

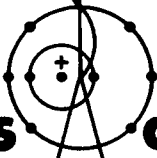
LA-5286-MS

INFORMAL REPORT

ER Record I.D.# 0004975

21

Underground Movement of Tritium from Solid-Waste Storage Shafts



los alamos
scientific laboratory

of the University of California

LOS ALAMOS, NEW MEXICO 87544



2936

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

In the interest of prompt distribution, this LAMS report was not edited by the Technical Information staff.

Printed in the United States of America. Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

LA-5286-MS
Informal Report
UC-41

ISSUED: May 1973



Underground Movement of Tritium from Solid-Waste Storage Shafts

by

William D. Purtymun

the levels of tritium in moisture in the tuff. The moisture content of the tuff was determined and tritium analyses were made on water distilled from the tuff samples (Table I).

The water distilled from the tuff contained above-background concentrations of tritium, indicating leakage of tritium from the wastes in the nearby shafts 1 through 13. The largest concentrations of tritium were in moisture from samples collected in shafts 47 and 48. These two shafts are adjacent to shafts 12 and 13 (Fig. 1), which received an unknown amount of tritium associated with a classified shipment of wastes in July 1966 and September 1966.

A study was made in August 1970 to determine the extent of the movement of tritium with moisture into the tuff and to delineate some of the geologic factors related to this movement. Fourteen test holes were laid out and drilled, 12 to the

west of the burial shafts and 2 to the east. This concentration of observation points to the west was chosen because shafts 39 through 48 (to the east), which were open at the time, permitted air exchange from the tuff to the atmosphere and could have affected natural migration of tritiated moisture through the tuff.

The test holes, 6 in. in diameter and 50 ft deep, were drilled with a power auger. Tuff samples representing 5-ft intervals were collected from the auger cuttings. The holes were thoroughly cleaned before each 5-ft sample run, and the results of the tritium analyses indicated that there was little, if any, cross-contamination between samples from successive 5-ft intervals.

The samples of tuff cuttings were collected in plastic bags and returned to the laboratory for analysis. Each sample was placed in a 600-ml beaker, filling it to about 3 cm from the top. A 50-ml beaker was nestled in the top of the cuttings, and a large watch glass filled with ice was placed on the top of the 600-ml beaker. The beaker and tuff were heated to drive out the water vapor which condensed on the bottom side of the watch glass. The condensate dripped into the 50-ml beaker and was retained for further analysis. Preparation of surface soil and tuff samples were made in this same manner.

Water from vegetation in the area was analyzed to determine if the vegetation contained tritiated moisture or was transpiring it to the atmosphere. To establish tritium content, a sample of vegetation was collected, cut into small pieces, and subjected to the same distillation process as the soil and tuff samples. Transpired moisture was collected as a condensate in a plastic bag tied over a plant or a portion of the plant for 24 h.

A 0.5-ml aliquot of the water obtained from the soil, tuff, or vegetation was mixed with 15 ml of a dioxane-base liquid scintillation cocktail and counted in a liquid scintillation counter for 100 minutes. Instrument background was determined by assaying a similarly prepared sample containing

TABLE I
TRITIUM ANALYSES OF MOISTURE
FROM DRILL CUTTINGS
SHAFTS 39 THROUGH 49

Shaft No.	Depth (ft)	Moisture Content (% by wt)	Tritium (pCi/ml)
39	25	1.2	52,800
40	15	2.0	7,400
40	25	1.0	15,100
41	15	1.1	116,000
41	25	1.4	6,800
42	15	1.4	70,700
42	25	0.9	10,600
43	15	1.2	3,100
43	25	0.2	23,000
44	15	1.6	4,640
44	25	0.6	1,780
45	15	1.0	211,000
45	25	0.8	178,000
46	15	0.9	30,800
46	25	0.5	6,810
47	15	0.9	221,000
47	25	0.7	1,180,000
48	15	2.3	171,000
48	25	0.8	325,000

only blank reagents. The detection limit for the above method was about 5 pCi/ml of water.

PHYSICAL CHARACTERISTICS OF THE TUFF

The test holes and the burial shafts penetrated two ashflows. The upper flow is about 17 ft thick at the shafts, thinning slightly to the west due to erosion of its upper surface, and dipping somewhat to the southeast. The shafts were completed into the top of the lower ashflow which is about 40 ft thick.

The ashflows are similar in composition. Both are moderately welded light gray tuff composed of quartz and sanidine crystals and crystal fragments, with rhyolite, latite, and pumice rock fragments in a matrix of gray ash. Laboratory determinations indicate that porosity of the tuff ranges from 36 to 44% by volume. The contact between the upper and lower flows is characterized by a zone of reworked tuff fragments and by an increase in the size and number of the pumice fragments in the base of the upper ashflow. Within Area G, the zone of reworked tuff may be very thin or nonexistent, or as much as 3 ft thick.

During cooling, the ashflows were broken into a number of large blocks by vertical or near vertical joints. Since both flows cooled as a single unit, the joints extend through both. Joint openings may range to as much as 1/2 in., with those terminating at the surface of the mesa being filled with a light brown clay to a depth of 3 to 4 ft. The joint frequency is about 1 per 7 ft of lateral distance as observed in the walls of 11 storage pits at TA-54.

Measurements of moisture content of the samples indicated that precipitation had infiltrated into the tuff to a depth of over 10 ft. The moisture content in this 10-ft interval ranged from 3 to 8% by weight, decreasing with depth. From a depth of 10 ft to 50 ft the moisture content ranged from 0.4 to about 3% by weight. This low value lies in the regime where moisture transport is primarily by diffusion in the vapor phase. The movement of the tritium is by exchange with natural airborne moisture in the tuff.

UNDERGROUND MOVEMENT OF TRITIUM

Tritium concentrations were determined for water distilled from 140 samples of tuff collected from the 14 test holes (Table II). In general the tritium concentrations increased to a maximum between depths of 10 and 30 ft and then decreased with depth.

Isotritium contours were constructed at depths of 10 to 15 ft, 20 to 25 ft, 30 to 35 ft, and 40 to 45 ft (Figs. 2 and 3). Test holes and burial shafts were used for control points on Fig. 2 while only the test holes were used on Fig. 3. The irregularities in the isotritium contours just east of the shafts are due to the movement of the tritium through open joints which provide a much more rapid means of migration than movement through the tuff matrix. The effect of these joints is apparent only near the shafts due to the close spacing of control points used in construction of the contours. West of the shafts where control points (test holes) are located on 20 ft centers the effect of the joints on movement of the tritium is not apparent. The contours are elongated to the west of the shafts showing the principal movement of the tritium.

Isotritium contours were also constructed for a vertical plane extending east and west through shaft 13 (Fig. 4). The contours show that the major movement of tritium took place along the contact between the two ashflows. The abundance of pumice fragments in the lower part of the upper ashflow and the presence of the reworked tuff between the two flows causes a greater porosity and permeability in the contact region as compared to the matrix or joints in the overlying or underlying tuff. The tritiated moisture migrating along the contact serves as a source for movement into the upper and lower ashflows.

A comparison of the isotritium contours in planview (Figs. 2 and 3) just above and below the contact shows that migration of the tritium was not uniform along the contact but greater in a westerly direction. This is due to a thickening of the layer

TABLE II
TRITIUM ANALYSES OF MOISTURE FROM DRILL CUTTINGS
TEST HOLES A THROUGH N (pCi/ml)

Depth (ft)	A	B	C	D	E	F	G
0- 5	68,000	5,370	7,510	383	294	531	729
5-10	94,700	11,800	22,300	6,310	1,100	538	1,610
10-15	196,000	8,270	55,500	13,800	439	632	4,190
15-20	159,000	12,900	44,800	14,300	1,010	1,860	7,060
20-25	471,000	13,200	39,700	18,800	744	2,620	9,310
25-30	275,000	9,850	32,800	19,900	611	3,750	9,370
30-35	140,000	6,390	22,900	15,000	516	3,190	7,490
35-40	211,000	4,470	13,900	9,200	742	2,160	6,550
40-45	116,000	4,590	4,790	4,380	918	1,405	3,680
45-50	79,000	4,170	4,950	1,470	816	1,107	3,650

Depth (ft)	H	I	J	K	L	M	N
0- 5	42	41	77	42	37	4,830	250
5-10	30	144	267	154	157	6,680	2,410
10-15	131	182	3,720	405	249	6,920	6,140
15-20	197	320	5,490	876	205	7,480	8,790
20-25	445	195	7,210	912	145	5,780	9,400
25-30	430	207	6,340	1,060	127	4,100	9,820
30-35	512	196	969	1,060	122	3,690	9,290
35-40	330	140	1,230	728	87	2,780	7,760
40-45	278	144	966	801	57	3,020	6,250
45-50	235	136	844	569	44	2,550	4,710

of reworked tuff lying along the contact which increases the rate of movement in that direction.

The tuff in the ashflows is not homogenous. It is broken by joints, and porosity varies within the tuff matrix. The contact between the two ashflows adds to the inhomogeneity. Thus the extent of the migration can only be approximated. About 4 yr after the wastes were stored in shafts 12 and 13, a 100 pCi/ml contour has moved to a distance of about 105 ft west of the shafts along the contact between the two ashflows (Fig. 4), and the 100 pCi/ml contour extrapolated beneath the shafts is at a depth of about 97 ft below the surface of the mesa. There is a second contact beneath the lower ashflow (Fig. 4), however, and this contact would slow the vertical migration of tritiated water vapors through the tuff by allowing them to move laterally along the more permeable contact. In general, the

volume of tuff containing the tritiated moisture has assumed the shape of an irregular lens, shortened to the east and elongated to the west (Fig. 4).

Little water from precipitation infiltrates through the soil into the underlying tuff.¹ There is not enough water to leach the tritium from the wastes and move it through the 240 ft of dry tuff and 610 ft of dry volcanic rocks and sediments to the main aquifer that lies at a depth of about 850 ft at Area G.

RELEASE OF TRITIUM TO THE ATMOSPHERE

Intake of air during periods of high atmospheric pressure and exhaust of air during periods of low pressure have been noted for burial shafts and test holes in the tuff in Area G. The soil and weathered surface tuff forms a partial barrier against the exchange of air between the underlying

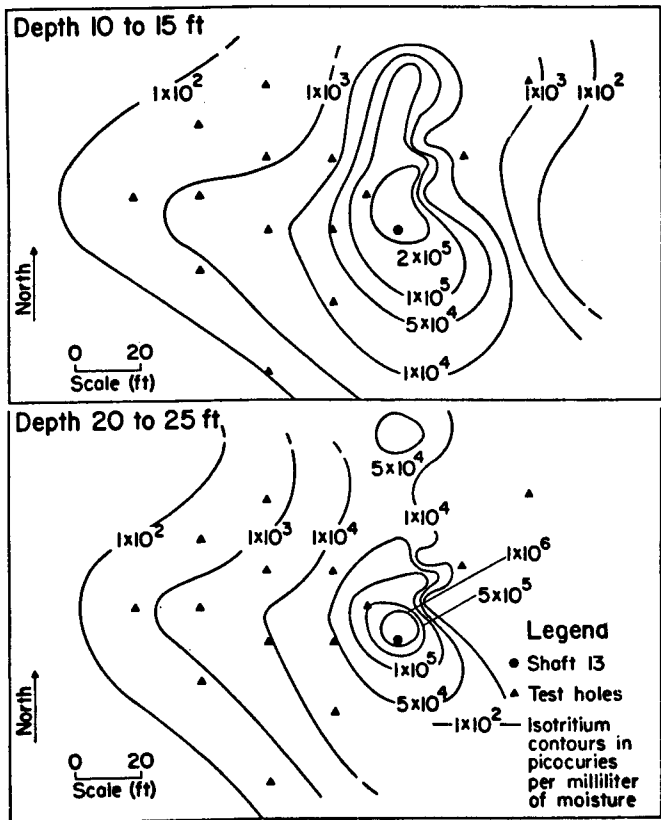


Fig. 2. Isotritium concentrations in picocuries per milliliter of moisture in planview at depths of 10 to 15 ft and 20 to 25 ft.

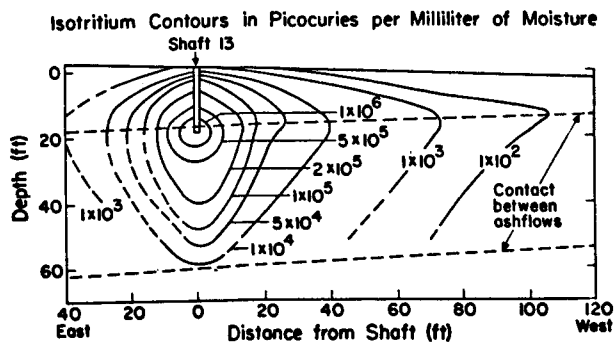


Fig. 4. Isotritium concentrations in picocuries per milliliter of moisture in section east and west of shaft 13.

tuff and the atmosphere.² Air was pumped from shaft 47 to determine if measurable amounts of tritium could be released to the atmosphere during an atmospheric low.

The air in the shafts is nearly saturated with water vapor. The measured relative humidity

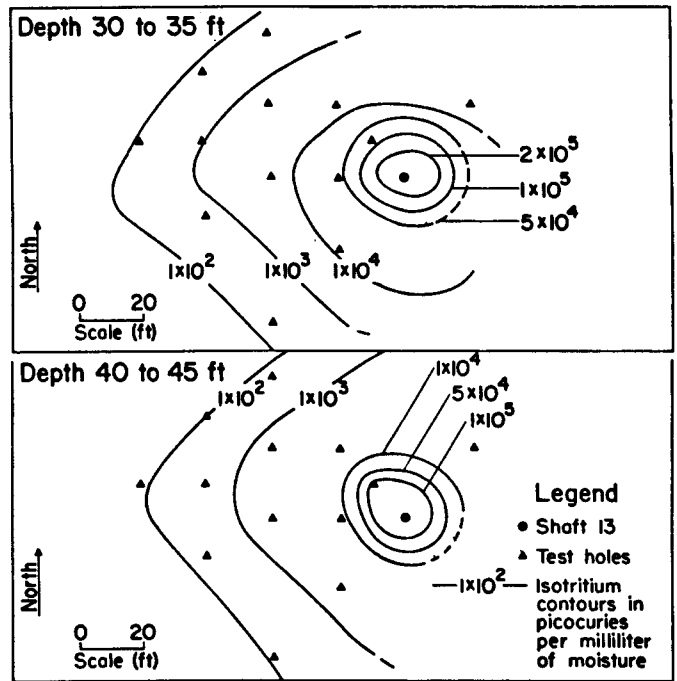


Fig. 3. Isotritium concentrations in picocuries per milliliter of moisture in planview at depths of 30 to 35 ft and 40 to 45 ft.

varied from 94 to 98% at a temperature of about 70°F. Prior to receiving wastes, shaft 47 was sealed at the surface and air pumped out of the unlined shaft at a rate of about 2.5 cfm for 24 h. Moisture was collected in a condenser at the pump and analyzed for tritium. The tritium concentration was 577,000 pCi/ml of water, about half the concentration found in moisture from a sample of tuff collected from a depth of 25 ft (Table I).

A number of samples of surface soil were collected west of shaft 13 out to a distance of 120 ft (Table III). Tritiated moisture concentrations generally decreased with distance from shafts 12 and 13. The sample collected at 80 ft consisted of tuff since there was no soil cover at that point. We therefore conclude that tritium is being released to the atmosphere as a result of evaporation of soil moisture near the shafts.

In an effort to determine whether or not vegetation has any influence on the transfer of tritiated moisture from the disposal shafts to the atmosphere, five samples of vegetation and moisture

TABLE III

MOISTURE CONTENT AND TRITIUM ANALYSES
OF MOISTURE FROM SOIL AND TUFF
WEST OF SHAFT 13, AUGUST 5, 1970

Distance (ft)	Moisture Content (% by wt)	Tritium (pCi/ml)
5	8.8	1,100
10	17.3	327
20	10.2	131
40	7.0	40
60	6.4	68
80	2.7	122
100	7.1	30
120	8.2	21

transpired from vegetation were collected in the immediate vicinity of the disposal shafts (Table IV). These analyses indicate that there is an uptake of tritium by plants from the soil and tuff and that tritiated moisture is being transpired to the atmosphere. Measurements from two samples collected about 0.5 miles west of the shafts are included for comparison. No attempt was made to determine the amount of tritiated moisture being released to the atmosphere due to transpiration from plants and evaporation from surface soil adjacent to the shafts.

STORAGE OF TRITIUM-CONTAMINATED WASTES

Containment is the goal of every waste storage operation. To attain this goal, new packaging

techniques for tritium-contaminated wastes were initiated. Small items are placed in 55 gal drums, which are then filled with asphalt, sealed, and lowered into the shafts. Large objects present a more complex problem. The walls and bottom of the receiving shaft are coated with asphalt, and the waste is emplaced in the shaft and encased in asphalt.

Additional measures would provide even better containment performance for these disposal shafts. Location and depth should be chosen to preclude the penetration of two ashflows. This may not always be possible due to the location of the disposal site. Furthermore, the frequency of joints and fractures in the ashflow tuffs makes it difficult to locate shafts where open joints and fractures do not intersect the walls. Therefore, the walls of all shafts should be coated by spraying with asphalt to help retain the tritium by sealing any contact between ashflows as well as any open joints or fractures, and by decreasing the porosity of the tuff matrix.

SUMMARY

The transport of tritium is by exchange with the natural water in the tuff and subsequent movement of this moisture in the vapor phase. Primary movement from the shafts was along the contact between two ashflows, and secondary movement was through open joints and through the tuff matrix.

TABLE IV

TRITIUM ANALYSES OF MOISTURE FROM PLANTS

Location	Type of Plant	Type of Sample	Tritium (pCi/ml)
40 ft South of Shaft 13	Ragweed	DIS	392
40 ft South of Shaft 13	Tumble Weed	DIS	398
At Shaft 13	Unknown	SWT	101,000
85 ft North of Shaft 13	Chamisa	SWT	17,700
At Shaft 14	Unknown	DIS	7,970
0.5 Mile West of Area G	Chamisa	SWT	<5
0.5 Mile West of Area G	Unknown	SWT	<5

DIS - Analyses made of moisture distilled from plant by heating.
SWT - Analyses made of moisture transpired from plant.

This migration has caused 100 pCi/ml concentrations of tritiated moisture to be displaced about 105 ft west of the shafts in 4 yr. Since the tuff is not homogenous, the tritium-contaminated volume has formed the shape of an irregular lens truncated to the east of the shaft and elongated to the west.

Tritiated moisture has migrated to the surface of the mesa where evaporation from the soil and exposed tuff and transpiration from plants is releasing it to the atmosphere. This effect is most noticeable in an area adjacent to the shafts and decreases with distance outward.

New storage techniques include encasing tritium-contaminated wastes in asphalt to prevent migration from the shafts. Additional precautions could be taken by spraying the walls of all shafts with asphalt in order to seal contacts between ash-flows and open joints and fractures and to reduce the porosity of the tuff matrix.

REFERENCES

1. W. D. Purtymun, "Geology and Hydrology of Mesita del Buey," Los Alamos Scientific Laboratory report LA-4660 (1971).
2. J. L. Kunkler, "Measurement of Atmospheric Pressure and Subsurface Gas Pressure in the Unsaturated Zone of the Bandelier Tuff, Los Alamos, New Mexico," U.S. Geol. Survey Prof. Paper 650-D (1969).

☆ U. S. GOVERNMENT PRINTING OFFICE: 1973 — 784-278/23