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Conf-820418--5

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STUDY OF TECHNETIUM UPTAKE IN VEGETATION IN THE VICINITY OF THE PORTSMOUTH GASEOUS DIFFUSION PLANT

By

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*To be presented at the 1982 UCCND-GAT Environmental Protection Seminar to be held in Gatlinburg, Tennessee, April 5-7, 1982.

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STUDY OF TECHNETIUM UPTAKE IN VEGETATION IN THE VICINITY OF THE PORTSMOUTH GASEOUS DIFFUSION PLANT

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KEY WORDS: Technetium, Vegetation, Soil, Concentration, Uptake, Dose

ABSTRACT

Technetium-99 was measured in vegetation and soil collected on and near the Portsmouth Gaseous Diffusion Plant to obtain an estimate of the soil-to-vegetation concentration factors. The concentration factors appear to be lognormally distributed with a geometric mean of 3.4 (Bq/kg dry wt. tissue per Bq/kg dry wt. soil) and a geometric standard deviation of 4.7. A dose commitment was calculated using a hypothetical 3.7×10^{10} Bq Tc-99/year release and the actual CY-1981 concentration release of Tc-99. The radiological significance of Tc-99 in the terrestial food chain is substantially less than previously believed.

INTRODUCTION

Technetium-99, a low energy beta emitter, is probably the most notable of the several isotopes of technetium, all of which are radioactive. Technetium-99 is a fission waste product that is present in spent nuclear reactor fuels. It has a half-life of 2.12×10^5 years and emits a beta particle with an energy of 0.292 MeV. Approximately 0.84 kg of technetium-99 is produced per metric ton of reactor fuel in a typical pressurized water reactor.[7] The nuclear fuel cycle accounts for most of the technetium-99 released to the environment and the reprocessing segment of the cycle is accountable for the majority of technetium released from this source. Other sources include medical diagnostic practices, atmospheric detonation of atomic weapons, and the natural fission of uranium-238 in the environment.

The atmospheric form of technetium will generally be as the pertechnetate ion, TcO_4^-. Although technetium will exhibit valencies of 0 to +7, the +7 valence state, the pertechnetate ion, is the most common and exists in aqueous solutions over a wide pH range. Pertechnetate is the basic chemical form of technetium used in nuclear medicine and the final form following the reprocessing of spent reactor fuel elements.[2]

There is relatively little information available concerning what happens to technetium in soils and plants. However, the literature does suggest a variety of mechanisms by which technetium may react with the organic and mineral components of natural soil systems. These mechanisms include ion-exchange, precipitation/coprecipitation and complexation/chelation.[2] Studies performed in soil systems have shown that precipitation may occur in soils with high concentrations of free iron, aluminum, and silica. Some of the studies were performed in non-soil systems and under somewhat idealized conditions, contributing to the difficulties in characterizing the fate technetium in soil and vegetation.[2]

Background

In 1975 and 1976, two studies were conducted and results published in an effort to characterize the uptake of technetium by plants. Wildung, <u>et. al.</u>, presented the results of an experiment in which they studied the uptake of technetium in wheat and soybeans

(Table 1).[1] In the study, the soils were watered with solutions of various technetium concentrations. It was found that at low concentrations of technetium (≤0.01 g Tc-99/g soil), the plants were similar in appearance to the controls. However, at higher concentrations (≥0.1 g Tc-99/g soil) toxic symptoms began to appear. At the concentrations of technetium used, it was not clear whether the toxic symptoms were due to radiation effects or chemical effects. To gain some insight as to what produced the toxic effects, anion competition studies were performed and indicated that pertechnetate ions may be taken up as a nutrient analog. However, this does not eliminate radiation effects as the cause of toxicity.

Table 1 Concentration Factors of Wheat and Soybeans at Various Tc-99 Soil Concentrations[1]

Soil Tc-99 Concentration	Concentration Factor (Bq/kg dry wt. tissue per Bq/kg dry wt. soil) Wheat	Soybeans
0.001	340	380
0.01	145	138
0.1	173	67
1.0	*	376
5.0	*	380

* No growth occurred.

In 1976 Gast, et. al.,[2] published the results of an experiment in which wheat seedlings were planted in various samples of Minnesota soils (Table 2). These seedlings were watered with technetium solutions ranging in concentration from 0 to 11.6 g Tc-99/ml of solution. Additionally, studies were performed to determine the toxic effects of technetium at very low concentrations, ranging from 0 to 3.7×10^4 Bq Tc-99 per 50 ml of solution.

Table 2 Concentration Factors for Wheat Seedlings in Samples Minnesota Soils (Bq/kg dry wt. tissue per Bq/kg dry wt. soil)

Sample Locations	Concentration Factor Unfertilized	Fertilized
Bearden	830	460
Hegne	1,120	760
Hibbing	925	715
Nicollet (surface)	1,200	875
Nicollet (subsurface)	1,065	995
Omega	655	445
Bergland	955	800
Arveson	685	250
Waukegan	1,160	875
Zimmerman	1,055	825

Because these experiments were inconclusive as to whether the toxicity was due to chemical or radiation effects, Gast made further efforts to determine the cause of toxicity. The dose rate was calculated at 1.6×10^{-1} Sv at the highest level of added technetium. These dose rates appear quite low when compared to the dose rates required in other species of plants to inhibit growth. Other studies have shown that x-radiation exposures of 4.0-5.0 Sv are required to produce a 20% growth inhibition.[2]

Prior to the mid-1970s when these two reports were published, most of the assessment literature had been using a concentration factor of 0.25 (Bq/kg wet weight plant tissue per Bq/kg dry weight soil). This value was based upon a series of assumptions relating the behavior of technetium to the behavior of iodine in the environment.[7] The concentration factors determined by Wildung[1] and Gast[2] were as much as three orders of magnitude greater than the assumed value of 0.25. These reports caused some concern among those agencies responsible for calculating dose assessments.

Two responses were forthcoming from this concern. One response was to get out into the field and actually take measurements from the vicinity of the reprocessing plants. The other response pointed out some possible discrepancies between the laboratory experiments and field conditions.[3, 7] The three major discrepancies are: 1) In the laboratory experiments, the plants were not allowed to grow to maturity. 2) The concentration of technetium was significantly higher than one would expect to find in the vicinity of a reprocessing plant. 3) No effort was made to reach an equilibrium level between the soil and plant.

In 1979, a report[7] was written using the generally assumed concentration factor of 0.25 and an author-assumed factor of 50 (Bq/kg wet weight tissue per Bq/kg dry weight soil). The factor of 50 represented a middle-of-the-road value between 0.25 and the values found in the two previously mentioned laboratory experiments. A dose rate for individuals was calculated using both factors and an assumed release of 3.7×10^{10} Bq Tc-99/year. The dose rates calculated at a concentration factor of 50 were found to approach/exceed the dose rate limits promulgated by the US EPA in 40 CFR 190[4] for two target organs (Table 3).

Table 3 50 Year Dose Committment (Sv)[7]

Organ	Concentration Factor 0.25	Concentration Factor 50	US EPA Limit
Total Body	8.6×10^{-8}	1.2×10^{-5}	2.5×10^{-4}
Bone	1.5×10^{-7}	2.0×10^{-5}	2.5×10^{-4}
Kidneys	1.9×10^{-7}	2.6×10^{-5}	2.5×10^{-4}
GI Tract	1.3×10^{-6}	1.8×10^{-4}	2.5×10^{-4}
Thyroid	5.7×10^{-6}	8.0×10^{-4}	7.5×10^{-4}

In 1980, the results of a study of the concentration of technetium in soils and vegetation in the vicinity of the three gaseous diffusion plants were published (Table 4).[3] The gaseous diffusion plants are located at Paducah, KY; Oak Ridge, TN; and Portsmouth, OH. The concentration factors at the three plants ranged from 1.4 to 44 (Bq/kg dry weight tissue per Bq/kg dry weight soil). The data was lognormally distributed and had a geometric mean of 9.5 and a geometric standard deviation of 2.4. The mean concentration factor of 9.5 represents the concentration of technetium in the dry weight of tissue. If expressed as the concentration per wet weight of tissue the concentration factor becomes 2.4 (assuming 75% of the tissue weight is water). This factor alone would reduce the dose to the thyroid by approximately twenty times as compared to the concentration factor of 50.

Table 4 Statistical Summary of Soil-to-Plant Concentration Factors Combined from All Sampling Sites[3]

Number of Values	24
Maximum Value (Paducah)	44
Minimum Value (Portsmouth)	1.4
Arithmetic Mean	14
Standard Deviation	10
Standard Error	2.0
Geometric Mean	9.5
Geometric Standard Deviation	2.4
Geometric Standard Error	1.2

RESULTS

The concentrations of technetium-99 in vegetation sampled at the Portsmouth Gaseous Diffusion Plant ranged from 34 to 27,200 Bq/kg dry weight tissue. The technetium concentrations in the soil ranged from 34 to 12,800 Bq/kg dry weight soil. The geometric means and standard deviations are 533 Bq/kg and 100 for vegetation, 145 Bq/kg and 52 for soil. The large standard deviations are influenced by the higher concentrations obtained from sampled points RIV-19 and RIV-26.

The concentration factors were calculated by dividing the soil concentration into the vegetation concentration. The highest concentration factors were associated with sample points 19 and 26, with the highest value at each point being 108 and 289, respectively. The concentration factors found at the Portsmouth plant ranged from 0.1 to 289 with a geometric mean of 3.4 (Bq/kg dry weight tissue per Bq/kg dry weight soil) and standard deviation of 4.7 (Table 5). The wide range of data accounts for the large standard deviation.

A histogram plot of the collected data is shown in Figure 1 and appears to most closely resemble an exponential curve.[11] If a exponential curve is the correct plot, one should be able to plot a straight line on logarithmic paper. A log-probability plot of the concentration factors is shown in Figure 2. The assemblage of factors appears to be reasonably lognormal. A correlation factor of 0.97 was obtained using a calculator program developed for normal and lognormal distributions.[12] A statistical summary is shown in Table 6.

Table 6 Statistical Summary of Concentration Factors at the Portsmouth Gaseous Diffusion Plant (Bq/kg dry wt. tissue per Bq/kg dry wt. soil)

Number of Values	61
Maximum Value (Point 26)	289
Minimum Value (Point 29)	0.10
Arithmetic Mean	14.0
Arithmetic Standard Deviation	40.1
Arithmetic Error	5.1
Geometric Mean	3.4
Geometric Standard Deviation	4.7
Geometric Error	1.2
Correlation Factor "r" for Lognormal Plot	0.97

Table 5 Concentration Factors (Bq/kg dry wt. Tissue per Bq/kg dry wt. soil)

SITE	NOV	DEC	JAN	FEB	MAR	APR	MAY	JUN	JULY	AUG	SEP	OCT	TOTAL
3	1.33	5.5					0.32		*	0.67	2.23		5
5									*				0
12	0.86	1.5	0.92	0.52		1.42	1.27	1.01	*		4.6	0.87	9
15									*				0
17									*				0
19	9.09	2.71	20.4			35.3	4.7	29.6	*	21.3	108.6	1.21	9
22	0.69	4.5		0.76			4.64	0.65	*	0.04	4.29	0.96	8
25				10.0					*			3.0	2
26	2.73	1.47			289	52.2	33.3	8.26	*	3.96	19.7	59.1	9
26A	1.33		13.2		1.49		1.57	2.10	*			4.81	6
29		5.5		1.31			9.52	38.0	*	0.78	0.10	2.34	7
32	1.33	2.73				1.70	0.68		*		1.35	1.62	6
TOTAL	7	7	3	4	2	4	8	6		5	7	8	61

* No sample collected.

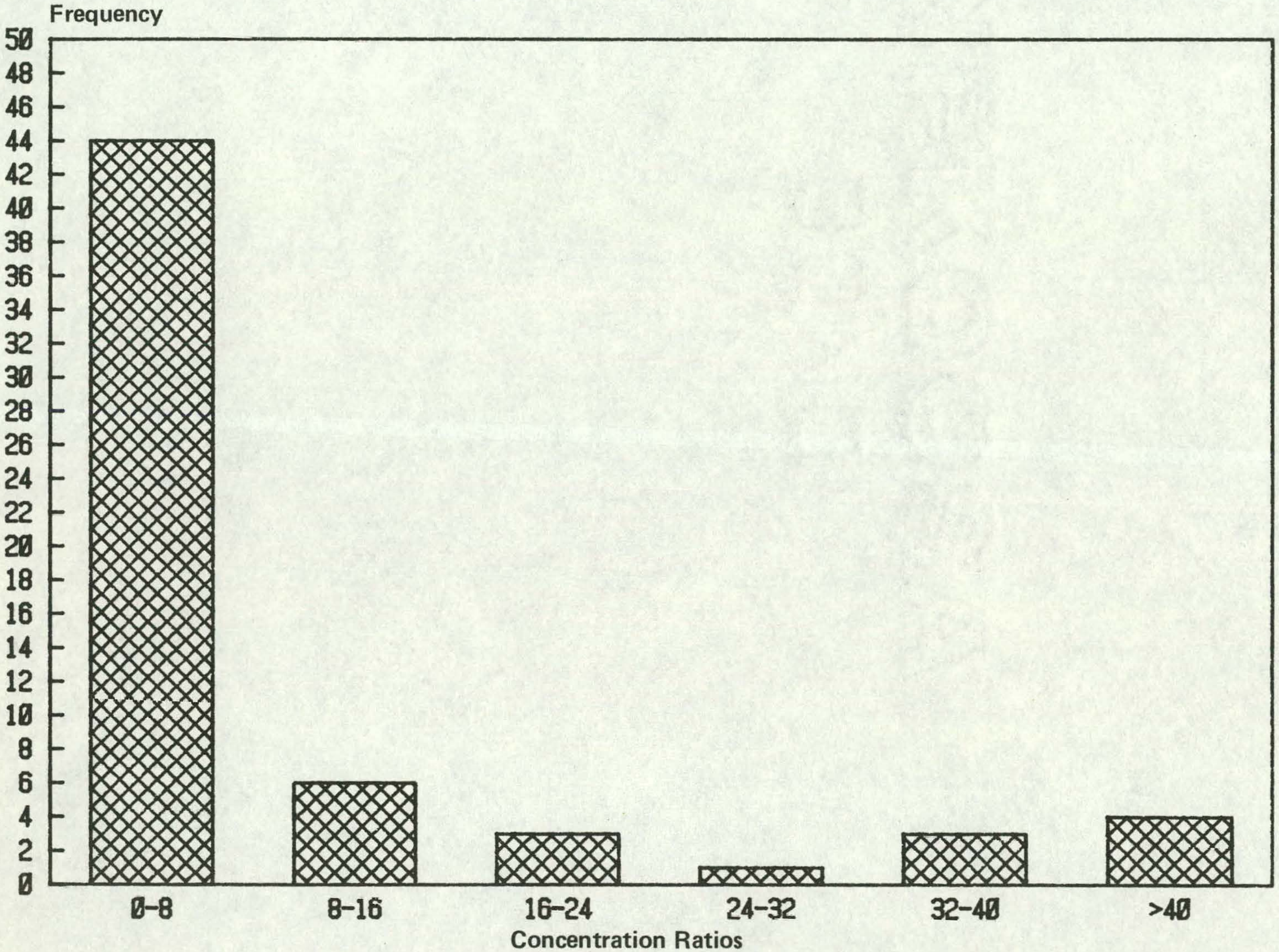

Figure 1 Distribution of Ratios

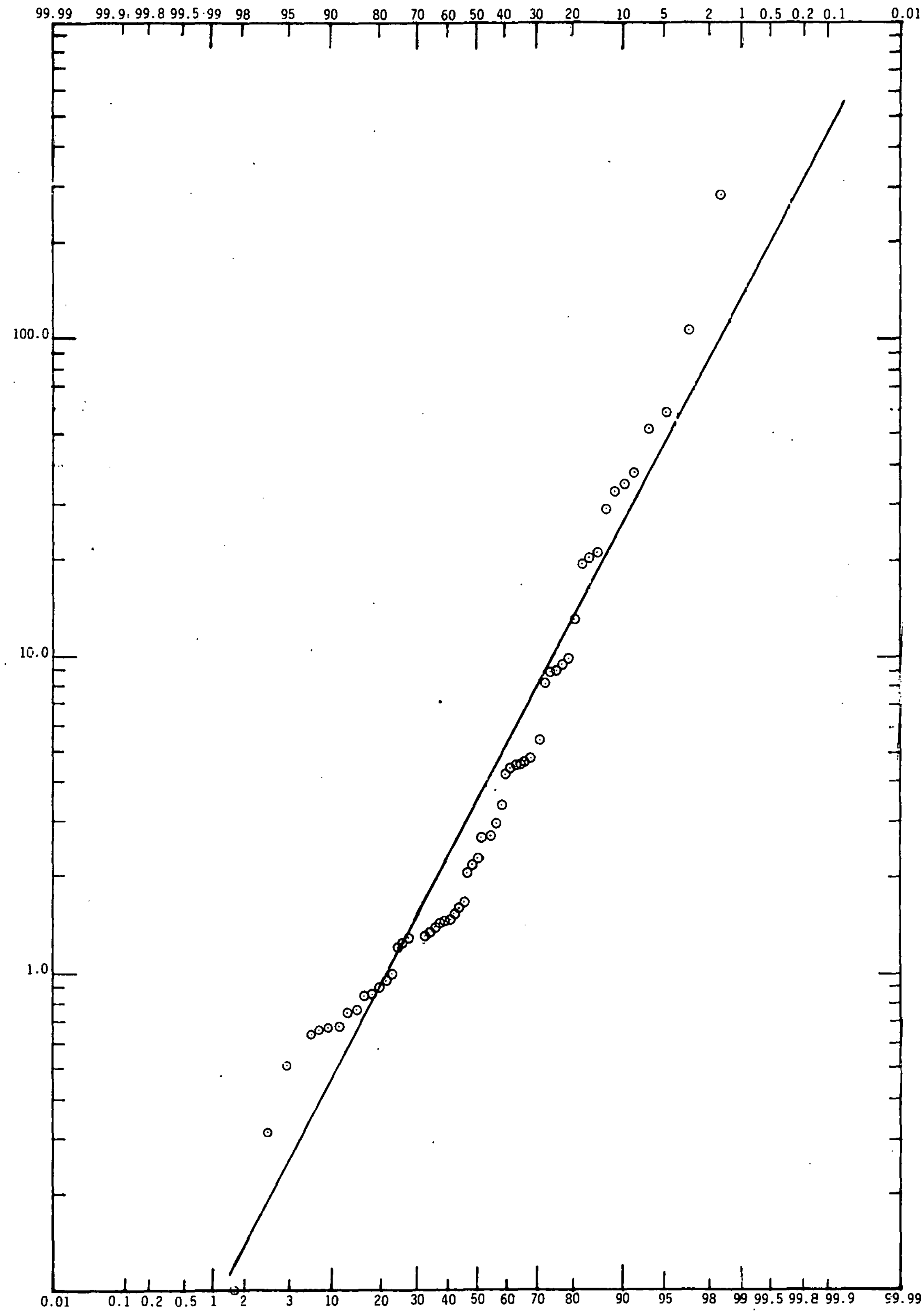

Figure 2 Lognormal Probability Plot of Concentration Factors Found at the Portsmouth Gaseous Diffusion Plant

DISCUSSION

Sampling and Analytical Procedures

At the Portsmouth GDP, the Environmental Control Department routinely samples vegetation and soil both on and off the DOE reservation. These samples are analyzed by laboratory personnel for various chemical and radiological parameters.

The routine vegetation and soil monitoring program consists of a series of monthly samples collected within the DOE reservation boundary. These samples are identified using Routine Internal Vegetation (RIV) and Routine Internal Soil (RIS) sample numbers. On a semi-annual basis, vegetation and soil samples are collected outside the DOE reservation boundary. These samples are identified using Semi-Annual Vegetation/Soil (SAV/SAS) sample numbers. The sample locations are shown in Figures 3 and 4.

The sampling of vegetation involved the collection of above-ground vegetation using shears to cut the sample. A minimum of 500 grams of wide blade grasses is collected. No soil, roots, or foreign matter are collected with the sample as these materials may contaminate the sample. As large an area as needed is sampled to provide the minimum sample quantity.

Soil samples are collected using a shovel to obtain a representative sample. The soil samples consist of approximately the top two inches of soil, as free of roots and stones as possible. The minimum sample consists of a 0.1 square meter area. Again, as large an area as needed is sampled to provide enough soil to fill two 1/2-liter containers.

A method was developed at Goodyear Atomic for the determination of technetium-99 in a variety of samples including vegetation and soils (Figure 5). The method includes destruction of the organic

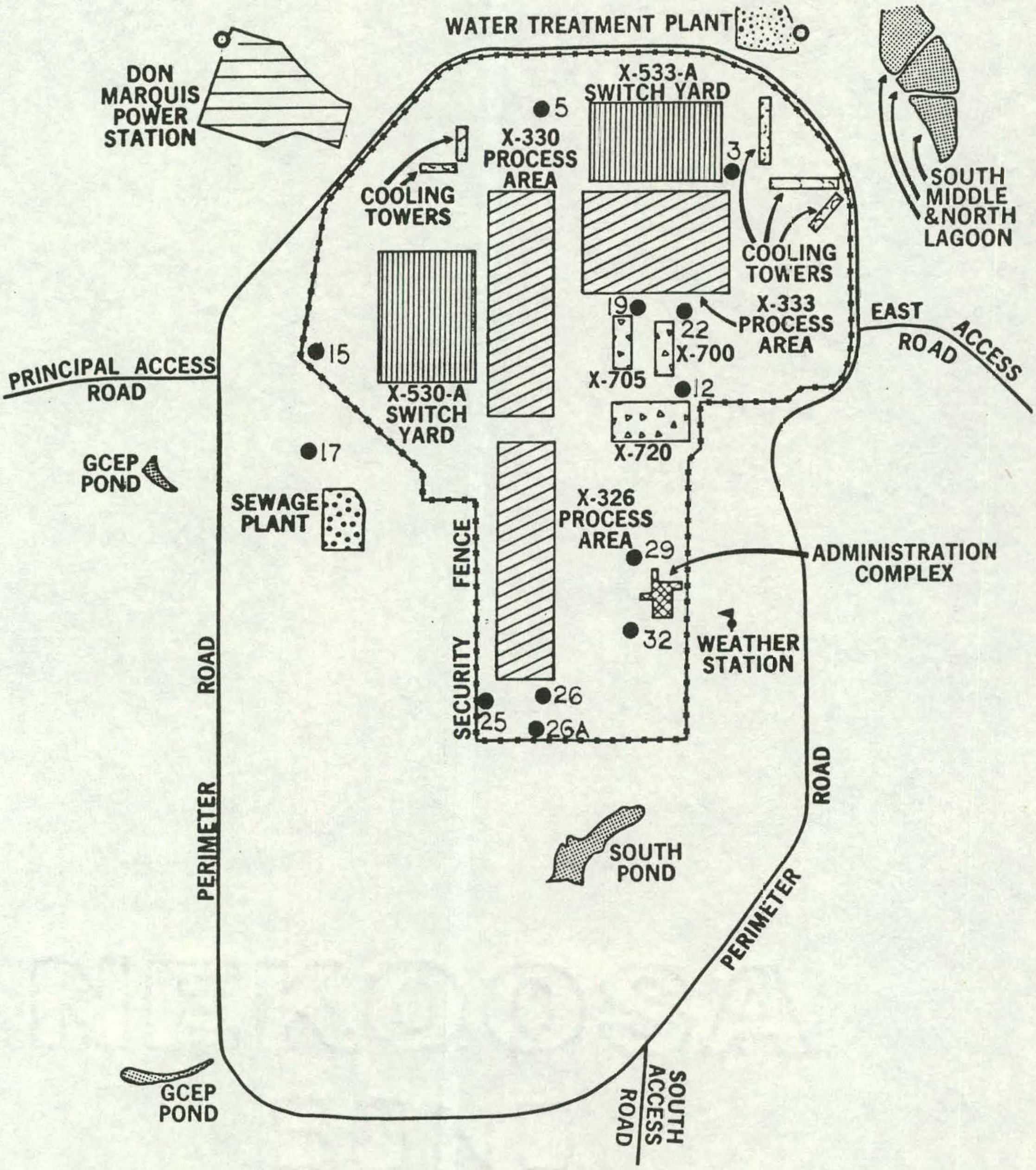

Figure 3 Vegetation and Soil Sampling Locations within the DOE Reservation (RIV and RIS)

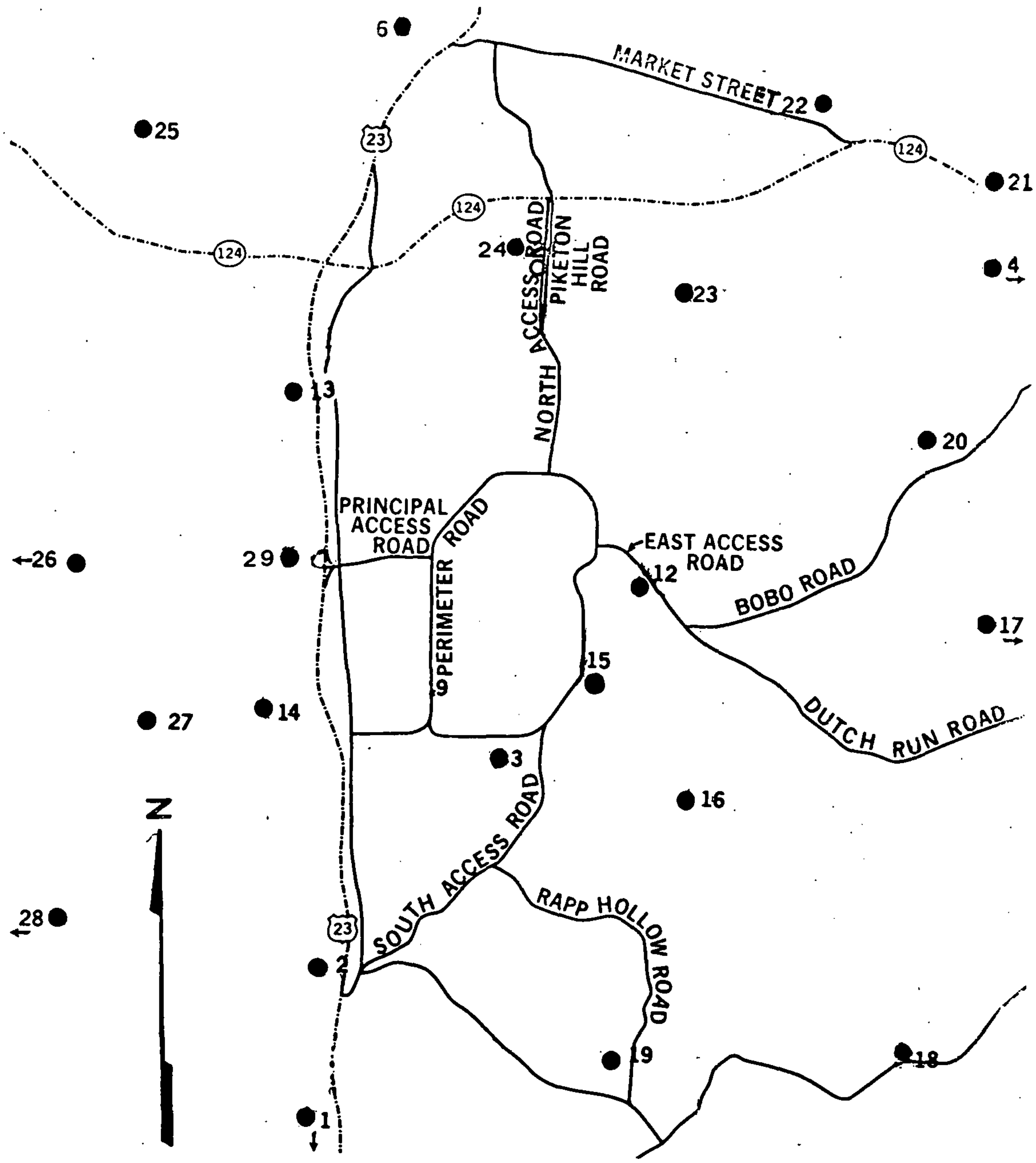

Figure 4 Offsite Vegetation and Soil Sampling Locations (SAV and SAS)

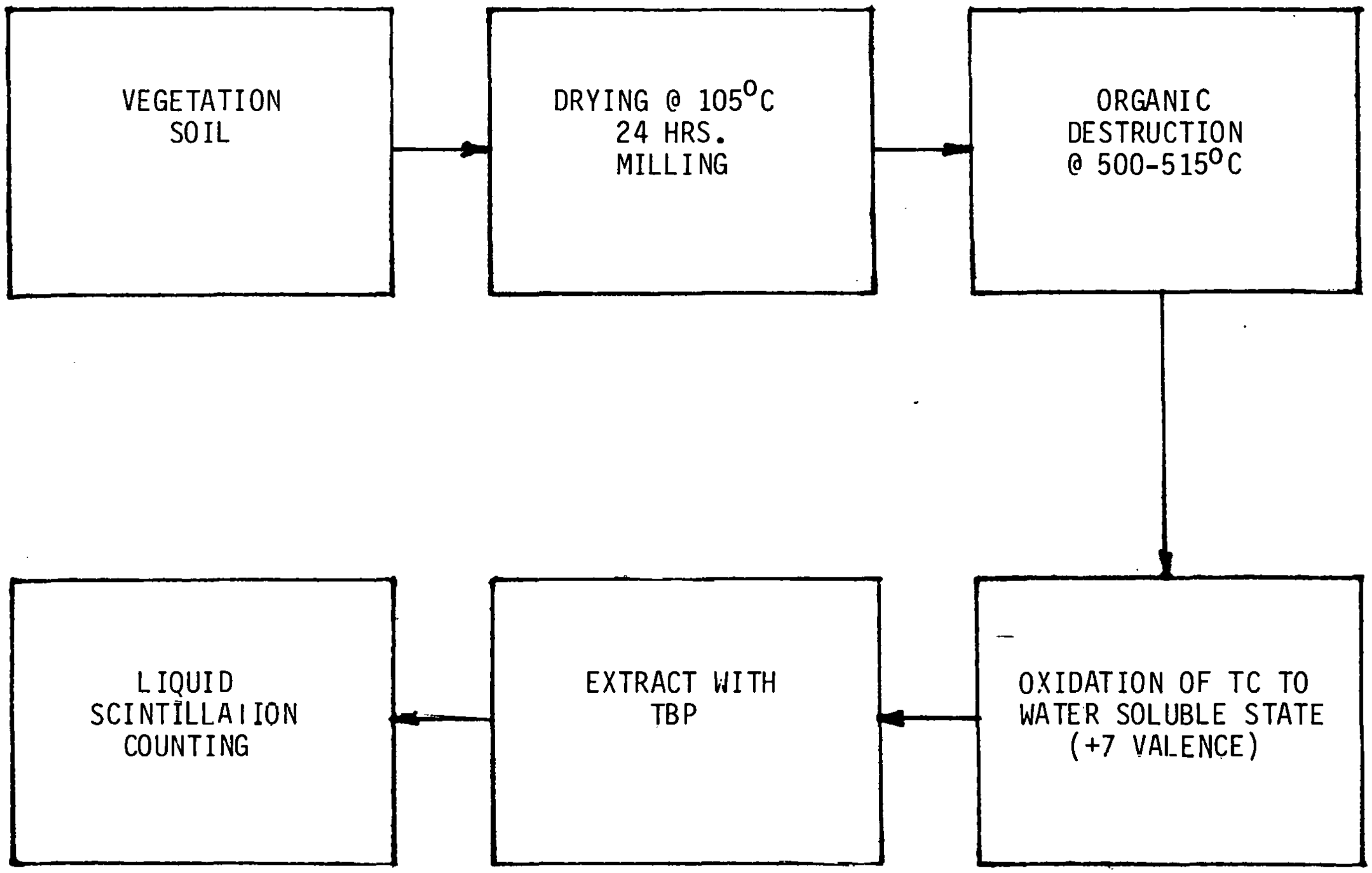

Figure 5 Basic Schematic for Determination of Technetium--99 in Vegetation and Soil

materials, oxidation of all the technetium to the +7 valence state and extraction of the technetium from the aqueous media with tributyl phosphate. A portion of the extract is added to a scintillation cocktail and technetium-99 activity is measured by liquid scintillation counting.[5]

Dose Assessment

In a study performed by Till, et. al.,[7] concentration factors of 0.25 and 50 were used in the calculation of a dose assessment. The atmospheric transport and deposition factors given in Table 7 are based on an assumed 3.7×10^{10} Bq technetium per year release. The calculated dose at each concentration factor was presented earlier. The dose commitment for an individual was recalculated using the same assumed release; however, the concentration factors of 9.5 (obtained from an earlier study) and 3.4 were used. But before being used in the calculations, the factors must be converted from the dry weight tissue factors to the wet weight tissue factors. Assuming the vegetation is 75% water, the concentration factors became 2.4 and 0.85, respectively.

Table 7 Atmospheric Transport and Deposition of Technetium[7]

Release Rate	3.7×10^{10} Bq/yr.
Release Height	20 m
χ/Q at 1600 m	9.9×10^{-7} sec./m^3
Average Annual Deposition Velocity for Wet and Dry Processes	1.1×10^{-2} m/sec.
Deposition Rate at 1600 m	1.12 Bq/m^2 day

After these changes were made, the calculations to determine the dose were performed using the procedure outlined in USNRC Regulatory

Guide 1.109.[8] In calculating the dose commitment, the USNRC assumes a chronic buildup of technetium for fifteen years. However, recent evidence shows there may be an effective half-life of technetium in the soil and vegetation.[3] Forage, harvesting, wind, rain runoff and leaching would tend to remove technetium from the area in question. While these effects may not change the concentration factors, they would lower the dose received by an individual by lowering the amount of technetium received by the individual. However, for calculations, the fifteen year buildup has been assumed. Table 8 shows the calculated concentration of technetium in vegetables, meat, and milk and Table 9 shows the amount of technetium ingested by an individual over a period of one year. The dose commitment is shown in Table 10 for all concentration factors cited. At the assumed release rate, an individual would receive a fifty year dose of approximately 2-6% of the limits set by the US EPA.

Using the actual release rate of technetium at the Portsmouth Gaseous Diffusion Plant, the technetium concentration would be that shown in Table 11 along with the amount of technetium ingested by an individual in one year. Based on this data, an individual would receive the dose commitment shown in Table 12. Using the concentration factor and release rate at the Portsmouth plant, the average individual dose commitment would be less than 1% of the US EPA limits stated in 40 CFR 190.

All of the calculations, thus far, have been performed on vegetation found on the DOE reservation. Semi-annual soil and vegetation samples are collected from thirty locations at varying distances from the reservation. In the past three calendar years, only two semi-annual vegetation samples and three semi-annual soil samples have been found to have greater than 0.03 Bq/gram dry weight (0.03 Bq/gram is the detection limit). The maximum vegetation concentration is 0.07 Bq/gm dry weight and the maximum soil

Table 8 Concentration of Tc-99 in Vegetable, Meat, and Milk at 1600 m

Concentration Ratio		Vegetable	Meat	Milk
Dry Tissue	Wet Tissue	Bq/kg	Bq/kg	Bq/L
1.0	0.25	8.5	4.1	4.8
3.4	0.85	24.0	11.0	13.0
9.5	2.4	63.0	28.0	33.0
200.0	50.0	1,250.0	560.0	630.0

Table 9 Intake Rate of Tc-99 (Bq/yr)

Concentration Ratio					
Dry Tissue	Wet Tissue	Vegetables	Meat	Milk	Total
1.0	0.25	5.6×10^2	4.4×10^2	5.2×10^2	1.5×10^3
3.4	0.85	1.5×10^3	1.3×10^3	1.4×10^3	4.1×10^3
9.5	2.4	4.1×10^3	3.1×10^3	4.1×10^3	1.1×10^4
200	50	3.1×10^4	5.9×10^4	2.1×10^5	3.0×10^5

Table 10 50 Year Dose Committment (Sv)

Concentration Factor						
Dry Tissue	Wet Tissue	Total Body	Bone	Kidneys	GI Tract	Thyroid
1.0	0.25	8.6×10^{-8}	1.5×10^{-7}	1.9×10^{-7}	1.3×10^{-6}	5.7×10^{-6}
3.4	0.85	2.3×10^{-7}	4.0×10^{-7}	5.1×10^{-7}	3.5×10^{-6}	1.6×10^{-5}
9.5	2.4	6.3×10^{-7}	1.1×10^{-6}	1.4×10^{-6}	1.0×10^{-5}	4.2×10^{-5}
200	50	1.2×10^{-5}	2.0×10^{-5}	2.6×10^{-5}	1.8×10^{-4}	8.0×10^{-4}
US EPA Limits		2.5×10^{-4}	2.5×10^{-4}	2.5×10^{-4}	2.5×10^{-4}	7.5×10^{-4}

Table 11 Yearly Intake of Technetium 99 by an Individual Concentration Factor - 0.85 Bq/kg Wet Wt. Tissue per Bq/kg Dry Wt. Soil Release Rate - 8.5×10^{-3} Bq/yr.

	Vegetables	Meat	Milk	Total
Concentration of Tc-99	2.56 Bq/kg	1.24 Bq/kg	1.42 Bq/l	NA
Intake of Tc-99	1.63×10^2 Bq/yr.	1.36×10^2 Bq/yr.	1.55×10^2 Bq/yr.	4.54×10^3 Bq/yr.

* Concentrations of Tc-99 were calculated using a concentration factor of 0.85 and the total CY-1981 release of 4.0×10^9 Bq Tc-99.

Table 12 50 Year Dose Committment (Sv)

	Total Body	Bone	Kidneys	GI Tract	Thyroid
Calculated* Dose	2.58×10^{-8}	4.43×10^{-8}	5.66×10^{-8}	3.94×10^{-7}	1.72×10^{-6}
US EPA Limit	2.5×10^{-4}	2.5×10^{-4}	2.5×10^{-4}	2.5×10^{-4}	7.5×10^{-4}

* Dose was calculated using concentration factor of 0.85.

concentration is 0.3 Bq/gm dry weight. These detectable concentrations of technetium did not occur at the same sample time nor at the same sample locations. Therefore, a concentration factor could not be calculated.

CONCLUSION

There is no evidence that shows technetium has been found off of the DOE reservation. The dose commitment presented earlier indicates that there is no apparent risk to an individual as the result of the deposition of airborne technetium.

ACKNOWLEDGMENT

Special acknowledgment should be given to the people in the Radiochemistry and Spectrochemistry Section and the Environmental and Industrial Hygiene Chemistry Section of the Chemical Analysis Department for their analytical work.

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