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1967-1978*

LOS ALAMOS NATIONAL LABORATORY



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**Radiochemical Quality of Water
in the Shallow Aquifer
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RADIOCHEMICAL QUALITY OF WATER IN THE
SHALLOW AQUIFER IN MORTANDAD CANYON
1967-1978

by

W. D. Purtymun, W. R. Hansen, and R. J. Peters

ABSTRACT

Mortandad Canyon receives treated industrial liquid effluents that contain trace amounts of radionuclides. The effluents, other waste water, and storm runoff recharge a shallow aquifer in the alluvium of the canyon. The aquifer lies within the Los Alamos National Laboratory boundaries. Analyses for gross alpha, gross beta, ^{137}Cs , ^{238}Pu , ^{239}Pu , ^{241}Am , ^{90}Sr , ^3H , and total U have been made of water in the aquifer from 1967 through 1978. Average concentrations of the radionuclides in solution decrease downgradient in the canyon with the exception of ^3H . Average ^3H concentrations were highest in the Middle Canyon. Inventories of most radionuclides in the water indicate that in 1978 less than 1% of the total amount released with the effluents in the canyon from 1963 through 1978 remained in solution. The amount of total U in solution in 1978 was about 16% of the total amount released. If there is no significant change in the amounts of radionuclides received at the treatment plant and methods of treatment remain the same, the projected estimates of radionuclide concentrations in the aquifer will increase about 80% from 1978 to 1990. The average concentrations in 1978 and projected concentrations in 1990 of gross alpha, ^{137}Cs , ^{238}Pu , ^{239}Pu , ^{241}Am , ^{90}Sr , ^3H , and total U are less than 1% of the Department of Energy's concentration guides (CG) for areas with controlled public access. Gross beta radioactivity in 1978 was 2% of the CG and is projected to increase to 3% of the CG by 1990.

I. INTRODUCTION

Industrial liquid wastes containing radionuclides from operations of the Los Alamos National Laboratory are collected and processed at the Industrial Waste Treatment Plant at TA-50 (Technical Area 50). After treatment that removes most of the radionuclides, the effluents are released into Mortandad Canyon. Release of effluents from TA-50 and waste water from TA-48

causes perennial flow in the upper reach of the canyon. Occasional storm runoff adds to the surface flow in the canyon.

The perennial surface flow and storm runoff recharge a shallow aquifer in the alluvium of the canyon that is perched (ground water separated from an underlying main body of ground water by an unsaturated zone) on the underlying tuff (Fig. 1). As the water in the shallow aquifer moves downgradient, losses occur from

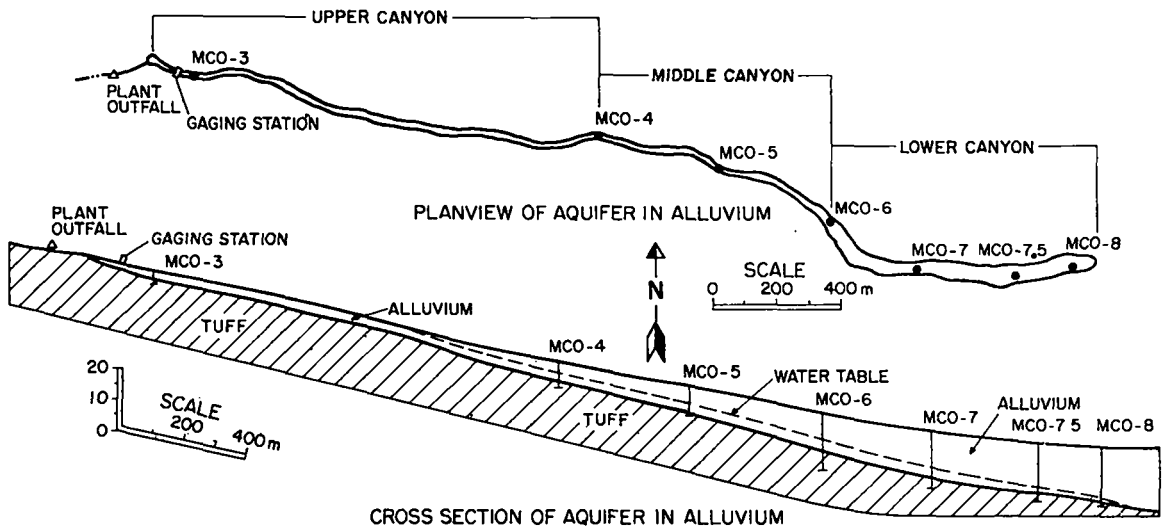


Fig. 1. Planview and cross section of aquifer in alluvium of Mortandad Canyon.

evapotranspiration and infiltration into the underlying tuff. The aquifer is of limited extent and forms a shallow ground water body in the canyon within the confines of the Laboratory property. The top of the main aquifer lies at a depth of 290 m below the perched aquifer.

The plant at TA-50 became operational in 1963 and has released low-level radioactive effluents into the canyon. Hydrologic studies initiated in 1960 have continued as part of the overall Laboratory environmental monitoring program. Before 1967, the water from the shallow aquifer in Mortandad Canyon was analyzed for gross beta-gamma activity, total plutonium (Pu), and total uranium (U); however, the minimum detection limits were not low enough to include with data collected during the period 1967 to 1978. During this latter period, analyses were made routinely for gross alpha, gross beta, ^{238}Pu , ^{239}Pu , ^3H , and total U, while periodic analyses were made for ^{137}Cs , ^{241}Am , and ^{90}Sr . The purpose of this study was to investigate annual changes in radiochemical quality of water in the shallow aquifer as the result of the release of industrial effluents from TA-50 and to project effects on the aquifer to 1990 if treatment methods at the plant remain the same.

Mortandad Canyon, a southeast-trending canyon, is about 15 km long. The canyon heads on Pajarito Plateau and is a tributary to the Rio Grande in White Rock Canyon. The upper 5.5 km of the canyon on the Pajarito Plateau is within Laboratory boundaries. The area of study is the shallow aquifer in the alluvium (Fig. 1). The aquifer extends from near the plant outfall to near obser-

vation hole MCO-8 (Mortandad Canyon observation hole).

The canyon is cut into the Bandelier Tuff, which forms the surface of the plateau.¹ The canyon floor is narrow at the plant outfall and widens eastward. The canyon walls are steep and in places are near vertical. The alluvium in the canyon is derived from erosion and weathering of the tuff. The alluvium thickens eastward from about 1 m at the gaging station to more than 24 m at MCO-8. The shallow aquifer in the alluvium occupies less than 10% of the volume of alluvium. There has been no storm runoff to transport radionuclides out of the canyon since hydrologic studies began in 1960 because of the small drainage area and large volume of unsaturated alluvium.

To facilitate the study, the canyon was divided into three sections: the Upper Canyon, Middle Canyon, and Lower Canyon (Fig. 1). The hydrologic characteristics of each section are slightly different.

The Upper Canyon is narrow, filled with underbrush, shrubs, pine, fir, box-elder, and oak trees. The alluvium thickens from less than 1 m at plant outfall to about 6 m at MCO-4. The stream in this section of canyon originates at the plant outfall. There is some return flow near MCO-4 (Fig. 1). Major recharge to the shallow aquifer occurs in this reach of the canyon.

The Middle Canyon widens and alluvium thickens from 6 m at MCO-4 to about 12 m at MCO-6. The stream channel is well defined, but surface flow is intermittent. The underbrush thins out and the canyon floor is covered by pines.

The Lower Canyon becomes progressively wider and the alluvium continues to thicken to about 24 m at MCO-8. The stream channel is discontinuous, braiding out on the canyon floor. The number of pines decreases eastward from the Middle Canyon with a transition to scattered piñon-juniper trees.

Alluvium in the canyon consists of two distinguishable units that affect the hydrologic characteristics of the aquifer. Water in the aquifer west of MCO-5 is in a sand unit and is transitional into a silty clay unit at MCO-6. East of MCO-6 the aquifer is within the lower silty clay. The velocity of water movement in the sand unit is about 18 m/day; in the transition from sand to silty clay unit, about 6 m/; and in the silty clay unit, about 2 m/day.²

A. Industrial Effluent, Storm Runoff, and Waste Water

Surface water entering the canyon is from two sources: (1) industrial effluents from TA-50 and (2) storm runoff and waste water from TA-48. The volume of industrial effluents was taken from plant records. The effluents are released twice daily from TA-50 in volumes of 70 to 90 m³ for each release. During the period 1967 to 1978, the annual volume of industrial effluents ranged from 4.0 to 6.0 × 10⁴ m³ with an average of 4.9 × 10⁴ m³ (Table I). In general, the trend for the period has shown a decrease in the volume of effluent released (Fig. 2).

The volume of storm runoff and waste water entering the canyon was determined from flow measurements made at the gaging station (Fig. 1). The volume of storm runoff was highly variable, whereas the volume of waste water from TA-48 remained about the same ranging from about 5 to 6 × 10³ m³ annually.

Annual storm runoff and waste water from TA-48 ranged from 1.0 to about 9.3 × 10⁴ m³ with an average of about 3.5 × 10⁴ m³ for the period 1967 to 1978 (Fig. 2). The volume of storm runoff entering the canyon has declined considerably in the past few years when compared with the volume of runoff from 1967 to 1970. This has resulted in less recharge to the shallow aquifer and a general decline in storage in the aquifer.

Continuous release of waste water maintains surface flow into the Upper Canyon. The volume and rate of discharge, along with storm events, govern the distance of surface flow down the canyon. The intermittent release of industrial effluents from TA-50 causes the normal waste water flow to extend a greater distance down the canyon from the plant outfall.³

B. Storage and Loss of Surface and Ground Water in the Shallow Aquifer

The volume of alluvium in the Upper, Middle, and Lower Canyons was determined by drilling lines of test holes across the canyon (to outline cross-sectional area of saturation) at selected intervals from the gaging station to MCO-8. The volume in storage (saturation) was determined from water level measurements in seven observation wells and the known volume of the alluvium (Fig. 1). The storage was computed at the end of December for each year for each of the three sections of the canyon (Table I).

The storage in the aquifer for the period 1967 to 1978 ranged from 1.5 to 3.0 × 10⁴ m³ (Fig. 3). The volume in storage has declined from 1967 to 1978 as a result of less storm runoff. The volume available for storage increases downgradient in the canyon as the alluvium thickens and widens. The storage in Upper Canyon ranged from 3 to 7 × 10³ m³, in Middle Canyon from 3 to 9 × 10³ m³, and in Lower Canyon from 5 to 18 × 10³ m³ for the period of study (Table I).

The loss of surface and ground water from the shallow aquifer is from evapotranspiration and infiltration into the tuff underlying the aquifer. The distribution of losses to evaporation and infiltration are unknown; however, losses to infiltration into the tuff are apparently low. The shallow aquifer is underlain by about 290 m of unsaturated volcanic (tuff and basalt) and sedimentary rocks above the main aquifer. Water analyzed from the main aquifer from a test well in the Middle Canyon from 1963 to 1978 has shown no change in chemical quality and no detectable radionuclides associated with the industrial effluents as found in the shallow ground water body in the alluvium.

A water balance was made using effluent and surface water inflow, storage, and annual change in storage. The surface and ground water losses ranged from 0.5 to about 1.5 × 10⁵ m³ (Fig. 4).

The annual inflow of surface water (effluents, storm runoff, and waste water) is about equal to surface and ground water losses in the canyon. An increase in surface water inflow into the canyon results in an increase in storage in the aquifer and increases water losses. The reverse occurs with a decrease in surface water inflow. Thus, with annual changes in volume of storm runoff, storage in the aquifer varies as do the annual water losses in the canyon (Table I).

TABLE I
BALANCE OF WATER IN MORTANDAD CANYON
(10³ m³)

Year	Surface Water				Storage in Aquifer				Annual Surface and Ground Water Loss
	Effluent TA-50	Waste Water and Storm Runoff	Total	Upper	Middle	Lower	Total		
1967	60	79	139	7	5	18	30	129	
1968	60	52	112	5	5	14	24	118	
1969	54	93	147	3	5	17	25	146	
1970	53	50	103	3	3	14	20	108	
1971	46	29	75	5	7	17	29	66	
1972	57	26	83	4	5	14	23	89	
1973	54	37	91	3	4	12	19	95	
1974	40	25	65	3	4	12	19	65	
1975	40	15	55	3	3	13	19	55	
1976	40	10	50	3	9	5	17	52	
1977	42	12	54	4	4	7	15	56	
1978	40	15	55	3	3	12	18	52	

Distribution of Storage, 1967 to 1978

	(10 ³ m ³)			Percentage of Average
	Minimum	Maximum	Average	
Upper	3	7	4	18
Middle	3	9	5	23
Lower	5	18	13	59

II. RADIOCHEMICAL QUALITY OF WATER IN SHALLOW AQUIFER

The radiochemical quality of the industrial effluents changes as they move into the canyon because of dilution with waste water from TA-48, dilution with storm runoff, and adsorption of radionuclides within the alluvium.⁴ One of the major factors facilitating adsorption, retention, or exchange of radionuclides with the alluvium is the high pH (average about 11) of the in-

dustrial effluent and the clay minerals (montmorillonite and illite) in the alluvium derived from weathering of the tuff.

As the industrial effluents released into the canyon move downgradient, the radionuclides are adsorbed or bound to bed sediments, reducing the amount of radioactivity in the surface flow. Radioactivity decreased downgradient in the sediments from the plant outfall. A high build-up of activity at the outfall does not occur as periodic storm runoff transports and disperses sediments

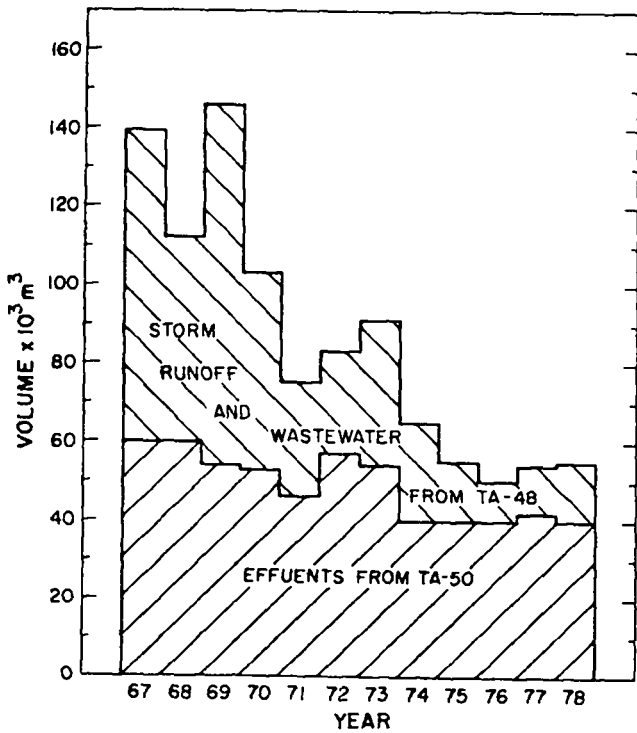


Fig. 2. Volume of storm runoff, waste water from TA-48, and effluents from TA-50 entering the canyon, 1967 to 1978.

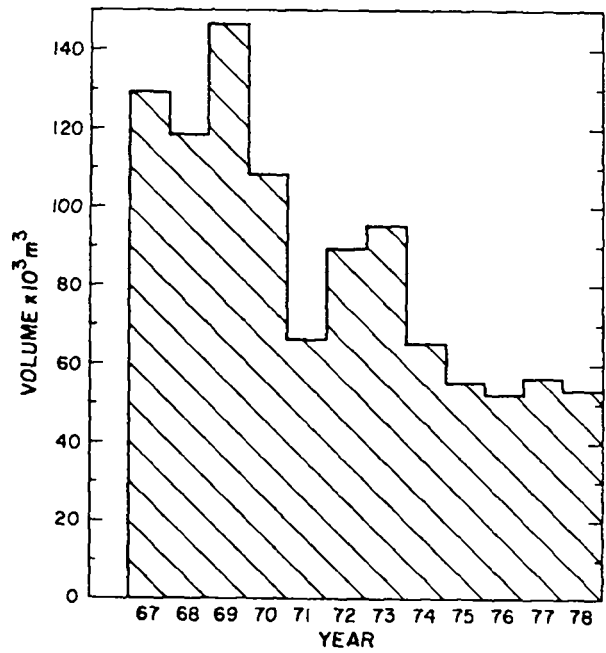


Fig. 4. Annual surface and ground water losses in the canyon, 1967 to 1978.

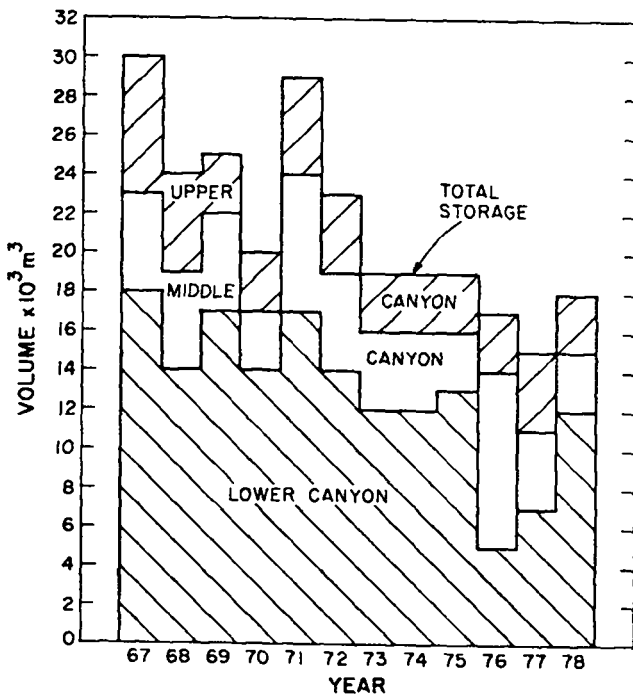


Fig. 3. Storage in the aquifer in the Upper, Middle, and Lower Canyons, 1967 to 1978.

and radionuclides down the channel in the canyon.⁴ Adsorption of radionuclides in sediments reduces the concentrations in the water of the shallow aquifer. Similar adsorption and retention of nuclides occur in the aquifer as the water moves downgradient.

Transit time for water to move through the aquifer from the Upper Canyon to the end of the Lower Canyon is about 1 yr. Based on chemical quality data in the shallow aquifer and the fact that the water balance shows that inflow and losses in the canyon are about equal, there is a "turnover" of water in the aquifer each year.⁵ The turnover of water is a continuous process with inflow and losses occurring daily. Because the aquifer is a changing system, trends in radionuclide concentration, distribution, and inventory were compiled annually.

Samples for radiochemical analyses were collected one to four times annually from the seven observation holes (MCO-3, -4, -5, -6, -7, -7.5, and -8) during the period 1967 to 1978. The annual average for two or more analyses from the individual observation holes is used for interpreting the quality of water in the shallow aquifer as the water is in transit storage (water in the aquifer is in continuous movement). The radiochemical data from individual observation holes is recapped in the Appendix.

The water was filtered through 0.45- μ m-pore membrane filters and analyzed for gross alpha, gross beta,

^{137}Cs , ^{238}Pu , ^{239}Pu , ^{241}Am , ^{90}Sr , ^3H , and total U. Solution is defined as the analyses of the filtered water. Detailed methods of analyses are presented in Ref. 6.

Minimum limits of detection (MDL) for the various radionuclides have varied for the period 1968 to 1978 but have generally decreased because of improved analytical technique. The MDL for each type of analysis are indicated in the Appendix.

A. Distribution of Radionuclides

The plant at TA-50 began operations in 1963. Continued release of effluents into the canyon caused a general increase in most radionuclides in the water of the shallow aquifer as shown by the average concentrations from 1967 to 1978 (Table II).

The average annual gross alpha activity in the water has increased from 1.5 pCi/l in 1967 to 27.5 pCi/l in 1978, whereas gross beta activity increased from 33 pCi/l in 1967 to 169 pCi/l in 1978. The concentration of ^{137}Cs has varied for the period of record (5 yr) showing a concentration of 27 pCi/l in 1978. The average concentration of ^{238}Pu in the water increased from 0.05 pCi/l in 1967 to 3.82 pCi/l in 1978. Similarly, the ^{239}Pu concentrations have increased from 0.07 pCi/l in 1967 to 0.68 pCi/l in 1978.

The total amount of plutonium released into the canyon from 1963 to 1978 was 96.8 mCi. Of that total, 84.2 mCi were released from 1967 to 1978 or about 87% of the total amount released (Table III). It is evident from the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio in the shallow aquifer that more ^{239}Pu than ^{238}Pu was released with the effluents from 1963 to 1969. Since 1969, most of the plutonium has been ^{238}Pu (Table III).

The average concentration of ^{241}Am for the period of record (4 yr) has varied showing no particular trend (Table II). The concentration of ^{90}Sr for period of record (2 yr) has declined from 36.4 pCi/l in 1976 to 15.3 pCi/l in 1978.

Concentrations of ^3H have varied considerably from 1967 to 1978 (Table II). The ^3H part of the water molecule is not affected by adsorption or ion exchange with clay minerals. Variation in concentrations is a result of large amounts released in 1967, 1970, and 1976.

The concentration of total U has varied considerably from 1967 to 1978 with a general increase from 2.0 $\mu\text{g}/\text{l}$ in 1967 to 11.1 $\mu\text{g}/\text{l}$ in 1978.

The highest average concentrations of gross alpha, gross beta, ^{137}Cs , ^{238}Pu , ^{239}Pu , ^{241}Am , ^{90}Sr , and total U occurred in the Upper Canyon, decreasing downgradient in the aquifer into the Lower Canyon. The decrease in concentrations with distance from the plant outfall is due mainly to adsorption or ion exchange of radionuclides with clay minerals in the channel and aquifer. Minor decreases are due to dilution with waste water and storm runoff.

During the period 1967 to 1978, the greatest concentrations of ^3H occurred in the Middle Canyon. The ^3H is mobile, moving with water in the aquifer.

B. Inventory of Radionuclides

The annual inventory of radionuclides in solution in the aquifer was computed in three sections, Upper, Middle, and Lower Canyons, using the average concentrations and volume of storage in each section. Inventories of each section were combined for the total in the aquifer. Cumulative totals of radionuclide released with the liquid wastes from 1963 to 1978 are compared with the amounts in storage in 1978.⁷

The gross alpha activity in the aquifer has increased from 46 μCi in 1967 to 495 μCi in 1978 (Table IV). The largest increase occurred in 1976 to 1978 (Fig. 5). Water in the aquifer contains a certain amount of naturally occurring alpha emitters. In a similar aquifer free from industrial effluents, the concentrations average about 0.8 pCi/l. The natural alpha activity in the aquifer has declined from 24 μCi in 1967 to 14 μCi in 1978 (Fig. 5). The amount of gross alpha activity released with effluents from the plant at TA-50 (1963 to 1978) was 42.8 mCi (Table V). The 495 μCi in the aquifer in 1978 represents about 1% of the total amount released with effluents.

Gross beta activity in the aquifer increased from 994 μCi in 1967 to 3036 μCi in 1978 (Table IV). Inventories have varied, increasing from 1967 to a high in 1972, then declining in 1973, and increasing to 1978 (Fig. 6). The high that occurred in 1971 was 8457 μCi and is related to a release of a large amount of beta-emitting radionuclides released from the treatment plant. Naturally occurring beta activity in water in a similar aquifer is about 3.3 pCi/l. The inventory of naturally occurring beta activity has ranged from 99 μCi in 1967 to about 49 μCi in 1978. The gross beta activity released with effluents (1963 to 1978) has been about 8453 mCi

TABLE II

CONCENTRATIONS AND DISTRIBUTION OF RADIONUCLIDES IN THE
UPPER, MIDDLE, AND LOWER CANYONS
(pCi/l, except as noted)

Concentrations 1967 to 1978										
Year	Gross Alpha	Gross Beta	¹³⁷ Cs	²³⁸ Pu	²³⁹ Pu	Total Pu	²⁴¹ Am	⁹⁰ Sr	³ H ^a	Total U ^b
1967	1.5	33	-	0.05	0.07	0.12	-	-	293	2.0
1968	2.3	44	-	0.09	0.38	0.47	-	-	129	1.1
1969	2.9	40	-	0.06	0.10	0.16	-	-	56	0.8
1970	3.6	74	-	0.15	0.12	0.27	-	-	485	0.8
1971	3.2	291	-	0.36	0.06	0.42	-	-	72	1.9
1972	3.7	174	-	0.56	0.18	0.74	0.36	-	78	3.3
1973	5.9	63	65	1.51	0.18	1.69	0.80	-	63	-
1974	4.0	92	61	1.78	0.26	2.04	-	-	42	5.2
1975	6.3	80	-	1.52	0.21	1.73	1.31	-	47	5.4
1976	23.0	124	14	2.05	0.28	2.33	0.35	36.4	1206	6.8
1977	29.2	198	72	2.48	0.34	2.82	-	-	521	10.1
1978	27.5	169	27	3.82	0.68	4.50	-	15.3	211	11.1

Distribution of Concentrations 1967 to 1978

	Gross Alpha	Gross Beta	¹³⁷ Cs	²³⁸ Pu	²³⁹ Pu	Total Pu	²⁴¹ Am	⁹⁰ Sr	³ H ^a	Total U ^b
Average Concentrations										
Upper Canyon	17	338	50	3.66	0.56	4.22	1.61	65.5	234	4.8
Middle Canyon	10	122	41	1.42	0.34	1.76	0.58	35.5	296	4.2
Lower Canyon	7	42	46	0.30	0.10	0.40	0.44	2.1	227	3.5
Percentage of Distribution										
Upper Canyon	50	67	36	68	56	66	61	64	31	38
Middle Canyon	29	24	30	26	34	27	22	34	39	34
Lower Canyon	21	9	34	6	10	17	17	2	30	28

^apCi/ml.^bμg/l.

TABLE III
INVENTORY OF PLUTONIUM RELEASED INTO MORTANDAD CANYON AND
RATIO OF $^{238}\text{Pu}/^{239}\text{Pu}$ IN EFFLUENT AND SHALLOW AQUIFER

Year	mCi				Ratio $^{238}\text{Pu}/^{239}\text{Pu}$	
	^{238}Pu	^{239}Pu	Total Pu		Effluent TA-50	Shallow Aquifer
			Annual	Cumulative ^a		
1967	-	-	4.2	12.8	-	0.4
1968	-	-	2.6	15.4	-	0.3
1969	-	-	6.8	22.2	-	0.8
1970	-	-	5.0	27.2	-	2
1971	-	-	6.9	34.1	-	13
1972	7.7	0.3	8.0	42.1	26	4
1973	8.4	0.6	9.0	51.1	14	11
1974	11.4	0.4	11.8	62.9	28	8
1975	14.8	0.7	15.5	78.4	21	8
1976	7.5	1.0	8.5	86.9	8	7
1977	2.6	1.5	4.1	91.0	2	8
1978	4.0	1.8	5.8	96.8	2	6

^aSince 1963.

(Table V). The 3036 μCi in the water in 1978 is only 0.04% of the amount released into the canyon.

The amount of ^{137}Cs (5 yr of record) has varied from 1160 μCi in 1974 to 232 μCi in 1976. The amount in storage in the aquifer in 1978 was 480 μCi (Table IV). This represents 0.04% of the total amount, 1215 mCi, released into the canyon from 1963 through 1978.

The amount of ^{238}Pu has increased from <1.5 μCi in 1967 to 68.8 μCi in 1978 (Table IV, Fig. 7). The ^{238}Pu was a minor part of the total plutonium released into the canyon before 1970 (Table III). Since 1970 the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio indicates that there is more ^{238}Pu than ^{239}Pu released with the effluents. The amount of ^{238}Pu released with the effluents (1963 to 1978) is about 55 mCi (Table V). Of this total, 68.8 μCi or about 0.1% has remained in solution in the aquifer in 1978.

The inventory of ^{239}Pu has varied from 1.7 μCi in 1971 to 12.3 μCi in 1978 (Table IV, Fig. 7). The amount of ^{239}Pu released with the effluents (1963 to 1978) has been about 41 mCi (Table V). Of this total, 12.3 μCi or 0.03% remained in solution in the aquifer in 1978.

The inventory of ^{241}Am was determined for four separate years (1972, 1973, 1975, and 1976). The

amount increased from 8.2 μCi in 1973 to 24.9 μCi in 1975 and then decreased to 5.9 μCi in 1976 (Table IV). The amount of ^{241}Am released into the canyon (1963 to 1976) was about 5.1 mCi. Of the 5.1 mCi released into the canyon by 1976, only 5.9 μCi or 0.1% remained in solution in the aquifer in 1976 (Table IV).

The inventory of ^{90}Sr in the aquifer was measured for 1976 (619 μCi) and 1978 (276 μCi). The amount of ^{90}Sr released into the canyon through 1978 was 304 mCi. Of this total, 276 μCi or 0.09% remained in solution in the aquifer.

The inventory of ^3H in the aquifer indicates three periods during which large amounts of ^3H were in storage: 8.8 Ci in 1967, 9.7 Ci in 1970, and 20.5 Ci in 1976. The three high periods do not represent the ^3H routinely found in the effluents but are the result of decontamination of ^3H -contaminated equipment (Fig. 8). The ^3H is generally released over a 2- or 3-day period with the effluents. It is estimated that the 1967 release contained 15 to 20 Ci. The amount released in November 1969 was about 20 Ci. Of this total, about 9.7 Ci or 49% remained in the aquifer at the end of 1970. By 1974, only 0.8 Ci or 4% of the 20 Ci released remained

TABLE IV

INVENTORIES AND DISTRIBUTION OF INVENTORIES OF
RADIONUCLIDES IN UPPER, MIDDLE, AND LOWER CANYONS
(μCi , except as noted)

Inventory 1967 to 1978										
Year	Gross Alpha	Gross Beta	^{137}Cs	^{238}Pu	^{239}Pu	Total Pu	^{241}Am	^{90}Sr	$^3\text{H}^a$	Total U ^b
1967	46	994	-	<1.5	2.1	3.6	-	-	8.8	60.3
1968	55	1069	-	2.2	9.2	11.4	-	-	3.1	26.3
1969	73	1002	-	1.5	2.4	3.9	-	-	1.4	20.8
1970	72	1482	-	3.1	2.3	5.4	-	-	9.7	16.5
1971	94	8457	-	10.3	1.7	12.0	-	-	2.1	57.2
1972	84	4012	-	12.8	4.2	17.0	8.2	-	1.8	75.7
1973	112	1196	846	28.7	3.5	32.2	15.1	-	1.2	-
1974	77	1757	1160	33.9	4.9	38.8	-	-	0.8	99.5
1975	120	1522	-	28.8	3.9	32.7	24.9	-	0.9	103
1976	392	2112	232	34.8	4.7	39.5	5.9	619	20.5	116
1977	410	2768	1011	34.7	4.8	39.5	-	-	7.3	141
1978	495	3036	480	68.8	12.3	81.1	-	276	3.8	200

Distribution of Inventory 1967 to 1978

	Gross Alpha	Gross Beta	^{137}Cs	^{238}Pu	^{239}Pu	Total Pu	^{241}Am	^{90}Sr	$^3\text{H}^a$	Total U ^b
Average Inventory										
Upper Canyon	58	1303	196	11.9	1.9	13.8	7.0	197	0.8	17.5
Middle Canyon	44	595	172	6.4	1.4	7.8	2.8	236	1.8	19.8
Lower Canyon	68	554	417	3.4	1.4	4.8	8.7	16	2.5	40.0
Per Cent Distribution										
Upper Canyon	34	53	25	55	40	52	15	44	16	23
Middle Canyon	26	24	22	29	30	30	21	53	35	25
Lower Canyon	40	23	53	16	30	18	64	3	49	52

^aCuries.

^bGrams.

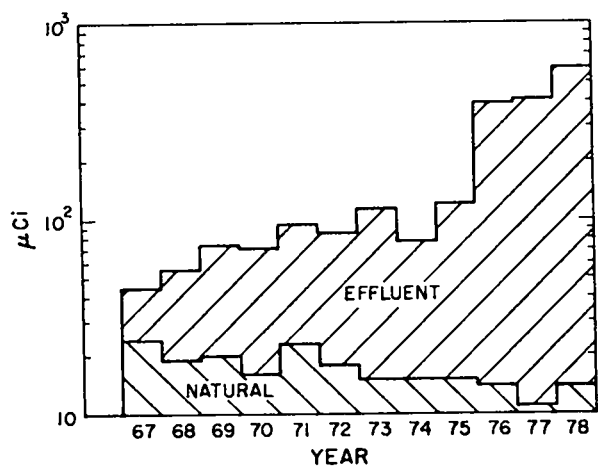


Fig. 5. Amount of gross alpha radioactivity in the aquifer, 1967 to 1978.

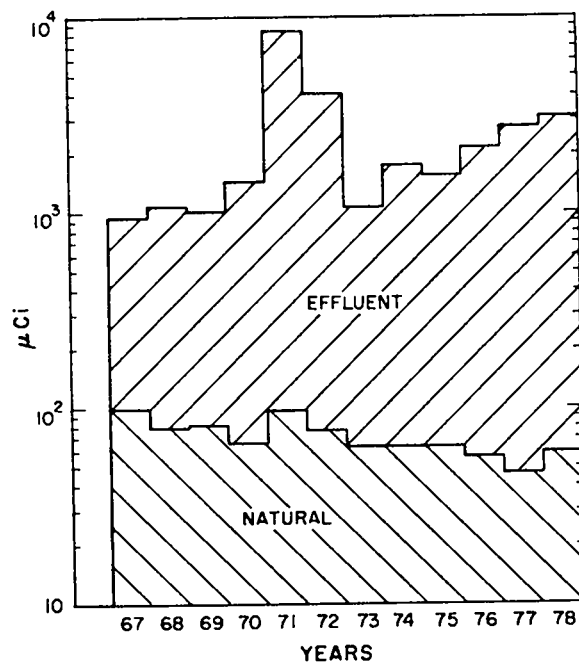


Fig. 6. Annual amounts of gross beta radioactivity in the canyon, 1967 to 1978.

TABLE V

CUMULATIVE TOTAL OF RADIONUCLIDES RELEASED WITH EFFLUENTS FROM TA-50 AND CONCENTRATIONS AND TOTALS IN SHALLOW AQUIFER IN 1978

Radionuclide	Cumulative TA-50 (mCi)	Shallow Aquifer	
		Average Concentration (pCi/l)	Total (μCi)
Gross alpha (unidentified)	42.8	27.5	495
Gross beta (unidentified)	8453	169	3036
¹³⁷ Cs	1215	27	480
²³⁸ Pu	55.4	3.8	68.8
²³⁹ Pu	41.4	0.68	12.3
Total Pu	96.8	4.5	81.1
²⁴¹ Am ^a	5.1	0.35	5.9
⁹⁰ Sr	304.4	15	276
³ H	263 300	211 000	3810
Total U	1225 ^b	11.1 ^c	200 ^b

^a1976.

^bGrams.

^cμg/l.

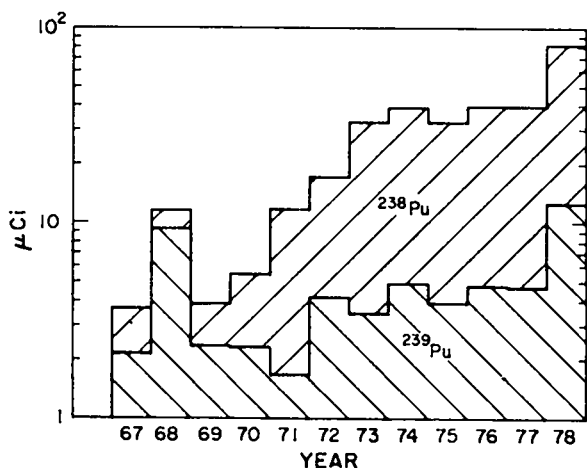


Fig. 7. Amount of ^{238}Pu and ^{239}Pu in aquifer, 1967 to 1978.

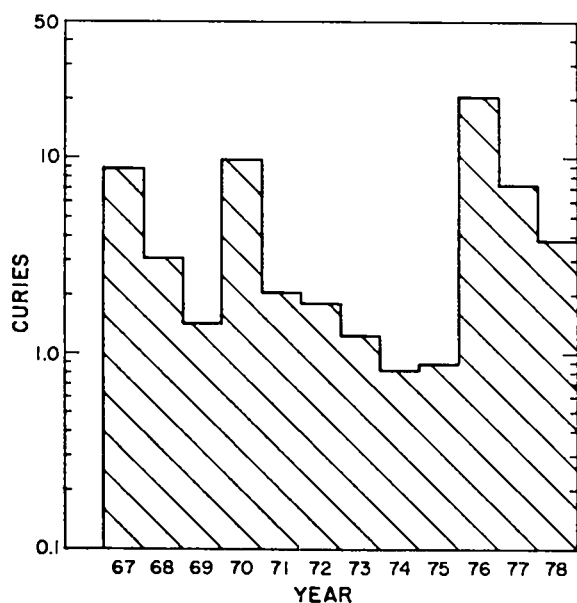


Fig. 8. Amount of tritium in the aquifer, 1967 to 1978.

in the aquifer. The dispersion and movement of ^3H and its use as a tracer in this release are described in Ref. 2. About 40 Ci of ^3H were released with the effluents in March 1975. By the end of the year, 20.6 Ci or 52% of the 40 Ci released remained in the aquifer.

The cumulative total of ^3H released through 1978 was 263.3 Ci. In 1978, 3.8 Ci or about 1% remained in the aquifer. The amount of ^3H in the aquifer is reduced by

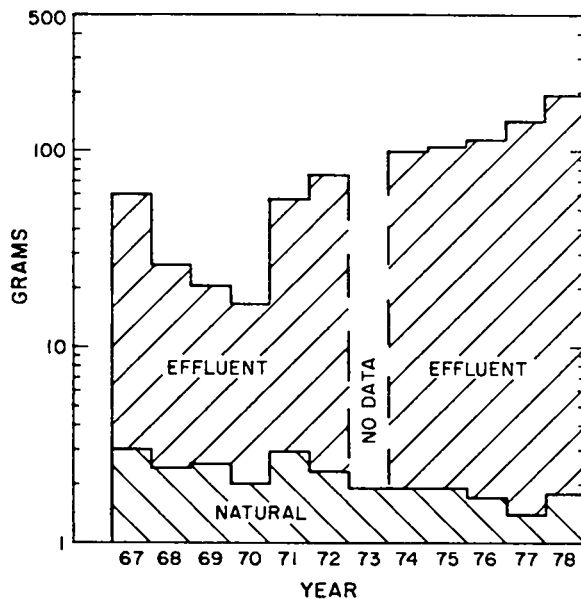


Fig. 9. Annual amount of total U in the aquifer, 1967 to 1978.

radioactive decay (half-life, 12.25 yr) and evaporation and transpiration through vegetation.

The inventory of total U varied considerably from 1967 to 1973 but showed a general decline from 60.3 g in 1967 to 16.5 g in 1970 (Table IV, Fig. 9). The inventory increased from 16.5 g in 1970 to 200 g in 1978. The natural total U in water of a similar aquifer is about $1 \mu\text{g}/\ell$; thus, the amount of natural total U in the water has ranged from 14 g to 30 g, depending on the volume in storage in the aquifer. The amount of total U released from the plant (1963 to 1978) was 1225 g. Of that total, 200 g or 16% remains in the water. Of the 200 g in the water, 18 g is attributed to natural uranium.

C. Distribution of Inventory in the Canyon

The volume of water in storage in the aquifer increased from the Upper Canyon into the Lower Canyon. The division of storage in 1967 to 1978 was 18% in the Upper Canyon, 23% in the Middle Canyon, and 59% in the Lower Canyon (Table I). The distribution of the inventory was made using the mean of 1967 to 1978 inventories for each section of canyon (Fig. 10).

The larger inventories of gross beta (53%), ^{238}Pu (55%), and ^{239}Pu (40%) occurred in the Upper Canyon,

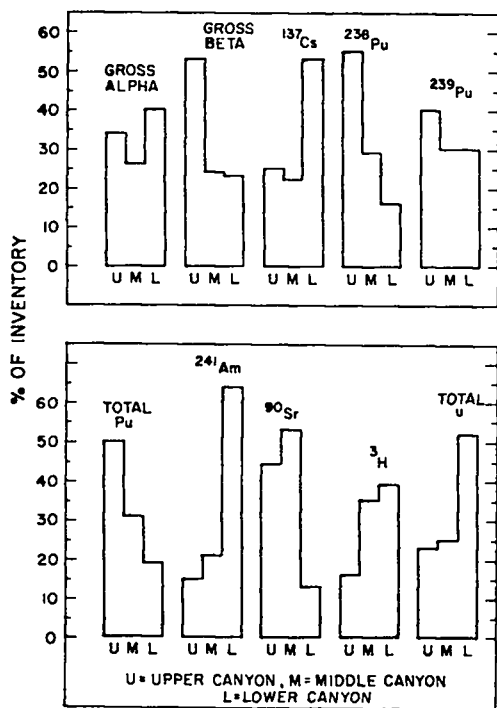


Fig. 10. Distribution of radioactivity in Upper, Middle, and Lower Canyons.

decreasing into the Middle and Lower Canyons. The larger inventories for ⁹⁰Sr (53%) occurred in the Middle Canyon (Table IV). The larger inventories for gross alpha (40%), ²⁴¹Am (64%), ³H (49%), ¹³⁷Cs (53%), and total U (52%) occurred in the Lower Canyon.

The variation in the distribution of the inventories in the three sections of the canyon is related in part to adsorption or ion exchange of radionuclides with silts and clay in the aquifer. The larger inventories of gross beta, ²³⁸Pu, and ²³⁹Pu in solution in the Upper Canyon indicate a greater adsorption of those radionuclides with silts and clays than occurs with ²⁴¹Am, ⁹⁰Sr, ¹³⁷Cs, or total U. These observations are consistent with other reported movements of radionuclides in soils by water.

III. THE PROJECTED RADIONUCLIDE INVENTORY AND CONCENTRATIONS IN SHALLOW AQUIFER IN 1990

The projected total of radionuclides released with effluents from TA-50 into the canyon was compiled using the average annual release from 1963 to 1978 extrapolated to 1990 (Table VI). The projected release is

TABLE VI

PROJECTED TOTAL OF RADIONUCLIDES RELEASED WITH EFFLUENTS FROM TA-50 AND PROJECTED CONCENTRATIONS AND TOTALS IN SHALLOW AQUIFER IN 1990

Radionuclide	Cumulative TA-50 (mCi)	Shallow Aquifer	
		Average Concentration (pCi/l)	Total (μCi)
Gross alpha (unidentified)	77	49	890
Gross beta (unidentified)	15 215	304	5465
¹³⁷ Cs	2 187	48	864
²³⁸ Pu	99	6.9	124
²³⁹ Pu	74	1.2	22
Total Pu	173	8.1	146
²⁴¹ Am	16	1.1	19
⁹⁰ Sr	548	28	497
³ H	527 000	424 000	7626
Total U	2 205 ^a	20 ^b	360 ^a

^aGrams.

^bμg/l.

based on past plant operations and assumes that there will be no significant change in amounts of radionuclides received for treatment and that methods of treatment will remain about the same.

The inventory of the amounts of radionuclides in solution in the water of the shallow aquifer was projected to 1990 based on the percentage of radionuclides in the aquifer in 1974 to the total amounts released from 1963 to 1978. The average concentrations in 1990 were determined using the volume in storage in 1978 of 1.8×10^4 m³. Estimated inventories and concentrations in the shallow aquifer will increase about 80% from 1978 to 1990 if the controlling physiochemical interactions remain the same (Tables V and VI). These interactions between water and alluvium in the perched aquifer and the effluents from the treatment plant are controlled by parameters that have not been studied. Behavior of radionuclides in this particular aquifer may be unique to the chemical treatment process. Application of the data to other liquid effluents from other treatment processes will require additional studies of the chemical characteristics of the aquifer and chemical states of the radionuclides.

IV. CONCENTRATION GUIDES FOR RADIOACTIVITY IN WATER

Concentration guides (CG) for radioactivity in water are set forth in the Department of Energy Order 5480.1.⁸ These CGs are used for comparison of the radiochemical quality of water from the shallow aquifer, with the exception of total U. The DOE standard does not consider the chemical toxicity of total U. Thus, for this report the more restrictive CG of the International Commission on Radiological Protection is used as a guide.⁹ The CG for controlled areas is used for comparison of the average concentrations of radioactivity in the shallow aquifer as the aquifer is of limited extent, entirely within the DOE project area.

Average concentrations of gross alpha, ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, ⁹⁰Sr, ³H, and total U in the shallow aquifer in 1978 and projected average concentrations in 1990 are less than 1% of the CG for controlled areas (Table VII). Gross beta radioactivity was about 2% of the CG in 1978 and the projected concentration for 1990 was estimated at 3% of the CG.

TABLE VII
COMPARISON OF AVERAGE CONCENTRATION OF RADIONUCLIDE
IN THE AQUIFER IN 1978 AND PROJECTED CONCENTRATIONS
IN 1990 WITH CG FOR CONTROLLED AREAS

Radionuclide	1978		Projected 1990		
	Average Concentration (pCi/l)	Per Cent CG	Average Concentration (pCi/l)	Per Cent CG	CG (pCi/l)
Gross alpha (unidentified)	27.5	<1	49	<1	1×10^5
Gross beta (unidentified)	169	2	304	3	1×10^4
¹³⁷ Cs	27	<1	48	<1	4×10^5
²³⁸ Pu	3.8	<1	6.9	<1	1×10^5
²³⁹ Pu	0.68	<1	1.2	<1	1×10^5
²⁴¹ Am ^a	0.35	<1	1.1	<1	1×10^5
⁹⁰ Sr	15	<1	28	<1	1×10^4
³ H	211 000	<1	424 000	<1	1×10^8
Total U ^b	11.1	<1	20	<1	6×10^4

^a1976.

^bμg/l.

V. SUMMARY

Inflow of surface water (effluents from TA-50, waste water from TA-48, and storm runoff) is about equal to the surface and ground water loss from the shallow aquifer. Storage in the aquifer has varied depending on the amount of surface water inflow and recharge ranging from 1.5 to 3.0×10^4 m³ from 1967 to 1978. The average distribution of storage from 1967 to 1978 was 18% in the Upper Canyon, 23% in the Middle Canyon, and 59% in the Lower Canyon.

Continued release of effluents accounts for 80 to 99% of the increased concentrations of gross alpha, gross beta, ²³⁸Pu, ²³⁹Pu, and total U observed from 1967 to 1978. Concentrations of ¹³⁷Cs, ²⁴¹Am, ⁹⁰Sr, and ³H have varied, showing some decline for the period of record. The higher concentrations of gross alpha, gross beta, ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, ⁹⁰Sr, and total U occur in the Upper Canyon near the effluent outfall and decrease in concentration downgradient in the aquifer. The decrease in concentration with distance from the plant outfall is due primarily to adsorption or ion exchange of radionuclides with clay minerals in the channel and aquifer and secondarily to dilution with storm runoff and waste water from TA-48. The ³H concentrations were slightly higher in the Middle Canyon in the aquifer during the period 1967 to 1978. The ³H is part of the water molecule and is not affected by adsorption or ion exchange.

Adsorption or ion exchange of radionuclides in the effluent with clay minerals in the channel or aquifer greatly reduces the amount found in water of the shallow aquifer when compared with the amount of radionuclides released with the effluent from 1963 to 1978. Generally, less than 1% of the total amount of gross alpha, gross beta, ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu, and ⁹⁰Sr released with effluents remained in solution in the aquifer in 1978. Of the inventory of ²⁴¹Am in 1976, less than 1% of the amount released with the effluents from 1963 to 1976 remained in solution. The amount of total U in the water in 1978 was about 16% of the total released with effluents from 1963 to 1978.

Less than 1% of ³H remained in the aquifer when compared with the amount released. Tritium is unaffected by adsorption or ion exchange but is removed from the aquifer by evaporation and transpiration through plants and trees.

The distribution of the inventory of radionuclides in storage from 1967 to 1978 indicated the largest amounts

of gross beta, ²³⁸Pu, and ²³⁹Pu were in the Upper Canyon, whereas the larger amount of ⁹⁰Sr was in the Middle Canyon. The Lower Canyon contained the largest inventories of gross alpha, ²⁴¹Am, ³H, ¹³⁷Cs, and total U.

If the amount of radionuclide received at the treatment plant and the methods of treatment remain the same, projected estimated amounts and concentrations in the shallow aquifer will increase about 80% from 1978 to 1990.

The average solution concentrations in the aquifer in 1978 and projected concentrations in 1990 were less than 3% of the concentration guide levels for controlled areas as set forth by the Department of Energy.

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APPENDIX

AVERAGE ANNUAL RADIONUCLIDE CONCENTRATIONS OF WATER
FROM THE SHALLOW AQUIFER, 1967 TO 1978
(pCi/l, except as noted)

Year	Number of Analysis	Observation Holes							
		MCO-3	MCO-4	MCO-5	MCO-6	MCO-7	MCO-7.5	MCO-8	
Gross alpha	1967	1	1.2	0.5	2.0	2.0	2.0	3.0	0.3
	1968	2	1.0	1.0	1.0	6.0	1.0	2.0	0.5
	1969	2	7.0	3.0	0.5	4.0	5.0	1.0	0.9
	1970	6	6.0	3.0	4.0	4.0	2.0	7.0	0.7
	1971	5	21	3.0	1.0	2.0	0.5	1.0	-
	1972	4	11	9.0	3.0	2.0	2.0	1.0	1.0
	1973	3	29	7.0	6.0	5.0	2.6	3.0	0.8
	1974	4	5.5	5.6	3.2	4.8	2.4	3.4	-
	1975	3	8.9	9.0	3.5	6.7	4.5	6.7	4.9
	1976	3	76	17	11	20	14	46	8.0
	1977	3	25	46	11	30	6.4	42	7.0
	1978	2	21	90	14	18	12	22	-
Gross beta	1967	1	116	83	9	7	2	11	10
	1968	2	166	83	45	27	10	9	8
	1969	2	93	129	29	18	9	39	22
	1970	6	505	141	25	31	17	32	10
	1971	4	1470	418	186	168	105	68	-
	1972	4	612	343	112	132	73	70	46
	1973	3	343	18	50	47	43	46	33
	1974	4	210	220	51	77	41	45	-
	1975	3	460	100	33	29	24	33	75
	1976	3	320	275	41	40	28	32	18
	1977	3	490	413	47	54	15	64	25
	1978	2	304	790	66	59	18	42	-
¹³⁷ Cs	1973	3	10	50	30	10	80	50	50
	1974	4	50	70	70	20	20	90	-
	1976	3	9	23	9	-	12	3	8
	1977	3	30	30	87	43	120	93	130
	1978	2	35	75	15	21	15	15	-

APPENDIX (cont)

	Year	Number of Analysis	Observation Holes						
			MCO-3	MCO-4	MCO-5	MCO-6	MCO-7	MCO-7.5	MCO-8
²³⁸ Pu	1967	1	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
	1968	2	0.27	0.13	0.10	0.07	<0.01	0.10	<0.01
	1969	2	0.27	0.14	0.05	<0.01	<0.01	0.07	<0.01
	1970	6	1.0	0.11	0.05	<0.01	<0.01	0.32	<0.01
	1971	5	3.1	0.23	0.09	<0.01	0.08	0.12	-
	1972	4	3.9	0.93	0.13	0.08	0.09	0.08	0.10
	1973	3	7.9	2.5	2.3	0.80	0.13	0.10	0.93
	1974	4	6.1	4.5	2.2	0.99	0.74	0.21	-
	1975	3	5.4	4.1	1.3	0.55	0.59	1.0	0.51
	1976	3	5.8	4.9	0.55	0.34	0.41	0.23	0.15
	1977	3	6.5	5.4	1.2	0.28	0.12	0.30	0.28
	1978	2	5.4	19	0.78	2.2	0.06	0.29	-
²³⁹ Pu	1967	1	0.14	0.06	<0.05	0.09	0.05	0.06	<0.05
	1968	2	0.22	0.43	0.63	0.79	0.10	0.32	0.08
	1969	2	0.35	0.12	0.08	0.06	0.05	0.15	<0.01
	1970	6	0.47	0.08	<0.01	<0.01	<0.01	0.37	<0.01
	1971	5	0.44	0.07	<0.01	<0.01	<0.01	<0.01	-
	1972	4	0.35	0.16	0.08	0.05	0.06	0.11	0.52
	1973	3	0.46	0.38	0.17	0.30	<0.01	0.05	<0.01
	1974	4	0.41	0.84	0.28	0.13	0.09	0.10	-
	1975	3	0.45	0.70	0.21	0.06	0.07	0.12	0.10
	1976	3	0.57	0.72	0.09	0.04	0.03	0.05	0.04
	1977	3	0.59	0.96	0.09	0.04	0.04	0.01	0.01
	1978	2	0.59	3.8	0.19	0.28	0.02	0.06	-
²⁴¹ Am	1972	1	0.50	0.10	0.19	0.22	0.08	1.1	0.31
	1973	1	-	2.4	0.62	0.40	0.80	0.44	0.66
	1975	1	0.36	0.84	0.34	0.47	1.1	0.90	0.24
	1976	1	7.4	1.3	<0.02	0.10	0.04	0.08	-
⁹⁰ Sr	1976	1	62	84	2.5	-	5.1	<0.1	-
	1978	1	36	80	2.6	2.8	0.2	1.6	-

APPENDIX (cont)

Year	Number of Analysis	Observation Holes							
		MCO-3	MCO-4	MCO-5	MCO-6	MCO-7	MCO-7.5	MCO-8	
³ H ^a	1967	10	56	128	119	94	204	389	942
	1968	8	27	38	88	89	132	171	342
	1969	5	36	25	34	34	43	59	141
	1970	45	688	592	695	755	460	326	97
	1971	4	40	49	64	51	84	122	-
	1972	4	47	64	78	60	69	49	130
	1973	3	99	65	51	48	53	61	84
	1974	4	76	44	42	42	38	36	-
	1975	3	195	49	44	24	26	33	41
	1976	3	211	2000	1030	1760	266	205	25
	1977	3	316	371	510	515	240	950	630
	1978	2	95	303	240	305	105	180	-
Total U ^b	1967	1	1.3	<0.4	<0.4	4.6	1.5	3.6	<0.4
	1968	2	0.7	1.2	0.5	1.3	0.8	0.8	1.7
	1969	2	0.9	1.7	0.7	0.5	0.7	0.8	0.9
	1970	6	1.9	1.5	0.5	0.9	<0.4	<0.4	0.8
	1971	4	9.0	3.7	1.4	0.7	0.7	0.7	-
	1972	4	7.6	6.0	2.7	3.5	1.8	1.8	0.9
	1974	4	4.0	9.0	2.7	6.1	4.5	3.6	-
	1975	3	3.6	2.6	2.6	5.4	3.2	11	6.0
	1976	3	3.2	13	4.6	5.1	3.6	6.6	3.2
	1977	3	8.8	18	5.2	9.2	1.9	8.8	4.7
	1978	2	4.3	10	14	8.0	8.2	20	-

^apCi/ml.

^bµg/l.

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