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ASSESSMENT OF TECHNETIUM IN THE SAVANNAH RIVER SITE ENVIRONMENT^(U)

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Table of Contents

CONTENTS

<i>Executive Summary</i>	<i>i</i>
<i>Chapter 1. Introduction</i>	1-1
References	1-3
<i>Chapter 2. Origin and Disposition of Technetium at SRS</i>	2-1
Origin of Technetium at SRS	2-3
Disposition of Technetium at SRS	2-3
Measurement of Technetium in the Environment	2-3
Technetium in the Reactor Moderator	2-3
Technetium in Fuel and Targets Irradiated in Production Reactors	2-3
Technetium in Certain Fuels Irradiated Offsite	2-4
Technetium in Fuel and Targets Irradiated in Test Reactors	2-4
References	2-4
<i>Chapter 3. Releases and Stored Inventories of Technetium at SRS</i>	3-1
Atmospheric Releases	3-3
Liquid Releases	3-3
Stored Inventories	3-5
Waste Tanks	3-5
Solid Waste Disposal Facility	3-5
Uranium	3-5
References	3-5
<i>Chapter 4. Technetium Concentrations and Transport Mechanisms</i>	4-1
SRS Groundwater System	4-3
Technetium-99 in SRS Groundwater	4-6
Geochemistry of Technetium	4-6
Origin of ⁹⁹ Tc Contamination	4-7
General Separations Area Seepage Basins	4-7
F-Area Seepage Basins	4-7
H-Area Seepage Basins	4-7
Solid Waste Disposal Facility	4-8
Other Measurements of ⁹⁹ Tc	4-8
General Observations	4-9
References	4-9

Table of Contents

Chapter 5. Assessment of Dose/Risk from SRS Technetium Releases	5-1
Relationship of Dose to Risk and Health Effects.....	5-3
Ionizing Radiation	5-3
Cancer Risk Estimates	5-3
Technetium Exposure and Dose to Man	5-3
Distribution and Retention of Technetium in the Human Body	5-3
Technetium Dosimetry	5-3
Models of Technetium Transport and Dose	5-3
Modeling Atmospheric Dispersion of Radioactive Releases.....	5-4
MAXIGASP	5-5
POPGASP	5-5
Modeling Doses from Liquid Releases	5-5
Validation of Transport Models Using Monitoring Data	5-6
Atmospheric Releases	5-6
Liquid Releases	5-6
Impact of SRS Technetium Releases on the Offsite Population	5-6
Doses from Atmospheric Releases	5-6
Doses from Liquid Releases	5-7
Comparisons of Technetium Doses Near SRS with Applicable Regulations	5-8
Atmospheric Releases	5-7
Liquid Releases	5-7
Summary of Dosimetric Impacts	5-8
References	5-12
Appendix 1. Physical Properties of Technetium	A1-1

Table of Contents

List of Tables

3-1	Fission product ratios	3-3
3-2	Calculated ^{99}Tc releases	3-3
3-3	Technetium concentrations measured on the SRS	3-4
3-4	Technetium concentrations in the Savannah River	3-4
5-1	Site-specific parameters for atmospheric releases	5-9
5-2	Site-specific parameters for liquid releases	5-10
5-3	Additional site-specific parameters for liquid releases	5-11
5-4	Offsite doses from atmospheric releases of ^{99}Tc	5-12
5-5	Offsite doses from liquid releases of ^{99}Tc	5-12
A1-1	Characteristics of Radioactive Technetium Isotopes that may be present at SRS because of Site Operations	A1-1

List of Figures

2.1	SRS Operating Facilities	2-2
4.1	Geological Cross-Section Showing the Main Aquifers	4-3
4.2	Stratigraphic and Hydrostratigraphic Nomenclature Used on the Savannah River Site	4-4
4.3	Updip Limits of the Confining Systems in the Coastal Plain Sediments	4-5
4.4	Hydraulic Head Differences Across Confining Units Near the Cretaceous/Tertiary Boundary	4-5
4.5	Eh-pH diagram showing equilibrium between pertechnetate ion (1000 pCi/L) and solid TcO_2 (thermodynamic data from Colton, 1965). Circles are Eh-pH conditions measured in the P-series of groundwater wells (Strom and Kaback, 1992). Filled circles represent water table aquifers; open circles represent deeper aquifers	4-6
4.6	Location Map of Waste Disposal Sites in the General Separations Area	4-8
5.1	Simplified Pathways Between Radioactive Materials Released to the Atmosphere and Man	5-4
5.2	Simplified Pathways Between Radioactive Materials Released to Ground or Surface Waters and Man	5-5

Table of Contents

Executive Summary

Assessment of Technetium in the Savannah River Site Environment is published as a part of the Radiological Assessment Program (RAP). It is the last in a series of eight documents on individual radioisotopes released to the environment as a result of SRS operations. The earlier documents describe the environmental consequences of tritium, cesium, iodine, uranium, plutonium, strontium, and carbon. These are living documents and current plans call for revising and updating each one every two years.

Technetium does not have a stable isotope and only exists naturally in the environment in minute quantities associated with uranium ore deposits. Its transport and metabolism have been studied by the nuclear industry because it is a fission product of uranium, and by the medical community because ^{99m}Tc commonly is used as a diagnostic imaging agent in nuclear medicine.

Technetium has been produced at SRS during the operation of five production reactors. The only isotope with environmental significance is ^{99}Tc . Because of the small activities of ^{99}Tc relative to other fission products, such as ^{90}Sr and ^{137}Cs , no measurements were made of releases to either the atmosphere or surface waters. Dose calculations were made in this document using conservative estimates of atmospheric releases (found in scientific literature) and from a few measurements of ^{99}Tc concentrations in the Savannah River.

Technetium in groundwater has been found principally in the vicinity of the separation areas seepage basins. Technetium is soluble in water and follows groundwater flow with little retardation. While most groundwater samples are negative or show little technetium, a few samples have levels slightly above the limits set by the EPA for drinking water.

The overall radiological impact of SRS ^{99}Tc releases on the offsite maximally exposed individual during 38 years of operations can be characterized by maximum individual doses of 0.1 mrem (atmospheric) and 0.008 mrem (liquid), compared with a dose of 13,680 mrem from non-SRS sources during the same time period. Technetium releases have resulted in a negligible risk to the environment and the population it supports.

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Chapter 1. Introduction

Technetium does not exist in nature, except for minute quantities in the presence of uranium. Because ^{99}Tc is a fission product, a significant inventory of the isotope has been injected into the atmosphere from nuclear weapons testing. Some of this radioactivity was deposited on the Savannah River Site (SRS) and the Savannah River drainage basin along with other fission products. Technetium in the oxide form is highly mobile in the environment and readily taken up by plants through their root systems (Murphy, 1990) and through direct deposition (Turcotte, 1982).

^{99}Tc has a low specific activity and decays by the emission of a low-energy beta. Its concentration is low relative to other fission products, and the technetium analysis is difficult because of the low-energy beta and lack of gamma radiation. Consequently, technetium generally has been ignored by the nuclear industry and SRS. Effluent monitoring data does not exist, except for a few creek and river measurements, and much of the analysis in this document rests on conservative assumptions and calculations.

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References

Murphy, C.E., 1990, "Lysimeter Study of Vegetative Uptake from Saltstone," WSRC-RP-90-421, Westinghouse Savannah River Company, Aiken, SC

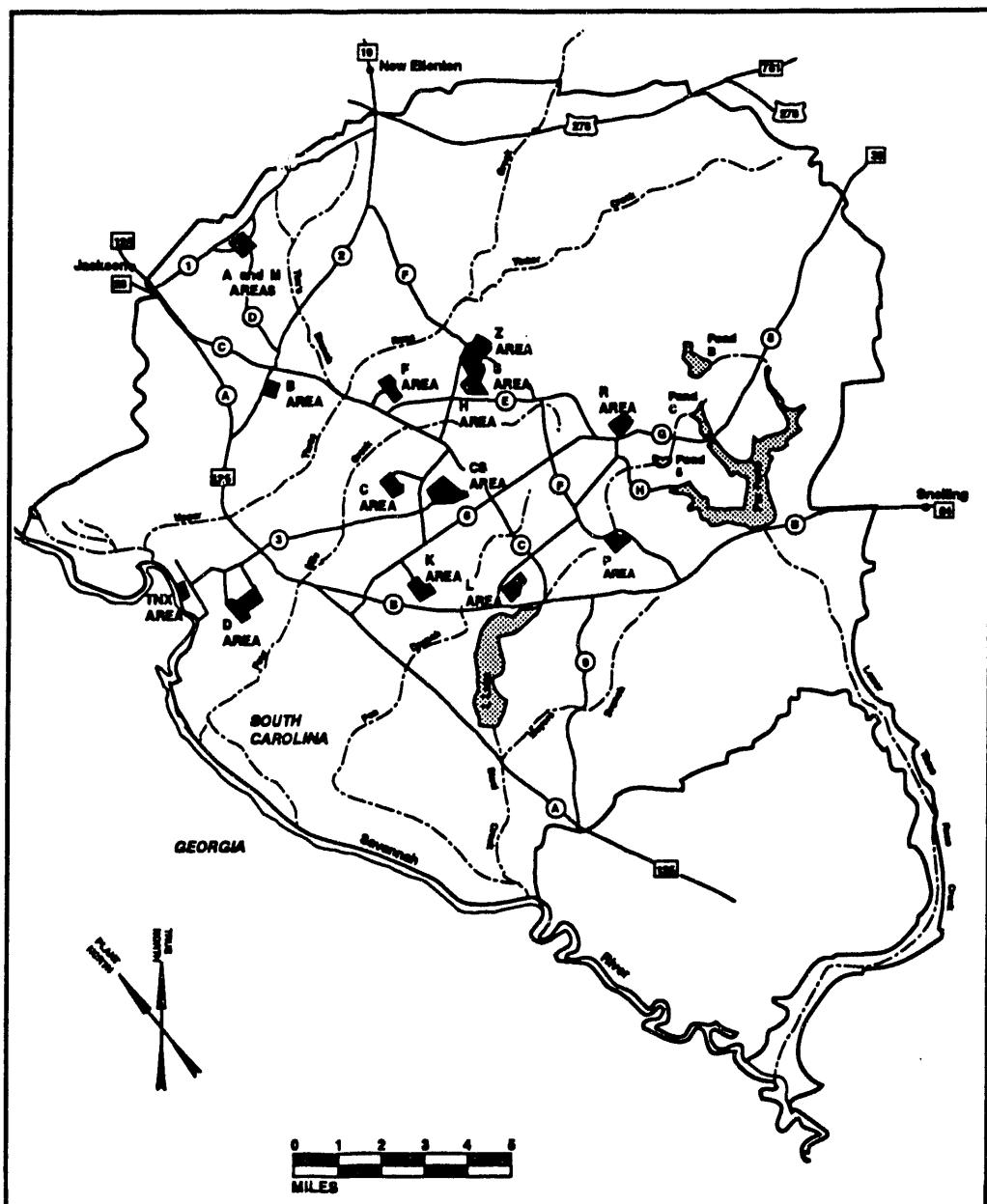
Turcotte, M-D.S., 1982, "Environmental Behavior of Technetium-99," DP-1644, Savannah River Laboratory, E. I. du Pont de Nemours & Co. Aiken, SC.

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Chapter 2.

Origin and Disposition of Technetium at SRS

This chapter gives a general overview of the origin and disposition of technetium at SRS under normal operating conditions. The locations of SRS operating facilities are shown in Figure 2.1. All the facilities that irradiated fuel or handled irradiated fuel had the potential to release technetium. The release of ^{99}Tc occurred during the operation of reactor and chemical separations facilities.



Origin of Technetium at the Savannah River Site

^{99}Tc is produced in minute quantities in nature by spontaneous fission in uranium ore deposits. No stable isotopes of the element technetium exist. The name technetium is derived from the Greek *technetos*, meaning artificial, because it is only produced by artificial means. Three Tc isotopes of relatively long half-life are known: ^{97}Tc (2.6×10^6 y), ^{98}Tc (1.5×10^6 y), and ^{99}Tc (2.1×10^5 y). Neither ^{97}Tc nor ^{98}Tc are formed during the fission of ^{235}U or ^{239}Pu and thus are not encountered in normal reactor separations facility operations. The isotope ^{99}Tc is a fission product of both uranium and plutonium, having a mass yield comparable to ^{90}Sr . Because of its long half-life and subsequent low specific activity (0.017 Ci/g), the curie inventory of ^{99}Tc is much lower than that of ^{90}Sr . The fact that ^{99}Tc emits no gamma radiation, coupled with its relatively weak beta energy ($E_{\text{max}} = 0.292$ MeV), has caused this isotope to be overlooked in most radiological assessments.

The primary source of SRS-produced ^{99}Tc was that formed in the production reactors as a fission by-product. It was formed in enriched uranium fuels, and in natural and depleted uranium targets.

Lesser amounts of technetium were produced in several small nuclear reactors in use at two SRS locations from the 1950s to the 1970s. The Heavy Water Components Test Reactor (HWCTR), located in B Area, was used in the early 1960s to test prototype fuels for a proposed heavy-water-moderated power reactor. The other test reactors, located in M Area, were used from the mid-1950s to the late 1970s. The Process Development Pile and the Lattice Test Reactor were used as zero-power mock-up facilities to test components for the production reactors. The Subcritical Experimental Pile also was used to test component designs. The Standard Pile provided neutrons for experiments such as neutron radiography and neutron activation.

Certain fuels irradiated at offsite non-commercial facilities were shipped to SRS for reprocessing. While awaiting reprocessing, the fuel was stored in the Receiving Basin for Offsite Fuel (RBOF), located in H Area. This fuel contained varying amounts of ^{99}Tc , depending on the fuel type and irradiation history.

Disposition of Technetium at SRS

Technetium-99 produced in SRS reactors may be released to the environment by several means. Trace amounts may be exhausted to the atmosphere as particulates (principally from the separation areas ventilation systems), to the aqueous environment by way of liquid waste in waste tanks, and to the Solid Waste Disposal Facility (SWDF) on the ion exchange resin (demineralizers) used to maintain moderator purity.

Measurement of Technetium in the Environment

Routine analyses were not made to quantify releases of ^{99}Tc to the environment. Analysis for ^{99}Tc is difficult because of the low specific activity, the low β^- energy, and the lack of a stable isotope to use as a carrier. A mass spectrometry technique has been developed to detect low levels of technetium in the environment (Beals, 1992), but the technique is dependent on production of a ^{97}Tc tracer in SRS reactors.

Technetium in the Reactor Moderator

^{99}Tc is produced in the reactors as a byproduct of the fission of uranium and plutonium. Most of the ^{99}Tc remains in the fuel or target tubes and is not released until it is processed in the separation areas. Small amounts of technetium may escape the fuel and target assemblies through microscopic cracks or holes in the cladding. This technetium would be trapped on demineralizers and buried in the Solid Waste Disposal Facility.

Technetium-99 could possibly be released to the environment through a heat exchanger coolant leak. However it will be demonstrated in Chapter 3 that other fission products, such as ^{137}Cs and ^{90}Sr are more than 1000 times as abundant as ^{97}Tc . Since these more abundant radioisotopes were never detected leaking in significant quantities through this pathway, the leakage of ^{97}Tc was truly insignificant.

Technetium in Fuel and Targets Irradiated in Production Reactors

During the chemical separation process, fuel or target elements are dissolved in nitric acid. Any ^{99}Tc present is oxidized to pertechnetate ion. The $[\text{TcO}_4]^-$ ion normally follows the other soluble fission products in the aqueous waste stream, but studies indicate that a fraction of the $[\text{TcO}_4]^-$ ions partition into the organic phase and follow the uranium through the extraction process (Orebaugh, 1984). Thus, some ^{99}Tc is present in UO_3 powder produced in the F-Area A-Line facilities. Most of the technetium is concentrated in the waste streams routed to high-level-waste tanks. Traces of entrained ^{99}Tc follow the evaporator condensate and are routed to the Effluent Treatment Facility (ETF). This activity formerly was routed to the seepage basins. Technetium reaching the ETF is only partially removed by a reverse osmosis unit (there is no anion exchange bed in the ETF). Very small amounts are discharged with the decontaminated waste stream to Upper Three Runs Creek and the Savannah River. ETF effluent is not monitored for ^{99}Tc .

Technetium in Certain Fuels Irradiated Offsite

Irradiated fuel received from certain offsite facilities was stored underwater in RBOF until it was ready for chemical separation. The fuel then entered the regular process stream in H Area. The disposition of technetium is the same as described for fuel irradiated in SRS production reactors.

Technetium in Fuel and Targets Irradiated in Test Reactors

Fuel and targets from the various test reactors were sent primarily to the RBOF for cooling before chemical separation. Some were sent to SRTC for research or to reactor materials fabrication facilities. Reactor materials fabrication facilities only received fuel or targets that were not irradiated and therefore contained no ^{99}Tc .

References

Beals, D.M., 1992, "Determination of Technetium-99 in Aqueous Samples by Isotope Dilution Inductively Coupled Plasma-Mass Spectrometry." Presented at the Third International Conference on Nuclear and Radiochemistry, Vienna Austria, September, 1992 and submitted for publication in the Journal of Radioanalytical and Nuclear Chemistry.

Orebaugh, E.G., 1984, "Analysis of Technetium in SRP Uranium Product Streams," DPST-84-385, Savannah River Laboratory, E.I. du Pont de Nemours & Co., Aiken, SC.

Chapter 3. Releases and Stored Inventories of Technetium at SRS

In the early days of nuclear energy, it was assumed that the radiation dose from ^{99}Tc was so small relative to other isotopes, such as ^{137}Cs and ^{90}Sr , that it could be ignored. In the late 1970s, interest was rekindled in the biological effects of this isotope (Till, Hoffman, and Dunning, 1979), and research on environmental pathways of technetium was stimulated. At SRS, no measurements have been made of atmospheric or liquid releases and only a few have been made of liquid transport. The wisdom of ignoring the dose contribution of ^{99}Tc will be explored in Chapter 5.

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Atmospheric Releases

There have been no measurements of atmospheric releases of technetium, but rough estimates can be made in two ways.

Till, et al., (1979), projected atmospheric releases to the environment from a commercial reprocessing plant to be about 0.14 Ci/yr. Because there are two chemical separation facilities at SRS, the atmospheric release would be 0.28 Ci/yr. As discussed in Chapter 2, there is little potential for atmospheric releases from SRS reactors.

Another rough estimate can be made because approximately the same number of atoms of ^{99}Tc , ^{90}Sr , and ^{137}Cs are produced by the fission process. Table 3-1 lists the ratios of fission product activities that exist immediately after uranium fuel discharge

from a pressurized water reactor after a burnup of 33,000 MWd/MT (Till, 1984). SRS atmospheric releases (1954-1989) of ^{137}Cs and ^{90}Sr have been quantified (Carlton et al, 1992a, 1992b) and are 3.5 and 1.5 Ci respectively. Applying fission ratios to the known releases results in calculated ^{99}Tc releases of 0.0003 to 0.0005 Ci (Table 3-2), much less than the commercial reprocessing plant. In Chapter 5, the dose will be calculated using the hypothetical release rate of 0.28 Ci/yr.

A potential source of atmospheric ^{99}Tc releases occurs during the enrichment of uranium in a gaseous diffusion plant, however, this process does not exist at SRS and will not be considered further.

Table 3-1. Fission product ratios.

<u>Isotope</u>	<u>Activity (Ci)</u>	<u>Activity relative to Tc-99</u>
Tc-99	14.4	1.0
Sr-90	7.6×10^4	5.3×10^3
Cs-137	1.1×10^5	7.6×10^3

Table 3-2. Calculated ^{99}Tc releases

<u>Isotope</u>	<u>Measured Quantity Released (Ci)</u>	<u>Calculated Tc-99 Released (Ci)</u>
Sr-90	1.5	0.0003
Cs-137	3.5	0.0005

Liquid Releases

There have been no measurements of liquid releases and only a few of concentrations in seepage basins and surface water. The very limited data has been published (Beals, 1992) and is reproduced in Table 3-

3. This data was obtained from dip samples, and each value represents the concentration at a single point in time.

Assessment of Technetium In the Savannah River Site Environment

Table 3-3. Technetium concentrations measured on the SRS.

<u>Location</u>	<u>Site Description</u>	<u>Activity (pCi/L)</u>	<u>Ref.</u>
Road F	Upstream of H Area near source of creek	0.22 \pm 0.01	1
Between F and 4	Upstream of H Area	0.18 \pm 0.03	1
Road 4	Below H Area	3.11 \pm 2.51	1
	Seepline 1988	2.67	2
	Seepline 1989	<7	2
Road C	Below F Area	2.05 \pm 1.45	1
	Seepline 1988	3.41	2
	Seepline 1989	64 \pm 14	2
	H-4 seepage basin (enters btwn Rd 4 & C)	90	3
	F-3 seepage basin (enters above Rd C and above Rd 3)	700	3
Road 3	Below F and H Areas	3.11 \pm 1.83	1
Road A	Below inflow from C Area	0.75 \pm 0.30	1
Highway 301	Savannah River below SRS, 1992	0.72 \pm 0.29	4
	Savannah River below SRS, 1979	0.0076	3

¹Beals, 1992b

²Annual environmental monitoring reports

³Anderson, 1979

⁴Beals, 1992a

The most recent measurements in the Savannah River are shown in Table 3-4. These values are near the limit of detection and have large uncertainties

associated with them. The SRS contribution is only an estimate, and more measurements are needed to adequately quantify technetium concentrations.

Table 3-4. Technetium concentrations in the Savannah River

<u>Location</u>	<u>Quarter CY92</u>	<u>Conc (pCi/L)</u>	<u>Error (2 σ)</u>	<u>Avg Conc (pCi/L)</u>	<u>SRS Contribution (pCi/L)</u>
Shell Bluff	1	0.41	0.23	0.42	0.16
	2	0.31	0.13		
	3	0.54	0.2		
Above Vogtle	1	0.43	0.34	0.58	0.11
	2	0.65	0.08		
	3	0.65	0.29		
Below Vogtle	1	0.39	0.19	0.53	0.16
	2	0.53	0.17		
	3	0.66	0.26		
Highway 301	1	0.42	0.09	0.58	0.11
	2	0.72	0.29		
	3	0.61	0.27		

The 1992 SRS contribution of 0.16 pCi/L is equivalent to a transport of 1.4 Ci/yr in the Savannah River (assuming a flow of 10,000 ft³/s), considerably greater than the 1979 measurement of 0.05 Ci/yr

(Anderson, 1979). The difference may be due to the difficulty inherent in the measurement of ⁹⁹Tc in the environment. The higher value will be used in Chapter 5 to calculate dose.

Stored Inventories

Waste Tanks

Storage tanks for high-level radioactive liquid waste, located in both F and H Areas, received waste products from the chemical separation process and are known to contain inventories of technetium. Fowler et al. (1984), compiled sampling data from several waste storage tanks and estimated the ⁹⁹Tc inventory to be 31,050 Ci. This technetium will be processed through the Defense Waste Processing Facility (DWPF) and entombed in glass and concrete.

Solid Waste Disposal Facility

Radioactive solid waste generated during SRS operations has either been burned in the low-level radioactive waste incinerator or placed in the SWDF. Because of the bulky nature of the waste, precise measurement of the isotopic content was not possible. Estimates were made from radiation surveys of the exterior of packages using conversion factors based

on the known or assumed distribution of radioisotopes. Since ⁹⁹Tc emits only a weak beta, it could never be measured directly. An estimate of the quantity of ⁹⁹Tc in the SWDF can be made from known fission product yield ratios by assuming that an equal fraction of Tc and total fission products was sent to the SWDF. This approach results in an estimate of 20 Ci (Jaegge, et al. 1985).

Uranium

Some technetium follows uranium through the chemical separation process in the F and H canyons (Orebaugh, 1984). Enriched uranium fuel, processed through the H canyon had a higher concentration of ⁹⁹Tc than depleted uranium targets processed through the F canyon. When processing was completed, the uranium fuel was shipped to Oak Ridge while the depleted uranium was drummed and stored. The ⁹⁹Tc is not released to the SRS environment and constitutes a lesser radiological hazard than the uranium.

References

Anderson, T.J., 1979, "Development of Chemical Isolation and Concentration Techniques for Technetium-99 Analysis by Resin-bead Mass Spectrometry," in Radioelement Analysis. Progress and Problems, Edited by W.S. Lyons, Ann Arbor Science, Ann Arbor, MI.

Beals, D.M., 1992a, "Tc-99 Results from the Savannah River," SRTC-ETS-92172, Westinghouse Savannah River Company, Aiken, SC.

Beals, D.M., 1992b, "Tc-99 Results from Four Mile Creek," SRTC-ETS-92208, Westinghouse Savannah River Company, Aiken, SC.

Carlton, W.H., L.R. Bauer, A.G. Evans, L.A. Geary, C.E. Murphy, Jr., J.E. Pinder, and R.N. Strom, 1992a, "Cesium in the Savannah River Site Environment,"

WSRC-RP-92-250, Westinghouse Savannah River Company, Aiken, SC.

Carlton, W.H., A.G. Evans, L.A. Geary, C.E. Murphy, Jr., and R.N. Strom, 1992b, "Strontium in the Savannah River Site Environment," WSRC-RP-92-984, Westinghouse Savannah River Company, Aiken, SC.

Fowler, J.R., B.A. Hamm, and J.R. Cook, 1984, "Technetium-99 in SRP High-level Waste and Saltstone," DPST-84-334, Savannah River Laboratory, E.I. du Pont de Nemours & Co., Aiken, SC.

Jaegge, W.J., N.L. Kolb, B.B. Looney, I.W. Marine, O.A. Towler, and J.R. Cook, 1985, "Environmental Information Document - Radioactive Waste Burial

Assessment of Technetium In the Savannah River Site Environment

Grounds," DPST-85-694, Westinghouse Savannah River Company, Aiken, SC.

Orebaugh, E.G., 1984, "Analysis of Technetium in SRP Uranium Product Streams," DPST-84-385, Savannah River Laboratory, E.I. du Pont de Nemours & Co., Aiken, SC.

Till, J.E., F.O. Hoffman, and D.E. Dunning, Jr., 1979, "A New Look at ^{99}Tc Releases to the Atmosphere," *Health Physics*, 36:21-30.

Till, J.E., 1984, "Source Terms for Technetium-99 from Nuclear Fuel Cycle Facilities," in Technetium in the Environment, Ed. By G. Desmet and C. Myttenaere, Elsevier Applied Science Publishers, New York.

Chapter 4. Technetium Concentrations and Transport Mechanisms

Air, surface water, and groundwater are the three modes by which technetium can be transported through the environment and reach humans. Only groundwater will be considered in this chapter because no SRS data exists for air transport, and extremely limited data exists for surface water transport of technetium. Because ^{99}Tc causes low doses in comparison to other released radionuclides and natural background radiation, the lack of air and surface water monitoring is justifiable (see dose calculations in Chapter 5).

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SRS Groundwater System

SRS lies on the Atlantic Coastal Plain, about 20 miles southeast of the edge of the Piedmont Physiographic Province. The Coastal Plain is underlain by a seaward-dipping wedge of sediments that thins to the northwest, where it contacts the exposed crystalline rocks of the Piedmont (Figure 4.1). Early investigations of the site hydrogeology (Siple, 1967) established three geologic and hydrogeologic systems:

- crystalline basement rocks composed of metamorphic and intrusive igneous rocks
- well-indurated Triassic-aged sediments in the Dunbaron Basin
- overlapping, poorly consolidated Cretaceous to Recent Coastal Plain sediments.

The crystalline basement rocks have very low permeabilities. Water is stored primarily in the fractures and joints in the rocks. The permeability of the Triassic rocks also is likely to be relatively low.

Two test wells drilled into the Triassic rocks showed that the water in these rocks is geopressured and that the hydraulic head of the water is above land surface. The origin of the overpressuring is uncertain but has been ascribed to osmotic pressure across the overlying impermeable confining units (Marine, 1974).

Both the crystalline basement and Triassic rocks are overlain by varying thicknesses of weathering residue containing degraded minerals and clay. Overlying these old soils is a depositional blanket of indurated, poorly sorted clayey sediments of the Cape Fear Formation (Bledsoe, 1988). This unit isolates the younger, sedimentary materials from the Triassic sediments and crystalline basement rocks.

The third hydrogeologic system consists of the Cretaceous and younger Coastal Plain sediments. The sediments were deposited largely in shallow marine and fluvial environments (Gohn, 1988). The thickness of the sedimentary units increases toward the southeast across the site, ranging from about 700

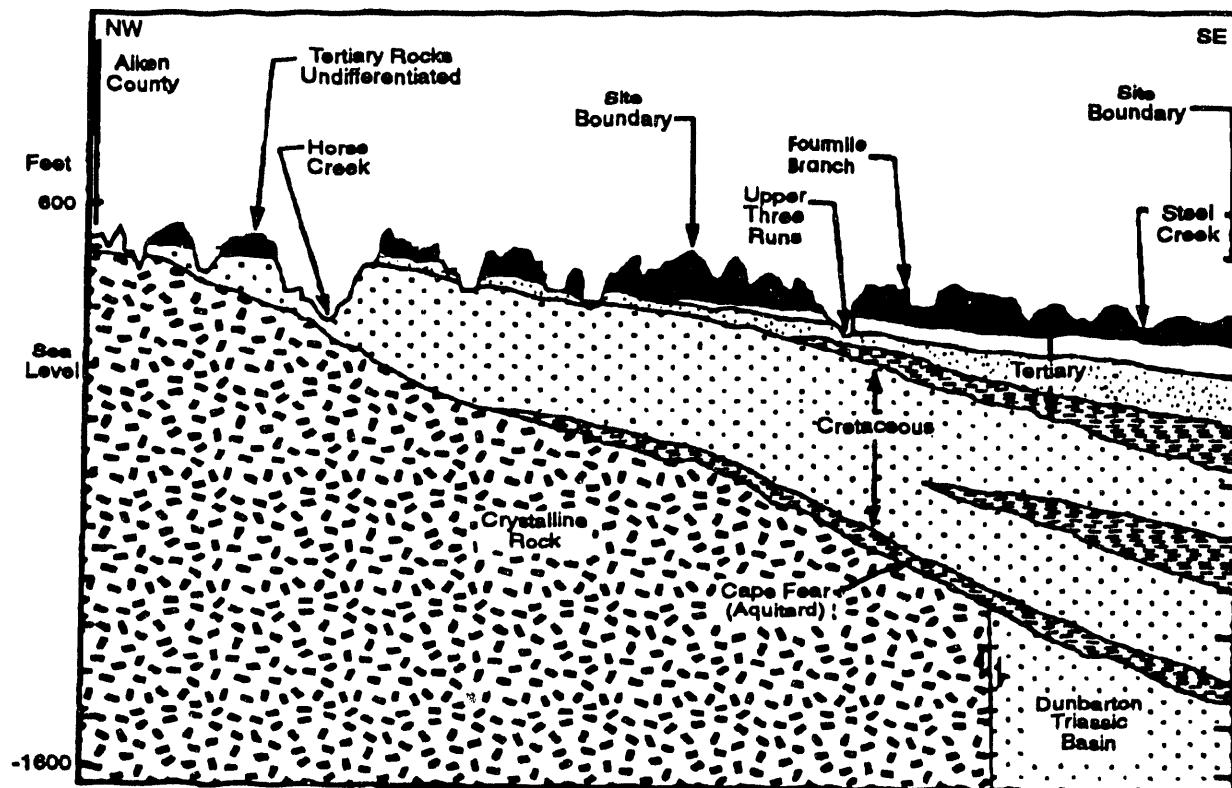


Figure 4.1. Geological Cross-Section Showing the Main Aquifers

feet at the northwest boundary to about 1,400 feet at the southeastern boundary. The stratigraphic names applied to the various units have been modified as more detailed information has become available. The principal aquifers frequently have been named after the principal formations; for example, the Barnwell Formation and the Barnwell Aquifer. As stratigraphic names have changed, the names of the aquifers have been changed.

Figure 4.2 compares some of the changes that have occurred in the hydrostratigraphic nomenclature since the mid-1960s. The currently recommended nomenclature (Aadland et al., 1992) takes into account the progressive thickening of the aquifers across the site and the effectiveness of the confining units that separate the aquifer units. The recent changes avoid tying the aquifer nomenclature to the lithostratigraphic names and extend existing hydrostratigraphic nomenclature from surrounding regions into SRS. A map view, Figure 4.3, shows the approximate limits of the coastal plain aquifer systems across SRS.

At the base of the Coastal Plain sediments is an aquitard, the Appleton Confining System, composed of residual soils of the basement rocks and dense

clays of the Cape Fear Formation. Overlying this confining unit are two aquifers in Cretaceous sands, McQueen Branch and Crouch Branch. The sands generally are considered to be prolific water producers and generally are the source for water supply wells on the site. They are separated from each other by the relatively less permeable beds that make up the Black Creek Formation.

The Tertiary aquifers are separated from the underlying Cretaceous aquifer units by confining beds of the Black Mingo Group, especially the Ellenton Formation. On the northwestern corner of the site, the separation is not as effective as in the southern portion.

In the southeastern area, the Tertiary section can be divided into two separate aquifers, the Gordon (formerly called the "Congaree Aquifer") and Upper Three Runs (frequently referred to as the "Barnwell Aquifer" or "Watertable Aquifer"). The aquitard or confining bed that separates these two units is a glauconitic clayey zone of the McBean Formation locally referred to as the "green clay." This clay pinches out and becomes more sandy toward the northwest.

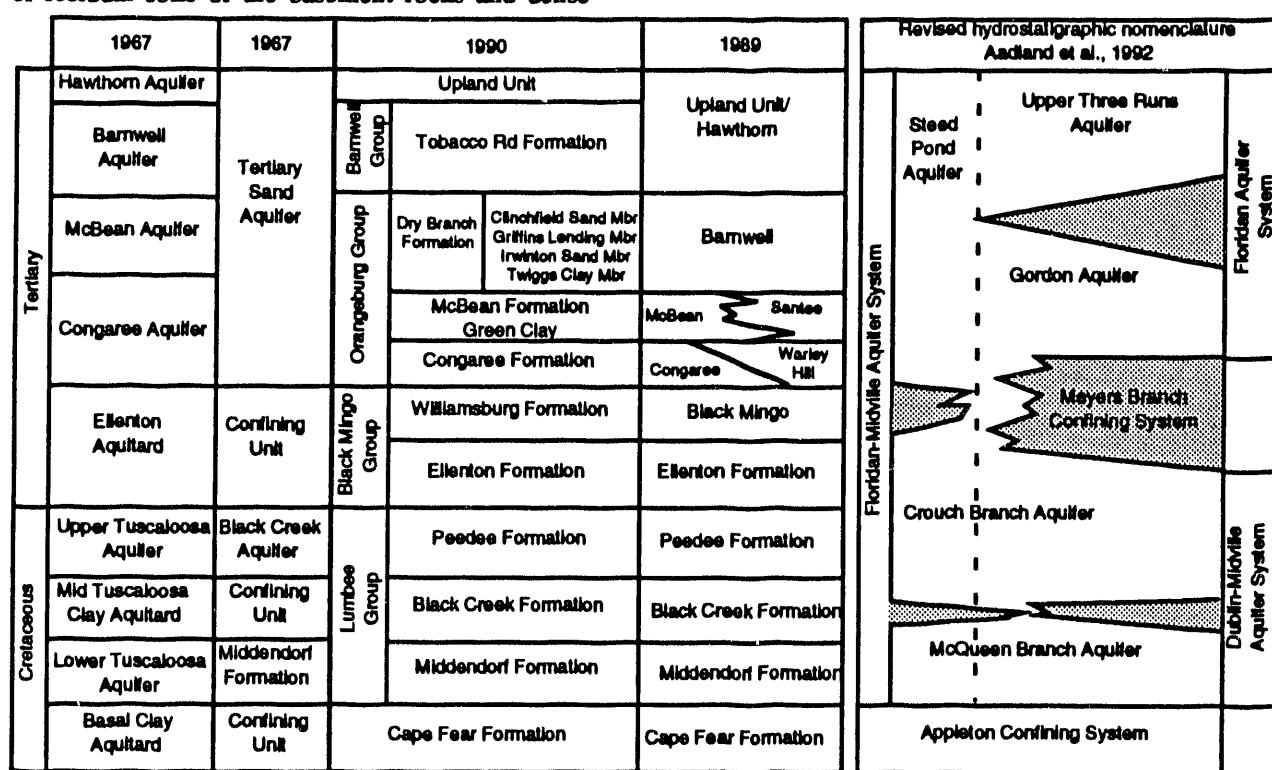


Figure 4.2. Stratigraphic and Hydrostratigraphic Nomenclature Used on the Savannah River Site

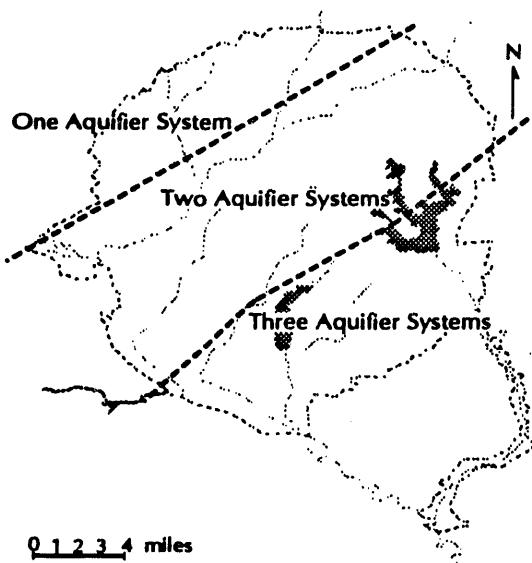


Figure 4.3. Updip Limits of the Confining Systems in the Coastal Plain Sediments

In some areas of the site, it is necessary to modify the hydrostratigraphic nomenclature because of local changes in lithologies and topography. For example, in some portions of the site, an aquitard unit referred to as "tan clay" subdivides the uppermost aquifer into two separate zones. In the southern portion of the site, the "tan clay" unit is thin or sporadic and does not form a consistent aquitard.

The water table level is controlled primarily by local topographic features. The surface of the Atlantic Coastal Plain at the site is a relatively flat plateau (the Aiken Plateau) dissected by stream erosion. The incision of the streams has left relatively isolated, flat-lying surfaces separated by 90-feet-deep to 125-feet-deep stream valleys. The depth to the water table below land surface varies from 0 feet, when it outcrops in the stream valleys or wetlands, to 125 feet below the remnant plateau areas. The water table depth usually is controlled by the elevation of the nearest Savannah River tributary stream. At many localities on site, the water table is situated in Tertiary sediments of low water-producing capabilities, and perched water tables exist sporadically across the site.

The direction of groundwater flow at any locality may change or even reverse in successively deeper aquifers. The aquifers in the Tertiary sediments receive local recharge, and flow generally is toward

nearby stream valleys. Typically, flow at the water table is toward minor tributaries, while deeper Tertiary aquifers flow toward major tributaries of the Savannah River. The deepest aquifers at SRS, the Dublin-Midville Aquifer System, receive recharge in the outcrop areas of the Cretaceous sediments north of the site. Groundwater flow beneath the site in this system is toward the Savannah River.

Over much of the site, the potentiometric surface, or hydraulic "head" of the deeper aquifers in the Cretaceous sediments is higher than that of the overlying Tertiary aquifers (Christensen and Gordon, 1983). This upward gradient is an important characteristic of many of the waste disposal areas and has provided protection from downward transport of contaminants to the deeper aquifers on the site. Figure 4.4 outlines the areas where there is an upward hydraulic gradient across the confining units near the Cretaceous-Tertiary boundary.

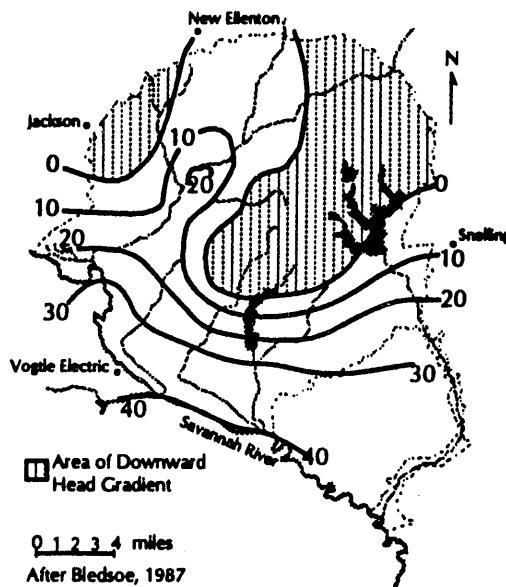


Figure 4.4. Hydraulic Head Differences Across Confining Units Near the Cretaceous/Tertiary Boundary

Technetium 99 in SRS Groundwater

Geochemistry of Technetium

On the Periodic Chart, Tc is between Mn and Re in the Group VIIA elements. The geochemical behavior of Tc is similar to that of these two elements, and that which remains unknown about Tc can be inferred from Mn and Re behavior. Tc is known to exist in six oxidation states 0, +2, +4, +5, +6, and +7, but +4 and +7 are the most common. Initially, Tc produced by fission exists in the metallic form, but during reprocessing, some is oxidized to Tc_2O_7 (Turcotte, 1982). In this form, Tc is soluble, and thus a potential mobile groundwater contaminant.

The mobility of Tc in groundwater is dependent on the groundwater chemistry. When exposed to water, Tc_2O_7 dissolves to form the weak acid $HTcO_4$. The weak acid then partially dissociates to the pertechnetate ion, TcO_4^- . This ion forms no insoluble salts, and thus Tc as pertechnetate is soluble and mobile in groundwater. Under more reducing conditions, however, Tc mobility is limited by formation of the insoluble phase $TcO_2 \cdot 2H_2O$. Figure 4.5 shows an Eh-pH diagram depicting the equilibrium line between the pertechnetate ion and $TcO_2(s)$. Also plotted on this diagram are the Eh-pH conditions of groundwater from the P-series of wells on site. Though reducing conditions may occur in isolated locations on site, SRS groundwater typically is oxidizing enough to sustain the pertechnetate ion. Thus, the mobility of Tc in SRS groundwater is not limited by precipitation of insoluble phases.

Several studies indicate that adsorption does not significantly limit the mobility of Tc in groundwater (Turcotte, 1982). Equilibrium adsorption is quantified using K_d values. These values usually are measured in the lab and are the ratio of the amount of solute adsorbed onto a substrate to the concentration of the solute in solution. Turcotte (1982) compiled summary tables of K_d values of Tc, measured by several workers, for a variety of rock media and soils. All of the natural substrates tested had low K_d values, but Tc did show some affinity for organic matter. That the highest K_d reported ($K_d=47 \text{ mL/mg}$) was for the clay-rich Eleana Argillite is consistent with the observation by Hoeffner (1985) that in SRS soils there is a loose positive correlation between the K_d value of Tc and clay content of the soil. Nevertheless, Hoeffner (1985) concluded that in the soils of SRS, Tc will have migration rates that range from 13 per cent to more than 90 per cent of the groundwater velocity.

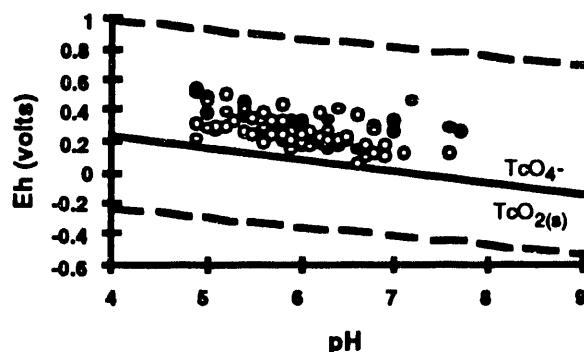


Figure 4.5. Eh-pH diagram showing equilibrium between pertechnetate ion (1000 pCi/L) and solid TcO_2 (thermodynamic data from Colton, 1965). Circles are Eh-pH conditions measured in the P-series of groundwater wells (Strom and Kaback, 1992). Filled circles represent water table aquifers; open circles represent deeper aquifers.

Thus, Tc in the groundwater of SRS should be relatively inert and travel at near-groundwater velocities. The geochemical character of the SRS groundwater favors the pertechnetate ion, which is soluble and not significantly retarded by adsorption.

Origin of ^{99}Tc Contamination

^{99}Tc is a by-product of the fission of ^{235}U and ^{239}Pu , and thus the fabrication of materials for nuclear weapons has resulted in some ^{99}Tc contamination in SRS groundwater. This contamination probably resulted from discharges to seepage basins, but atmospheric releases could have contributed some ^{99}Tc to the groundwater. As airborne ^{99}Tc reacts with the atmosphere it becomes soluble and can enter the groundwater as precipitation. At SRS, however, this mechanism of ^{99}Tc groundwater contamination is considered negligible compared to discharges to seepage basins.

General Separations Area Seepage Basins

During the period 1954 to 1988, seepage basins were used at SRS for the disposal of wastewater containing low concentrations of chemicals and radionuclides. The seepage basins were intended to delay the release of radionuclides that could not otherwise be contained and to allow time for radioactive decay to decrease the activity (Reichert, 1968). Several of these basins were located in the General Separation area near F Area and H Area (see Figure 4.6).

F-Area Seepage Basins

The first seepage basin on site was constructed in 1954 just north of F Area, but the seepage rate was inadequate to handle the volume of wastewater coming from the F-Area facilities. Three additional basins were constructed south of F Area and received effluent during the period 1954 to 1988. Water infiltrating from the F-Area seepage basins encountered a perched water table 15 to 25 feet below the surface and a normal water table at a depth of 60 to 65 feet below the land surface. This water produced a contamination plume that can be identified in monitor wells by its high conductivity and tritium activities. Flow of water at the water table aquifer is toward Fourmile Branch (also known as Four Mile Creek) at a rate estimated to be about 0.5 feet/day (Stone and Christensen, 1983). Some of this water intersects the surface at a seepline along Fourmile Branch (Looney et al., 1988).

^{99}Tc has been detected in the groundwater and the soils at the Fourmile Branch seepline near F Area. Haselow et al. (1990) sampled soils and water along the seepline near both F and H areas. They found a maximum ^{99}Tc activity of 1200 pCi/L in the seepline water near F Area. This was the only sample collected by Haselow et al. (1990) that exceeded the EPA primary drinking water standard (DWS) guideline for ^{99}Tc , 900 pCi/L, (EPA, 1976). The other samples ranged from below the detection limit to 330 pCi/L.

The Environmental Monitoring Section of the Environmental Protection Department of SRS has measured ^{99}Tc in some wells since 1990. These analyses are reported in quarterly environmental monitoring reports and in the annual SRS Environmental Report. Activities of ^{99}Tc above the detection limit were reported in the groundwater of several F-Area seepage basin monitoring wells. However, the highest activity, 340 pCi/L, was less than the DWS guideline.

H-Area Seepage Basins

The four H-Area seepage basins covered an area of 42,700 m², and were in use from 1955 to 1988. Liquids discharged to these basins contained low-level radioactivity from the H-Area separation facility. In addition, the effluents to these basins contained heavy metals, nitrate in the form of HNO₃, and NaOH (Killian et al., 1985). The basins were closed in 1988 and have since been capped.

Though in close proximity to the F-Area basins, the hydrogeology at the H-Area basins is distinct. The depth to the water table is only about 20 feet, and the basins are located closer to the groundwater discharge zone along Fourmile Branch. Much of the groundwater flow in this area of the plant appears to occur in narrow, high permeability channels in the sediments. While a small percentage of the seepage basin waters penetrate into a deeper aquifer, the majority migrate to the seepline adjacent to Fourmile Branch.

^{99}Tc has been detected in the water at the H-Area seepline. Samples measured by Haselow et al. (1990) had ^{99}Tc activities that ranged from less than the detection limit to 280 pCi/L. All ^{99}Tc activities measured in groundwater from monitoring wells around the H-Area seepage basin were below the DWS guideline and ranged from below the detection limit to 340 pCi/L.

Solid Waste Disposal Facility

At the Savannah River Site, solid radioactive waste is stored at a single, centrally located Solid Waste Disposal Facility (SWDF) between F and H separations areas (Figure 4.6). The original 76-acre burial site operated from 1953 until 1972 and is designated as 643-E (Old Burial Ground). A second area, of 119 acres, later was opened to receive solid radioactive wastes and was designated as 643-7E (New Burial Ground).

The new SWDF is located on a water table divide. The groundwater beneath about half of the 643-7E area and the groundwater beneath the old SWDF flows south toward Fourmile Branch. Beneath the northern half of the 643-E SWDF, the groundwater flows toward Upper Three Runs. ^{99}Tc activity has been measured at low levels in some of the groundwater monitoring wells surrounding the old SWDF. The highest activity measured was 14 pCi/L, but ^{99}Tc in most of the wells was below detection limits.

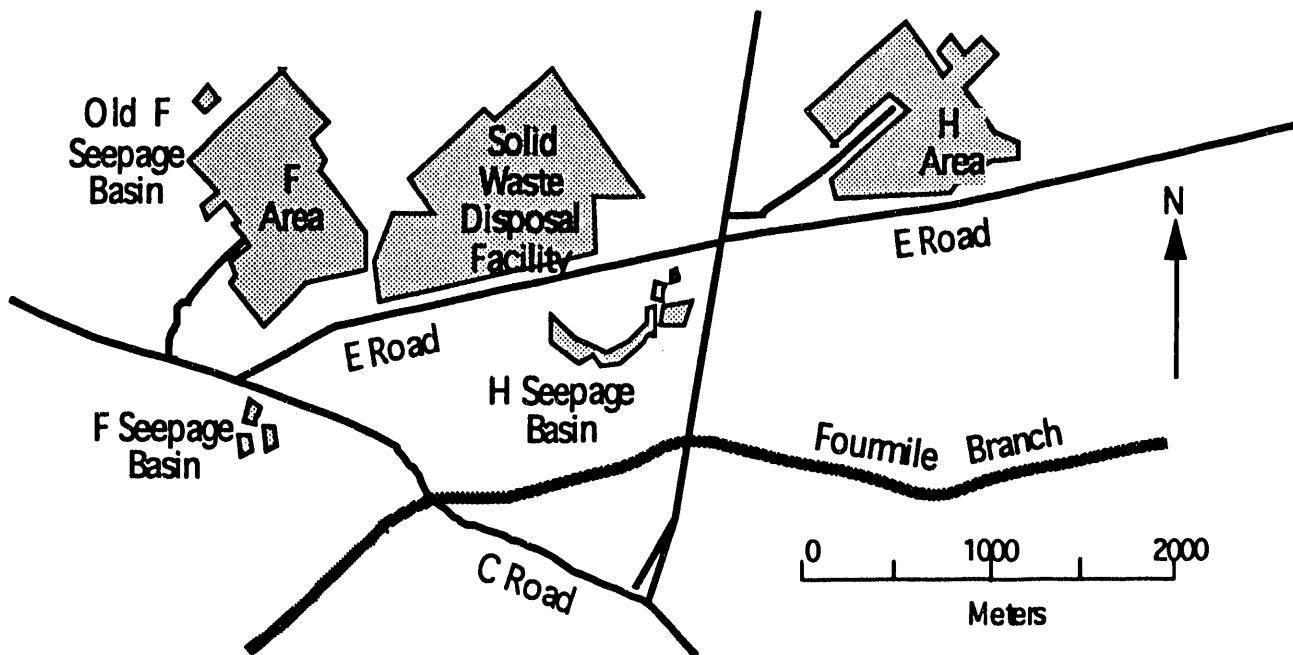


Figure 4.6. Location Map of Waste Disposal Sites in the General Separations Area

Other Measurements of ^{99}Tc

In SRS areas other than the General Separation Area, measurements of ^{99}Tc activity have been sparse. In 1992, samples of groundwater from two downgradient monitoring wells (LCO-1 and LCO-2) at the L-Area Oil and Chemical Basin were analyzed for ^{99}Tc and found to have activities that approached or exceeded the DWS guideline. These high ^{99}Tc activities ranged from 200 to 1100 pCi/L and occurred for three consecutive quarters in samples from well LCO-1. This contamination resulted from

small amounts of radioactive oil and chemical waste discharged to this basin from 1961 until 1979.

Low ^{99}Tc activities were reported in samples from two other monitoring wells at SRS. A sample from well ZBG-1 (Z-area Saltstone Facility) had a ^{99}Tc activity of 4.5 pCi/L, but this measurement was followed by three consecutive quarters during which the activity was below detection limits. Likewise, a first quarter 1992 sample from a monitoring well (XSB-1) at the Old TNX Seepage Basin had a ^{99}Tc activity of 92 pCi/L but was followed by two consecutive quarters of activity below detection limits.

General Observations

^{99}Tc in groundwater beneath SRS is a result of the disposal of materials associated with nuclear fission. ^{99}Tc remains mobile in the subsurface of SRS because of its geochemical behavior and the nature of

the groundwater. Though ^{99}Tc activity exceeds the DWS guideline in two locations — the F-Area seep line and the L-Area oil and chemical basin — it is well below the DWS in the other samples measured.

References

Aadland, R.K., P.A. Thayer, and A.D. Smits, 1992, "Hydrostratigraphy of the Savannah River Site Region, South Carolina and Georgia," in Wallace Fallaw and Van Price, eds: Geological Investigations of the Central Savannah River Area, South Carolina and Georgia, Carolina Geological Field Trip Guide Book, Nov. 13-15, 1992, South Carolina Geological Survey, Harbison Forest Road, Columbia, SC. 29210-40898, pp. CSG-B-X-1 to 6.

Bledsoe, H.W., 1988, "SRP Baseline Hydrogeologic Investigation - Phase III," DPST-88-627, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Christensen, E.J., and D.E. Gordon, 1983, "Technical Summary of Groundwater Quality Protection Program at Savannah River Plant, Volume I," DPST-83-829, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Colton, R., 1965, "The Chemistry of Rhenium and Technetium," Interscience Publishers, London, 185p.

EPA, 1976, "National Interim Primary Drinking Water Regulations," EPA-570/9-76-003, Environmental Protection Agency.

Gohn, G.S., 1988, Late Mesozoic and Early Cenozoic Geology of the Atlantic Coastal Plains; North Carolina to Florida, The Geology of North America, Boulder, Colo., Geologic Society of America, pp 107-130.

Haselow, J.S., M. Harris, B.B. Looney, N.V. Halverson, and J.B. Gladden, 1990, "Analysis of Soil and Water at the Fourmile Branch Seepline Near the F&H Areas of SRS (U)," WSRC-RP-90-0591, Westinghouse Savannah River Company, Aiken, SC.

Hoeffner, S.L., 1985, "Radionuclide Sorption on Savannah River Plant Burial Ground Soil — A Summary and Interpretation of Laboratory Data," DP-1702, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Killian, T.H., et al., 1985, "Environmental Information Document - H Area Seepage Basins," DPST-85-706, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Looney, B.B., J.E. Cantrell, and J.R. Cook, 1988, "Sampling and Analysis of Surface Water in the Vicinity of the F- and H- Area Seepage Basins," DPST-88-229, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Marine, I.W., 1974, "Geohydrology of Buried Triassic Basin at Savannah River Plant, South Carolina," American Association of Petroleum Geologists Bulletin 58: 1825-1837.

Reichert, S.O., 1968, "Geology Plays an Important Role in Radioactive Waste Management," Mining Engineering (September): 98-103.

Siple, G.E., 1967, "Geology and Groundwater of the Savannah River Plant and Vicinity, South Carolina," U.S. Geological Survey, Washington, DC.

Stone, J.A., and E.J. Christensen, 1983, "Technical Summary of Groundwater Quality Protection Program at Savannah River Plant, Volume II - Radioactive Waste," DPST-83-829, E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC.

Strom, R.N., and D.S. Kaback, 1992, "SRP Baseline Hydrogeologic Investigation: Aquifer Characterization — Groundwater Geochemistry of the Savannah River Site and Vicinity," WSRC-RP-92-450, Westinghouse Savannah River Company, Aiken, SC.

Turcotte, M-D.S., 1982, "Environmental Behavior of Technetium-99," DP-1644, E.I. du Pont de Nemours & Co., Savannah River Plant and Laboratory, Aiken, SC.

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Chapter 5. Assessment of Dose/Risk from SRS Technetium Releases

This chapter describes the health impacts associated with exposure to technetium. The methodology used to produce dose estimates is presented. The results of a dose assessment for technetium releases during the 1955—1992 period are discussed.

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Relationship of Dose to Risk and Health Effects

Ionizing Radiation

Ionizing radiation is that radiation which has enough energy to remove electrons from the atoms through which it passes. The interaction of ionizing radiation with biological systems can induce a series of chemical reactions that can cause permanent changes in the genetic material of cells. These changes (mutations) may cause abnormal functioning within the cell or may lead to cell death.

The nature of radiation-induced cellular changes depends on the magnitude of the dose and the rate at which it is received. For the low doses and dose rates encountered in the environment from SRS releases, the most significant effect is cancer induction. This is believed to be a stochastic effect, i.e., an increase in dose increases the probability of the effect, but the severity of the effect is independent of dose.

A characteristic of stochastic risks is the absence of a threshold. In other words, it is conceivable that any

dose of radiation, no matter how small, might give rise to a cancer. On the other hand, there is no way to be certain that a given dose, no matter how large, will cause a cancer in an individual.

Cancer Risk Estimates

The most comprehensive estimates of cancer induction by exposure to ionizing radiation come from studies of the atomic bomb survivors at Hiroshima and Nagasaki. Less definitive studies include those of medical patients exposed to therapeutic and diagnostic radiation. Studies of laboratory animals have increased the understanding of dose-effect relationships. The International Commission on Radiological Protection (ICRP) has evaluated all these studies and concluded that the best estimate of lifetime risk of fatal cancer for members of the general population is approximately 500 cases per 1,000,000 person-rem (ICRP, 1991). This is equivalent to one case per 2000 person-rem.

Technetium Exposure and Dose to Man

Distribution and Retention of Technetium In the Human Body

Technetium does not exist in nature except in minute quantities associated with spontaneous fission and is not a natural constituent of the human body. The metabolic fate of technetium in the form of the pertechnetate ion, TcO_4^- , has been studied extensively because of its use as a diagnostic agent in nuclear medicine. For the calculation of dose, technetium is assumed to be uniformly distributed in the body except for the thyroid, stomach wall, and liver, where it is concentrated (ICRP, 1980).

Technetium Dosimetry

The U.S. Department of Energy (DOE) issued internal dose conversion factors in 1988 to ensure that doses are calculated in a consistent manner at all DOE facilities (DOE, 1988). The factors are based on ICRP recommendations (ICRP, 1981). These adult dose factors are used, in conjunction with the models described in the subsection on models of technetium transport and dose, to calculate all the doses reported in this document.

Future changes are anticipated in the dose calculation methodology. The ICRP (ICRP, 1989) has issued age-specific dose factors for ingestion of some of the more common isotopes and is developing age-specific dose factors for inhalation of radioisotopes.

Models of Technetium Transport and Dose

With the exception of tritium, most of the radioactive materials released from SRS have such low concentrations in the offsite environment that they are

not detectable by conventional monitoring techniques (i.e. methods used by the Environmental Monitoring Section). Therefore, radiation doses to offsite

individuals and populations are calculated with mathematical models. These models use known transport mechanisms for atmospheric and liquid releases and known major pathways of exposure to man. Modeled atmospheric and aqueous dispersion are periodically verified using environmental tritium measurements. Tritium is released during normal SRS operations.

The first models used at SRS for calculating offsite doses were developed by the Savannah River Technology Center (Cooper, 1975). These models, MREM (atmospheric releases) and RIVDOSE (liquid releases), were used first in 1972. In 1982, MREM and RIVDOSE were replaced with the more technologically advanced models now in use.

SRS annual offsite doses are calculated with the transport and dose models developed for the commercial nuclear industry (NRC, 1977a, and NRC, 1977b). The models are implemented at SRS in the following computer programs:

Atmospheric Releases

- **MAXIGASP**—calculates maximum and average doses to offsite individuals
- **POPGASP**—calculates offsite population collective dose

MAXIGASP and POPGASP are SRTC-modified versions of the Nuclear Regulatory Commission (NRC) programs XOQDOQ (Sagendorf et al., 1982) and GASPAR (Eckerman et al., 1980). The modifications were made to meet the requirements for input of physical and biological data specific to SRS. The basic calculations in the XOQDOQ and GASPAR programs have not been modified.

Modeling Atmospheric Dispersion of Radioactive Releases

The routine atmospheric transport of radioactive materials from SRS is evaluated on the basis of meteorological conditions measured continuously at seven onsite towers. The towers relay wind speed, direction, and atmospheric stability information at 1.5-second intervals to SRTC via the WIND (Weather Information and Display) system. A database of this information containing the 60-minute average values for the period 1982—1986 is accessed by dispersion codes to estimate downwind concentrations of released radionuclides.

Offsite doses from atmospheric releases have been calculated using H-Area meteorology and assuming

the releases occurred at the geographic center of the site. It has been demonstrated that using data from one of the other onsite meteorological towers has little effect on the maximum individual dose and no effect on the 50-mile population dose (Hamby and Parker, 1991).

The dispersion of an atmospheric release from SRS is modeled using XOQDOQ, which computes concentrations in the plume as a function of downwind distance and compass sector. The plume was depleted by dry deposition and/or radioactive decay. No credit was taken for plume rise resulting from thermal buoyancy and/or momentum effects. XOQDOQ has undergone a comprehensive review in association with Westinghouse Savannah River Company (WSRC) quality assurance requirements for software (Bauer, 1991).

The plume concentration information generated by XOQDOQ is used by the dose modeling program GASPAR to estimate doses to offsite individuals and populations. GASPAR estimates doses from a number of pathways, which are illustrated in a general sense in Figure 5.1.

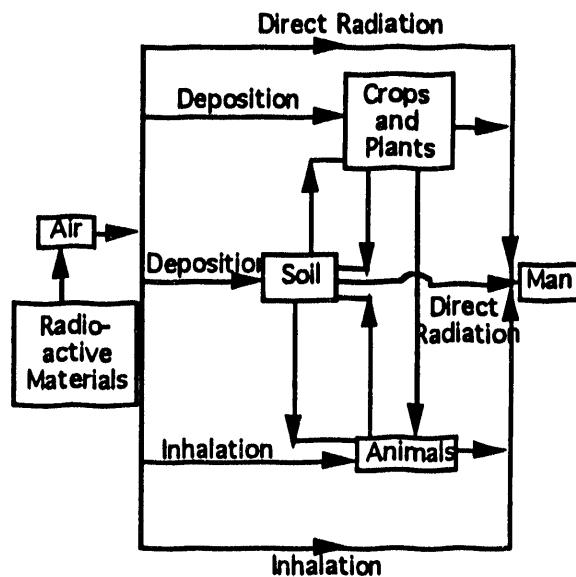


Figure 5.1. Simplified Pathways Between Radioactive Materials Released to the Atmosphere and Man

The doses estimated by GASPAR are reported on a pathway-specific basis as follows:

- **Plume** - external dose from radioactive materials suspended in the atmosphere
- **Ground** - external dose from radioactive materials deposited on the ground
- **Inhalation** - internal dose from inhalation of radioactive materials present in the plume
- **Vegetation** - internal dose from consumption of contaminated crops
- **Milk** - internal dose from milk produced in a contaminated area
- **Meat** - internal dose from consumption of meat produced in a contaminated area.

MAXIGASP

The calculations required by XQDOQ and GASPAR to estimate maximum and average individual doses are performed at SRS using the computer program MAXIGASP. MAXIGASP calculates annual average ground-level air concentrations and 50-yr committed doses at a number of points along the site boundary in each of 16 compass sectors.

The main output from the program is the maximum dose equivalent to an individual along the SRS perimeter. The maximally exposed individual is assumed to reside continuously at the location of highest exposure and to have living and eating habits which maximize dose. These assumptions provide a ceiling on doses from atmospheric releases, as no such individual is believed to exist.

POPGASP

The calculations required by XQDOQ and GASPAR to estimate population doses from atmospheric releases are performed at SRS using POPGASP, which calculates annual average ground-level air concentrations and annual doses for each of 160 regions (16 wind direction sectors at 10 distances per sector) within an 80-kilometer radius of the release location.

In addition to compass sector-specific meteorological information, POPGASP uses sector-specific data on population distribution and composition. Comparable data on milk, meat, and vegetable production and consumption also are used in the code. These databases are quite extensive and are available for review in the annual SRS Environmental Report. The site specific parameters (Hamby, 1991) used to calculate doses with MAXIGASP are presented in Table 5-1.

Modeling Doses from Liquid Releases

The consequences of liquid releases from SRS are modeled using LADTAP XL (Liquid Annual Doses To All Persons). The potential pathways of exposure from liquid releases to the environment are shown in Figure 5.2. The pathway-specific doses calculated by LADTAP XL are grouped into the following four categories:

- **Potable drinking water** - internal dose from consuming drinking water of Savannah River origin
- **Sport fish and commercial fish** - internal dose from consuming fish of Savannah River origin
- **Saltwater invertebrates** - internal dose from consuming shellfish from estuaries of the Savannah River
- **Recreation** - external dose from recreation (boating, swimming, and shoreline) activities in and along the Savannah River

LADTAP XL estimates individual doses at specific downstream locations. The only removal mechanism included in the transport model as it is used at SRS is radioactive decay. No credit is taken for adsorption on stream sediments.

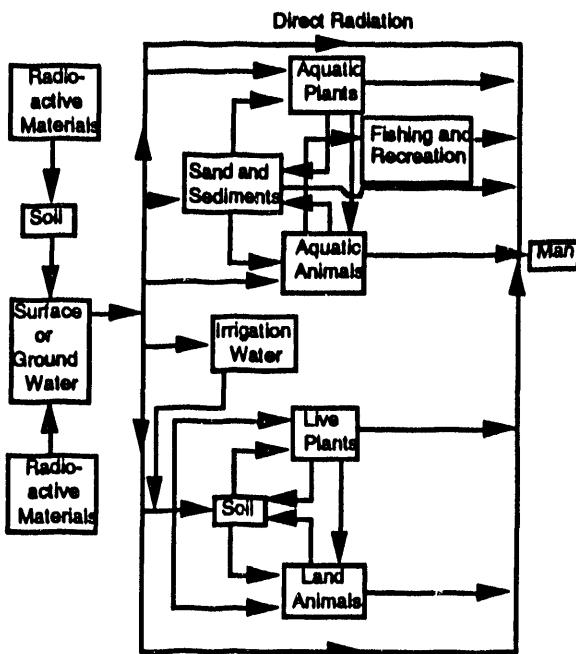


Figure 5.2. Simplified Pathways Between Radioactive Materials Released to Ground or Surface Waters and Man

The major assumption inherent in the application of LADTAP XL to SRS releases is that liquid discharges undergo complete mixing in the Savannah River before reaching potentially exposed populations. This assumption is supported by repeated measurements indicating that complete mixing occurs in the river between SRS and the Highway 301 sampling station (Cummins et al., 1990).

LADTAP XL generates maximum individual and population doses for all the exposure pathways identified above. LADTAP XL calculations are performed with SRS-specific information to the extent that it is available. Summary tables of principal input values used in LADTAP XL are shown in Tables 5-2 and 5-3.

Validation of Transport Models Using Monitoring Data

Atmospheric Releases

The radionuclide concentrations predicted by XOQDOQ are compared routinely with measured values of tritium concentrations in air to evaluate the performance of the code. (Tritium is the only radionuclide released by SRS that can be detected routinely offsite using conventional measuring techniques.) Predicted values tend to exceed observed values, but not to a degree that would indicate an excessively conservative approach.

Other comparisons of predicted and measured concentrations have been made (Marter, 1984) and have exhibited similar results. The available data suggest that calculated concentrations of tritium in air generally are conservative estimates of actual offsite values.

Liquid Releases

Throughout the period 1955—1992, drinking water doses from routine SRS releases have been dominated by tritium. Measured, rather than estimated, tritium concentrations therefore are used for the downriver locations most important in terms of dose calculations: (1) just below SRS, (2) at the Beaufort-Jasper water treatment plant, and (3) at the Port Wentworth water treatment plant. In addition to supplying measured tritium concentrations, such data allow calculation of Savannah River dilution factors for these locations. The availability of accurate estimates of river dilution characteristics significantly enhances LADTAP's ability to predict downriver concentrations of other radionuclides.

Impact of SRS Technetium Releases on the Offsite Population

The computer codes MAXIGASP and POPGASP have been used to calculate effective dose equivalents that theoretically have been received by segments of the offsite population from technetium exposure during site operations. *Since the ^{90}Tc releases have never been monitored, the dose is calculated based on a hypothetical atmospheric release of 0.28 Ci/yr (see Chapter 3), which is equivalent to 10.6 Ci over 38 years.* The results are shown in Table 5-4.

Doses from Atmospheric Releases

The effective dose equivalent (EDE) theoretically received by the "maximum" adult during the 1955—1992 time period at the site perimeter was 0.1 mrem (Table 5-4).

"Maximum" individuals are hypothetical persons who lived at the SRS boundary and subsisted on diets of locally produced milk, meat, and vegetables. No such individuals are known to exist. This theoretical dose is the upper bounding case and would make a minor contribution to the overall dose received during that time period.

A person living in the Central Savannah River Area (CSRA) would have received an effective dose of approximately 11,000 mrem from exposure to natural sources of radioactivity and an additional 2,500 mrem from medical practices and various consumer products during the 38-year period (Cummins et al., 1990). Therefore, the cumulative dose contribution to this individual from SRS atmospheric technetium releases can be estimated as 0.0007 percent.

Because the contribution of SRS technetium releases to any one individual's total radiation dose is so small, it is necessary to pool the radiation exposures from a given population if an assessment of potential health risks is desired. The population dose within an 80-kilometer radius is the figure of merit frequently used to make such an assessment.

The population doses reported in Table 5-4 are based on 1980 census data (555,100 people within 80 km) and 1982-86 meteorology and current dose factor data (DOE, 1988). If it is assumed that this population has lived in the SRS vicinity throughout the period of site operation, the total collective effective dose received by that population through 1992 would be 6.5 person-rem.

The risks associated with this collective dose are quite small. The risk estimate using the ICRP factor for the number of excess fatal cancers potentially induced by a collective dose of 6.5 person-rem is 0.003. Conversely, in that same population, at the current fatal cancer frequency of 16 percent (EPA, 1989), about 90,000 fatal cancers will occur from all other causes. Therefore, it is impossible to demonstrate that a relationship exists between any of the cancer deaths occurring in this population and the release of technetium to the atmosphere from SRS.

Doses from Liquid Releases

Dose equivalents potentially received by downstream consumers of Savannah River water and fish are shown for ⁹⁹Tc in Table 5-5. The dose calculations are based on the total number of curies released directly to onsite streams from 38 years of SRS operations. The activity released (53 Ci) assumes that the maximum measured value (Chapter 3) was the same for each year of operation from 1955 through 1992. The dose was calculated for a "maximum" individual living just below SRS who subsisted on a diet of untreated Savannah River water

and fish of Savannah River origin. Such a dose is believed to represent the bounding case for technetium releases to surface water. Drinking water doses also have been calculated for "maximum" individuals at both of the downstream water treatment plants and for the populations served by the plants.

The effective dose equivalent to the maximum individual for the 38 year period has been estimated as 0.008 mrem. Since this individual's dose from non-SRS sources of radiation for that same time period exceeded 13,000 mrem, it may be concluded that the contribution to downstream individuals' doses by SRS technetium releases is insignificant. An effective dose equivalent of ≤ 1 mrem/yr has been termed a Negligible Individual Risk Level by the National Council on Radiation Protection and Measurements (NCRP, 1987).

With respect to the effect of liquid technetium releases on the populations downstream of SRS, drinking water doses for users of the Beaufort-Jasper (50,000 customers) and Port Wentworth, (15,000 effective consumers) water treatment plants also have been estimated. Different terminology is used to describe the two populations to reflect the difference in their compositions. The Beaufort-Jasper plant services residential areas and therefore provides full-scale domestic water service. The Port Wentworth facility serves a commercial complex in which contact with treated Savannah River water is limited to industrial workers who consume tap water at work.

If the cumulative effective doses received by these populations are summed, the collective dose equivalent is about 0.24 person-rem. Using the ICRP nominal risk factor (ICRP, 1991), the predicted impact of this collective dose is an estimated 0.00012 excess fatal cancers in a population of 65,000 people—10,400 of whom, at the current fatal cancer rate, are projected to succumb to cancer from all other sources.

Comparisons of Technetium Doses Near SRS with Applicable Regulations

Atmospheric Releases

The hypothetical dose received by the maximally exposed individual due to atmospheric releases of technetium from SRS over 38 years was 0.1 mrem (Table 5-4). The current EPA *annual* limit on doses

to members of the public due to atmospheric releases is 10 mrem from all released radioisotopes.

Liquid Releases

The hypothetical dose received by the maximally exposed individual due to liquid releases of

technetium from SRS over 38 years was 0.008 mrem (Table 5-5). The current EPA *annual* limit on doses

to members of the public due to drinking water consumption is 4 mrem.

Summary of Dosimetric Impacts

The overall radiological impact of SRS technetium releases (1955—1992) on the offsite maximum individual can be characterized by a total dose of 0.1 mrem. During this same period, however, such an individual received a dose of 13,700 mrem from non-SRS sources of ionizing radiation present in the environment.

The impact of SRS technetium releases on offsite populations also has been evaluated. The total

collective dose from atmospheric technetium releases (1955—1992) is estimated as 6.5 person-rem distributed among 555,100 individuals. The total collective dose from liquid releases (1955—1992) is estimated as 0.24 person-rem distributed among 65,000 individuals.

⁹⁹Tc releases from SRS present a negligible risk to the offsite environment and the population it supports.

Table 5-1. Site-specific parameters for atmospheric releases

Population Group

50-mile radius 555,100

Maximum Individual (MAXIGASP)

Exposure Pathway

Inhalation (m³/yr) 8000

Ingestion

Cow's milk (L/yr)	230
Meat (kg/yr)	81
Leafy vegetables (kg/yr)	43
Fruits, grains, and other vegetables (kg/yr)	276

External exposure

Transmission factor for shielding from buildings	0.7
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General Population (POPGASP)

Exposure Pathway

Inhalation (m³/yr) 8000

Ingestion

Cow's milk (L/yr)	120
Meat (kg/yr)	43
Leafy vegetables (kg/yr)	21
Fruits, grains, and other vegetables (kg/yr)	163

External exposure

Transmission factor for shielding from buildings	0.5
--	-----

Table 5-2. Site-specific parameters for liquid releases.

Maximum Individual Dose Assessments (LADTAP XL).

Site Parameters

Savannah River flow rate (ft ³ /sec)	6,000 or measured average
Transit time from SRS to Savannah River (hr)	24
Shore-width factor	0.2

Maximum Individual

Human Parameters

	Adult
Water consumption (L/yr) (a)	370 (730)
Fish consumption (kg/yr)	19
Shellfish consumption (kg/yr)	8
Shoreline recreation (hr/yr)	23
Swimming (hr/yr)	8.9
Boating (hr/yr)	21

(a) The values in parentheses are EPA parameters mandated for use when calculating maximum individual doses to Beaufort-Jasper and Port Wentworth water users.

Table 5-3. Additional site-specific parameters for liquid releases.

Average Individual/Population Dose Assessments (LADTAP XL)

Site Parameters

Savannah River flow rate (ft ³ /sec)	10,000 or measured average
Transit time from SRS to Savannah River (hr)	24
Transit time from SRS to water treatment plants (hr)	72
Retention time in water treatment system (hr)	24
Shore-width factor	0.2
River dilution in estuary	3
Aquatic food harvest, edible portions(kg/yr)	
Sport fish	35,000
Commercial fish	2,700
Saltwater invertebrates	390,000

Demographic Data

Population Group

<u>Population Group</u>	<u>Size</u>
50-mile radius	555,100
Beaufort-Jasper water users	50,000
Port Wentworth water users	15,000

Average Individual

Human Parameters

Adult

Water Consumption (L/yr)	370
Fish Consumption (kg/yr)	9
Shellfish Consumption (kg/yr)	2

Usage (person-hr)

Shoreline Recreation (hr/yr)	960,000
Swimming (hr/yr)	160,000
Boating (hr/yr)	1,100,000

Assessment of Technetium In the Savannah River Site Environment

Table 5-4. Offsite doses from atmospheric releases of ^{99}Tc

Release 1955-92 (Ci)	Site Perimeter CEDE(a) mrem		EDC (b) person-rem 80-km Pop
	Max Ind	Average Ind	
10.6	0.1	0.06	6.5

(a) CEDE = Committed effective dose equivalent

(b) EDC = Environmental dose commitment

Table 5-5. Offsite doses from liquid releases of ^{99}Tc

Release 1955-92 (Ci)	Downriver CEDE(a) mrem		EDC (b) person-rem 80-km Pop & drinking water
	Max Ind River	Max Ind WTP	
53	0.008	0.006	0.24

(a) CEDE = Committed effective dose equivalent

(b) EDC = Environmental dose commitment

References

Bauer, L.R., 1991, "Modeling Chronic Atmospheric Releases at the SRS: Evaluation and Verification of XOQDOQ(U)," WSRC-RP-91-320, Westinghouse Savannah River Company, Aiken, SC.

Cooper, R.E., 1975, "Computer Programs at SRL to Evaluate Environmental Effects of SRP Operations and Postulated Accidental Releases," DPST-75-384, Savannah River Laboratory, E.I. du Pont de Nemours & Co., Aiken, SC.

Cummins, C.L., D.K. Martin, and J.L. Todd, 1990, "Savannah River Site Environmental Report for 1989," WSRC-IM-90-60, Westinghouse Savannah River Company, Aiken, SC., p. xv.

DOE (U.S. Department of Energy), 1988, "Internal Dose Conversion Factors for Calculation of Dose to the Public," DOE/EH-0071, Washington, DC.

Eckerman, K.F., F.J. Congel, A.K. Roecklein, and W.J. Pasciak, 1980, "User's Guide to GASPAR Code," NUREG-0597, U.S. Nuclear Regulatory Commission, Washington, DC.

EPA, 1989, 40 CFR Part 61, "National Emission Standards for Hazardous Air Pollutants; Regulation of Radionuclides; Final Rule and Notice of Reconsideration," Federal Register, Vol. 54, No. 240.

Hamby, D.M., 1991, "Land and Water Use Characteristics in the Vicinity of the Savannah River Site (U)," WSRC-RP-91-17, Westinghouse Savannah River Company, Aiken, SC.

Hamby, D.M., and M.J. Parker, 1991, "Gaussian Dispersion and Dosimetric Modeling Sensitivity to Area-Specific 1982-1986 Meteorological Data Collected at the Savannah River Site (U)," WSRC-

RP-91-909, Westinghouse Savannah River Company, Aiken, SC.

ICRP (International Commission on Radiological Protection), 1980, "Statement and Recommendations of the 1980 Brighton Meeting of the ICRP, Part 2, Limits for Intakes of Radionuclides by Workers," ICRP Publication 30, Annals of the ICRP.

ICRP, 1981, "Limits for Intake of Radionuclides by Workers," ICRP Publication 30, Supplement to Part 2, Annals of the ICRP.

ICRP, 1989, "Age-Dependent Doses to Members of the Public from Radionuclides," ICRP Publication 56, Annals of the ICRP.

ICRP, 1991, "Risks Associated with Ionizing Radiations," Vol. 22, No. 1, Annals of the ICRP.

Marter, W.L., 1984, "Environmental Dosimetry for Normal Operations at SRP," DPST-83-270, Rev. 1, Savannah River Laboratory, E.I. du Pont de Nemours & Co. Aiken SC.

NCRP (National Council on Radiological Protection and Measurements), 1987, "Recommendations on Limits for Ionizing Radiation Exposure," NCRP Report No. 91, Washington, DC

NRC (U.S. Nuclear Regulatory Commission), 1977a, "Methods for Estimating Atmospheric Transport and Dispersion of Gaseous Effluents in Routine Releases from Light-Water-Cooled Reactors," Regulatory Guide 1.111, Rev. 1, Washington, DC.

NRC, 1977b, "Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I," Regulatory Guide 1.109, Rev. 1, Washington, DC.

Sagendorf, J.F., J.T. Goll, and W.F. Sandusky, 1982, "XOQDOQ: Computer Program for the Meteorological Evaluation of Routine Effluent Releases at Nuclear Power Stations," NUREG/CR-2919, U.S. Nuclear Regulatory Commission, Washington, DC.

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Appendix 1.

Physical Properties of Technetium

All isotopes of technetium are radioactive. Eleven radioactive isotopes of technetium may be present at SRS because of site operations. Each isotope's half-life and primary source at SRS are tabulated in Table A1-1. Half-lives range from seconds to millions of years. The isotopes can be present at SRS from two sources—as fission products from fission nuclear reactions and as activation products from either neutron capture or (n,2n) nuclear reactions.

Some of the isotopes may exist in their lowest energy level and in an excited energy level if this excited level does not have a very short half-life. In the latter case the isotope designation includes the letter "m" to indicate a metastable level; for example, ^{97m}Tc indicates that ^{97}Tc exists in an excited energy level.

^{99}Tc is the isotope with the greatest impact on the SRS environment.

Table A1-1. Characteristics of radioactive technetium isotopes that may be present at SRS because of site operations.

<u>Technetium Isotope</u>	<u>Primary Source</u>	<u>Half-Life</u>
^{97}Tc	AP	2.6×10^6 years
$^{97\text{m}}\text{Tc}$	AP	91 days
^{98}Tc	AP	1.5×10^6 years
^{99}Tc	FP	2.1×10^5 years
$^{99\text{m}}\text{Tc}$	FP	6.0 hours
^{100}Tc	AP	16 seconds
^{101}Tc	AP	14 minutes
^{102}Tc	FP	4.5 minutes
^{103}Tc	FP	50 seconds
^{104}Tc	FP	18 minutes
^{105}Tc	FP	7.7 minutes
^{106}Tc	FP	37 seconds
^{107}Tc	FP	29 seconds

AP = activation product; FP = fission product.

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