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HW--74536

DE92 005507

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FISSION PRODUCT TRITIUM IN SEPARATIONS WASTES AND IN THE GROUND WATER

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Chemical Effluents Technology
CHEMICAL RESEARCH AND DEVELOPMENT

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HANFORD LABORATORIES OPERATION

August 1, 1962

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General Electric Company, Richland, Washington. Work performed under Contract No. AT(45-1)-1350 for the U. S. Atomic Energy Commission.

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FISSION PRODUCT TRITIUM IN SEPARATIONS WASTES AND IN THE GROUND WATERINTRODUCTION

The fairly recent discovery that tritium is a product of uranium fission^(1,2) prompted the analysis of separations plant waste streams and the local ground water for this radionuclide. The purpose of the sampling and analytical program was to determine the concentrations and relative distribution of tritium in cribbed-waste streams and its concentration and disposition in the local ground water regime. These factors are of interest from the environmental monitoring standpoint; they are also potentially of real value in evaluation of the regional ground water hydrology.

Albenesius⁽²⁾ reported that eight samples of natural uranium irradiated to exposures between 300-1600 MWD/ton gave a fission-to-tritium atom ratio of $1.05 \pm 0.09 \times 10^4$. The calculated yields for the lower and upper exposure limits, respectively, are 3.8 and 20.3 curies of tritium/ton of irradiated uranium. The magnitude of these yields, together with the assumption that the tritium released by fuel dissolution would be prevalent in aqueous waste streams, made it apparent that there should be little difficulty in detecting the radionuclide in process wastes discharged to ground; such was found to be the case.

This report presents the initial results of the process waste and ground water tritium-sampling program. Two-dimensional comparisons of the ground water tritium analyses with routine gross beta analyses are also presented.

SUMMARY

Analyses of composited waste samples from the Purex and Redox separations plants indicate that about 47 to 64 per cent of the theoretical quantity of fission product tritium entering the plants is discharged to ground in process waste streams. Although a precise material balance cannot be calculated at present because of insufficient data, most of the remaining 36 to 53 per cent of charged fission product tritium is believed to be in a gas phase, which is released to the atmosphere, and in high level liquid wastes, which are stored in underground tanks.

Ground water contamination patterns based on tritium concentrations are similar to those based on gross beta emitter and nitrate ion concentrations. The comparatively rapid movement of tritium in the ground water has assisted in defining the preferential paths which other radionuclides may later follow.

Tritium concentrations exceeding the recommended maximum concentrations in drinking water for continuous non-occupational exposure, $1 \times 10^{-3} \mu\text{c/cc}(3)$, were observed in monitoring wells located up to six miles southeast of the 200-East Area.

DISCUSSIONTritium in Separations Waste Streams

Composited samples of condensate wastes from the Purex and Redox Plants were analyzed for tritium to determine the relative distribution and concentrations in streams discharged to ground. Table I shows the percent of the theoretical amount of fission product tritium charged to the dissolvers which can be accounted for in the condensate waste streams discharged to ground.

TABLE ITritium Distribution In Purex And Redox Condensate
Wastes Discharged to Ground

<u>Stream</u>	<u>Per cent of tritium charged appearing</u> <u>in waste</u>	
	<u>Purex</u>	<u>Redox</u>
Process condensate to crib	42-54	27-37
Tank farm condensate to crib	1- 2	27
Acid fractionator condensate to swamp	4- 8	--
Total	47-64	54-64

These values were computed from analytical and material balance data for the last two months in 1961. Corroboration of the analytical data was made with composited samples of selected waste streams collected over the first five months of 1962.

Quite consistent analytical results were obtained on tank farm condensate samples. This was expected since slight variations in tritium concentrations in high-level wastes from month to month would be inconsequential considering the years of storage represented by the waste already in the Purex and Redox tank farms. Although no high-level waste samples were analyzed, it is probable that the tritium concentration in the boiling tanks is nearly identical to the concentration in the condensate. The probability that the overheads and bottoms are nearly identical is supported by results reported by Schumacher⁽⁴⁾.

Of special interest was the tritium content of the stack exhaust from the separations plants. Samples of condensed vapor were collected from the Redox stack over

a 22.5 hour period which encompassed the coating removal step and first dissolver cut⁽⁵⁾. Analyses of the samples indicated that about one per cent of the charged fission product tritium is released to the atmosphere via the stack. About 75 per cent of that released was in the vapor phase, and the remainder was in the gas phase. Albenesius⁽²⁾ reported that 25 per cent of the tritium charged to the dissolvers was found in the gas phase in both laboratory and plant investigations. The variations in these results indicate the advisability of additional stack gas sampling at the Purex and Redox plants.

A sample of coating removal waste was found to contain about one per cent of the charged fission product tritium, which indicates no significant release of the radionuclide as HTO or T₂O during the coating removal step.

Several other wastes (i.e., cell drainages, steam condensates, silica gel rinses, 300-Area cribbed wastes, and UO₃ Plant process condensates) contained low but measurable concentrations of tritium. Due to their relatively small volumes and/or low concentrations, none of these miscellaneous streams accounted for more than one per cent of the tritium charged to the prime separations plants.

The tritium material balance, based on the limited data available, shows that the greatest percentage of the fission product tritium charged is in plant condensate wastes. The tritium content of the stack exhaust and that in the high level wastes probably account for most of the balance. Coating wastes, cell drainages, steam condensates, silica gel rinses, 300-Area cribbed wastes, and the UO₃ Plant process condensates collectively do not account for more than perhaps five per cent of the total tritium charged.

Tritium In The Regional Ground Water

The tritium ground water contamination map, Figure 1, shows contamination patterns resulting from the disposal of wastes to ground in the 200 Areas. Concentrations in the wells are based on the average analyses of samples collected between January and June, 1962; most of the wells were sampled four or five times during this period. Table II presents average tritium analyses for pertinent wells shown on Figure 1.

In general, the contamination patterns outlined on this map are in good agreement with those noted on the gross beta emitter (essentially all Ru^{106} - Rh^{106}) map for the same period, Figure 2. Major differences are noticeable in the northern part of 200-East Area. The source of radiocontaminants at that location is the 216-BY scavenged waste cribsite. Evidently, the concentration of tritium in scavenged wastes was sufficiently low to preclude detection at the current routine detection limit of 1×10^{-5} μc tritium/cc of water. The zone of contamination just south of Gable Mountain is probably attributable to the B-swamp. The Purex acid fractionator condensate (Table I) is the only contributor of tritium to this site.

Ground water contamination patterns to the southeast of 200-East Area show the preferential directions of movement toward the Columbia River. The noticeable "forking" of the contamination about midway to the river is in agreement with hydrologic and geologic studies which show a division of the permeable glaciofluvial sediment in this area⁽⁶⁾ with the more permeable material extending to the east of the dividing point.

FIGURE 1.
Probable Extent Of Ground Water Tritium Contamination, June 1962

FIGURE 1.
Probable Extent Of Ground Water Gross Beta Contamination, June, 1962

Samples of water taken at riverbank locations adjacent to the most eastward contaminated ground water plume did not contain tritium above the detection limit of 1×10^{-5} $\mu\text{c/cc}$. Efforts are now being made, however, to lower the detection limit to 1×10^{-8} by using a more sensitive analytical technique^(7,8). These data should then greatly extend our knowledge of the flow pattern in this general region and aid in detecting the movement of tritium into the Columbia River from separations plants disposal sites.

Relationships of Tritium Concentrations to Radiation Protection Standards⁽⁹⁾

At the present time the ground water under the Hanford reservation is not tapped for use for drinking water supplies. The possibility that this may occur at some future time was recognized, however, in the establishment of disposal criteria for radioactive liquid waste and stated in Radiation Protection Standard 7.2⁽³⁾. This standard also takes into account the probability that liquid wastes discharged into the ground will appear in the Columbia or Yakima Rivers.

Radiation Protection Standard 7.2 states that, "Disposal of a waste to a particular (ground) disposal facility shall cease when certain long-lived radionuclides are detected in the ground water below the facility in concentrations which can be expected to result in concentrations at the point of potential intake (e.g., human consumption) that are above the controls noted in RPS 3.1." For tritium in the absence of other radioactive contaminants a maximum permissible concentration of 1×10^{-3} $\mu\text{c/cc}$ is implied by RPS 3.1 for drinking water used continuously by persons not considered as occupational workers.

The existing standard (RPS 7.2) was developed principally for long-lived radionuclides such as Sr^{90} and Cs^{137} which are effectively retained by the soil column beneath the disposal sites. Although tritium is not appreciably retained in the soil column, the basic objective of the standard " dispersion of the wastes in such a manner as to assure safety of the human population, preserve plant and animal resources, and minimize restrictions on the use of land and water," remains unchanged. The existing concentrations of tritium in the ground water within a few miles of the disposal sites (see Figure 1) are sufficiently high that the tapping of the water from these zones for unrestricted drinking purposes would be inadvisable. Tritium introduced into the ground water from the chemical separations plants has not been detected in any water sources now used for supplies, and on the basis of existing knowledge no significant contamination of such supplies is anticipated from current disposal practices.

ACKNOWLEDGMENTS

The sample preparation work of D. P. Argyle and others in the Radiological Analysis Unit and the sample counting performed by the Internal Dosimetry Unit are gratefully appreciated.

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TABLE IIAverage Tritium Concentration In Well Water Samples, January - June, 1962Detection limit is 1.0×10^{-5} $\mu\text{c}/\text{cc}$ at 90 per cent C.L.

<u>200-West Area Wells</u> (prefixed by 299)	<u>Concentration</u> (units of 10^{-5} $\mu\text{c}/\text{cc}$)	<u>600 Area Wells</u> (prefixed by 699)	<u>Concentration</u> (units of 10^{-5} $\mu\text{c}/\text{cc}$)
W6-1	< detection	17-5	< detection
W10-1	< detection	18-48	< detection
W11-7	< detection	18-43	< detection
W11-11	41	20-20	20
W15-2	< detection	20-EL4	7
W15-4	120	24-33	50
W19-2	150	25-55	1
W22-14	18,000	25-70	38
W22-19	1,400	25-80	< detection
W22-20	20	26-15	30
W23-4	100,000	27-8	7
		30-31	200
<u>200-East Area Wells</u> (Prefixed by 299)		31-65	< detection
E23-2	20	32-70	< detection
E24-1	1,100	34-39A	280
E24-4	< detection	34-51	< detection
E25-3	400	34-88	< detection
E25-4	< detection	35-9	38
E25-6	50	36-61B	< detection
E25-12	16	37-42	180
E26-4	200	37-82	< detection
E28-6	50	38-70	60
E28-8	< detection	39-79	< detection
E28-10	2	40-1	< detection
E32-1	< detection	40-24	2
E33-4	< detection	40-33	22
		42-12	< detection
		42-42	100
		45-20	< detection
		45-42	25
		45-69	< detection

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600 Area Wells
(prefixed by 699)

Concentration
(Units of 10^{-5} $\mu\text{c/cc}$)

S12-3	< detection
S3-E12	< detection
1-18	< detection
2-3	< detection
8-17	2
8-32	< detection
9-E8	< detection
15-15	< detection
15-26	220

600 Area Wells
(prefixed by 699)

Concentration
(Units of 10^{-5} $\mu\text{c/cc}$)

47-35	< detection
47-46	3
48-71	< detection
49-55	7
49-57	< detection
49-79	< detection
50-28	1
50-30	1
50-42	2
50-53	4
50-85	< detection
51-63	< detection
51-75	1
53-55	1
54-42	4
54-75	1
55-50	< detection
55-76	< detection
55-89	< detection
57-83	< detection
59-80B	< detection
60-60	< detection
61-66	< detection
62-43	< detection
63-90	< detection
65-50	< detection
65-72	< detection
71-84	< detection

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