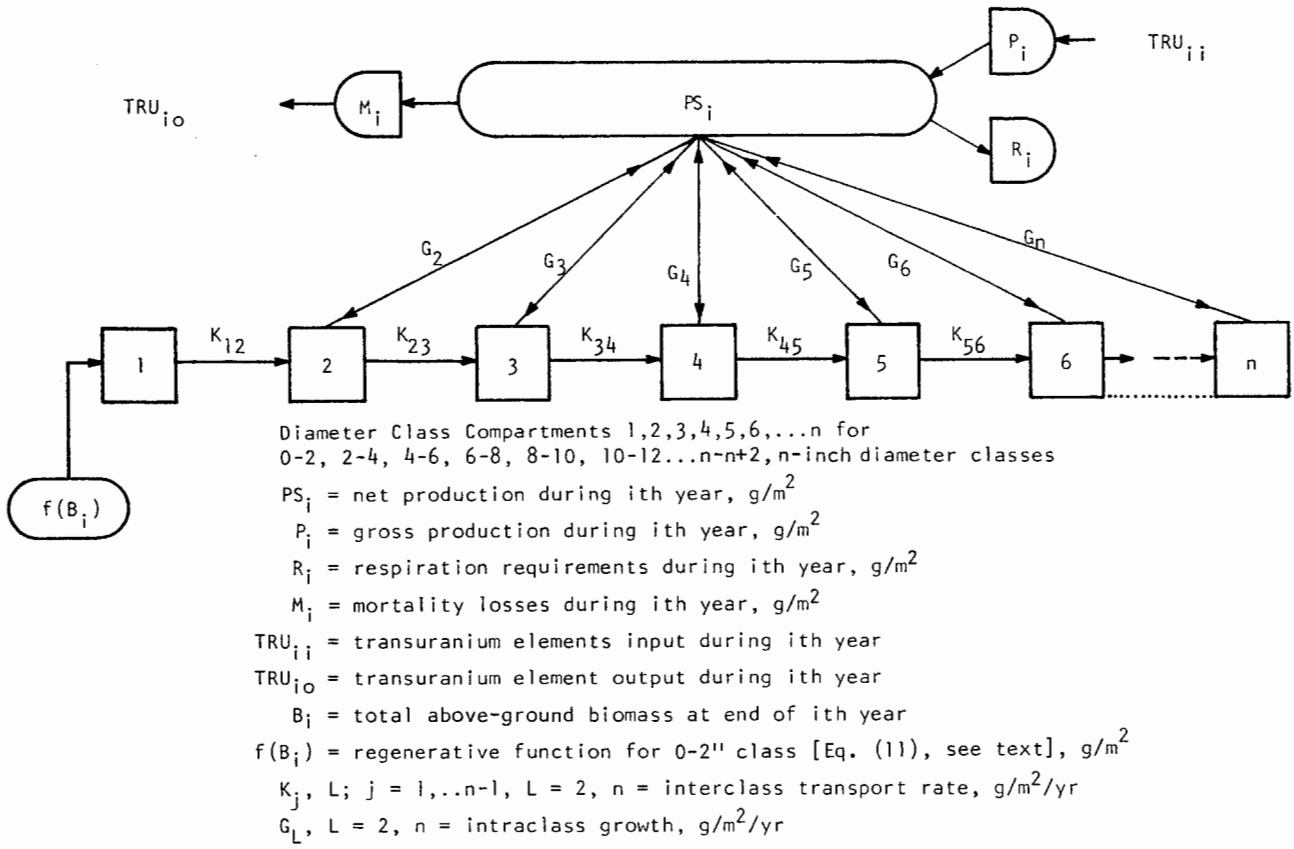


Fig. 5. Biomass and TRU allocation in forest with N diameter classes.

$\bar{M}$ = geometric standard deviation of tree stand, inches; and

σ = geometric standard deviation of stand, inches.

The geometric σ was determined by tuning to equal exp (1) while other values derived by similar procedures were

$$M_o = M/\sigma^{3.3},\ M_\infty = M\sigma^{1.2},\ \text{and}$$

$$\bar{M} = 0.00118\ B_i,$$

where B_i = above-ground biomass, g/m^2, at the end of the ith year. The number of trees in this regenerative class were derived from class and individual tree volume considerations.[2] The log-normal portion of the model thus responds to changes in above-ground biomass, B_i, and serves as a link between the forest structure submodule and the plant submodule.

The compartmental portion of the forest structure submodule is highly dependent on two important transport phenomena, intraclass growth and interclass transport. These two factors were determined in three steps:

1. tuning the compartment model structure generator with the log-normal distribution generator for all diameter classes,[2] using constant positive growth increments;
2. interfacing the dynamic growth characteristics of the plant module, allowing unequal growth increments of either sign to perturb the model; and
3. interfacing the forest management submodule to allow perturbations of greater magnitude, such as forest fires or timber cuts.

Interclass growth was found to be highly sensitive to two relationships related to crown volume and tree diameter at breast height.

$$VF_i = \Pi(1.597+1.013\ M_i/2)^2 \times 90 \times (M_i/24)^{0.094} \times T_i, \quad (14)$$

$$DF_i = (1 - \exp(-0.693\ M_i/M_{j\infty})\ )^3; \quad (15)$$

where

VF_i = volume factors, ft^3 of ith diameter class;

M_i = diameter at breast height, inches of ith diameter class;

T_i = number of trees of ith diameter class;

DF_i = diameter factor for i^{th} diameter class; and

MJ_∞ = maximum diameter for j^{th} year.

The relationship for crown area, and the height are derived from Larson.[17] Biomass resulting from net annual production, PS_j, is allocated according to the following relationship.

$$G_L = \frac{PS_j \times VF_L \times DF_L}{\sum_{i=2}^{n} VF_i \times DF_i}, \quad L = 2,n;\ i = 2,n \text{ (diameter class)} \quad (16)$$

where G_L = intraclass growth for L^{th} diameter class. Both negative and positive values of PS_j are treated identically; transuranics are also allocated in this manner.

Interclass transports are based primarily on previous-year-realized growth increments for all classes. Negative growth increments are related to mortality losses and, hence, involve tuned curve shaping techniques that simulate a forest structure using a log-normal distribution generator.[2] In addition, interclass transport was limited to values less than or equal to the corresponding intraclass growth estimates. Both intraclass and interclass processes are still being refined with the hope of simplifying and gaining a better understanding of the relationships.

The forest management submodule is a collection of cutting strategies which remove selected diameter classes and adjust the above ground biomass estimate and forest structure for subsequent year growth. The options now included within this module are: choice of cut [any diameter(s)], sawtimber (12-40 in. diam), pulpwood (6-10 in.), precommercial (1-4 in.), clear (all diameters), reverse-J cut,[17] and forest fire (selected diameters). All cutting or removal options maintain a material balance of transuranics as in the submodule previously described. An example of forest management operation is presented in Fig. 6 where a Ponderosa pine forest (LASL, Region 49,[1] 25 in. ppt) is compared with an unmanaged stand. The reverse-J cut option was exercised every 40 yr during the 300-yr simulation period. The results of the simulation

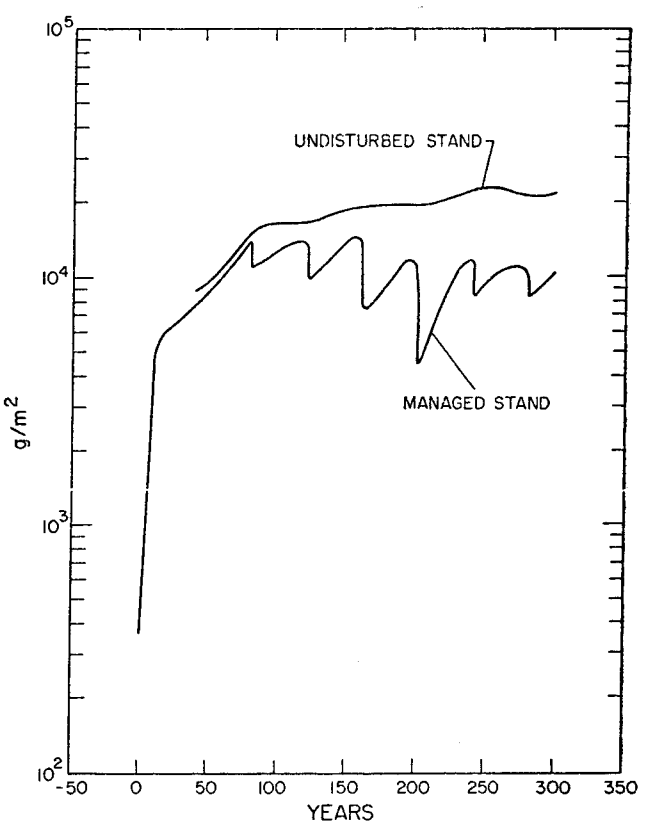


Fig. 6. Ponderosa Forest biomass.

show that by management an excess of 13 675 g/m^2, or about 1928 ft^3, was harvested over the unmanaged forest. In addition, 25 025 g/m^2 (3528 ft^3) of wood containment transuranics were removed from the forest during the simulation period under management.

The structure of the unmanaged forest previously discussed is presented in Fig. 7. The simulation indicates a transfer of wood volume inventory to larger diameters until an optimum tree size is reached, then declining to older larger trees; the simulation is given in volume/acre. Finally, the relative number of trees/acre of any given diameter can be estimated by comparison of Figs. 7 and 8. The volume estimates for trees of a set diameter were obtained by tuning with Beaver Creek data.[18] Efforts are now under way to obtain similar estimates for other conifer types.

3. Herbivore Module

The herbivore module has not received as much attention as the previously described modules and submodules as most of the effort in this regard has been devoted to obtaining the proper format for coding all of the livestock information gathered on cow-calf operations and movement, and human consumption estimates on an intra-and interregional basis. Most of these data are now coded in proper form. A food preference scheme for domestic livestock and deer is now being devised but most of this will be considered after satisfactory operation of a plant-succession submodule now in the planning stage.

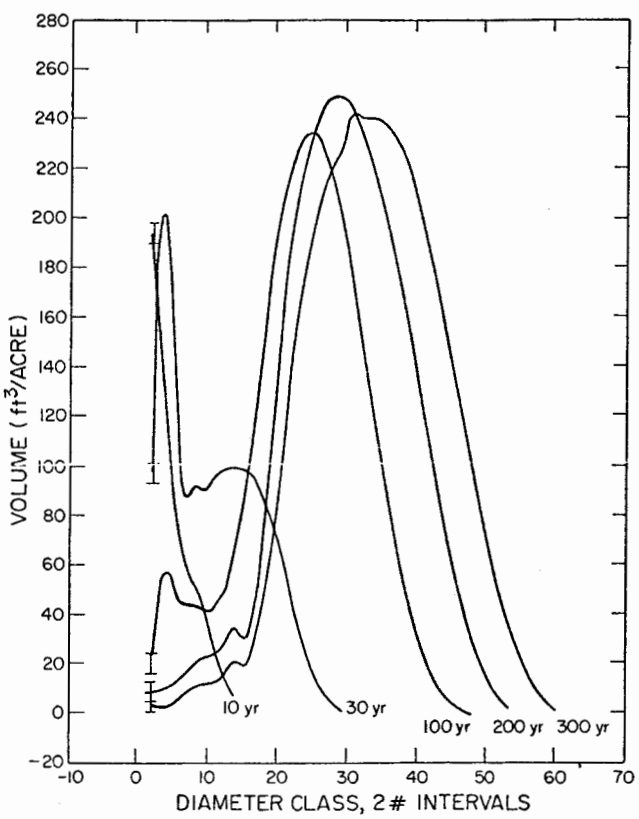


Fig. 7. Forest volume by diameter class.

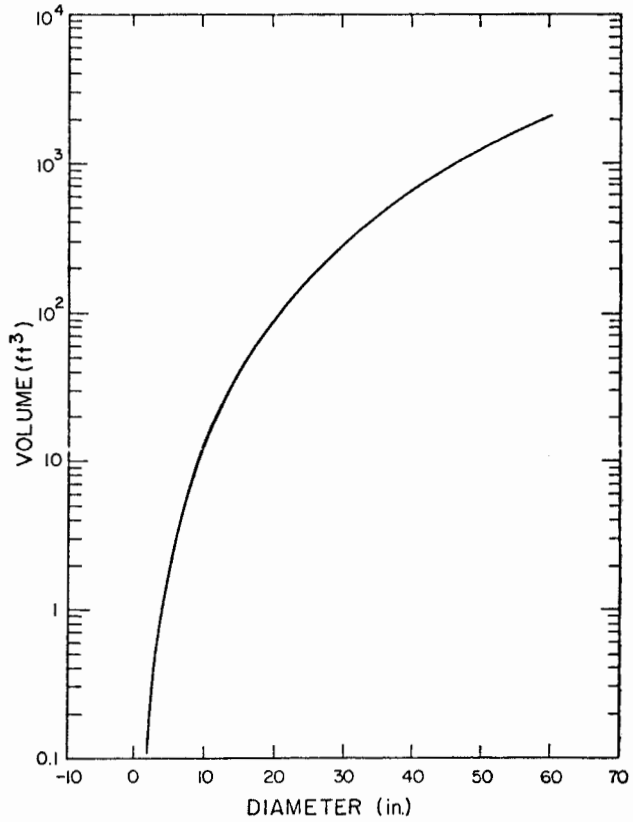


Fig. 8. Ponderosa tree volume by diameter.

B. Interregional Transport Model

An interregional transport code is being formulated which will allow simultaneous simulation of the 75 regions of the study area.[1] Presently, it will successfully simulate the growth of up to 21 plant categories and track transuranics through their various compartments. The atmospheric module[2] and herbivore modules have been successfully interfaced, although more development of the latter is required for

proper operation. Efforts are under way to interface both the forest structure and forest management submodules for regions where forests occur. The rather large data base for agricultural and livestock operations is now almost integrated into the model. With the exception of the plant succession submodule now under planning, and for other submodules such as the human module[2] and the aquatic module which will receive more attention in the future, the interregional transport model is very close to the testing stage for determining concentrations of transuranics along the food chain to man in the study area.

REFERENCES

1. "Semiannual Report. Transuranic Solid Waste Management Programs. July-December, 1974," Los Alamos Scientific Laboratory report LA-6100-PR (1975).

2. "Semiannual Report. Transuranic Solid Waste Management Research Programs. January-June, 1975," Los Alamos Scientific Laboratory report LA-6264-PR (1975).

3. S. W. Running, R. H. Waring, and R. A. Rydell, "Physiological Control of Water Flux in Conifers," Oecologia (Berl.) 18, 1-16 (1975).

4. W. H. Adams, "Solubility of $^{238}PuO_2$ in Los Alamos Tap Water," Los Alamos Scientific Laboratory unpublished data, 1968.

5. B. J. Gilbert, "Ranges IV Grassland Simulation Model. Regional Analysis of Grassland Environmental Systems, Federal Grant No. ESR-72-03396-A02, Colorado State Univ., Ft. Collins, CO, report No. 17-18 (1974).

6. W. E. Martin, S. G. Bloom, and R. J. Yorde, "NAEG Plutonium Study Modeling Program: Plutonium Transport and Dose Estimation Model. The Dynamics of Plutonium in Desert Environments," Nevada Applied Ecology Group, USAEC, Prog. Rpt. NVO-142, pp. 338 (1974).

7. E. M. Romney, H. M. Mork, and K. H. Larson, "Persistence of Plutonium in Soil, Plants, and Small Mammals," Health Phys., 19, 487-491 (1970).

8. R. L. Edmonds, Ed., "An Initial Synthesis of Results in the Coniferous Forest Biome, 1970-1973," Coniferous Forest Biome, Ecosystems Analysis Studies US/IBP, Bulletin No. 7, 40 (1974).

9. W. H. Langham, "Plutonium Distribution as a Problem in Environmental Science," Proc. of Environ. Plutonium Symp., Los Alamos Scientific Laboratory report No. LA-4756 (1971).

10. D. W. Wilson, "A Mathematical Model of the Transport of Cs-137 from Fallout to Milk," Ph.D. Dissertation, Colorado State University, Ft. Collins, CO, p. 139, (1968).

11. Jen-Hu Chang, Climate and Agriculture (Aldine Publishing Co., Chicago, IL, 1968) pp. 211-224.

12. C. C. Grier and R. H. Waring, "Conifer Foliage Mass Related to Sapwood Area," Forest Science 20, 3, pp. 205-206 (1974).

13. P. L. Altman and D. S. Dittmer, "Growth," Federation of Am. Soc. for Experimental Biology, p 379, (1962).

14. M. H. Zimmerman and C. L. Brown, Trees - Structure and Function (Springer-Verlog, New York, NY, 1974).

15. H. E. Brown, M. B. Baker, J. J. Rogers, W. P. Clarey, J. L. Kooner, F. R. Larson, C. C. Avery, and R. E. Campbell, "Opportunities for Increasing Water Yields and Other Multiple Use Values on Ponderosa Pine Forest Lands," USDA Forest Service, Research Paper RM-129 (1974).

16. F. R. Larson, "Simulating Growth and Management of Ponderosa Pine Stands," Ph.D. Dissertation, Colorado State University, Ft. Collins, CO (1975).

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