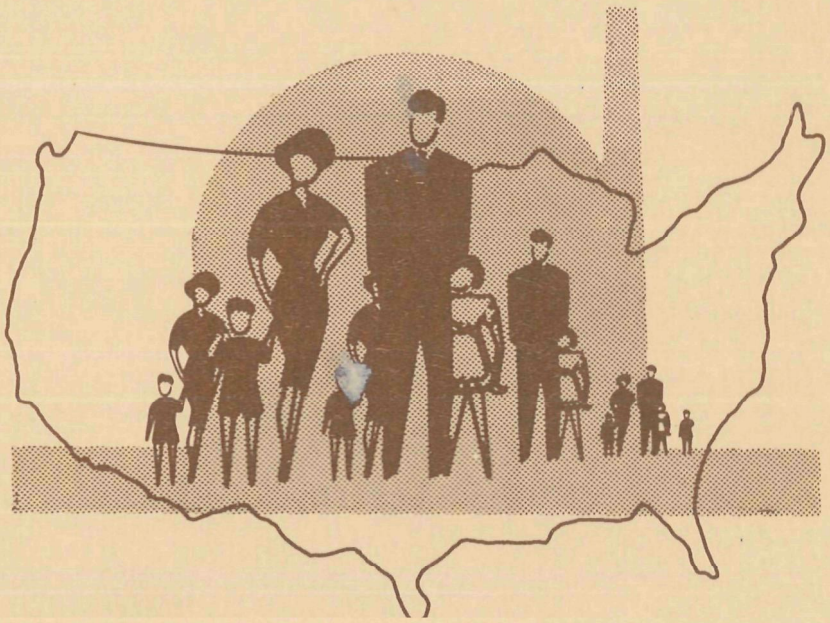
RADIOLOGICAL SURVEILLANCE STUDIES AT A PRESSURIZED WATER NUCLEAR POWER REACTOR



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RADIOLOGICAL SURVEILLANCE STUDIES AT A PRESSURIZED WATER NUCLEAR POWER REACTOR

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Foreword

The Environmental Protection Agency has the responsibility of carrying out a national program for measuring the population exposure to ionizing and nonionizing radiation and for assessing the radiological quality of the environment. The Radiation Research group conducts a program to determine the presence and examine the effects of radiation in order to form the scientific base for protecting man and his environment. Part of this research includes the development of means for identifying radionuclides, and methods for performing field studies at nuclear power stations and related facilities to quantitate discharged radionuclides and to measure radionuclides in the environment.

The projected increase in the use of nuclear power for generating electricity has placed an increased emphasis on nuclear surveillance programs at both the state and federal levels. The Environmental Protection Agency is engaged in studies at routinely operating nuclear power stations to provide information on the concentration of radionuclides in effluents and throughout the environment.

The data for this study were obtained at the pressurized water nuclear power reactor operated by the Yankee Atomic Electric Company at Rowe, Massachusetts. The results reported here are intended to provide an initial base for performing radiological surveillance at pressurized water nuclear power stations. Additional studies are planned at newer and larger stations to provide applicable information and to evaluate the effect of other operational and environmental conditions on radiation exposures to the population.

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1. Introduction

1.1 Need for Study

Each of the many nuclear power stations that will soon be operating in the United States requires an effective radiological surveillance program to assure that radiation exposure to the population is within acceptable limits. The Radiochemistry and Nuclear Engineering Branch of the Environmental Protection Agency (EPA)--formerly a part of the Bureau of Radiological Health, Public Health Service -- has, therefore, undertaken a program of studies at commercial nuclear power stations to suggest surveillance guidelines. The studies are intended to provide the following information: (1) identity and amount of radionuclides in effluents, (2) influence of station operation on radionuclide discharges, (3) degree of dispersion or concentration of radionuclides in the environment, (4) relative importance of specific radionuclides and vectors in exposing population groups, (5) magnitude of radiation exposure in the environment, and (6) applicability of various monitoring and measuring techniques.

In the future, much of this information should be available in response to the recent requirements by the Atomic Energy Commission (AEC) that nuclear power stations report semiannually the quantities of discharged radionuclides and the environmental levels of radiation and radioactivity that result from plant operation.[1] Until now, stations have reported discharges mostly in terms of gross activity and tritium.[2-7] Few of the environmental surveillance reports by the stations are publicly available, and most of these, while indicating the absence of significant radiation exposure through "less-than" values, provide little guidance in planning other monitoring programs. On the other hand, much general information is available on environmental surveillance for radionuclides (see footnote, Section 1.3), including several recent publications concerning nuclear facilities.[8,9]

The work described here was performed at the Yankee Nuclear Power Station, a pressurized water reactor (PWR). Yankee was built at Rowe, Massachusetts by the Westinghouse Electric Corp., and operates at a maximum power of 185 megawatts electric (MWe) and 600 thermal megawatts (MWt). It had produced more than 1×10^7 megawatt-hours between 1960 and 1969, and had passed through seven fuel cycles. The fuel is enriched (4.9 percent ^{235}U) uranium oxide (UO_2) pellets, clad in stainless steel. The operation of the station has been described by several authors.[10-12]

The study at Yankee follows one performed at the Dresden Nuclear Power Station,[13] a boiling water reactor (BWR) that began operation in 1959 and has been producing power at a rated capacity of 210 MWe. At present, a study is in progress at one of the newer and larger PWR's, and one is being planned at a large new BWR. In the meantime, it is believed that many of the reported observations are applicable to planning radiological surveillance at the newer BWR's and PWR's. Caution should be exercised, however, in applying the reported discharge data to newer stations, because aspects of both design and operation tend to differ among stations. For example, even gross activity values indicate that, among commercial nuclear power stations, Yankee discharges unusually small amounts of radionuclides other than tritium.[7]

1.2 Description of Study

The study at the Yankee Nuclear Power Station was planned and performed by the Radiochemistry and Nuclear Engineering Branch, supported by staff of the Divisions of Surveillance and Inspection, and of Technology Assessment, in the Office of Radiation Programs, EPA. The Yankee Atomic Electric Company, which operates the station, the Massachusetts Department of Public Health (MDPH), and the Division of Compliance of the AEC cooperated in the study. A field trip to Yankee was undertaken on June 3-4, 1969; other samples were obtained on October 4, 1968, April 1, 1969, July 10

and 29, 1969, June 4 and 10, 1970, November 19, 1970, and February 9, 1971. Participants in the study are listed in Appendix A.

As in the study at Dresden, measurements of radionuclides at the station, in effluents, and in environmental media were coordinated to attempt to show relative magnitudes among these three categories, critical radionuclides or pathways, and indicator radionuclides or media. Detailed descriptions are provided to demonstrate monitoring procedures. At Yankee, however, the amounts of discharged radionuclides were so small that only in the Sherman Reservoir, which receives liquid wastes, could radionuclides attributable to station effluents be detected. Although results of radionuclide analyses in other environmental media are reported, the most detailed discussion of environmental sampling, therefore, pertains to the aquatic environment.

Planning was guided by the available data on radionuclides in effluents and the environment, and an attempt was made to avoid duplicating ongoing programs. Monthly operating reports by Yankee Nuclear Power Station contain gross beta-gamma and tritium discharge values. Gross alpha activity, gross beta activity, tritium concentrations, and some gamma-ray spectral analyses are reported annually by Yankee's contractor for environmental surveillance. The MDPH reports gross beta activity in water and concentrations of photon-emitting radionuclides in benthal deposits. These data are cited in the appropriate sections of this report.

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*References that provide guidance for environmental surveillance, information on waste management at nuclear facilities, and discussion of the transfer of radionuclides in the environment are listed in Section 1.3 of Reference 13.

2. Radionuclides in Water on Site

2.1 Water Systems and Samples

2.1.1 *General.* A PWR such as Yankee has three consecutive cooling systems, shown schematically in Figure 2.1. In the main or primary system, water is heated under pressure in the reactor, circulates through four parallel steam generators, and returns at lower temperature to the reactor. In the secondary system, steam formed in the steam generators passes through the turbine to produce power and is then cooled to form water in the condenser. The water is then returned to the steam generators. In the third system, circulating coolant water is pumped from the bottom (25-m depth) of Sherman Reservoir through the secondary-system condenser at the rate of 530,000 liters/min (140,000 gal/min), and returned to the surface of the reservoir.(1)

2.1.2 *Main coolant system.* (1, 2) The main coolant is 64,000 kg of water that circulates approximately once every 12 seconds. In addition to the four high pressure loops for steam generation, the system includes the lines, shown in Figure 2.1, by which water is added or withdrawn for pressure control, chemical adjustments, continuous purification, and sample collection. At the time of the study, the flow rate through the purification filter and demineralizer was 113 kg/min (30 gal/min), (2) which results in a mean turnover period of 64,000 kg ÷ 113 kg/min = 570 min (3.4×10^4 sec) for main coolant water.

The water for the system is taken from Sherman Reservoir and demineralized. During two-thirds of the operating cycle of approximately 16 months, the main coolant water contains boron (in the form of boric acid) to control the neutron flux, and is, as a consequence, at a pH value of approximately 5. The boron concentration is decreased gradually during this period from 1,300 parts per million (ppm) to 0 ppm. During the final "power stretchout" period of the operating cycle, the water contains no boron, but ammonium hydroxide is added to maintain the pH value at approximately 9 for corrosion control.

Nitrogen gas is added to the system to maintain the concentration of oxygen from ambient air at a low concentration, and hydrogen gas is added to depress below 0.1 ppm the concentration of oxygen formed by the radiation-induced decomposition of water. The concentration of nitrogen is approximately 4 cc/kg of water at standard temperature and pressure (STP) in the absence of ammonium hydroxide, and 12 cc/kg of water at STP with ammonium hydroxide in the coolant. The concentration of hydrogen is approximately 35 cc/kg of water at STP.

During refueling, the space above the opened reactor vessel is flooded, and fuel is carried to the fuel transfer pit on a carriage through the transfer chute. Upper and lower lock valves in the transfer chute reduce the movement of water from reactor to fuel transfer pit. The water is continuously purified by passage through the shutdown demineralizer.

2.1.3 *Secondary coolant system.* (1,2) The secondary coolant is 190,000 kg (420,000 lb) of water that circulates approximately once every 10 minutes. The water is obtained from Sherman Reservoir and demineralized. Accumulation of salt in the steam generators is minimized by a relatively small continuous blowdown and a more massive blowdown for one to two hours every night. The rate of continuous blowdown depends on the salt content of water in the steam generators, and averages approximately 2,000 liters/day; the once-nightly blowdown is approximately 12,000 liters/day.

2.1.4 *Paths of radionuclides from main and secondary systems.* The radionuclides in the main coolant water are fission products and activation products. The fission products in the water are formed within the uranium oxide fuel and enter the water through small imperfections in the stainless steel cladding of the fuel elements. Other possible sources of fission products--apparently minor at Yankee--are fuel that contaminates the surface of new fuel elements ("tramp uranium") and fuel that reaches the main coolant water from failed fuel elements. Activation products in water are formed by neutron

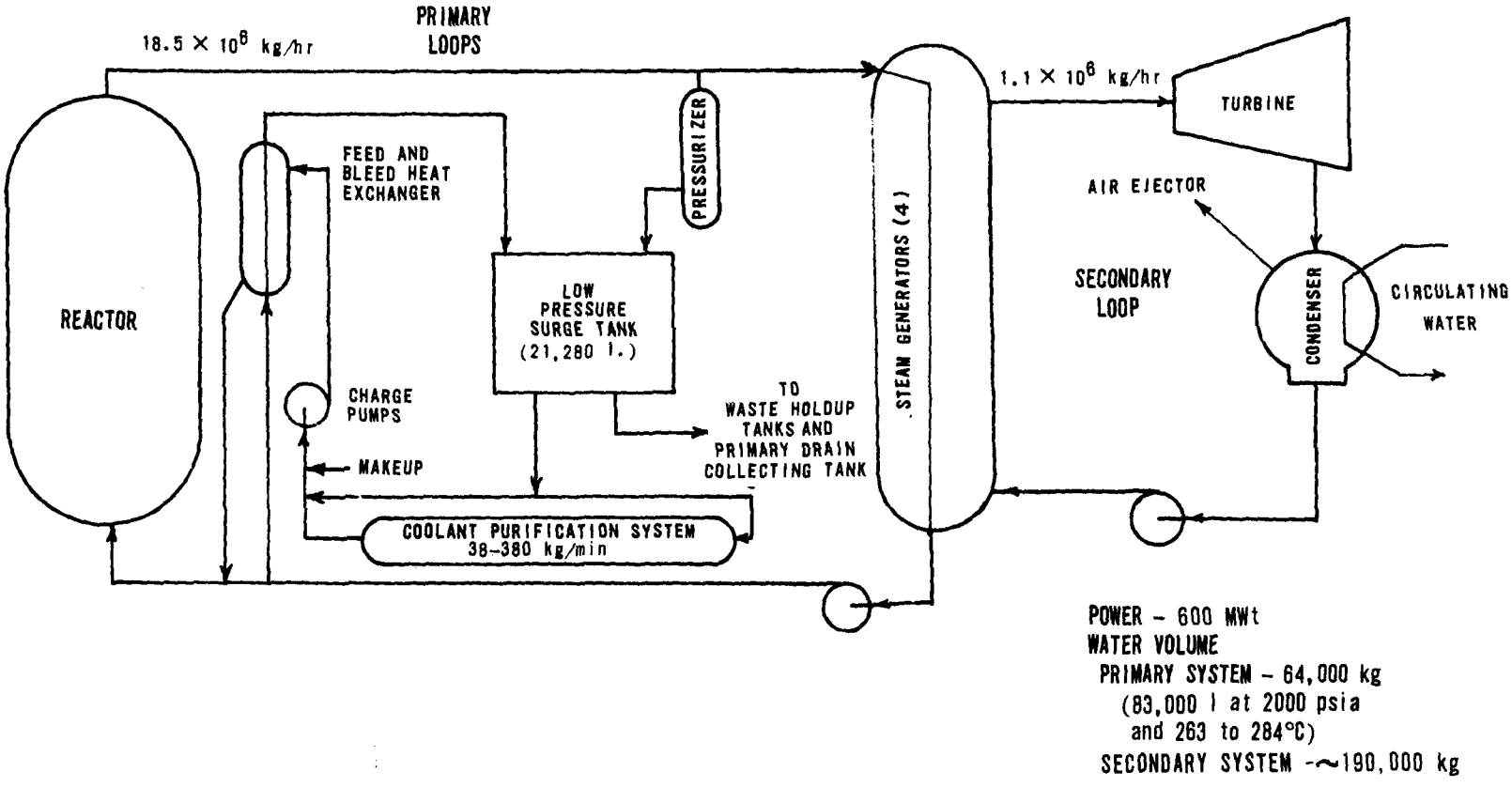

Figure 2.1. Coolant Flow Schematic of Yankee PWR.

irradiation of the water and its contents (including gases and dissolved or suspended soils) and of reactor materials that subsequently corrode or erode.

The radionuclides in main coolant water circulate and decay within the system, deposit as crud (which may later recirculate), are retained by the purification filter and demineralizer (which are periodically replaced and shipped off-site as solid waste), or leave the system with gases and liquids. Paths from the system to the environment are shown schematically in Figure 2.2.

Under routine operation, water and associated gases leave the main coolant system at leaks and by intentional discharge from the low-pressure surge tank for pressure control, boron-concentration adjustment and sample collection. The water passes through the primary drain collecting tank and is stored in a waste holdup tank. When a sufficient volume has accumulated, the liquid waste is treated in an evaporator. The distillate is discharged to effluent circulating coolant water, and the residue is shipped as solid waste. The water released during refueling is also processed in the evaporator. Gas from the liquid waste system is collected in the gas surge drum. The gaseous waste system is described in Section 3.1 and the liquid waste system, in Section 4.1.

Liquid waste from the reactor plant was estimated to consist of the following constituents in 1969:

main system leakage	0.2×10^6 liters/year
routine operation of main system	0.9 "
refueling	0.8 "
incinerator rotoclone (2)	0.4 "
decontamination (2)	0.1 "
tank moats (2)	0.2 "
total reactor plant (see Section 4.1)	2.6×10^6 liters/year

The main system leakage was an average of 1 percent per day of the system contents. (2) The discharge during routine operation includes of the order of 1×10^5 liters for each major reduction of boron concentration during startup or for power stretchout.

Radionuclides enter the secondary coolant system through leaks in the steam generators. Normally, the leakage rate is only a few liters per day in each of the four loops, but occasionally the leakage rate increases rapidly until the faulty tubes are plugged. As a rule of thumb, tubes are plugged before the leakage rate approaches 4,000 liters/day; high leakage rates, requiring loop isolation and tube repair, have occurred on four occasions in the period 1960-1969.(2).

Secondary system liquid waste consisted of 15×10^6 liters/year in 1969 and 1970, of which approximately 37 percent was blowdown, 57 percent was leakage, and 6 percent, discharge from the spent fuel pit and waste tank moats. Through combined leakage and blowdown, 19 percent per day of the system water was discharged. The water is discharged directly, without storage or treatment, to effluent circulating coolant water.

2.1.5 *Other liquids on site.* During and after refueling, used fuel elements are stored in demineralized water in the fuel transfer pit. The water is circulated through the fuel-pit ion-exchanger for purification, and through a cooler to control the temperature. After use, water from the fuel transfer pit is discharged with secondary system liquid wastes.

The waste holdup system consists of two tanks with a total capacity of 570,000 liters (150,000 gal). Reactor plant liquid wastes are pumped into these tanks from the primary drain collecting tank, and are stored until treated in the evaporator. An 18,000-liter (5,000-gal) gravity drain tank collects other reactor plant liquid wastes for treatment in the evaporator.

Component cooling water is circulated in the neutron shield tank and other components at the station. The water contains approximately 400 ppm potassium chromate as corrosion inhibitor. It has not been discharged from the station. (2)

Safety injection water, containing 1 percent (by weight) boric acid, is stored in a 470,000-liter (125,000-gal) tank to be available for cooling the reactor core during a major loss-of-coolant accident. It is used to flood the shield-tank cavity during refueling and is returned to the storage tank after refueling. It is not normally discharged.(2)

The incinerator at Yankee utilizes a mechanical centrifugal scrubber (rotoclone) to moisten and retain dust particles from the exhaust air steam. In 1969, approximately 4×10^5 liters of water were used in the scrubber. (2) This water is stored and processed by evaporation with main coolant liquid waste (see Section 2.1.4).

The sanitary-system water at Yankee is passed into a septic tank on the site. Normally, it would not be contaminated with radioactivity.

Because Yankee is located between a ridge and the Sherman Reservoir, rain water runs off across the site. Two yard drains lead into the reservoir (see Section 4.2.3). In addition, some water collects in storage tank moats and is treated as necessary (see Section 2.1.4). In 1969, approximately 200,000 liters of

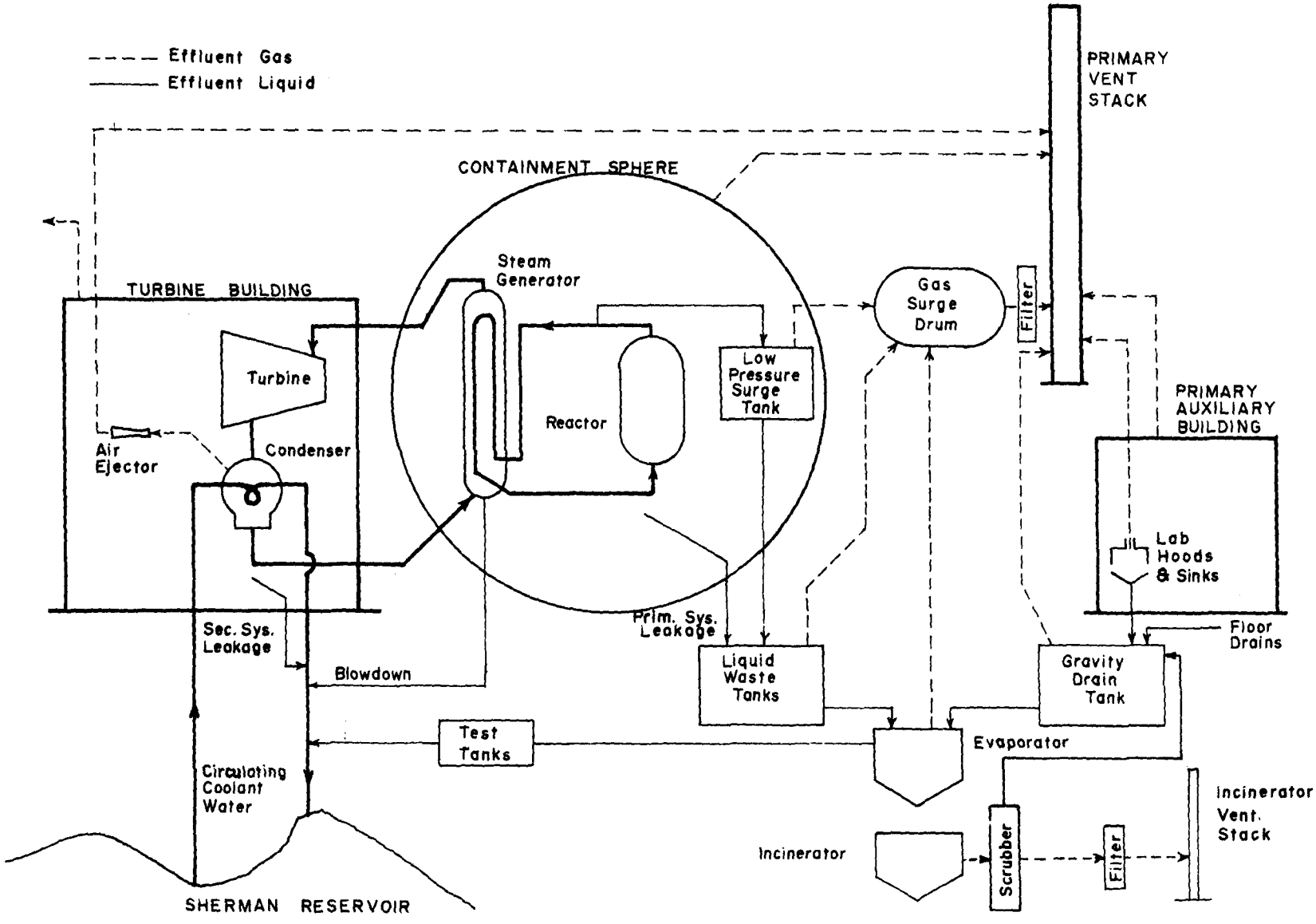

Figure 2.2. Paths of Effluents at Yankee PWR.

water from the moats were stored and evaporated with reactor plant wastes, and 300,000 liters were discharged with secondary plant liquid waste.[2]

2.1.6 *Samples.* To identify potential radioactive effluents, liquids at the Yankee Nuclear Power Station were sampled within the plant, where radionuclides were at much higher concentrations and therefore more easily detected than at the point of release. The following water samples were provided by Yankee staff in plastic bottles:

(1) main coolant, 1 liter, collected Oct. 4, 1968 at 1300;

(2) main coolant, 2 liters, collected July 10, 1969 at 0840;*

(3) main coolant, 2 liters, collected June 10, 1970 at 0945;

(4) waste holdup tank, 1 liter, collected Oct. 4, 1968 at 1000;

(5) continuous steam generator blowdown, 1 liter, collected Oct. 4, 1968 at 0950;

(6) continuous steam generator blowdown, 3.5 liters, collected June 10, 1970 at 1000;

(7) secondary system condensate, 2 liters, collected July 10, 1969 at 0855;*

(8) secondary system condensate discharge, 1 liter, collected June 10, 1970 at 1120;

(9) component cooling system, 3.5 liters, collected June 10, 1970 at 1100; and

(10) safety injection tank, 3.5 liters, collected June 10, 1970 at 1045.

One liter each of Samples (2), (3), and (7) was acidified with 100 ml conc. HNO_3 to reduce deposition of radionuclides on the walls of the bottle; the other liter of sample remained unacidified to prevent loss of radioiodine. Sample (6) was filtered at collection, and the filter and filtrate were analyzed separately.

2.2 Analysis

2.2.1 *General approach.* Aliquots of all samples were first counted for gross alpha and beta activity, then examined with gamma-ray spectrometers, and finally analyzed radiochemically. Analyses were performed for high-yield fission products and common activation products in reactor water. Relatively short-lived radionuclides could not be measured because of radioactive decay between sampling and analysis: the main coolant water of Oct. 4, 1968 was first analyzed 20 hours after sampling; the other two main-coolant samples, after 2 days; and the other samples, usually after 1 week. Aliquot volumes ranging from less than 1 ml to 200 ml were used.

A special effort was made to measure radionuclides that, because they emit only weak beta particles, tend to be underestimated by gross beta counting and are not detected by gamma-ray spectrometry. These radionuclides were 12.3-yr ^{3}H (maximum beta particle energy, 18 keV), 5,730-yr ^{14}C (158 keV), 88-d ^{35}S (167 keV), 92-yr ^{63}Ni (67 keV), and 1.6 x 10^{7}-yr ^{129}I (150 keV).

Radionuclide concentrations were computed from count rates obtained with counters calibrated as functions of gamma-ray or average beta-particle energies. All values were corrected for radioactive decay and are given as concentrations at sampling time. The values of decay rates and branching ratios are from recent publications.[4, 5, 6]

Loss of radionuclides from the samples by volatilization and adsorption on container walls was given special consideration.[7] Concentration values for radioiodine and ^{14}C were obtained with unacidified samples. For other radionuclides in main-coolant water, data from analyses of the acidified samples and *aqua-regia* leaches of the containers were combined. Results of analyzing the filtered blowdown water and the filter (sample No. 6) were also combined.

2.2.2 *Gamma-ray spectrometry.* Radionuclides that emit gamma rays were identified in aliquots of main coolant water by multichannel spectrometry with a Ge(Li) detector (see Figures 2.3 and 2.4).

*We thank G. J. Karches, Northeastern Radiological Health Laboratory, Public Health Service (NERHL, PHS), for obtaining these samples.

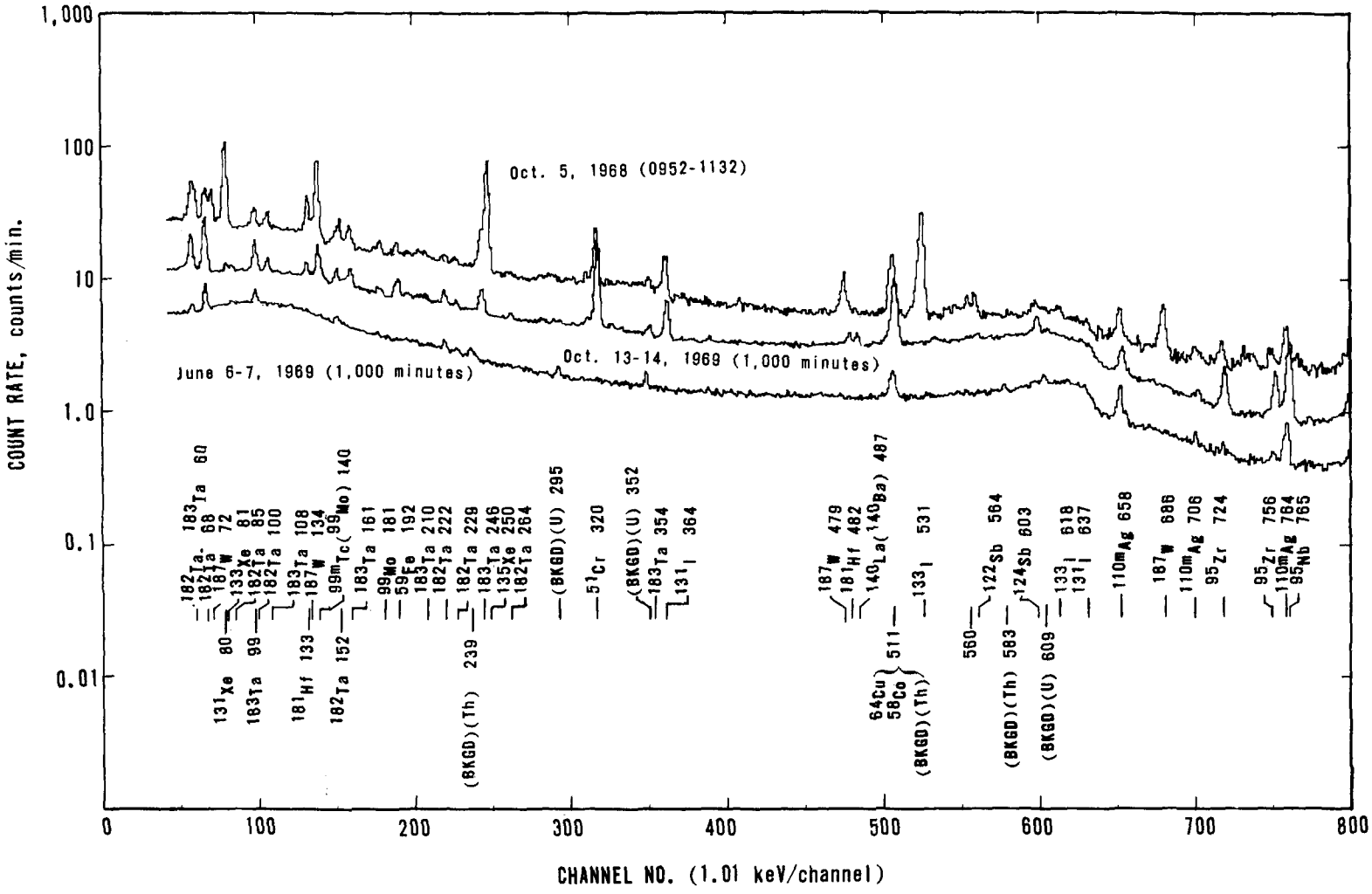

Figure 2.3. Gamma-ray Spectra of Main Coolant Water, 40 – 808 keV.

Detector: *Ge(Li), 10.4 cm² × 11 mm, trapezoidal*

Sample : *35 ml (including 1 ml acid), collected Oct. 4, 1968 at 1300.*

Counts : *At times indicated on spectra; background (bkgd) not subtracted.*

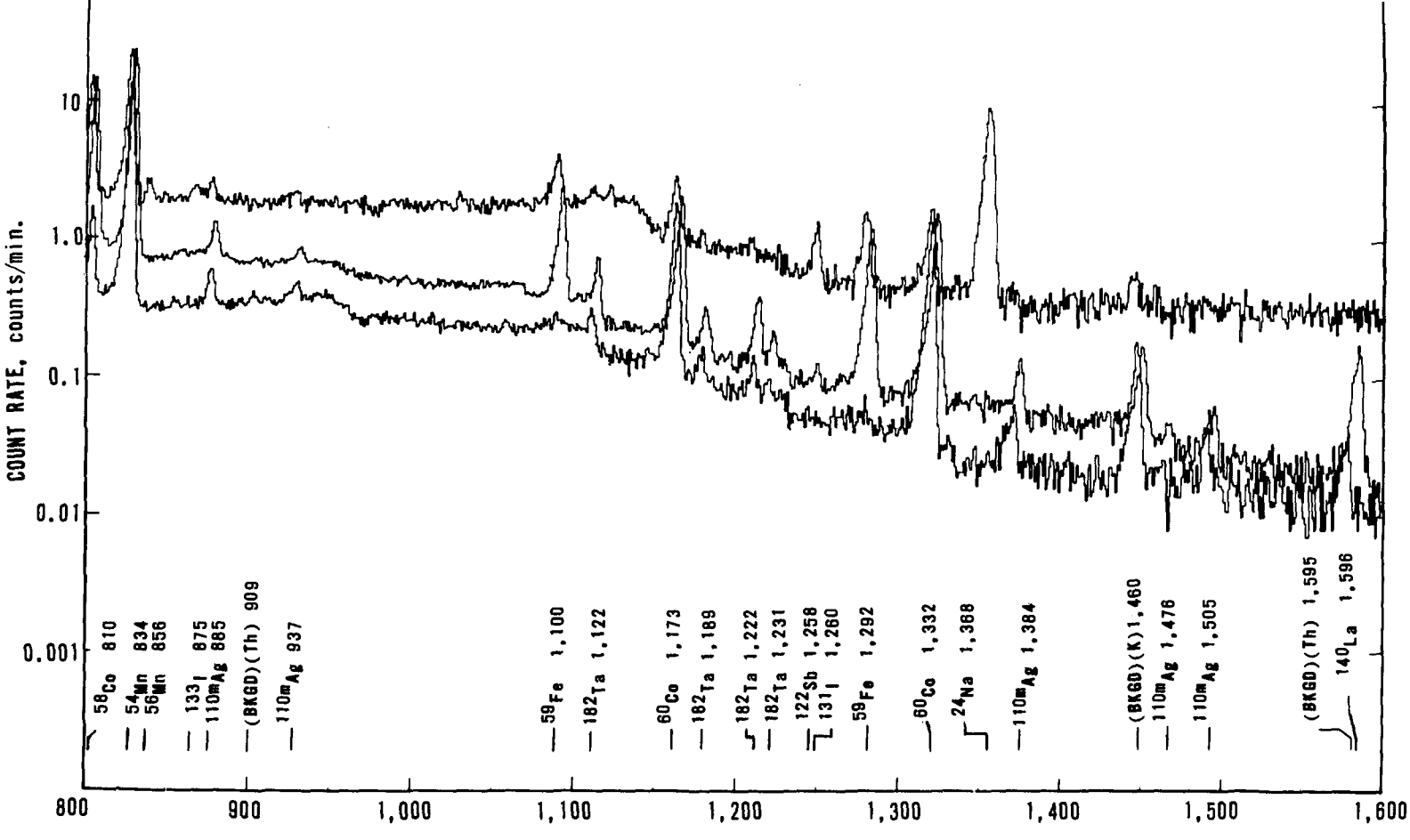

Figure 2.4. Gamma-ray Spectra of Main Coolant Water, 808 - 1,616 keV.

Detector: Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal

Sample : 35 ml (including 1 ml acid), collected Oct. 4, 1968 at 1300.

Counts : At times indicated on spectra; background (bkgd) not subtracted.

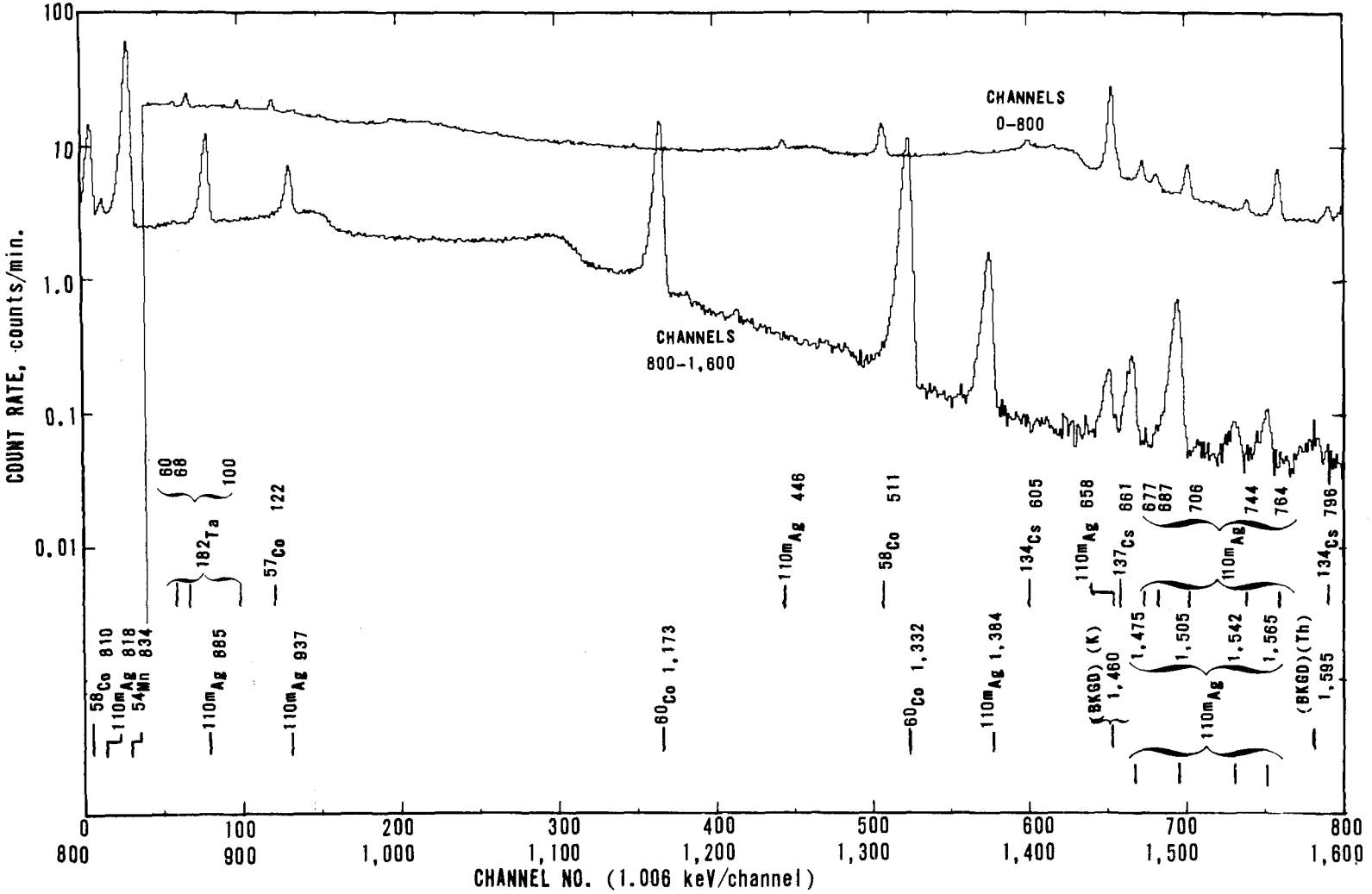

Figure 2.5. Gamma-ray Spectrum of Liquid in Waste Holdup Tank.

Detector: Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal

Sample : 35 ml (including 1 ml acid) collected Oct. 4, 1968.

Count : Oct. 10-11, 1968 (1,000-minute background not subtracted).

Spectral analyses were repeated at appropriate intervals to measure long-lived radionuclides in the main coolant without interference by short-lived ones and to measure half lives for confirming the identity of the radionuclides. The minimum half life of the measured radionuclides was 6 hours, and the maximum, 30 years. Minimum detectable concentrations at these half lives were approximately 1×10^{-4} and 1×10^{-5} microcurie per milliliter (μCi/ml), respectively.

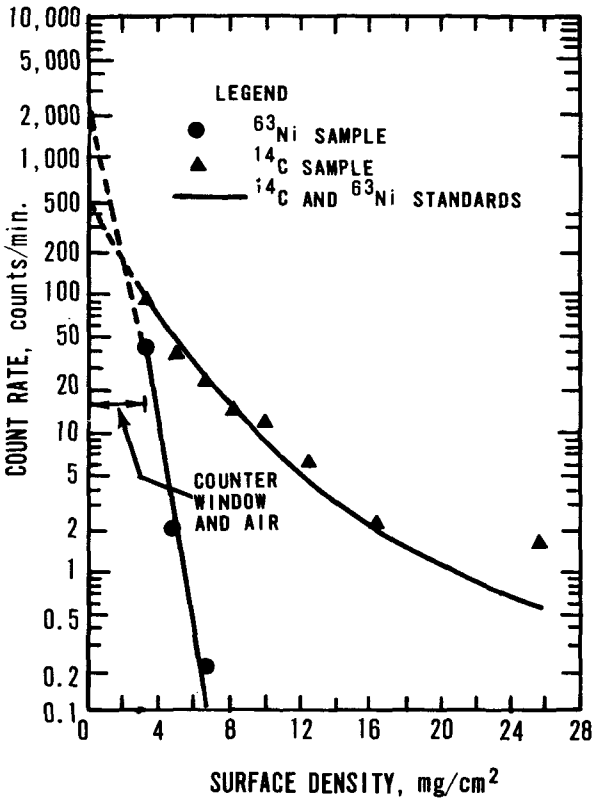

Figure 2.6. Aluminum Absorber Curves of ^{14}C and ^{63}Ni Separated from Yankee Waste Holdup Tank Liquid.

Detector: Low-background G-M end-window.

Samples : ^{14}C: unacidified 5 ml aliquot of sample collected Oct. 4, 1968; ^{63}Ni: acidified 25 ml aliquot.

Counts : April 14-15, 1970, 200 min. at each point.

The main-coolant sample of Oct. 4, 1968, was analyzed by obtaining 6 spectra in the interval from 0.9 to 250 days after sample collection. Main coolant samples collected on July 10, 1969, and June 10, 1970, were analyzed similarly, but gamma rays from relatively short-lived radionuclides were obscured because the initial spectrum could not be obtained so soon after sampling and the ^{24}Na content was relatively high. Analysis by Ge(Li) spectrometry was also performed for the sample from the waste holdup tank (see Figure 2.5).

All other water samples were analyzed by multichannel spectrometry with a 10 cm x 10 cm NaI(Tl) detector. These samples contained fewer radionuclides at much lower levels of radioactivity. Hence, the higher energy resolution of the Ge(Li) detector was generally unnecessary, and the higher counting efficiency of the NaI(Tl) detectors was advantageous.

2.2.3 *Radiochemistry.* Radiochemical analysis was performed to confirm spectral identification by gamma-ray energy and half life, measure radionuclides more precisely and at lower concentrations than by instrumental analysis of a mixture, and detect radionuclides that emit only obscure gamma rays or none at all. After chemical separation, the following detectors were used: NaI(Tl) crystal plus spectrometer for photon-emitting radionuclides; low-background end-window Geiger-Mueller (G-M) counter for ^{14}C, ^{32}P, ^{35}S, ^{89}Sr, ^{90}Sr, ^{129}I, and ^{185}W; liquid scintillation detector plus spectrometer for ^{3}H and ^{63}Ni; and xenon-filled proportional counter plus spectrometer for ^{55}Fe. Measurements with the G-M detector includes observation of the effect of aluminum absorber on count rates to determine maximum beta-ray energies (see Figure 2.6) and thus confirm radionuclide identification.

2.3 Results and Discussion

2.3.1 *Radioactivity in main coolant water.* Tritium was by far the most abundant of those radionuclides with half lives longer than 6 hours (see Table 2.1). At second highest concentration was ^{24}Na, some of which may have been formed in sodium salt added at times by Yankee staff as leak tracer. (2) The sum of all other measured radionuclides was between 0.003 and 0.005 μCi/ml. The average gross activity (without ^{3}H) reported by Yankee (see below) was

Table 2.1

Radionuclide Concentration in Main Coolant Water, μCi/ml*

Radionuclide	Oct. 4, 1968	July 10, 1969	June 10, 1970
		from fuel	
12.3 -yr ^{3}H †	5.0	4.0×10^{-1}	1.8
50.5 -d ^{89}Sr	7×10^{-6}	$<5 \times 10^{-7}$	1×10^{-6}
28.5 -yr ^{90}Sr	2×10^{-7}	8×10^{-7}	2×10^{-7}
9.7 -hr ^{91}Sr	9.8×10^{-5}	NA	NA
65 -d ^{95}Zr †	4.5×10^{-5}	6×10^{-6}	2×10^{-6}
35.1 -d ^{95}Nb†	5.0×10^{-5}	6×10^{-6}	1×10^{-6}
66.2 -hr ^{99}Mo†	1.1×10^{-4}	4.6×10^{-5}	1.9×10^{-4}
8.06-d ^{131}I	5.2×10^{-5}	3.0×10^{-5}	5.5×10^{-5}
20.9 -hr ^{133}I	6.6×10^{-4}	4.0×10^{-4}	8.0×10^{-4}
6.7 -hr ^{135}I	9.1×10^{-4}	NA	NA
5.29-d ^{133}Xe	2.4×10^{-4}	1.8×10^{-5}	$<1 \times 10^{-5}$
9.1 -hr ^{135}Xe	2.5×10^{-4}	NA	NA
2.07-yr ^{134}Cs†	3×10^{-7}	8×10^{-7}	3×10^{-7}
30 -yr ^{137}Cs	2×10^{-7}	3×10^{-6}	1×10^{-7}
12.8 -d ^{140}Ba	1.2×10^{-5}	$<1 \times 10^{-6}$	1×10^{-6}
	from activation of water, cladding, and construction materials		
5730 -yr ^{14}C	NA	1.5×10^{-5}	8×10^{-6}
15.0 -hr ^{24}Na	1.6×10^{-3}	1.8×10^{-2}	3.8×10^{-2}
14.3 -d ^{32}P	1.0×10^{-5}	2.5×10^{-5}	6.5×10^{-5}
88 -d ^{35}S	NA	3×10^{-6}	3×10^{-6}
27.7 -d ^{51}Cr	8.5×10^{-4}	2.1×10^{-4}	5.0×10^{-5}
313 -d ^{54}Mn	5.4×10^{-4}	2.4×10^{-4}	4.0×10^{-5}
2.7 -yr ^{55}Fe	1.0×10^{-4}	3.7×10^{-4}	1.0×10^{-4}
44.6 -d ^{59}Fe	1.9×10^{-4}	8.9×10^{-5}	2.0×10^{-5}
270 -d ^{57}Co	$\sim 6 \times 10^{-7}$	2×10^{-6}	$<1 \times 10^{-6}$
71.3 -d ^{58}Co	3.4×10^{-4}	1.0×10^{-3}	8.0×10^{-5}
5.26-yr ^{60}Co	9.0×10^{-5}	1.3×10^{-4}	1.2×10^{-5}
92 -yr ^{63}Ni	1.9×10^{-5}	4×10^{-6}	1×10^{-6}
12.8 -hr ^{64}Cu	$\sim 2 \times 10^{-4}$	1×10^{-5}	1.4×10^{-3}
253 -d ^{110m}Ag	1.9×10^{-5}	$<1 \times 10^{-6}$	1.7×10^{-5}
2.7 -d ^{122}Sb	4.9×10^{-5}	2×10^{-6}	2.5×10^{-4}
60.2 -d ^{124}Sb	2.0×10^{-5}	1×10^{-6}	4.0×10^{-5}
42.5 -d ^{181}Hf	$\sim 8 \times 10^{-6}$	$\sim 4 \times 10^{-6}$	NA
115 -d ^{182}Ta	$\sim 6 \times 10^{-5}$	$\sim 1.1 \times 10^{-5}$	$\sim 3 \times 10^{-6}$
5.1 -d ^{183}Ta	1.1×10^{-4}	1.5×10^{-5}	NA
75 -d ^{185}W	1.0×10^{-5}	1.4×10^{-5}	NA
24 -hr ^{187}W	4.8×10^{-4}	2.6×10^{-4}	NA

*Concentrations at time of sampling.

†^{3}H is also an activation product; ^{95}Zr, its daughter ^{95}Nb, and ^{99}Mo may also be activation products; ^{134}Cs is produced by the (n,γ) reaction with fission-produced ^{133}Cs.

Notes:
1. NA = not analyzed.
2. < values are 3σ counting error.
3. The following fission products were not detected ($< 1 \times 10^{-6}$ μCi/ml): ^{93}Y, ^{97}Zr, ^{103}Ru, ^{106}Ru, ^{127}Sb, ^{129}I, ^{132}Te, ^{141}Ce, ^{143}Ce, ^{144}Ce, ^{147}Nd. The radionuclides ^{65}Zn, ^{136}Cs, and ^{239}Np were also not observed at this minimum detectable level.
4. No gross alpha activity could be detected ($<1 \times 10^{-9}$ μCi/ml).

approximately 0.1 μCi/ml; this presumably consists mostly of radionuclides with half lives shorter than 6 hours.

Fission products contributed only a small fraction of the non-tritium radioactivity, and many relatively long-lived high-yield fission products could not be detected at the limiting sensitivity of approximately 1 x 10^{-6} μCi/ml (see footnote 3 to Table 2.1). Most of the other radionuclides are neutron activation products that have been reported earlier.[7,8] They are formed in water, steel, copper, silver (in the original Ag-In-Cd control rods), antimony (in the Sb-Be neutron source), hafnium (in new Hf control rods) and zirconium (in Zircaloy-2 cladding of control rods). In addition to activation products previously reported in power reactors, ^{14}C, ^{35}S, and ^{63}Ni were found at relatively low concentration. No radionuclides that emit alpha particles were detected (<1 x 10^{-9} μCi/ml).

Considerable differences among the samples in radionuclide concentrations were expected because, during a fuel cycle, the boron concentration decreases, the pH value increases, and the power level decreases toward the end (see Figure 2.7). The monthly average values reported by Yankee [9, 10, 11] at the sampling periods are:

	October 1968	July 1969	June 1970
core	VII	VII	VIII
month of cycle	6th	15th	9th
power level, MWe	182	130	169
boron, ppm	585	0	183
pH	5.3	9.4	6.7
radioactivity, μCi/ml	0.084	0.085	0.108

Radionuclide concentrations are also affected by many other variables, including the quality of the fuel elements, the occurrence of shutdowns, the rate of coolant-water purification and turnover, and the extent of accumulation of radioactive material within the coolant system.

2.3.2 *Tritium in main coolant water.* Measured concentrations are consistent with the reported monthly average values shown in Appendix B.1. The sources of tritium at Yankee are believed to be known, but the contribution of each has not been quantified. These sources are:

(1) ternary fission within the fuel. The generation rate of 85 μCi/sec at 600 MWt was computed from a fission yield of 9.5 x 10^{-5} (see Appendix B.3). Other values of the fission yield, [12] ranging from 8 x 10^{-5} to 13 x 10^{-5} would change the computed generation rate proportionally.

(2) the fast neutron reaction ^{10}B (n,2α) ^{3}H in main coolant water. A generation rate of 4 μCi/sec at 600 MWt (185 MWe) and a boron concentration of 1,300 ppm was derived from predicted values for a reactor at 1,000 MWe and 1,500 ppm B,[13] and also from values for a reactor at 1,000 MWt and 1,200 ppm B. [14]

(3) the two-step reaction ^{10}B (n,α) ^{7}Li (n, nα) ^{3}H in main coolant water. This reaction appears to contribute only a small fraction of the ^{3}H produced in the above-cited direct reaction, [15] although Ray[13] indicates that it is important if ^{7}Li accumulates in the coolant.

(4) the reaction ^{2}H (n,γ) ^{3}H in main coolant water. This was computed to produce ^{3}H at the relatively low rate of 0.06 μCi/sec in a slightly larger boiling water reactor. [7]

(5) reactions (2) and (3) in the eight shim rods that are located near the periphery of the reactor vessel and consist of Ziracaloy-clad steel with 1.2 percent boron. The remote location, small amounts of boron relative to that initially in the coolant, and the cladding, which appears to be a good barrier against ^{3}H,[7] suggest that this source is minor.

(6) other reactions, such as ^{3}He (n,p) ^{3}H (where the ^{3}He is produced by the beta-decay of ^{3}H) and ^{10}B (n,d) ^{9}Be (n, α) ^{6}Li (n,α) ^{3}H. The former accounts for less than 0.5 percent of reactions (2) and (3); [15] the latter is believed to produce a negligible amount of ^{3}H. [13]

On the basis of the above evaluation, only the first two-ternary fission combined with high fractional transfer from fuel to water, and the ^{10}B (n,2α) ^{3}H reaction are important sources of tritium at Yankee.

The 1,400 curies of tritium discharged annually in 1970 and 1969 (see Section 4.1.2), if produced during approximately 320 days (2.8 x 10^{7} sec) of operation per year, indicate an average generation rate of 50 μCi/sec. This value suggests that ternary fission produced more than 90 percent of the discharged ^{3}H, and that approximately one-half of

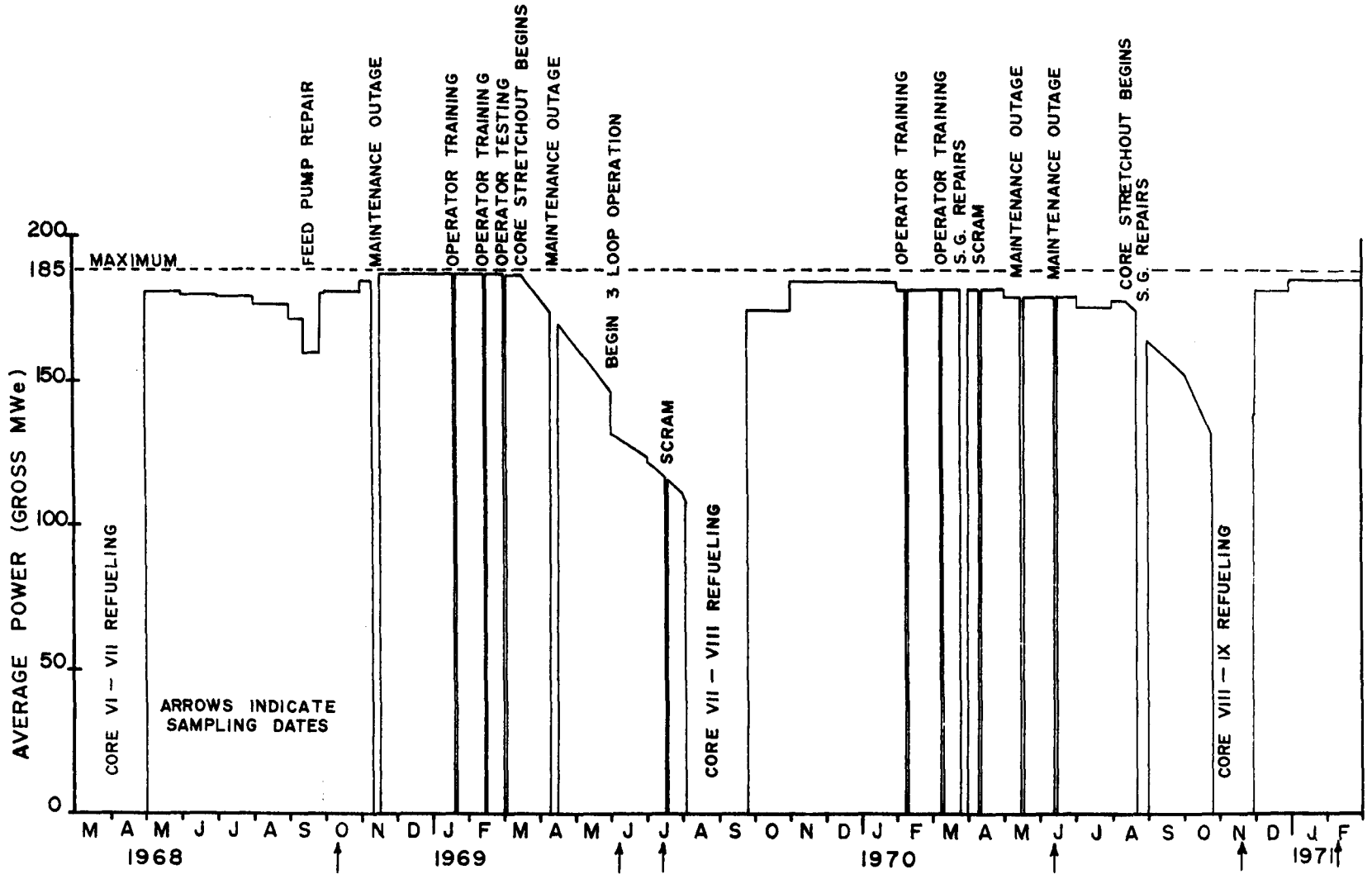

Figure 2.7. Yankee Electrical Loading, April, 1968 through February, 1971 (From Monthly Operation Reports).

the ^3H formed by ternary fission moved from fuel into coolant water. Alternatively, either the generation rate of 4 μCi/sec by the ^{10}B (n, 2α) ^3H reaction is vastly underestimated, at least one of sources (3) to (6) is not negligible, or another source of tritium exists in the reactor.

The monthly average ^3H concentrations measured in main coolant water by Yankee (see Appendix B.1) yield conflicting results concerning the major source of the ^3H. The concentration of ^3H is related to its rate of production in, or transfer to, the main coolant by:

$$C = R(1 - \exp -\lambda t)(V\lambda)^{-1} + C_o \exp - \lambda t \qquad (2.1)$$

where C : radionuclide concentration in main coolant, μCi/ml

R : rate of production in or transfer to main coolant, μCi/sec

λ : water turnover constant (1.2×10^{-7} sec^{-1} at 1 percent per day) + radionuclide decay constant (1.78×10^{-9} sec^{-1} for ^3H)

t : reactor operating period, sec

V : main coolant water volume (6.4×10^7 ml)

C_o : radionuclide concentration at t = 0, μCi/ml

Thus, after continuous operation for, say, five months at maximum power and boron concentration, the ^3H concentration from dissolved boron would be

$$C_B = 4\ (1 - 0.21)(6.4 \times 10^7 \times 1.2 \times 10^{-7})^{-1}$$
$$= 0.4\ \mu\text{Ci/ml} \qquad (2.2)$$

while that from ternary fission at 50 percent leakage from fuel would be

$$C_{fp} = 85 \times 0.5\ (1 - 0.21)(6.4 \times 10^7 \times 1.2 \times 10^{-7})^{-1}$$
$$= 4.4\ \mu\text{Ci/ml} \qquad (2.3)$$

These computed values are consistent with the magnitude of measured averages toward the beginning of the fuel cycle. Concentrations at the end of the fuel cycle–when no boron was in the coolant and special efforts had been made to change to fresh water–were considerably lower in 1969 and 1970, however, than would be computed by equation 2.3, even when the lowered power levels and briefer period of operation are taken into account.

This inconsistency may be due to the previously discussed alternative modes of tritium formation, or to the influence on the fractional transfer of ^3H from fuel to water by other factors, such as ^3H accumulation, local temperature, and surges of ^3H through cladding during start-up. Continuing observations of ^3H levels at Yankee, and a current study at the Ginna PWR [16] -- where the fuel elements are Zircaloy-clad–may provide quantitative information on the sources of ^3H.

2.3.3 *Fission products in main coolant water.* The ^{131}I concentrations and atom ratios of ^{131}I relative to ^{133}I in the three samples were comparable to average monthly values reported by Yankee: [9-11]

	^{131}I, μCi/ml	
	This report	Yankee monthly avg.
Oct., 1968	5.2×10^{-5}	2.3×10^{-5}
July, 1969	3.0×10^{-5}	2.0×10^{-5}
June, 1970	5.5×10^{-5}	3.4×10^{-5}

	$^{131}I/^{133}I$, atom/atom*	
	This report	Yankee monthly avg.
Oct. 1968	0.73	0.83
July, 1969	0.69	0.44
June, 1970	0.64	0.51

*$\left(\frac{\mu\text{Ci}}{\text{ml}} \times \text{half life}\right)\ ^{131}I / \left(\frac{\mu\text{Ci}}{\text{ml}} \times \text{half life}\right)\ ^{133}I$

The 6-hour ^{135}I could be measured only in the one sample that was analyzed promptly after collection. The concentration of 1.6×10^7-yr ^{129}I was below detectable levels in all three samples, as is expected from the low production rate (more than a billion-fold lower than ^{131}I).

According to the very low ratios of turnover rates of the fission products in main coolant water to production rates in fuel (see Appendix B.4), an extremely small fraction of these radionuclides moved from fuel to coolant. In the Oct. 4, 1968 sample, ratios ranged from 0.5×10^{-9} to 22×10^{-9} with an average of 8.2×10^{-9}; in the two other samples, most ratios were equal or lower. The similar ratios for ^{131}I, ^{133}I, and ^{135}I, despite their different half lives, suggest that the composition approached that of a "recoil mixture". This results from the rapid transfer of fission products from fuel to water.

The main-coolant water samples also contained the radioactive gases ^{133}Xe and ^{135}Xe. The values shown in Table 2.1 refer to concentrations in excess of those due to the decay of ^{133}I and ^{135}I in the

samples, and show that a fraction of these radioactive gases remains in water despite its high temperature and rapid movement. The values are not quantitative in view of the probable loss of xenon from water during sample collection and aliquot removal. Concentrations of ^{133}Xe and ^{135}Xe in the gas phase are given in Section 3.3.1 for the same June 10, 1970 sample whose aqueous phase is described in Table 2.1.

The measured concentrations of fission products were 10^2 - to 10^6- fold lower than was predicted by Yankee for 1 percent of fuel rods with pin holes or small cracks, at a 38-liter/min flow rate through the purification system:

Radionuclide	Predicted concentration, (1) μCi/ml	Measured (Oct. 4)/ predicted, percent
^{89}Sr	0.036	0.019
^{99}Mo	0.029	0.38
^{131}I	1.6	0.003
^{133}I	2.1	0.031
^{135}I	0.94	0.097
^{137}Cs	0.088	0.0002

Predictions for the other measured fission products are not available; on the other hand, the concentration of 78-hour ^{132}Te was predicted to be 2.2 μCi/ml, but none ($<1 \times 10^{-6}$ μCi/ml) was detected.

That some fission products were found and others, of similar fission yields, were not (see Table 2.1), may be attributable to the volatility of the detected ones or their radioactive precursors. Relatively little removal by the main coolant demineralizer or relatively high solubility in coolant water may also be responsible for the presence of these radionuclides in the water. Thus, radioiodine, the radiokrypton precursors of ^{89}Sr, ^{90}Sr, and ^{91}Sr, and the radioxenon precursors of ^{137}Cs and ^{140}Ba may have passed through the fuel cladding at higher rates than other radionuclides; or the radioisotopes of the rare earths, ruthenium, etc., may have been removed from the water very effectively by the demineralizer or by crud formation in the coolant system. In the case of ^{95}Zr, its daughter ^{95}Nb, and ^{99}Mo, neutron activation of Zircaloy and steel, respectively, may be responsible for the presence of these radionuclides in coolant water.

2.3.4 *Activation products in main coolant water.* The turnover rates of the longer-lived activation products, considering only their removal by demineralizing and decay (not by crud formation or coolant water discharge, for example), ranged from 0.001 to 1.6 μCi/sec according to calculations in Appendix B.5. These are in the same range as fission product turnover rates (see Appendix B.4).

The highest concentrations of measured activation products were between 0.1 and 0.5 percent of predicted concentrations for most radionuclides. The maximum concentration relative to predicted values was 25 percent for ^{24}Na. These predictions by Yankee were based on an overall monthly corrosion rate of 10 milligrams per square decimeter and a 38-liter/min flow rate through the purification system:(1)

Radionuclide	Predicted concentration, μCi/ml	Highest measured/ predicted, percent
^{24}Na	0.15	25.
^{51}Cr	0.8	0.11
^{54}Mn	0.116	0.46
^{55}Fe	0.12	0.31
^{59}Fe	0.052	0.37
^{58}Co	0.84	0.12
^{60}Co	0.077	0.17
^{64}Cu	0.019	7.4

The measured concentrations presumably include suspended (insoluble) radioactive material. The influence of the pH value in main coolant water on these radionuclide concentrations is suggested by comparing measured totals with Yankee data on average radionuclide concentrations in crud multiplied by crud concentrations of 0.4 ppm:(9,10)

	October 4, 1968	
Radionuclide	in crud, μCi/ml	crud/total
^{51}Cr	1.1×10^{-4}	0.13
^{54}Mn	5.3×10^{-6}	0.01
^{59}Fe	1.3×10^{-5}	0.07
^{58}Co	2.7×10^{-5}	0.08
^{60}Co	1.2×10^{-5}	0.13
^{110m}Ag	7.8×10^{-6}	0.41
	July 10, 1969	
Radionuclide	in crud, μCi/ml	crud/total
^{51}Cr	3.2×10^{-4}	1.5
^{54}Mn	2.8×10^{-4}	1.2
^{59}Fe	1.8×10^{-4}	2.0
^{58}Co	1.6×10^{-3}	1.6
^{60}Co	5.5×10^{-4}	4.2
^{110m}Ag	1.2×10^{-6}	<1.2

The listed radionuclides are relatively soluble at the low pH value of the Oct. 4 sample, but insoluble at the higher pH in the July 10 sample. The ratio is only qualitative, as indicated by ratios that exceed unity,

because crud radionuclide concentrations are average monthly values.

Analyses for the three activation products that emit only low-energy beta particles -- ^{14}C, ^{35}S, and ^{63}Ni -- showed that all three are present at relatively low concentrations (see Table 2.1). All may be formed by thermal-neutron activation of the elements. In addition, ^{14}C is formed by the ^{17}O (n, α) reaction (in water, for example) and the ^{14}N (n,p) reaction (in ammonia and nitrogen gas, for example).

2.3.5 *Radionuclides in secondary coolant water.* Samples of steam generator blowdown water and condenser water contained ^{3}H at concentrations between 0.02 and 0.002 μCi/ml, and several other fission and activation products at much lower concentrations (see Table 2.2). Except in the sample of Oct. 4, 1968, only a few radionuclides other than ^{3}H could be detected. The blowdown and condensate water samples of June 10, 1970 contained the same concentrations of ^{3}H and ^{131}I; other radionuclides could not be measured with sufficient sensitivity for comparison. Some of the radionuclides may have been in insoluble form, as suggested by the observation that all of the ^{54}Mn in the June 10, 1970 sample was removed from the water by filtering.

The leakage rate of water from the main into the secondary system was estimated by the equation:

$$\frac{\text{main-to-secondary leakage rate}}{\text{secondary turnover rate}} = \frac{^{3}\text{H concentration, secondary}}{^{3}\text{H concentration, main}}$$

Based on the tritium concentrations in Tables 2.1 and 2.2 and a makeup volume for secondary coolant water of 40,000 liters/day, leakage rates were as follows:

date of sample	^{3}H ratio, secondary / main	Calculated leakage rate, liters/day
Oct. 4, 1968	5×10^{-4}	20
July 10, 1969	1.6×10^{-2}	640
June 10, 1970	1.0×10^{-2}	400

Leakage rates between 370 and 580 liters reported by Yankee (11) for June, 1970, are consistent with the value for June 10, 1970. The calculation assumes equilibrium and is applicable only to constant or slowly changing leakage rates. That several other radionuclides have lower ratios than ^{3}H can be

Table 2.2

Radionuclide Concentration in Secondary System Water, μCi/ml

Radionuclide	Continuous steam generator blowdown		Condenser water	
	Oct. 4, 1968	June 10, 1970	July 10, 1969	June 10, 1970
^{3}H	2.5×10^{-3}	1.8×10^{-2}	6.3×10^{-3}	1.9×10^{-2}
^{14}C	$< 2 \times 10^{-7}$	$< 1 \times 10^{-7}$	3×10^{-7}	$< 2 \times 10^{-7}$
^{32}P	$< 2 \times 10^{-7}$	$< 1 \times 10^{-8}$	$< 2 \times 10^{-7}$	$< 1 \times 10^{-8}$
^{51}Cr	3×10^{-6}	$< 1 \times 10^{-7}$	NA	$< 5 \times 10^{-7}$
^{54}Mn	1×10^{-6}	1×10^{-7}	1.2×10^{-6}	$< 1 \times 10^{-7}$
^{55}Fe	5×10^{-8}	$< 5 \times 10^{-8}$	$< 5 \times 10^{-8}$	$< 5 \times 10^{-8}$
^{59}Fe	1×10^{-6}	$< 1 \times 10^{-7}$	$< 5 \times 10^{-8}$	$< 1 \times 10^{-7}$
^{58}Co	2×10^{-6}	$< 1 \times 10^{-7}$	8×10^{-7}	$< 1 \times 10^{-7}$
^{60}Co	5×10^{-7}	$< 5 \times 10^{-8}$	1×10^{-7}	$< 1 \times 10^{-7}$
^{63}Ni	2×10^{-7}	$< 1 \times 10^{-8}$	$< 1 \times 10^{-7}$	4×10^{-8}
^{89}Sr	$< 1 \times 10^{-8}$	$< 5 \times 10^{-8}$	$< 2 \times 10^{-8}$	$< 2 \times 10^{-8}$
^{90}Sr	5×10^{-9}	$< 2 \times 10^{-8}$	1×10^{-8}	$< 1 \times 10^{-8}$
^{95}Zr	8×10^{-7}	$< 1 \times 10^{-7}$	NA	$< 1 \times 10^{-7}$
^{95}Nb	6×10^{-7}	$< 1 \times 10^{-7}$	NA	$< 1 \times 10^{-7}$
^{110m}Ag	2×10^{-7}	$< 2 \times 10^{-8}$	$< 2 \times 10^{-7}$	$< 1 \times 10^{-7}$
^{124}Sb	NA	1×10^{-7}	NA	NA
^{131}I	$< 4 \times 10^{-8}$	5×10^{-7}	$< 5 \times 10^{-7}$	5×10^{-7}
^{134}Cs	$< 2 \times 10^{-8}$	$< 6 \times 10^{-8}$	$< 1 \times 10^{-7}$	$< 6 \times 10^{-8}$
^{137}Cs	1×10^{-8}	$< 1 \times 10^{-8}$	2×10^{-8}	$< 1 \times 10^{-8}$
gross beta	$< 1 \times 10^{-6}$	3×10^{-7}	2.2×10^{-7}	$< 1 \times 10^{-7}$

Notes:
1. NA - not analyzed.
2. Radionuclide concentrations are at time of sampling, gross beta values, 1 week later.
3. < values are 3 σ counting errors.

attributed to their removal by deposition either in the main or the secondary system; higher ratios–found only in the Oct. 4, 1968 sample–may indicate residues from earlier leakage.

2.3.6 *Radionuclides in other liquids.* The ^{3}H concentration in one of the two waste holdup tanks on Oct. 4, 1968 (see Table 2.3) was approximately an order of magnitude lower than in main coolant water; the other radionuclides were all relatively long-lived, and mostly at higher concentrations than in the main coolant at the same date. The specific sources of the waste are not known. The low gross beta activity in Table 2.3 indicates how misleading this measurement can be in the presence of radionuclides that emit few or no beta particles.

Safety injection water contained, at relatively low concentrations, some of the long-lived radionuclides detected in the main coolant, as shown in Table 2.4. These radionuclides presumably entered the water while it was in the shield tank cavity during refueling.

None of the radionuclides listed in Table 2.4 was found in component cooling water. The minimum detectable concentration was 1×10^{-6} μCi/ml for ^{3}H and between 1×10^{-7} and 1×10^{-8} μCi/ml for all others.

Table 2.3
Radionuclide Concentration in Waste Holdup Tank on Oct. 4, 1968

Radionuclide	Concentration, μ Ci/ml
^{3}H	3.8×10^{-1}
^{14}C	1.2×10^{-4}
^{32}P	$< 2 \times 10^{-7}$
^{51}Cr	$< 5 \times 10^{-7}$
^{54}Mn	1.4×10^{-3}
^{55}Fe	8.2×10^{-4}
^{59}Fe	1.0×10^{-6}
^{57}Co	6×10^{-6}
^{58}Co	2.9×10^{-4}
^{60}Co	6.4×10^{-4}
^{63}Ni	2.3×10^{-4}
^{89}Sr	3×10^{-7}
^{90}Sr	7×10^{-8}
^{110m}Ag	3.1×10^{-4}
^{131}I	$< 3 \times 10^{-8}$
^{134}Cs	2.0×10^{-5}
^{137}Cs	5.4×10^{-5}
^{182}Ta	1.6×10^{-5}
gross beta	5.0×10^{-4}

Notes: 1. Radionuclide concentrations are at time of sampling, gross beta activity is on Oct. 9, 1968.
2. $<$ values are 3 σ counting error.

Table 2.4
Radionuclide Concentration in Safety Injection Water, June 10, 1970

Radionuclide	Concentration, μCi/ml
^{3}H	2.2×10^{-2}
^{54}Mn	9.5×10^{-5}
^{55}Fe	5×10^{-6}
^{59}Fe	1×10^{-6}
^{57}Co	6×10^{-7}
^{58}Co	1.5×10^{-5}
^{60}Co	4.5×10^{-5}
^{110m}Ag	8×10^{-6}
^{124}Sb	2×10^{-6}
^{137}Cs	2×10^{-7}

Note: The following radionuclides were not detected ($< 1 \times 10^{-7}$ μCi/ml): ^{14}C, ^{32}P, ^{51}Cr, ^{89}Sr, ^{90}Sr, ^{131}I, and ^{134}Cs

2.4 References

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3. Radionuclides Released from Stack

3.1 Gaseous Waste System and Samples

3.1.1 *Gaseous waste system.* Gaseous radioactive wastes generated at Yankee are discharged to the air as depicted in Figure 3.1, which is based on descriptions by several authors. [1-3] Yankee wastes are classified as hydrogen-bearing and air-bearing. Hydrogen-bearing waste originates in the main coolant system; with one exception, it is collected in the gas surge drum at a compression of several atmospheres and held for radioactive decay. Three decay tanks are available to store additional gas under pressure. Transfer from main coolant to storage is either direct, at the low-pressure surge tank, or through venting the hydrogen-bearing liquid waste from collection tanks and the waste-evaporator condenser. The storage tanks are blanketed with nitrogen to prevent mixing hydrogen with air.

Gas from the surge drum is released at a nominal rate of 0.425 standard m^3/min through a deep-bed glass-fiber filter to the base of the 1.1-m dia., 46-m high, cylindrical primary vent stack. In the stack, the gas is diluted with ventilating air from the Primary Auxiliary Building, which is discharged at the nominal rate of 425 m^3/min. Surge drum gas is usually released once each year, [3] although releases were reported in February, March and April 1969, [4] and a special release was made for the measurements during the field trip on June 3, 1969.

Air-bearing waste consists of gases from the air ejector at the main condenser in the secondary coolant system, the gland seal condenser in the secondary system, tanks that contain secondary-system liquid wastes, and the evaporator when air-bearing liquid waste (from the gravity drain tank) is being processed. These gases are released directly into the primary vent stack for dilution by the ventilating air from the Primary Auxiliary Building. Vapor container air is also discharged to the stack whenever the containment building is opened; this occurred 15 times in a 4-year period (see footnote to Appendix B.2). Air from the Primary Auxiliary Building is discharged continuously through the stack. Air from the Turbine Building is discharged to outside air without passing through the stack.

Also released directly to the stack are two liters per day of hydrogen-bearing gas that pass from main-coolant sampling ports into the laboratory hoods when aliquots of main coolant water are collected for analysis. This usually occurs once daily. Yankee routinely reports values of ^{41}Ar, ^{133}Xe, and ^{135}Xe concentrations in the main coolant.[4]

Gases from burning solid waste in the incinerator are discharged through a wet-gas scrubber and deep-bed glass-fiber filter through a 20-cm dia., 2.4-m high, stack on top of the Primary Auxiliary Building. This effluent is reported to contain negligible radioactivity.[1]

The major components of the radioactivity released from the surge drum would be expected to be the fission-produced long-lived radioisotopes of krypton and xenon (see Appendix B.3 of the Dresden study) [5] and tritium. Any other gaseous or relatively volatile fission and activation products in this effluent would also be long-lived because of the long retention period.

The radionuclide content of the continuously discharged stack gases depends on the leakage rate from the main-coolant system and the extent of specific releases such as main coolant sampling and vapor container venting. Radioactive gases from the air ejectors and main coolant sampling would contain relatively short-lived isotopes. Some of these gas streams are unfiltered and may carry radioactive particles. Off-gas emissions from the air ejector at the condenser are monitored continuously by an anthracene detector; all stack discharges are monitored by 4 G-M tubes.

3.1.2 *Radionuclide release.* Radioactive gas discharges by Yankee are limited by the AEC as follows: "As determined at the point of discharge from the primary vent stack and averaged over a

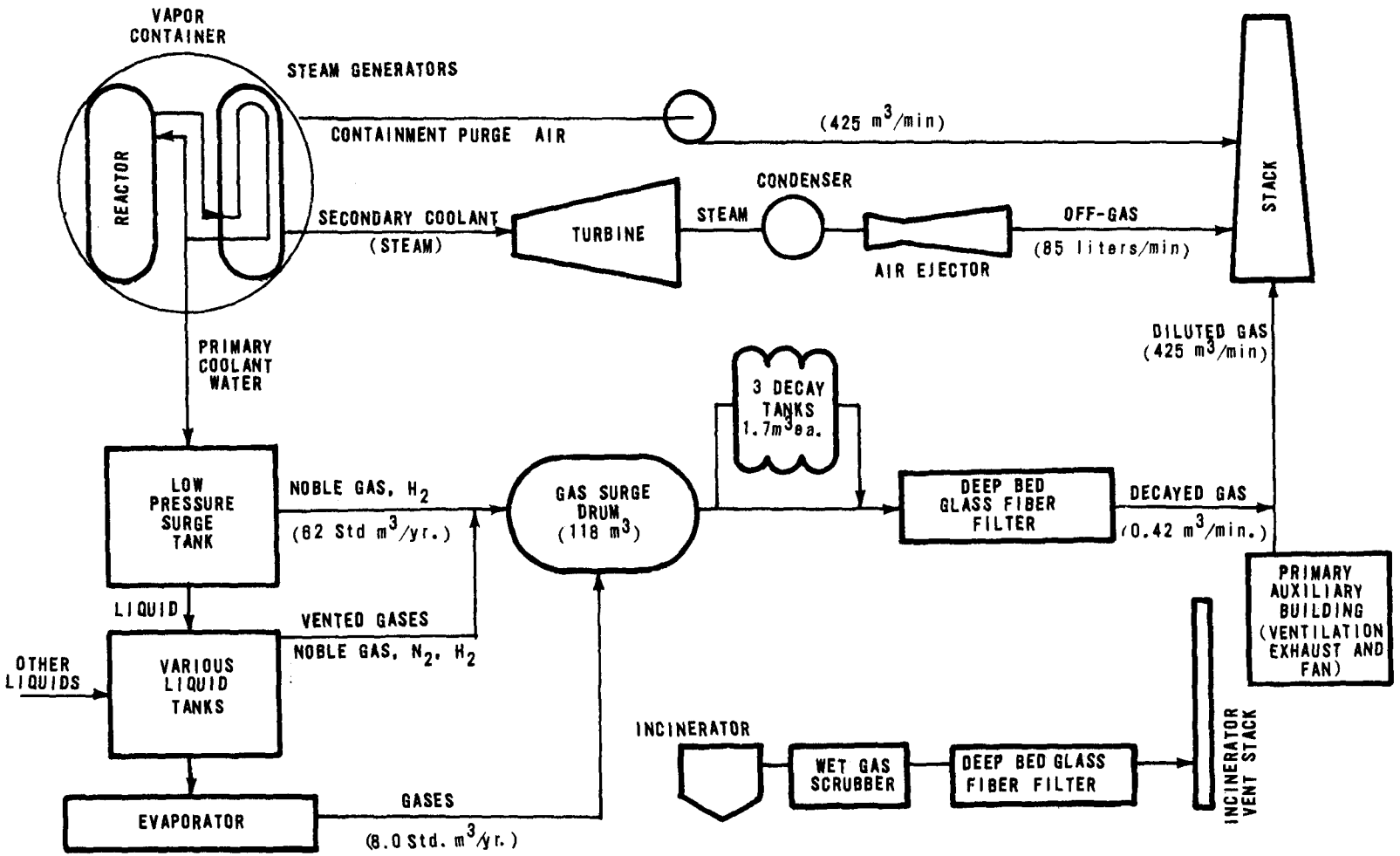

Figure 3.1. Sources of Airborne Effluent.

period not exceeding one year, the concentration of radioactive gaseous wastes discharged shall not be in excess of 1,000 times the limits specified in Appendix B, Table II, 10 CFR 20." (6) The values in Table II derive from Section 20.105 of 10 CFR 20, which limits the added radiation dose to individuals in unrestricted areas to 0.5 rem/year. The factor of 1,000 is allowed in consideration of atmospheric diffusion from the stack (1) to the boundary (at the 300-m perimeter) of the Yankee exclusion area. Limits for discharging individual radionuclides to air by Yankee are given in Section 3.3.8.

Yankee has reported the following annual releases: (1,4,7)

Radioactivity	1970	1969	1962 to 1968
$\beta\gamma$ in gas, Ci	17.2	4.13	0.7-22
^{3}H, Ci	9.0	9.19	8-16*
$\beta\gamma$ in particles, μCi	1.82	2.51	7.89+

*1965-1968; +1968

The highest annual release of gross beta-gamma activity represents 0.5 percent of the release limit of 4,500 Ci/yr for ^{87}Kr and ^{88}Kr, the most hazardous noble gas fission products. For ^{3}H, the highest annual release represents 0.04 percent of the 45,000 Ci/yr limit for tritiated water vapor (HTO). The particulate radioactivity is an extremely small component of the total radioactive discharge.

3.1.3 *Sample collection.* Samples of gas surge drum contents were obtained on October 4, 1968, April 1, 1969, and June 3, 1969. The first sample was withdrawn in triplicate at the sampling port of the surge drum into evacuated 9-cc glass serum bottles, sealed with rubber stoppers held by crimped aluminum holders. On the other two occasions, duplicate samples were collected in evacuated 0.85-liter gas cylinders.

A 144-m^3 volume of gas was discharged from the surge drum through the primary vent stack on June 3, 1969, from 1500 to 2145 hours. During this period, a sampling system was attached to a single-nozzle probe, centered in the stack. The system components were in the following sequence:

(1) membrane filter (Millipore Filter* type AA, 5-cm dia., in Unico holder) for sampling particles;
(2) carbon bed (26.6 g Columbia 6GC activated charcoal, type 10/20, 3.2-cm dia.) for sampling gaseous iodine;
(3) pressure-vacuum gauge;
(4) calibrated flowmeter (F and P Flowrator);
(5) vacuum pump (Gast Model 0406).

This sampling procedure was repeated on June 4, 1969, from 0910 to 1530, to measure radionuclide concentrations when no gas surge drum contents were being discharged. The sample volumes that passed through the filter and carbon bed were computed to be 2.0 m^3 on June 3 and 3.6 m^3 on June 4. At the beginning of each of the 2 sampling periods, an evacuated 8.2-liter gas bottle was filled with gas at the stack probe to measure the concentration of radioactive gases.

Beginning June 5, 1970, at 0930 hours, five consecutive 24-hr filter and carbon bed samples were obtained in the primary vent stack to measure the variability of particulate and radioiodine emissions. The sampling system was the same as that described above, except that the carbon bed was 5.0 cm in diameter. Sampling flow rates varied between 12 and 20 liters/min; the typical sample volume was 27 m^3.

The following samples were collected on June 10, 1970:

(1) air ejector off-gas from the main condenser in the secondary coolant system before dilution in the stack, 8.2 liters.
(2) vapor container atmosphere, 8.2 liters. Ambient temperature was 31°C; the relative humidity was 43 percent of saturation. (3)
(3) water from the dehumidifer in the vapor container, 4 liters. Yankee operates the dehumidifier to collect water samples for ^{3}H analysis, and reports ^{3}H concentrations in discharged vapor container air on the basis of these analyses. (3)
(4) main-coolant gas, during depressurizing for routine liquid sampling, 17 cc in two 9-cc serum bottles.

* Mention of commercial products does not constitute endorsement by the Environmental Protection Agency.

(5) 5-cm-dia. glass fiber filter used by Yankee to sample particulate emissions in incinerator stack, during operation from 2030 to 2130 hours, June 9, 1970. Sampling flow rate was 10 liters/min.

Other samples were as follows:

(6) vapor container atmosphere, 8.2 liters, on Nov. 19. 1970. Ambient temperature was 14°C; the relative humidity was 47 percent of saturation. (3) The vapor container was open to outside air during refueling.

(7) water from the dehumidifier in the vapor container, 100 ml, on Nov. 19, 1970.

(8) water from the dehumififier in the vapor container, 1 liter, on Nov. 30, 1970. Ambient temperature was 27°C; the relative humidity was 53 percent of saturation. (3) The vapor container had been sealed for 10 days prior to sampling.

(9) main coolant gas, two 9-cc serum bottles, on Feb. 9, 1971.

(10) air ejector off-gas from the main condenser in the secondary coolant system before dilution in the stack, 8.2 liters on February 9, 1971.

3.2 Analysis

3.2.1 *Gamma-ray spectrometry.* Radionuclides that emit gamma rays were routinely analyzed with a 10-cm x 10-cm cylindrical NaI(Tl) detector coupled to a 400-channel spectrometer (see Figures 3.2 and 3.3). Identification was confirmed by spectral analysis with a high-resolution 10.4-cm^2 x 1.1-cm Ge(Li) detector and a 1600-channel spectrometer or with a low-background dual 10-cm x 10-cm NaI(Tl) detector system in various coincidence/anticoincidence modes. Iron-55 was measured with a xenon-filled x-ray proportional counter and a 200-channel spectrometer.

The samples of main coolant gas and air ejector gas obtained on February 9, 1971, were first analyzed within 5 hours of collection to detect relatively short-lived ^{41}Ar, ^{87}Kr, and ^{88}Kr.* All other samples were counted one day after collection, hence only radionuclides with longer half lives (>6 hours) could be detected. As an exception, ^{41}Ar and ^{85m}Kr were detected in the sample of main coolant gas obtained on June 10, 1970, because of their relatively high initial concentrations.

Primary coolant gas and waste surge drum samples were analyzed in 9-cc glass serum bottles. Aliquots of 8.2-liter gas samples were counted in 209-cc

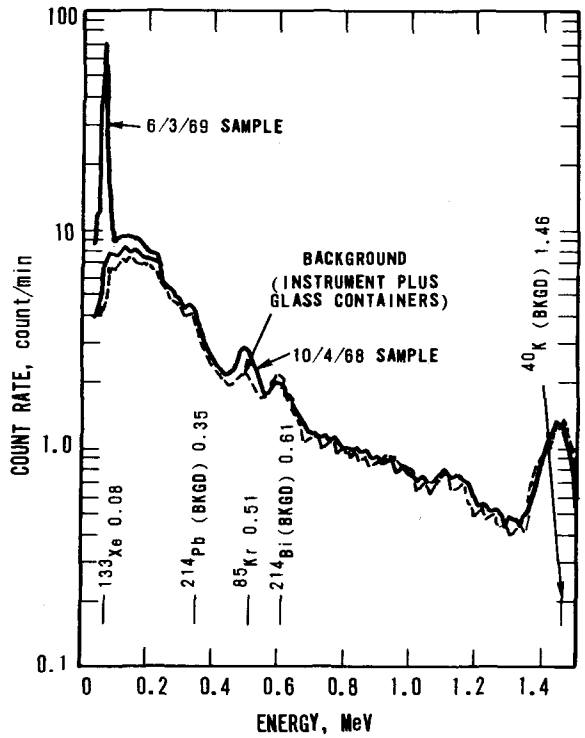

Figure 3.2. Gamma-ray Spectra of Gas Surge Drum Samples.

detector: *10 × 10-cm NaI(Tl)*

sample : *27 cc of gas in glass serum bottles, collected 1200 EDT October 4, 1968, and 1125 EDT, June 3, 1969.*

count : *Oct. 4 sample - 1600 Oct. 10 to 0840 EDT, Oct. 11, 1968. June 3 sample - 1700 June 6 to 0940 EDT, June 7, 1969.*

*We thank Messrs. G. J. Karches and C. Nelson, NERHL, PHS, for making possible the prompt analysis of these samples.

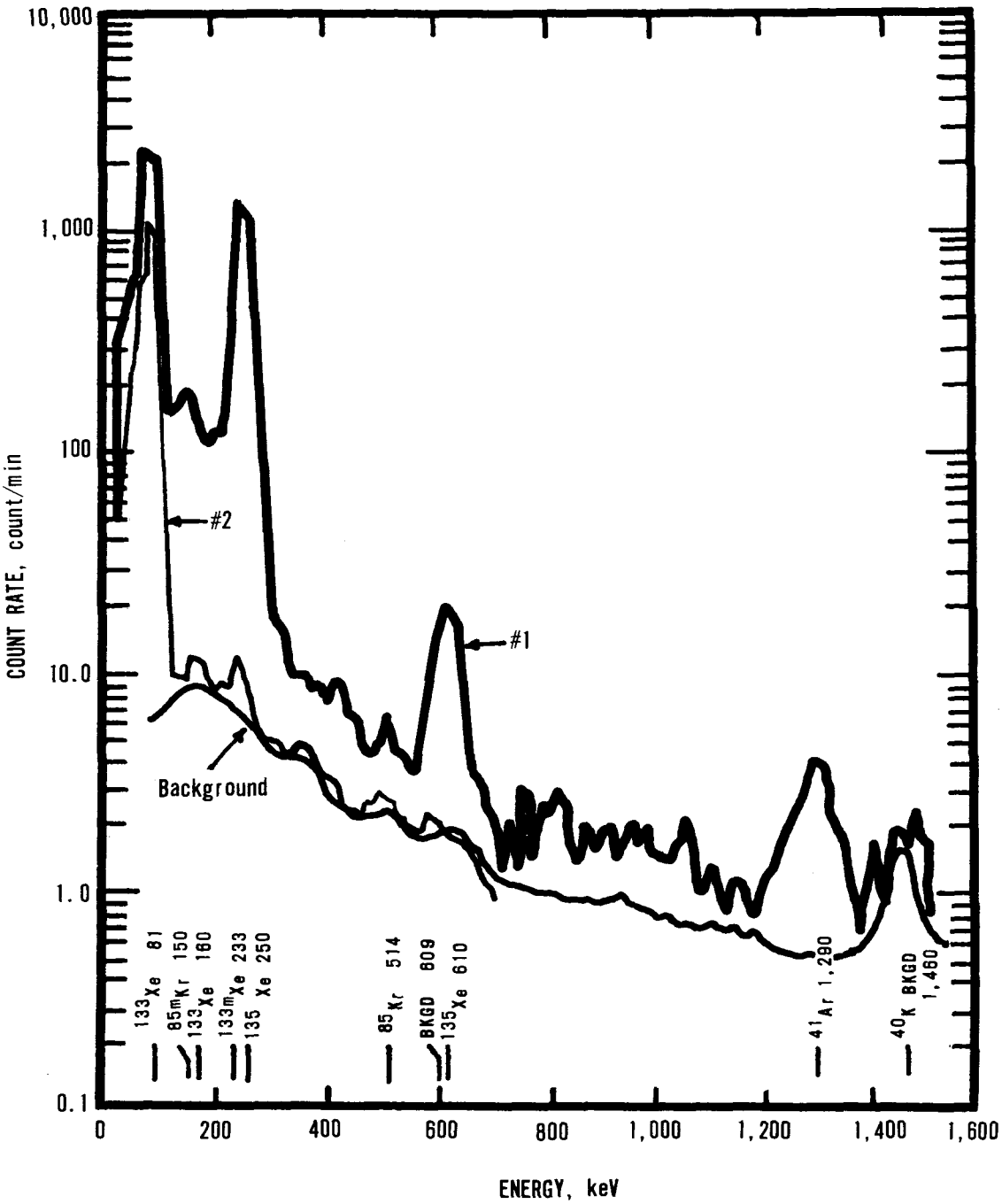

Figure 3.3. Gamma-ray Spectra of Gas Released in Sampling Main Coolant.

Detector: *NaI(Tl), 10 × 10 cm*

Sample : *9 cc bottle of gas collected 1030 hours EDT, June 10, 1970*

Counts : *#1 – 1515 hours EDT, June 11, 1970 (10 min)*
#2 – 1044 hours EDT, June 16, 1970 (50 min)

volumetric flasks. The serum bottles and flasks were sealed with rubber stoppers held by crimped aluminum seals. Activated charcoal and 440-ml aliquots of vapor container water were counted in plastic containers.

Detection efficiencies for the radionuclides, containers, sample volumes, and media of interest were determined with standardized radioactivity solutions or ^{85}Kr gas. Because glass contains ^{40}K and charcoal contains ^{40}K and ^{226}Ra, distinct backgrounds were measured for these materials.

Counting intervals and techniques were selected to provide, when possible, counting precision of ± 10 percent or better at the 95 percent confidence level. The usual counting duration for low-level activity was 1000 min. Samples were re-analyzed periodically to confirm identification of radionuclides by determining half lives, and to look for longer-lived radionuclides.

3.2.2 *Radiochemical analysis.* Strontium was chemically separated from one half of each particulate filter and from aliquots of the dehumidifier condensates. The radiostrontium content was measured by counting for 100-min intervals with low background G-M beta particle detectors. Strontium-90 was distinguished from ^{89}Sr by separating and counting the ^{90}Y daughter.

Krypton-85 at relatively low concentrations was determined by liquid scintillation counting. Approximately 3-cc aliquots of the gas surge drum samples were mixed with degassed PPO and bis-MSB liquid scintillator solution and measured for 50-min periods in a liquid scintillation counter with spectrometer. *(8) Aliquots of all samples obtained in June of 1969 and thereafter were analyzed by counting ^{85}Kr with 1-mm-dia. plastic scintillator spheres occupying 15 cc of the 25-cc vial volume. Samples of ^{85}Kr were either transferred directly to the counting vial or concentrated from 0.5 - to 1-liter aliquots by passing gas through charcoal at -78°C and then heating the charcoal to transfer ^{85}Kr to the counting vial.

During liquid scintillation analysis of the first gas surge drum sample for ^{85}Kr, an unexpected gaseous radionuclide at relatively high concentration was detected. This gas was identified as ^{14}C in the form of CO or an organic compound, but not CO_2, by observing its disintegration mode, beta-particle spectrum, and chemical behavior. Gas samples collected later were analyzed in duplicate for ^{14}C by passing aliquots mixed with CO carrier gas through an alumina-platinum (0.5 percent) catalyst heated to 550°C to convert the sample carbon to CO_2, and into a bubbler containing $BaCO_3$. The ^{14}C activity in the precipitate was counted for 10-to 100-min intervals in low-background beta counters. Identification was confirmed by aluminum absorber curves. Aliquot sizes ranged from 10 cc to 1 liter, depending on the ^{14}C concentration.

Tritium in HT, HTO vapor, or other gaseous form, was separated in the samples collected in June, 1969, and thereafter by passing aliquots mixed with H_2 carrier gas through a copper oxide bed heated to 550°C to oxidize hydrogen, and collecting the water in a trap at -78°C. The ^{3}H activity in the condensate was measured by liquid scintillation counting for 200-min periods. Aliquot sizes were the same as for ^{14}C analyses.

Tritiated water vapor concentrations in the gas samples were determined by liquid scintillation counting. To collect ^{3}H in this form, distilled water equivalent to 5 percent of the gas sample volume was injected into containers previously used for gamma-ray analysis and intermittently swirled for 2 to 3 days. The water was then removed and distilled, and aliquots were mixed with liquid scintillator for analysis.

3.3 Results and Discussion

3.3.1 *Gases released by sampling main coolant.* The radionuclides found in main-coolant gas (see Table 3.1) include all high yield fission-produced krypton and xenon isotopes whose half lives were longer than 1 hour. In addition, ^{3}H and the activation products ^{14}C and ^{41}Ar were detected. The ^{41}Ar was probably formed from argon in air within the system; production of ^{3}H and ^{14}C is discussed in Sections 2.3.2 and 2.3.4, respectively. Measurements by Yankee staff of the June 10, 1970, sample were as follows:

^{41}Ar	1.18	μCi/cc
^{133}Xe	4.05×10^{-3}	
^{135}Xe	4.74×10^{-3}	

The values for ^{41}Ar and ^{133}Xe in Table 3.1 are in agreement, while the concentration of ^{135}Xe is more than two-fold higher.

*We thank Dr. A. A. Moghissi, Mr. R. Shuping and staff at the Southeastern Radiological Health Laboratory (SERHL, EPA) for analyzing these samples.

Table 3.1

Radioactive Gases Released to Stack during Depressurizing Main Coolant for Sampling

Radionuclide	Concentration, μCi/cc		Release per Sample, μCi*		Estimated Average Annual Release, + Ci
	June 10, 1970	Feb. 9, 1971	June 10, 1970	Feb. 9, 1971	
12.3 -yr ^{3}H	$1.9 \pm 0.1 \times 10^{-4}$**	$1.4 \pm 0.1 \times 10^{-3}$	3.6×10^{-1}	2.6	5×10^{-4}
5730 -yr ^{14}C	$2.6 \pm 0.1 \times 10^{-3}$	$3.7 \pm 0.3 \times 10^{-3}$	4.9	7.0	2×10^{-3}
1.83-hr ^{41}Ar	1.0 ± 0.2	$3.8 \pm 0.1 \times 10^{-1}$	1.9×10^{3}	7.1×10^{2}	4×10^{-1}
4.4 -hr ^{85m}Kr	$5.4 \pm 0.6 \times 10^{-3}$	$6.7 \pm 0.1 \times 10^{-2}$	1.0×10^{1}	1.3×10^{2}	2×10^{-2}
10.7 -yr ^{85}Kr	$9 \pm 4 \times 10^{-5}$	$1.8 \pm 0.2 \times 10^{-3}$	1.7×10^{-1}	3.4	6×10^{-4}
76 -m ^{87}Kr	NA++	$7.2 \pm 0.7 \times 10^{-2}$	—	1.4×10^{2}	2×10^{-2}
2.8 -hr ^{88}Kr	NA	$8.8 \pm 0.3 \times 10^{-2}$	—	1.7×10^{2}	3×10^{-2}
2.3 -d ^{133m}Xe	$1.5 \pm 0.3 \times 10^{-4}$	$5.4 \pm 0.3 \times 10^{-3}$	3.0×10^{-1}	1.0×10^{1}	2×10^{-3}
5.29-d ^{133}Xe	$4.8 \pm 0.1 \times 10^{-3}$	$4.2 \pm 0.1 \times 10^{-1}$	9.2	7.9×10^{2}	1×10^{-1}
9.1 -hr ^{135}Xe	$1.2 \pm 0.1 \times 10^{-2}$	$2.1 \pm 0.1 \times 10^{-1}$	2.2×10^{1}	4.1×10^{2}	7×10^{-2}

* based on the release of 1900 cc of gas during sampling operation

\+ Average of two release values (approx. 10 μCi each assumed for ^{87}Kr and ^{88}Kr on June 10, 1970), multiplied by 320 days/yr estimated operating period.

** $\pm$ values indicate analytical error expressed at 2 σ .

++ NA - not analyzed

The concentrations and relative amounts of the radionuclides differed considerably in the two samples. In the earlier one, the proportions approach that of a diffusion mixture, while the later sample had higher concentrations of the longer-lived fission products. The earlier sample was collected midway in the eighth core cycle; the later sample, near the beginning of the ninth cycle.

The amounts of radionuclides released per sampling were calculated in Table 3.1 from the measured concentrations and an average gas release during daily sampling of 1,900 cc from the main coolant system through the hood ventilation system to the stack. The annual discharge of radionuclides from this source was then computed by multiplying the averages of the two sets of values in Table 3.1 by an estimated 320 operating days per year. The major constituent was ^{41}Ar at 0.4 Ci/yr; the sum of all other listed radionuclides was 0.2 Ci/yr (see Table 3.1).

Shorter-lived fission-produced gases would also be expected in the mixture. On the assumption that the gas is a diffusion mixture and had decayed for five minutes, the amounts of shorter-lived radionuclides would be as follows (see Appendix B.3 of the Dresden study) (5) relative to a value of unity for ^{135}Xe:

9.1-hr	^{135}Xe	1.0
3.2-min	^{89}Kr	3.2
15.6-min	^{135m}Xe	1.4
3.8-min	^{137}Xe	4.4
17. -min	^{138}Xe	5.0

The sum of the four short-lived radionuclides is thus estimated to be 14 times the amount of ^{135}Xe, or 14 x 0.07 = 1.0 Ci/yr. Short-lived 10-min ^{13}N probably is also in this effluent gas.

3.3.2 *Gaseous effluent from secondary coolant.* Tritium, ^{14}C, ^{133}Xe, and ^{135}Xe were observed in off-gas from the air ejector at the secondary-coolant main condenser (see Table 3.2). The concentrations of these radionuclides were so low in the sample of Feb. 9, 1971, that the shorter-lived xenon and krypton isotopes, if present in the proportions indicated in Table 3.1, would not have been detected after the 5-hour interval between sampling and counting. Only a small fraction of the tritium was in the form of water vapor; the largest fraction probably was associated with molecular hydrogen or an organic compound.

The concentrations of these radionuclides on June 10, 1970, were three to four orders of magnitude lower than in main-coolant gas; on Feb. 9, 1971, they were as much as six orders of magnitude lower (see Table 3.1). The lower ratios in the later sample suggest a much lower main-to-secondary leakage rate. Differences among the ratios of concentrations in the secondary system to those in main-coolant water may be caused also by differences in gas turnover rates in the secondary system, the solubility of the various gases in water, and the occurrence of chemical reactions.

Radionuclide release rates at the times of sampling were computed from the concentrations and gas flow

Table 3.2

Radioactivity Contents of Off-gas from Air Ejector at Main Condenser in Secondary Coolant System

Radionuclide	Concentration before dilution in stack, μCi/cc		Release rate, * μCi/sec		Estimated Average Annual Release, + Ci
	June 10, 1970	Feb. 9, 1971	June 10, 1970	Feb. 9, 1971	
^{3}H (total)	$2.2 \pm 0.1 \times 10^{-7}$**	$< 3 \times 10^{-8}$	3.1×10^{-4}	$< 4 \times 10^{-5}$	4×10^{-3}
^{3}H (water vapor)	$8 \pm 6 \times 10^{-9}$	$< 2 \times 10^{-8}$	1.1×10^{-5}	$< 2 \times 10^{-5}$	2×10^{-4}
^{14}C	$5.9 \pm 0.8 \times 10^{-7}$	$1.1 \pm 0.7 \times 10^{-8}$	8.4×10^{-4}	1.3×10^{-5}	1×10^{-2}
^{41}Ar	NA++	$< 4 \times 10^{-6}$	---	$< 5 \times 10^{-3}$	$< 1 \times 10^{-1}$
^{85}Kr	$< 1 \times 10^{-8}$	$< 1 \times 10^{-8}$	$< 1.5 \times 10^{-5}$	$< 1.2 \times 10^{-5}$	$< 4 \times 10^{-4}$
^{133m}Xe	$< 1 \times 10^{-6}$	$< 2 \times 10^{-6}$	$< 2 \times 10^{-3}$	$< 2 \times 10^{-3}$	$< 6 \times 10^{-2}$
^{133}Xe	$1.7 \pm 0.4 \times 10^{-6}$	$6.0 \pm 0.8 \times 10^{-7}$	2.4×10^{-3}	7.1×10^{-4}	4×10^{-2}
^{135}Xe	$7 \pm 1 \times 10^{-6}$	$< 4 \times 10^{-7}$	1×10^{-2}	$< 5 \times 10^{-4}$	1×10^{-1}

* Based on flow rates of 1420 and 1180 cc/sec on June 10, 1970 and Feb. 9, 1971, respectively.

+ Average of 2 release values, multiplied by 2.8×10^{7} sec/yr estimated operating period.

** $\pm$ values indicate analytical error expressed at 2 σ.

++ NA - not analyzed

rates (see Table 3.2); the annual radionuclide release was then calculated by averaging the two release rates and multiplying by a release period of 320 days per year. The major constituent among the measured radionuclides was ^{135}Xe, at 0.1 Ci/yr (see Table 3.2), and the sum of all other measured radionuclides was 0.05 Ci/yr. In addition, some shorter-lived radioactive gases may have been released. According to their ratios relative to ^{135}Xe (see Section 3.3.1), the total for these krypton and xenon radionuclides was approximately 2 Ci/yr, but this estimate is highly uncertain because relative concentrations of radioactive gases in the secondary system appear not to be the same as in the main coolant. Direct measurement of the short-lived radioactive gases by the station operator would, therefore, be of interest, especially because these short-lived gases may be the largest sources of external radiation exposure from effluent gases in the environment (see Section 3.3.8).

3.3.3 *Gas surge drum contents.* Relatively long-lived ^{3}H, ^{14}C, ^{85}Kr and ^{133}Xe were found in the stored gas (see Table 3.3). The gamma-ray spectrum of the sample showed no other radionuclides, hence the concentration of ^{131}I was below 1×10^{-7} μCi/cc. The amount of each radionuclide in the surge drum is the product of the listed concentrations, the pressure in the drum (a few atmospheres), and the drum volume (118 m^3).

Concentrations of ^{85}Kr decreased slightly from the first to the third sample, but ^{133}Xe concentrations increased. In June 1969, near the end of the fuel cycle, the primary coolant contained ammonia, which required relatively frequent venting of the low-pressure surge tank gas to the main surge

Table 3.3

Gas Surge Drum Contents, μCi/cc at 1 atm.

Radionuclide	Oct. 4, 1968	April 1, 1969	June 3, 1969
^{3}H (total)	NA	NA	$8.8 \pm 0.2 \times 10^{-5}$
^{3}H (water vapor)	$< 2 \times 10^{-6}$	$2.0 \pm 0.1 \times 10^{-6}$	$4 \pm 1 \times 10^{-7}$
^{14}C	P	P	$7.9 \pm 0.4 \times 10^{-4}$
^{85}Kr	$1.2 \pm 0.1 \times 10^{-4}$	$8.4 \pm 1.8 \times 10^{-5}$	$7.4 \pm 0.1 \times 10^{-5}$
^{133m}Xe	$< 1 \times 10^{-6}$	NA	$< 2 \times 10^{-6}$
^{133}Xe	$< 5 \times 10^{-7}$	$3.0 \pm 0.2 \times 10^{-5}$	$5.2 \pm 0.1 \times 10^{-5}$
^{135}Xe	$< 1 \times 10^{-6}$	NA	NA

Notes:

1. $\pm$ values are 2 σ counting error; $<$ values are minimum detectable concentrations at 3 σ counting error.
2. NA - not analyzed.
   P - observed but not quantified.

Table 3.4

Radioactivity in Vapor Container

Radionuclide	In air, μCi/cc		In dehumidifier condensate, μCi/ml		
	June 10, 1970	Nov. 19, 1970	June 10, 1970	Nov. 19, 1970	Nov. 30, 1970
^{3}H (total)	$1.8\pm0.2\times10^{-6}$	$8\pm2\times10^{-7}$	$9.8\pm0.2\times10^{-1}$	$2.1\pm0.01\times10^{-3}$	$1.0\pm0.01\times10^{-1}$
^{3}H (water vapor)	$5.3\pm0.8\times10^{-7}$	$5.5\pm0.6\times10^{-7}$	NA	NA	NA
^{14}C	$1.1\pm0.1\times10^{-6}$	$4\pm2\times10^{-9}$	$1.9\pm0.1\times10^{-6}$	$1.8\pm0.6\times10^{-6}$	$2\pm1\times10^{-7}$
^{24}Na	NA	NA	$8\pm2\times10^{-6}$	NA	NA
^{51}Cr	NA	NA	NA	$1.3\pm0.1\times10^{-4}$	$<8\times10^{-7}$
^{54}Mn	NA	NA	$4\pm1\times10^{-8}$	$1.6\times0.03\times10^{-4}$	$2.3\pm0.6\times10^{-7}$
^{57}Co	NA	NA	NA	$1.6\times0.02\times10^{-6}$	$<4\times10^{-7}$
^{58}Co	NA	NA	NA	$2.5\pm0.02\times10^{-4}$	$1.7\pm0.5\times10^{-7}$
^{60}Co	NA	NA	$2\pm1\times10^{-7}$	$2.6\pm0.03\times10^{-4}$	$4.3\pm0.9\times10^{-7}$
^{59}Fe	NA	NA	NA	$5.5\pm0.6\times10^{-5}$	$<3\times10^{-7}$
^{85}Kr	$<5\times10^{-9}$	$1.5\pm0.1\times10^{-7}$	NA	NA	NA
^{89}Sr	NA	NA	NA	$<1\times10^{-7}$	NA
^{90}Sr	NA	NA	NA	$3.2\pm0.3\times10^{-6}$	NA
^{110m}Ag	NA	NA	NA	$1.4\pm0.2\times10^{-5}$	$<1\times10^{-7}$
^{124}Sb	NA	NA	NA	$1.0\pm0.2\times10^{-5}$	$<1\times10^{-7}$
^{131}I	NA	NA	$4\pm1\times10^{-7}$	$<1\times10^{-5}$	$<8\times10^{-7}$
^{133}Xe	$<3\times10^{-7}$	$<3\times10^{-7}$	NA	NA	NA
^{134}Cs	NA	NA	NA	$3\pm1\times10^{-7}$	NA
^{137}Cs	NA	NA	NA	$6\pm2\times10^{-7}$	NA
^{182}Ta	NA	NA	NA	$1.7\pm0.2\times10^{-5}$	$<2\times10^{-7}$

Notes:
1. $\pm$ values indicate analytical error expressed at 2 σ, $<$ values are minimum detectable concentrations at 3 σ counting error.
2. NA - Not analyzed.
3. Water vapor concentration, g/m^3: June 10 - 14.1; Nov. 19 - 5.6; Nov. 30 - 14.0

drum.(3) Gaseous ^{14}C was observed at concentrations above 1 x 10^{-4} μCi/cc in all three samples, but was measured accurately only in the last sample. The ^{3}H in the sample of June 3, 1969, was mostly (> 99 percent) in the form of hydrogen gas (HT) or a gaseous organic compound.

3.3.4 *Radionuclide concentrations in the vapor container.* The only radionuclides found in vapor container air were ^{3}H (both as water vapor and gas), ^{14}C, and ^{85}Kr, at the concentrations in columns 2 and 3 of Table 3.4. The minimum detectable level of other radionuclides by gamma-ray spectrometry is indicated by the "less-than" value of ^{133}Xe. Condensed water vapor, from a dehumidifier which collects water samples for tritium analysis by Yankee, contained ^{3}H and relatively low concentrations of many of the fission and activation products found in main coolant water (see Table 3.4, columns 4 to 6).

The ^{3}H concentrations in air, computed from concentrations in the condensed water vapor and the moisture content in air (see note 3 in Table 3.4), do not agree with directly measured values:

Date	^{3}H in air (condensed water vapor)
June 10	$1.4\times10^{-5}\ \frac{ml}{cc} \times 9.8\times10^{-1}\ \frac{\mu Ci}{ml} = 1.4\times10^{-5}\ \frac{\mu Ci}{cc}$
Nov. 19	$5.6\times10^{-6} \times 2.1\times10^{-3} = 1.2\times10^{-8}$

	^{3}H in air (direct)
June 10	$1.8\times10^{-6}\ \frac{\mu Ci}{cc}$
Nov. 19	8×10^{-7}

On June 10, during reactor operation, the condensed water vapor indicated an 8-fold higher ^{3}H concentration than was found in air; on Nov. 19, 1970, while the building was open during refueling, it indicated a 70-fold lower concentration. The two types of samples were obtained at the same location, but the air was collected for a much shorter interval than the condensed water vapor. The presence of ^{14}C

and ^{85}Kr in air and the differences in ^{3}H values suggest that gas samples should be analyzed by Yankee to determine radionuclide releases while ventilating the vapor container. The detection of the other, nonvolatile, radionuclides in condensed water vapor indicates their presence, but air filter samples would be required to quantify their concentrations.

The two sets of measurements in air were used to estimate annual releases: the amount discharged immediately after a shutdown was taken to be the product of the concentration on June 10, the air volume in the vapor container (24,000 m^3) (3), and the number of shutdowns per year (say 4); the amount discharged during refueling was taken to be the product of the concentration on Nov. 19, the ventilation rate (425 m^3/min), the refueling period (say, 30 days per year). Thus, the annual release would be:

		^{3}H, Ci	^{14}C, Ci	^{85}Kr, Ci
accumulated radionuclides discharged immediately after reactor shutdown	=	0.17	0.11	<0.0005
radionuclides discharged continuously during refueling	=	13.	0.066	2.8
yearly total		13.	0.18	2.8

These calculations suggest that most of the effluent gaseous radioactivity at Yankee is released from the vapor container during refueling (see totals in Section 3.3.8), and that monitoring this effluent provides a significant portion of the annual release data. The above values, based on one sample each, serve only to indicate the magnitude of radionuclide releases from the vapor container.

3.3.5 *Particulate radioactivity and radioiodine in the primary vent stack.* The activation products ^{54}Mn and ^{60}Co and the fission product ^{90}Sr were the only particulate radionuclides detected on the stack sampler (see Table 3.5 and 3.6). All three radionuclides were at extremely low concentrations. Except possibly ^{60}Co, these radionuclides appear to be associated with continuous release, rather than surge-drum gas (see Table 3.6). No particulate or gaseous ^{131}I was detected in any sample. The 24-hour samples of June 1970 (see Table 3.5) provided a more sensitive test of ^{131}I concentrations during continuous discharge than those of June 1969 because gamma-ray spectrometry was initiated sooner (within 31 hours) after sampling.

The average release rates according to the seven values in Tables 3.5 and 3.6 (and less-than values for ^{131}I based on Table 3.5 only) were:

Radionuclide	Average stack release
^{54}Mn	5 pCi/sec
^{60}Co	8
^{90}Sr	8
^{131}I	<9

To compute the annual discharge of these radionuclides, multiply the release rates by 2.8×10^{7} sec/yr.

The amounts of ^{89}Sr and ^{137}Cs that are formed in environmental air by radioactive decay of their

Table 3.5
Stack Releases of Particulate Radionuclides and Gaseous Iodine-131, pCi/sec

Radionuclide	Date, 1970 June 5	June 6	June 7	June 8	June 9
Particles on membrane filter					
313 -d ^{54}Mn	7 ± 6	< 1	5 ± 2	< 1	< 1
2.7 -yr ^{55}Fe	< 1	< 1	< 1	< 1	< 1
71.3 -d ^{58}Co	< 2	< 2	< 2	< 2	< 2
5.26-yr ^{60}Co	10 ± 4	3 ± 2	8 ± 3	2 ± 1	2 ± 1
50.5 -d ^{89}Sr	< 1	< 1	< 1	< 1	< 1
28.5 -yr ^{90}Sr	3 ± 1	0.5 ± 0.2	0.8 ± 0.2	< 0.2	0.8 ± 0.1
8.06-d ^{131}I	< 11	< 4	< 9	< 2	< 3
Gaseous iodine on charcoal					
8.06-d ^{131}I	< 3	< 3	< 3	< 3	< 3

Notes: 1. Nominal stack flow rate is 7.1 m^3/sec.
2. < values are minimum detectable concentrations at 3 σ counting error; ± values are 2 σ counting error.
3. 10^6 pCi/sec = 1 μCi/sec.

Table 3.6

Stack Effluent Release Rates During and After Gas Surge Drum Release, μCi/sec

Radionuclide	Calculated from surge drum contents,* June 3, 1969	Measured during release, June 3, 1969	Measured after release, June 4, 1969
Gas			
12.3 -yr ^{3}H	0.62	0.46 ± 0.02	< 0.15
5730 -yr ^{14}C	5.6	3.2 ± 0.4	0.010 ± 0.002
10.7 -yr ^{85}Kr	0.52	0.42 ± 0.09	< 0.03
5.3 -d ^{133}Xe	0.37	< 0.7	< 0.7
Gaseous iodine on charcoal			
8.06-d ^{131}I	---	$< 50 \times 10^{-6}$	$< 30 \times 10^{-6}$
Particles on filter			
5.26-yr ^{60}Co	---	$20 \pm 5 \times 10^{-6}$	$14 \pm 3 \times 10^{-6}$
313 -d ^{54}Mn	---	$18 \pm 4 \times 10^{-6}$	$3 \pm 1 \times 10^{-6}$
50.5 -d ^{89}Sr	---	$< 1 \times 10^{-5}$	$< 1 \times 10^{-5}$
28.5 -yr ^{90}Sr	---	$25 \pm 2 \times 10^{-6}$	$28 \pm 3 \times 10^{-6}$

*Calculated for concentrations in Table 3.3 and release rate of 0.425 m^3 (STP) per minute from surge drum.

Notes:

1. Nominal stack flow rate is 7.1 m^3/sec (15,000 cfm).
2. $<$ values are minimum detectable concentrations at 3 σ counting error; $\pm$ values are 2 σ counting error of single analysis or the difference of results of duplicate analyses, whichever is greater.

respective gaseous precursors, ^{89}Kr and ^{137}Xe, would be even smaller. The maximum amounts per curie of precursor are 42 μCi ^{89}Sr and 0.25 μCi ^{137}Cs. Approximately 1 Ci each of the two gases may be released annually at the air ejector and through sampling the primary coolant system (see Section 3.3.1 and 3.3.2). Hence the amount of ^{89}Sr reaching the environment through this mode may be 42 μCi/yr, or 1.4 pCi/sec; and the amount of ^{137}Cs, 0.25 μCi/yr or 0.008 pCi/sec.

3.3.6 *Gaseous radioactivity in the primary vent stack.* As shown in Table 3.6, ^{14}C was measured both during and after the release from the gas surge drum. Its emission rate was considerably higher during the release than afterwards. Tritium and ^{85}Kr were detected only during the release.

The measured surge drum release rates shown -- i.e., differences between values in the third and fourth columns of Table 3.6 -- were approximately two-thirds of the rates computed from the surge drum gas analyses (Table 3.3, last column). The lower value may have been caused by imperfect sampling or a slower gas release than the rated 0.425 m^3/min. This rate pertains to conditions at initial discharge, when the stack sample was obtained; as the internal surge drum pressure decreases due to discharge, the flow rate and hence the emission rate to the stack may decrease. The total released radioactivity, based on the discharged volume of 144 m^3 and the concentrations listed in the last column of Table 3.3 was as follows:

Radionuclide	Total release, Ci
^{3}H	1.3×10^{-2}
^{14}C	1.1×10^{-1}
^{85}Kr	1.1×10^{-2}
^{133m}Xe	$< 3 \times 10^{-4}$
^{133}Xe	7.5×10^{-3}

This indicates the magnitude of the annual release if the waste was typical and is discharged once per year.

Compared to the β-γ gaseous release for June 1969 reported by Yankee (see Appendix B.2) of 0.445 Ci, the radionuclides from the surge drum accounted for approximately one-third of the activity. The tritium release from the surge drum was 10 percent of the reported monthly release of 0.13 Ci.

The ^{14}C release rate in the stack on June 4, 1969, was 12-fold higher than measured at the main condenser air ejector (Table 3.2) on two other occasions. The difference was probably due to a higher radionuclide release at the air ejector on June 4, 1969, because the leakage from main to secondary coolant was higher.(3)

3.3.7 *Particulate effluent from incinerator.* The only airborne particulate radionuclides observed in the effluent from the incinerator during combustion of solid wastes were ^{58}Co, ^{60}Co and ^{90}Sr, as shown

Table 3.7

Particulate Radioactivity Emitted from Incinerator Stack, June 9, 1970*

Radionuclide	Concentration, μCi/cc	Emission rate, μCi/sec	Estimated annual release, + Ci
^{55}Fe	$< 10 \times 10^{-12}$**	---	---
^{58}Co	$4 \pm 2 \times 10^{-12}$	6×10^{-10}	5×10^{-10}
^{60}Co	$10 \pm 3 \times 10^{-12}$	2×10^{-9}	2×10^{-9}
^{89}Sr	$< 2 \times 10^{-12}$	---	---
^{90}Sr	$2.5 \pm 0.6 \times 10^{-12}$	4×10^{-10}	3×10^{-10}
^{131}I	$< 4 \times 10^{-12}$	---	---

* operation from 2030 to 2130 hrs.; sampling rate and stack exhaust flow assumed to be 167 cc/sec.

+ computed for 241 hours of operation in 1970.(3)

** $<$ values indicate minimum detectable concentrations at 3 σ counting error; $\pm$ values are 2 σ counting error.

in Table 3.7. For computing emission rates, the sampling flow rate was assumed to be identical to the stack exhaust rate. Radionuclide concentrations and the estimated annual release were very low on the basis of these data.

3.3.8 *Release limits and estimated annual radionuclide releases.* The totals of the release values in Sections 3.3.1, 3.3.2, 3.3.4, 3.3.5, and 3.3.6 compare as follows with the limits established by the AEC at the Yankee stack (1,000 times the limits given in 10 CFR 20,(9) Appendix B, Table II,column 1 for unrestricted areas):

Radionuclides		Yankee limit, μCi/cc	Annual release limit,* Ci	Estimated annual release, Ci	Percent of limit
Gases					
12.3 -yr	^{3}H (as HTO)	2×10^{-4}	4.5×10^{4}	1.3×10^{1}	0.029
	(as HT)	4×10^{-2}	8.9×10^{6}		
5730 -yr	^{14}C (s)	1×10^{-4}	2.2×10^{4}	3×10^{-1}	0.001
	(as CO_2)	1×10^{-3}	2.2×10^{5}		
1.83-hr	^{41}Ar	4×10^{-5}	8.9×10^{3}	4×10^{-1}	0.004
4.4 -hr	^{85m}Kr	1×10^{-4}	2.2×10^{4}	2×10^{-2}	$<$ 0.001
10.7 -yr	^{85}Kr	3×10^{-4}	6.7×10^{4}	3	0.004
76 -min	^{87}Kr	2×10^{-5}	4.5×10^{3}	2×10^{-2}	$<$ 0.001
2.8 -hr	^{88}Kr	2×10^{-5}	4.5×10^{3}	3×10^{-2}	$<$ 0.001
2.3 -d	^{133m}Xe	3×10^{-4}	6.7×10^{4}	2×10^{-3}	$<$ 0.001
5.29-d	^{133}Xe	3×10^{-4}	6.7×10^{4}	1×10^{-1}	$<$ 0.001
9.1 -hr	^{135}Xe	1×10^{-4}	2.2×10^{4}	2×10^{-1}	0.001
Other fission gases, half lives $<$ 2 hr		3×10^{-5}	6.7×10^{3}	(3)+	(0.04)
Particles and ^{131}I					
313 -d	^{54}Mn (s & i)	1×10^{-6}	2.2×10^{2}	1×10^{-4}	$<$0.001
5.26-yr	^{60}Co (i)	3×10^{-7}	6.7×10^{1}	2×10^{-4}	$<$0.001
50.5 -d	^{89}Sr (s)	3×10^{-7}	6.7×10^{1}	$(4 \times 10^{-5})^{+}$	($<$0.001)
28.5 -yr	^{90}Sr (s)	3×10^{-8}	6.7	2×10^{-4}	0.003
8.06-d	^{131}I (s)	1×10^{-7}	2.2×10^{1}	$< 3 \times 10^{-4}$	$<$0.001
30 -yr	^{137}Cs (i)	5×10^{-7}	1.1×10^{2}	$(2 \times 10^{-7})^{+}$	($<$0.001)

***Based on a continuous stack discharge rate of 425 m^3/min**

+These values were estimated from ^{135}Xe measurements (see Sections 3.3.1, 3.3.2, and 3.3.5).

Notes:

1. **The individual limits apply in the absence of other radionuclides; if several radionuclides are present, the sum of individual percentages of the limit may not exceed 100.**
2. **s = soluble, i = insoluble.**

The estimated annual releases of ^{3}H and the sum of all other radionuclides shown above are within better than a factor of two of the 1969-1970 values in Section 3.1.3 reported by Yankee. The annual values by Yankee are based on many more measurements than the ones in this study; on the other hand, the station reports isotopic analyses only for ^{3}H.

The whole-body radiation dose to persons who remained at the exclusion boundary throughout the year would have been 0.08 percent of 500 mrem/yr--i.e., 0.4 mrem/yr -- according to the above estimates from measured radionuclide releases. At highest fraction of the limit were ^{3}H (assuming the worst case--that all tritium was in the form of water vapor) and the very short-lived noble gas fission products among gases, and ^{90}Sr among particles.

The actual population exposure would probably be lower than the estimated value at the boundary because the nearest town, Monroe Bridge, is approximately 1 km distant. A better value of the annual dose rate could be obtained by performing isotopic analyses of the various airborne effluents at the station and measuring with a tracer the degree of dispersion from the stack to ground-level air.

3.4 References

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4. Radionuclides in Liquid Effluent

4.1 Liquid Waste System and Samples

4.1.1 *Liquid waste system.* (1-3) Two classes of liquid waste are discharged by Yankee: reactor plant liquid waste, which may contain hydrogen gas added to the main coolant to minimize the decomposition of water in the reactor, and secondary plant water, which does not contain added hydrogen gas. The usual sources and directions of flow of these wastes are indicated in Figures 4.1 and 2.2. Interconnections in the storage and treatment system provide other options.

Reactor plant liquid waste consists mostly of water that had been used in the main coolant system or in refueling the reactor (see Section 2.1.4). It is stored in two Waste Holdup Tanks and a Gravity Drain Tank before treatment by batch evaporation. The condensed water from the evaporator is collected in the Test Tanks, analyzed by Yankee for gross beta-gamma activity and tritium concentration, and discharged into effluent circulating coolant water. The dilution factor during this discharge is 530,000 liters/min ÷ 113 liters/min = 4,700.

Secondary-plant liquid wastes are mostly system leakage and once-daily steam-generator blowdown water; some steam-generator blowdown water is also discharged continuously (see Sections 2.1.3 and 2.1.4). The water is passed through two 5,300-liter (1,400 gal) monitored waste tanks and discharged at rates up to 113 liters/min into effluent circulating coolant water.

4.1.2 *Radionuclide release.* The following liquid waste was discharged at Yankee during 1970 and 1969: (4,5)

Class	Volume, liters	Gross beta-gamma, Ci	Tritium, Ci
1970			
reactor plant	3.1×10^6	0.69×10^{-3}	1,212
secondary plant	15.8×10^6	33.15×10^{-3}	280
total	18.9×10^6	33.84×10^{-3}	1,492
1969			
reactor plant	2.6×10^6	0.89×10^{-3}	1,048
secondary plant	13.8×10^6	18.34×10^{-3}	173
total	16.4×10^6	19.23×10^{-3}	1,221

The total discharge of gross activity and ^3H was typical of operations at Yankee. (6) The volume of liquid waste and the radioactivity varied considerably from month to month, as shown in Appendix B.2. Average concentrations of gross activity were higher in secondary-plant waste, probably because it is usually untreated while reactor-plant waste is usually evaporated before discharge:

Class	Gross beta-gamma, μCi/ml		Tritium, μCi/ml	
	1970	1969	1970	1969
reactor plant	2.2×10^{-7}	3.4×10^{-7}	3.9×10^{-1}	4.0×10^{-1}
secondary plant	2.1×10^{-6}	1.3×10^{-6}	1.8×10^{-2}	1.3×10^{-2}

Tritium concentrations were considerably lower in secondary-plant waste.

Average radioactivity concentrations in effluent circulating coolant water during waste discharge in 1969 and 1970, based on the annual release data given above and the coolant water flow rate of 530,000 liters/min, were:

source	assumed release rate, liters/min	gross $\beta\gamma$, μCi/ml	^3H, μCi/ml
reactor plant	113	6×10^{-11}	8×10^{-5}
secondary plant	28	9×10^{-11}	8×10^{-7}

The assumed rates imply discharge of reactor plant (Test Tank) waste during 4.8 percent of the year, and continuous release of secondary plant waste.

Concentrations of radionuclides in effluents to unrestricted areas are limited by the AEC according to paragraph 20.106 of 10 CFR 20. (7) Concentrations above background in water, averaged

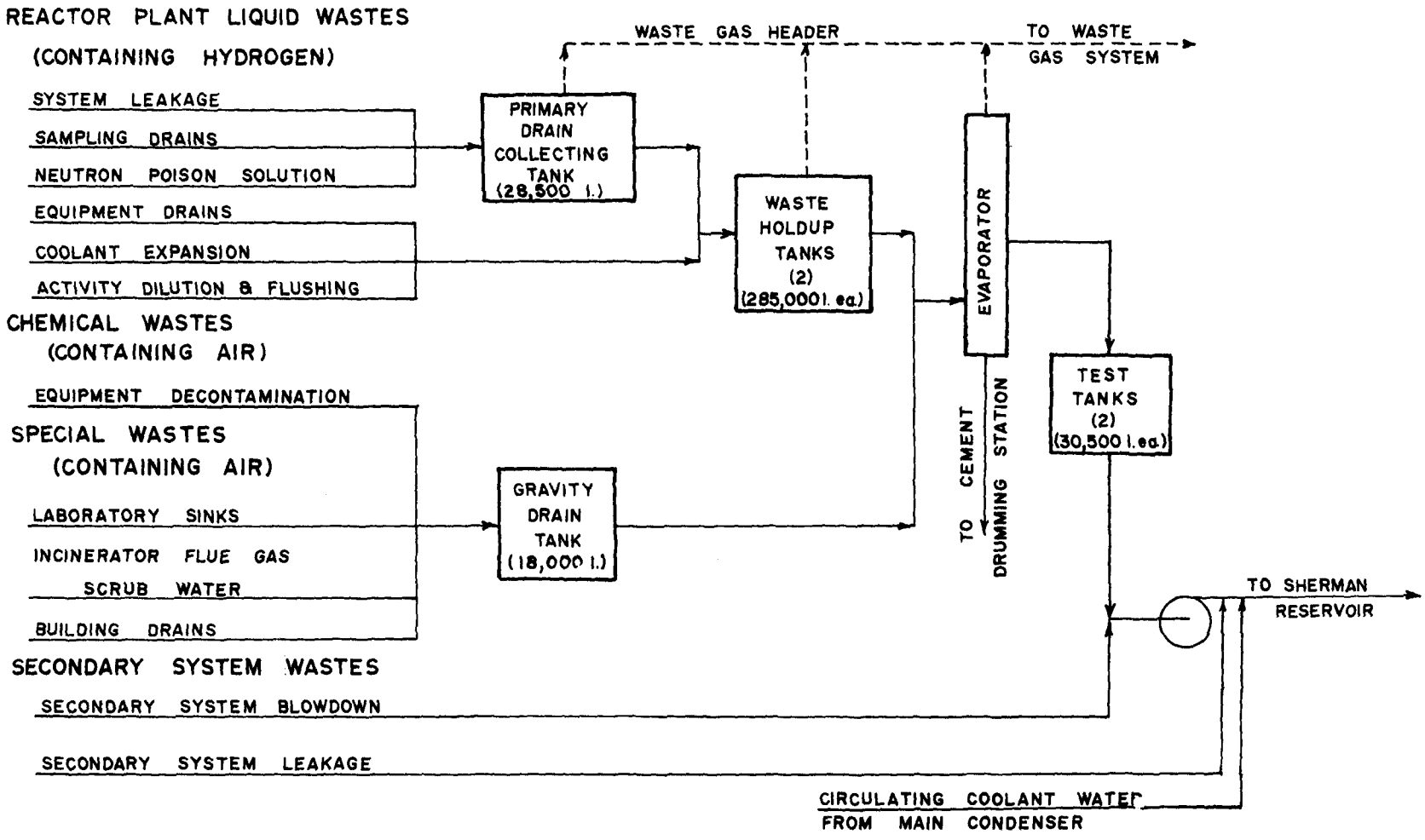

Figure 4.1. Liquid Waste Sources and Treatment.

over no more than 1 year, as listed in Appendix B, Table II, column 2 of 10 CFR 20, are applied at the boundary of the restricted area. The limit is 1×10^{-7} μCi/ml for an unidentified mixture containing no ^{129}I, ^{226}Ra, and ^{228}Ra. Limits for individual radionuclides are 3×10^{-3} μCi/ml for ^{3}H, the radionuclide at highest concentration in Yankee effluent, and 3×10^{-7} μCi/ml each for soluble ^{90}Sr and ^{131}I, which are usually the radionuclides with the lowest limits in reactor effluent. Higher limits are permissible under conditions of Subsection (b) of paragraph 20.106, or more stringent limits may be applied under Subsection (e).

Massachusetts has given temporary approval for daily releases of ^{3}H by Yankee at amounts not to exceed 10 Ci on the average, or 75 Ci at any time.(8) This is considerably lower than the limit of 2300 Ci/day computed at the normal flow rate of circulating coolant water according to Appendix B in 10 CFR 20.

4.1.3 *Samples.* Two 4-liter samples of a 27,400-liter (7,236-gal) reactor-plant waste solution in one of the two Test Tanks were obtained from the Yankee staff on June 3, 1969. This waste solution was condensate from the evaporator. One of the samples was acidified with 100 ml concentrated HCl to reduce possible sorption of radionuclides on the sides of the plastic container. The waste solution was released as usual into the effluent circulating cooling water at the flow rate of 113 liters/min (30 gal/min) between 1130 and 1530 on June 3, 1969.

To measure directly the radionuclide content of the effluent circulating coolant water, 200 liters were collected at the outlet weir in a steel drum at 1150-1200. The water was passed through an ion-exchange resin column at a flow rate of 100 ml/min to concentrate the ionic radionuclides on the column. A 4-liter aliquot of water from the drum was retained for measuring water hardness and radioactivity. For comparison, a 200-liter sample of service water, which is obtained at the same location in Sherman Reservoir as circulating coolant water, was collected in a steel drum from a tap in the pump house at 1015-1020 on June 3, 1969. A 180-liter volume of this water flowed through an ion-exchange resin column in a 29-hour period, and a 4-liter aliquot was retained for further analysis.

A second set of reactor-plant waste solution samples--4 liters acidified (10 percent HNO_3) and 1 liter unacidified--was obtained on Nov. 19, 1970. The reactor had been shut down for refueling on Oct. 24, hence most or all of the waste was from the refueling operation.

Four samples of water from the secondary plant--samples No. (5) to (8) in Section 2.1.6 -- were obtained from Yankee staff for analysis. Samples (5) and (6) were taken to represent blowdown discharges, and (7) and (8), secondary-plant leakage water.

Samples of flowing water and of a mixture of sand and gravel were collected on two occasions from the two yard drains that carry run-off water from the plant area:

(1) 4 liters water and 0.8 kg sand and gravel from east yard drain on June 3, 1969 at 1700;

(2) 0.8 kg sand and gravel from east yard drain on June 10, 1970 at 1000;

(3) 4 liters water and 0.8 kg sand and gravel from west yard drain on June 10, 1970 at 1000.

The west drain is located near the parking area and discharges into No. 5 Reservoir; the east yard drain is to the east of the pump house and discharges into Sherman Reservoir (see Section 5.1.2). Flow rates at the time of sampling were estimated to be 3 liters/min in the east drain and ten times as much at the west drain.

4.2 Analysis

4.2.1 *Test Tank solution.* The unacidified (at pH 6.1) and acidified solutions of the waste were analyzed spectrometrically with Ge(Li) and NaI(Tl) gamma-ray detectors. The samples were first counted within a week after collection and again several months afterwards to identify radionuclides by combining observations of gamma-ray energies and decay rates. The identified radionuclides were quantified by computing disintegration rates from count rates under characteristic photon peaks on the basis of prior counting efficiency calibrations of these detectors. The unacidified sample was analyzed radiochemically for ^{3}H, ^{14}C, ^{129}I, and ^{131}I, and the acidified sample, for ^{55}Fe, ^{63}Ni, ^{89}Sr, and ^{90}Sr. Thirty-ml aliquots of the samples were evaporated, measured with a low-background G-M counter to determine gross beta activity, and analyzed by counting with aluminum absorbers of increasing

thickness to indicate the beta energy of the major component and the effective counting efficiency (see Figure 4.2).

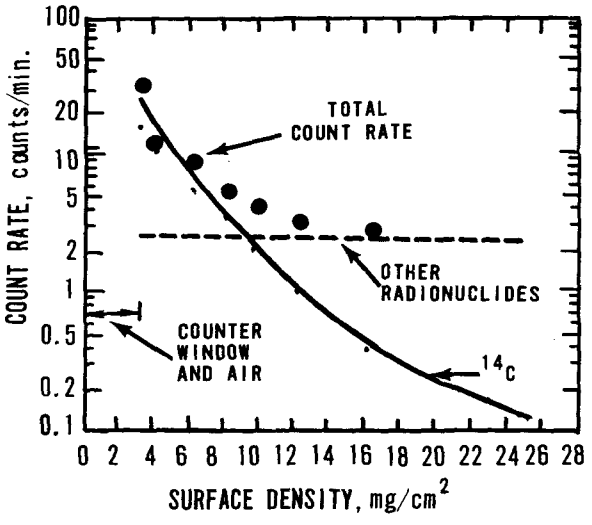

Figure 4.2. Aluminum Absorber Curve of Yankee Test Tank Sample.

Detector: Low-background G-M end-window.

Sample : 30-ml aliquot of sample collected June 3, 1969 evaporated on stainless steel planchet.

Counts : April 20, 1970, 100 min. at each point.

4.2.2 *Circulating coolant water.* Each of the two ion-exchange resin columns was separated into 6 parts: 3 cation-exchange resin sections, 2 anion-exchange resin sections, and a glass wool filter. (9) Each part was analyzed with a NaI(Tl) gamma-ray spectrometer for 1,000-minute counting periods. Every cation-exchange resin section was eluted with 1,200 ml 6 N HCl. The elutriants were analyzed radiochemically in sequence for strontium, cesium, and cobalt.

The two water samples were analyzed for hardness, gross beta activity, photon-emitting radionuclides, and a few individual radionuclides. Ten-ml aliquots were used to determine hardness and tritium. The tritium sample was distilled, and 4 ml were counted with a liquid scintillation detector. The remaining 4 liters of water were acidified with 10 ml conc. HNO_3 and evaporated to 45 ml, of which 15 ml were further evaporated to dryness for gross beta measurement and gamma-ray spectrometry with NaI(Tl) detectors, and 30 ml were analyzed sequentially for radiostrontium and radiocesium. These radionuclides and the gross-beta-activity samples were counted for 100 or 1000-min periods with G-M detectors at a background of approximately 1.5 counts/minute.

4.2.3 *Yard-drain samples.* The water samples were analyzed in the same manner as circulating coolant water by gamma-ray spectrometry and for tritium, radiostrontium, and radiocesium.

The sand and gravel were dried at 125°C, mixed, and analyzed in weighed 100-cc and 400-cc aliquots by gamma-ray spectrometry with Ge(Li) (see Figure 4.3) and NaI(Tl) detectors. The material was then separated with a U. S. No. 10 sieve and the larger and smaller particles were analyzed separately with a NaI(Tl) detector. Ten-gram samples of the larger and smaller particles were analyzed for ^{89}Sr and ^{90}Sr content by leaching with two 25-ml portions of hot 6N HNO_3, chemically separating strontium, and counting first total radiostrontium and, after 2 weeks, radiostrontium plus ^{90}Y.

4.3 Results and Discussion

4.3.1 *Radionuclides discharged to circulating coolant water.* The ^{3}H concentration in the Test Tank waste solution of June 3, 1969, was the same as in main coolant water on July 10, 1969 (see Tables 4.1 and 2.1), ^{14}C was approximately 3-fold lower in the waste, and all other radionuclides were lower in the waste by two to four orders of magnitude. Presumably, distillation in the evaporator reduced the concentrations of all other radionuclides. Tritium was the main radioactive component both during reactor operation on June 3, 1969, and during refueling on Nov. 19, 1970. Of the other radionuclides, ^{14}C and ^{55}Fe were at highest concentrations.

In this type of sample, analysis for specific radionuclides is particularly desirable because many of the radionuclides are not effectively counted with the usual beta-particle detectors, hence the gross activity value may be considerably lower than the actual radionuclide content. For example, the tritium concentration measured in the June 3 sample by Yankee staff (3) was 0.375 μCi/ml, in agreement with the value in Table 4.1, but the gross beta-gamma concentration was only 5.63×10^{-7} μCi/ml. For

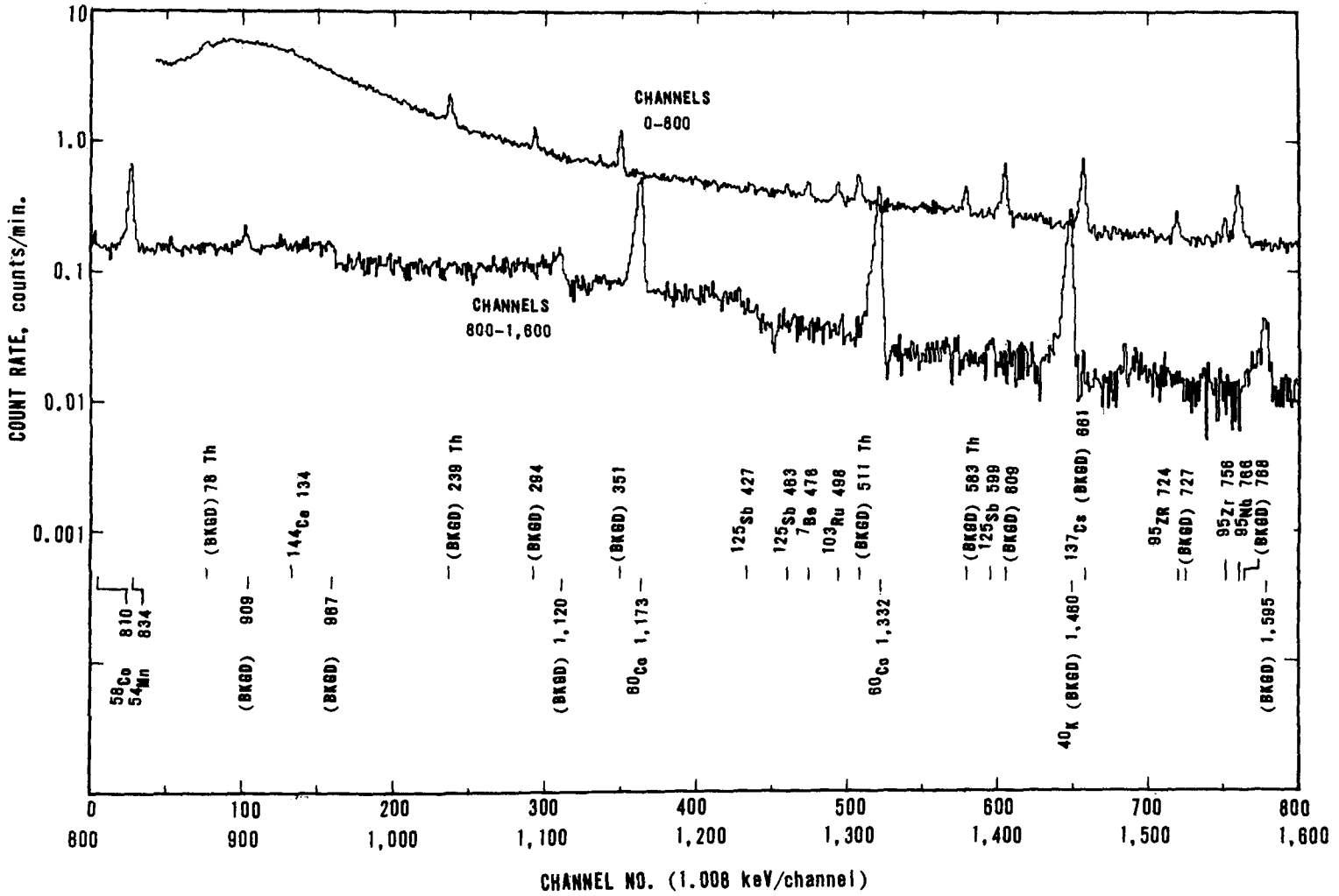

Figure 4.3. Gamma-ray Spectrum of Sand and Gravel from East Yard Drain.

Detector: *Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal*

Sample : *633 g dried wt (400 cc), collected June 3, 1969.*

Count : *July 1-2, 1969 (1,000 min.); Th refers to ^{232}Th and progeny, Bkgd refers to counter background (see background in Figures 5.4 and 5.5).*

comparison, gross-beta concentrations in 5 batches of waste in the Test Tanks during the first seven months of 1968 ranged from 4 x 10^{-8} μCi/ml to 1 x 10^{-6} μCi/ml.(10)

The radionuclide concentrations in secondary plant wastes (see columns 2 to 5 of Table 2.2) were generally two to three orders of magnitude lower than in the main coolant. As indicated in Section 2.3.5, the concentration of ^{3}H in secondary coolant water depends on the rates at which water leaks into and from the secondary system; hence, it may be one to four orders of magnitude below the main-coolant concentration. Several other radionuclides had the same secondary/main concentration ratios as ^{3}H. Compared to the 1970 annual average concentration in Section 4.1.2, ^{3}H values were the same on June 10, 1970, but the sum of other radionuclide concentrations was lower.

4.3.2 *Radionuclides in circulating coolant water.* Tritium was the only radionuclide measured in effluent circulating coolant water, while Test-Tank contents were discharged, that was attributable to release of this waste, as indicated in Table 4.2. The measured concentration of 7.9 x 10^{-5} μCi/ml was in excellent agreement with the value of 8.5 x 10^{-5} μCi/ml computed from the concentration in the Test Tank on June 3, 1970 (Table 4.1) and the dilution factor of 4,700. No tritium could be detected in intake water.

Even after concentrating ionic radionuclides from 200 liters of water on the ion-exchange resin column, only ^{90}Sr and ^{137}Cs could be detected. The two radionuclides were at the same concentrations in influent and effluent water, suggesting that these radionuclides originated in fallout from atmospheric nuclear weapon tests. As indicated by the calculated discharge values -- measured concentrations in the Test Tank divided by the dilution factor for circulating coolant water -- in Table 4.2, the concentrations added by Yankee to the effluent were within analytical uncertainty and thus not noticeable. All other radionuclides discharged by Yankee were below minimum detectable concentrations (<1 x 10^{-10} μCi/ml).

The ^{90}Sr concentrations measured directly in the water samples (see upper half of Table 4.2) were also the same in effluent and influent, but were higher than values obtained with the ion-exchange resins. The difference between the total and the ionic ^{90}Sr concentrations may be due to ^{90}Sr in suspended

Table 4.1

Radionuclide Concentration in Test Tank before Discharge at Yankee,* μCi/ml

Radionuclide	June 3, 1969	Nov. 19, 1970
^{3}H	4.0 x 10^{-1}	7.3 x 10^{-3}
^{14}C	5 x 10^{-6}	1.4 x 10^{-6}
^{32}P	5 x 10^{-8}	< 1 x 10^{-8}
^{54}Mn	2 x 10^{-7}	1.1 x 10^{-6}
^{55}Fe	1 x 10^{-7}	8 x 10^{-6}
^{59}Fe	< 1 x 10^{-7}	4 x 10^{-7}
^{58}Co	2 x 10^{-7}	9 x 10^{-8}
^{60}Co	7 x 10^{-8}	6 x 10^{-8}
^{90}Sr	7 x 10^{-9}	1.0 x 10^{-8}
^{110m}Ag	2 x 10^{-7}	< 1 x 10^{-8}
^{124}Sb	< 1 x 10^{-7}	5 x 10^{-8}
^{131}I	5 x 10^{-8}	6 x 10^{-8}
^{137}Cs	< 1 x 10^{-7}	9 x 10^{-8}
gross beta (unacidified)	5 x 10^{-6}	1.4 x 10^{-6}

* Radionuclide concentrations are at time of sampling; gross beta activity was obtained 5 days later.

Note:

^{51}Cr, ^{63}Ni, ^{89}Sr, ^{95}Zr, ^{95}Nb, ^{129}I, and ^{134}Cs were not detected. Minimum detectable levels were 5 x 10^{-7} μCi/ml for ^{51}Cr, 1 x 10^{-8} μCi/ml for ^{89}Sr, and 1 x 10^{-7} μCi/ml for all others.

Table 4.2

Radionuclide Concentration in Main-Condenser Circulating Coolant Water on June 3, 1969

Radionuclide	Intake, μCi/ml	Effluent, μCi/ml	Calculated discharge, μCi/ml
Water analysis			
^{3}H	$< 2 \times 10^{-6}$	7.9×10^{-5}	8.5×10^{-5}
^{14}C	NM	NM	1.1×10^{-9}
^{32}P	NM	NM	1.1×10^{-11}
^{55}Fe	NM	NM	2×10^{-11}
^{90}Sr	$1.5 \pm 0.6 \times 10^{-9}$	$1.4 \pm 0.6 \times 10^{-9}$	1.5×10^{-12}
^{110m}Ag	NM	NM	4×10^{-11}
^{137}Cs	$< 3 \times 10^{-10}$	$< 3 \times 10^{-10}$	$< 2 \times 10^{-11}$
gross beta	$2.4 \pm 0.5 \times 10^{-9}$	$2.4 \pm 0.5 \times 10^{-9}$	
Ion-exchange resin analysis			
^{54}Mn	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	4×10^{-11}
^{58}Co	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	4×10^{-11}
^{60}Co	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	2×10^{-11}
^{89}Sr	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	$< 2 \times 10^{-12}$
^{90}Sr	$3.1 \pm 0.3 \times 10^{-10}$	$3.3 \pm 0.3 \times 10^{-10}$	1.5×10^{-12}
^{131}I	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	1.1×10^{-11}
^{134}Cs	$< 1 \times 10^{-10}$	$< 1 \times 10^{-10}$	$< 2 \times 10^{-11}$
^{137}Cs	$5 \pm 1 \times 10^{-11}$	$5 \pm 1 \times 10^{-11}$	$< 2 \times 10^{-11}$

Notes: 1. Concentrations are corrected to collection date.
2. $\pm$ values are 2 σ counting error; $<$ values are 3 σ counting error; NM - not measured.
3. Calculated concentrations are concentrations in Test Tank (Table 4.1) divided by dilution factor of 4,700.

solids, but is considerably larger than the single measured value in suspended solids of 1.2×10^{-10} μCi/ml (Section 5.3.2). Total ^{90}Sr values are relatively uncertain because only 2.7 liters of water were analyzed, compared to the analysis of ionic ^{90}Sr in 160-liter and 180-liter samples.

Concentrations of longer-lived radionuclides (except ^{3}H) from the continuous secondary-system steam-generator blowdown were also too low for detection in effluent circulating coolant water. Calculated from the averages of the values in Table 2.2, the concentration of ^{3}H in circulating coolant water was 6×10^{-7} μCi/ml, and the concentrations of all other radionuclides were less than 1×10^{-10} μCi/ml each. A dilution factor of 19,000 was used to compute these values, based on a secondary-plant waste liquid discharge of 15×10^{6} liters/yr and the circulating coolant flow of 530,000 liters/min.

The continuous blowdown water may also contain short-lived radionuclides, although these were not detected in this study because of the relatively long interval between collecting and analyzing the sample. For the short-lived radionuclides that were measured in main coolant water (those with half lives of 6 hours or longer) the following concentrations at the point of discharge were computed at an average dilution factor of 19,000:

Radionuclide	Calculated concentration in secondary coolant,* μCi/ml				Calculated concentration at discharge, μCi/ml
	Oct. 10, 1968	July 10, 1969	June 10, 1970	Average	
^{24}Na	8.0×10^{-7}	2.9×10^{-4}	3.8×10^{-4}	2.2×10^{-4}	1×10^{-8}
^{64}Cu	$\sim 1 \times 10^{-7}$	1.6×10^{-7}	1.4×10^{-5}	4.7×10^{-6}	2×10^{-10}
^{99}Mo	5.5×10^{-8}	7.4×10^{-7}	1.9×10^{-6}	9.0×10^{-7}	5×10^{-11}
^{133}I	3.3×10^{-7}	6.4×10^{-6}	8.0×10^{-6}	4.9×10^{-6}	3×10^{-10}
^{135}I	4.6×10^{-7}	$\sim 9 \times 10^{-6}$	$\sim 1 \times 10^{-5}$	6×10^{-6}	3×10^{-10}

* From Table 2.1, for respective dilution factors of 2,000, 62, and 100; ^{135}I concentrations on July 10, 1969 and June 10, 1970 were estimated from ^{131}I and ^{133}I in main coolant.

Among these radionuclides, only ^{24}Na could have been readily detected. During 1966, a gamma-ray detector (with spectrometer) tested as an underwater monitor at the point of cooling-water discharge showed the presence of only one radionuclide -- ^{24}Na--at the concentration of 1.3×10^{-10} μCi/ml.(11)

4.3.3 *Performance of the ion-exchange columns for collecting radionuclides.* Relatively large volumes of water were passed through the columns because the hardness of the water was very low -- 9 mg/liter in terms of $CaCO_3$ in both inlet and outlet samples. On each cation-exchange resin, approximately 60 pCi ^{90}Sr and 9 pCi ^{137}Cs were retained. The distribution of these radionuclides on each column was:

section	^{90}Sr	^{137}Cs
top	81 ± 5*%	81 ± 5*%
middle	14 ± 4	12 ± 3
bottom	5 ± 1	7 ± 2

* ± values are one-half range of percent values for influent and effluent.

The sequential percentages suggest that, at most, 2 percent of the ionic strontium and 4 percent of the cesium were not retained on the columns. The devices are therefore useful for concentrating these radionuclides under the indicated conditions.

It was not necessary to wash suspended solids from these columns as was done during continuous sampling of coolant water (9) because the solids had settled in the barrels that held the water prior to passage through the columns. It would be desirable in future studies to collect and analyze the associated suspended solids.

4.3.4 *Radionuclides in yard-drain effluent.* Tritium and ^{60}Co were found in water samples from both drains; ^{54}Mn, ^{90}Sr, and ^{95}Zr were detected in water from the east yard drain only (see Table 4.3), possibly because water from the west yard drain was analyzed with lesser sensitivity. Of these radionuclides, ^{3}H and probably ^{54}Mn and ^{60}Co came from Yankee operations, and the others from fallout. Average concentrations of radionuclides in rainwater at Cincinnati during May and June, 1969, were:

^{54}Mn	$< 2 \times 10^{-9}$ μCi/ml	^{106}Ru	2×10^{-8} μCi/ml
^{60}Co	$< 2 \times 10^{-9}$	^{125}Sb	$< 2 \times 10^{-9}$
^{90}Sr	3×10^{-9}	^{137}Cs	3×10^{-9}
$^{95}Zr + ^{95}Nb$	5×10^{-8}	^{144}Ce	4×10^{-8}

Tritium concentrations in rain at nine locations in the U.S. were all below 2×10^{-6} μCi/ml during 1969.(12) Concentrations of ^{90}Sr, $^{95}Zr + ^{95}Nb$, ^{106}Ru, and ^{137}Cs in Cincinnati rainwater were considerably higher than detected or minimum detectable concentrations for the yard-drain water, possibly

Table 4.3

Radionuclide Concentration in Yard Drains

	East yard drain			West yard drain	
	Water, μCi/ml	Sand, pCi/g		Water, μCi/ml	Sand, pCi/g
Radionuclide	June 3, 1969	June 3, 1969	June 10, 1970	June 10, 1970	June 10, 1970
^{3}H	1.4×10^{-5}	NA	NA	7.5×10^{-6}	NA
^{54}Mn	4×10^{-9}	1.7	3.2	$< 5 \times 10^{-8}$	0.2
^{58}Co	$< 3 \times 10^{-9}$	0.1	< 0.1	$< 5 \times 10^{-8}$	< 0.1
^{60}Co	5×10^{-9}	3.0	5.1	3×10^{-8}	0.3
^{89}Sr	$< 1 \times 10^{-9}$	< 0.1	0.4	$< 2 \times 10^{-8}$	< 0.1
^{90}Sr	1.5×10^{-9}	0.1	0.5	$< 1 \times 10^{-8}$	0.3
^{95}Zr	2×10^{-9}	1.2	NA	$< 1 \times 10^{-8}$	0.9
^{106}Ru	$< 2 \times 10^{-9}$	0.4	NA	NA	NA
^{125}Sb	$< 2 \times 10^{-9}$	0.1	NA	$< 5 \times 10^{-8}$	NA
^{137}Cs	$< 2 \times 10^{-9}$	1.0	3.1	$< 1 \times 10^{-8}$	0.5
^{144}Ce	$< 2 \times 10^{-9}$	0.7	NA	NA	NA

Notes:
1. radionuclide concentrations are at time of sampling.
2. 1 pCi/g = 1×10^{-6} μCi/g
3. NA: not analyzed
4. < values are 3 σ counting error.
5. The radionuclides ^{59}Fe, ^{103}Ru, and ^{141}Ce were not detected; detection limits are approximately 2×10^{-9} μCi/ml water and 0.4 pCi/g of soil.

because these radionuclides were retained on soil during runoff of rainwater.

Tritium concentrations above background have been reported in the east and west storm drains by Yankee's contractor for environmental surveillance on several occasions during 1968 and 1969. (13) Values ranged from 2.2×10^{-4} to $< 2 \times 10^{-6}$ μCi/ml. A single beta activity value -- 2.38×10^{-8} μCi/ml -- was above background. The variation in reported ^{3}H values suggests that radionuclides from Yankee were only occasionally in the yard drain.

The ^{54}Mn, ^{58}Co and ^{60}Co in the sand and gravel over which the water flows (see Table 4.3) are attributed to Yankee, and were undoubtedly deposited from the water. The other radionuclides in the solids are at similar or higher concentrations in soil at other locations -- several were found in the samples listed in Table 6.3 -- and are attributed to fallout from atmospheric nuclear weapon tests. The radionuclides were found in both gravel and sand, but at somewhat higher concentrations in the smaller particles.

4.3.5 *Release limits and estimated annual radionuclide releases.* Amounts of individual radionuclides in liquid wastes were calculated by multiplying concentrations in reactor-plant liquid waste (Section 4.3.1) and secondary system steam generator blowdown (Section 2.3.5) by the volumes of waste water discharged annually (Section 4.1.2). The yard drains did not contribute significantly to these totals, according to Table 4.3. The releases compare as follows with the AEC limits for aqueous discharges:

	10 CFR 20 limit,* μCi/ml	Annual release limit, ** Ci	Estimated annual release, + Ci	Percent of limit
^{3}H (s,i)	3×10^{-3}	8×10^{5}	8×10^{2}	0.1
^{14}C (s)	8×10^{-4}	2×10^{5}	1×10^{-2}	<0.001
^{24}Na (i)	3×10^{-5}	8×10^{3}	(3)	(0.04)
^{32}P (s,i)	2×10^{-5}	6×10^{3}	8×10^{-5}	<0.001
^{51}Cr (s,i)	2×10^{-3}	6×10^{5}	2×10^{-2}	<0.001
^{54}Mn (s,i)	1×10^{-4}	3×10^{4}	1×10^{-2}	<0.001
^{55}Fe (s)	8×10^{-4}	2×10^{5}	1×10^{-2}	<0.001
^{59}Fe (i)	5×10^{-5}	1×10^{4}	4×10^{-3}	<0.001
^{58}Co (i)	9×10^{-5}	3×10^{4}	1×10^{-2}	<0.001
^{60}Co (i)	3×10^{-5}	8×10^{3}	2×10^{-3}	<0.001
^{63}Ni (s)	3×10^{-5}	8×10^{3}	1×10^{-3}	<0.001
^{64}Cu (i)	2×10^{-4}	6×10^{4}	(7×10^{-2})	(<0.001)
^{90}Sr (s)	3×10^{-7}	8×10^{1}	9×10^{-5}	<0.001
^{95}Zr (s,i)	6×10^{-5}	2×10^{4}	4×10^{-3}	<0.001
^{95}Nb (s,i)	1×10^{-4}	3×10^{4}	3×10^{-3}	<0.001
^{99}Mo (i)	4×10^{-5}	1×10^{4}	(1×10^{-2})	(<0.001)
^{110m}Ag (s,i)	3×10^{-5}	8×10^{3}	1×10^{-3}	<0.001
^{124}Sb (s,i)	2×10^{-5}	6×10^{3}	2×10^{-3}	<0.001
^{131}I (s)	3×10^{-7}	8×10^{1}	4×10^{-3}	0.005
^{133}I (s)	1×10^{-6}	3×10^{2}	(7×10^{-2})	(0.02)
^{135}I (s)	4×10^{-6}	1×10^{3}	(9×10^{-2})	(0.009)
^{137}Cs (s)	2×10^{-5}	6×10^{3}	2×10^{-4}	<0.001

* The most restrictive limit is listed.

** At annual flowrate of circulating coolant water = 5.3×10^{8} ml/min x 5.26×10^{5} min/yr = 2.8×10^{14} ml/yr.

\+ Average of test tank concentrations in Table 4.1 multiplied by average reactor plant discharge of 3×10^{9} ml/yr plus average of concentrations in Table 2.2 multiplied by secondary plant discharge of 15×10^{9} ml/yr. The values in parentheses are the average concentrations of short-lived radionuclides in secondary coolant water inferred from main coolant analyses (see Section 4.3.2), multiplied by the discharge rate of 15×10^{9} ml/yr.

Note: s = soluble, i = insoluble.

The estimated annual release of ^{3}H was 0.1 percent of the limit, that of ^{131}I was 0.005 percent, and all other measured radionuclides were at considerably lower percentages of the limit. Of the shorter-lived radionuclides whose concentrations in effluent water from the secondary system was inferred from analyses of main coolant water (see Section 4.3.2), the amounts of released ^{24}Na and ^{133}I were estimated to be, respectively, 0.04 and 0.02 percent of the limits; the others were at much lower percentages. The estimated annual release of ^{3}H is almost 2-fold lower than reported by Yankee (see Section 4.1.2), and the sum of all other radionuclides is several-fold higher. Note that these calculations are based on only a few sets of radioactivity data, and are therefore indications of the magnitude of individual radionuclide discharges rather than exact values.

These amounts of radionuclides in water at Yankee have no direct health implication because the Sherman Reservoir, the Deerfield River downstream from Yankee, and the Connecticut River below its confluence with the Deerfield River are not sources of public water supplies. The intake of radionuclides through eating fish caught in these waters is considered in Section 5.5.3.

4.4 References

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5. Radionuclides in the Aquatic Environment

5.1 Introduction

5.1.1 *Studies near Yankee.* A preliminary examination of release data in Sections 3.1.2 and 4.1.2 suggested that the only location in the environment where radionuclides from Yankee might be found was Sherman Reservoir near the circulating coolant water outlet. Efforts to detect and measure effluent radionuclides were therefore concentrated in this area. These studies are described in detail in Sections 5.2 to 5.6. In brief, they consisted of the following:

(1) Tritium concentrations in the Sherman Reservoir and Deerfield River below the Reservoir were measured during and after release of a batch of radioactive liquid waste by Yankee. As indicated in Table 4.2, tritium was the only radionuclide in this waste that could be detected at the point of discharge. Tritium concentrations above background were found just beyond the point of discharge and in the Deerfield River below the Sherman Reservoir.

(2) Water samples were also analyzed by gross beta, gamma-ray spectrometric, and radiostrontium measurements. Plankton samples were collected throughout Sherman Reservoir and analyzed in the same way, but only very small samples could be obtained. No radioactivity attributable to Yankee was detected in any sample.

(3) Radiostrontium and photon-emitting radionuclides were measured in a sample of water moss and a sample of dead leaves from Sherman Reservoir near Yankee. Both media apparently collected some of the radionuclides discharged by Yankee. Algae were looked for on June 3 and July 19, 1969, but were not found growing, presumably because the water was too cold.

(4) Radionuclide contents were compared in fish from Sherman Reservoir and from Harriman Reservoir, upstream from Yankee. Only ^{90}Sr, ^{137}Cs and traces of ^{22}Na were found in both sets of fish samples.

(5) Benthal samples -- mostly bottom mud -- were collected both by diver and with dredges from a boat, and examined for radionuclide content by gamma-ray spectrometry and ^{90}Sr analysis. The bottom of the southern end of Sherman Reservoir was monitored with an underwater NaI(Tl) probe connected to a portable gamma-ray spectrometry system. Radionuclides attributable to Yankee were found in the samples and with the probe throughout the southern end of Sherman Reservoir.

Radioactivity attributed to Yankee in water (tritium) and in benthal samples had been observed previously by Yankee's contractor for environmental surveillance.(1) Such radioactivity had also been detected in sediment samples by the Massachusetts Department of Public Health (MDPH). (2) Gross beta activity measured in Sherman Reservoir water during previous years (1,2) showed no increase due to Yankee, in accord with the observations in this study. Gamma-ray spectra from a NaI(Tl) detector immersed in water at the circulating coolant outlet on June 16, 1966, had shown naturally occurring ^{40}K, ^{226}Ra, and ^{232}Th, and a trace of ^{24}Na (0.13 ± 0.1 pCi/liter) from Yankee. (3)

At the Indian Point I PWR, low levels of ^{24}Na, ^{56}Mn and ^{131}I were observed in discharge water with the immersed NaI(Tl) detector; (3) and ^{54}Mn, ^{58}Co, ^{60}Co, ^{134}Cs, and ^{137}Cs were detected in sediment, aquatic vegetation, and fish below the outfall. (4) At the Dresden I BWR, ^{58}Co,^{60}Co, ^{89}Sr, ^{90}Sr, ^{131}I, ^{134}Cs, ^{137}Cs, and ^{140}Ba were found in effluent

coolant-canal water during waste discharges. (5)

5.1.2 *Deerfield River and Sherman Reservoir.* The Deerfield River is formed by several branches that arise in the Green Mountains of southern Vermont. It empties into the Connecticut River near Greenfield, Mass., 40 river miles* below the Sherman Dam. The river is used intensively for generating power, and its flow is closely controlled for this purpose at the large Somerset and Harriman Reservoirs, upstream from Sherman Reservoir. Water flows into the northern end of Sherman Reservoir from discharge at the Harriman hydroelectric station and/or Harriman Dam; it flows out of Sherman Reservoir through the intake of Sherman hydroelectric station, and/or the sluice and spillway of Sherman Dam. Approximately 0.7 miles below Sherman Dam is Dam No. 5, which impounds water for use by hydroelectric station No. 5 and a paper (glassine) manufacturer at Monroe Bridge. The Deerfield River is used for sport fishing but not for public water supply. (6) Flow data for the Deerfield River at the USGS Charlemont Gaging Station on the left bank near Deerfield River Mile 26 (DRM 26) from 1913 to 1966 are as follows: (7)

maximum daily, Sept. 21, 1938	**56,300 cfs (flood)**
minimum daily, June 17, 1921	**5**
mean daily in 1965	**528**

During the field trip described here, the average daily river flow was as follows:

Date, 1969	Sherman Station(8)	Charlemont gaging station (9)
June 2	701 cfs	831 cfs
June 3	728	993
June 4	560	750

The Sherman Reservoir, located at the Vermont-Massachusetts border, is approximately rectangular with a narrow neck at its northern end, as shown in Figure 5.1. The rectangle is approximately 8,000 ft (2,400 m) long and 850 ft (260 m) wide, and the lake extends to a depth of 80 ft (24 m). On the basis of these dimensions, it was estimated to have a capacity of 3×10^8 ft^3. The water is cold; on June 3, 1969, it was 54°F at the surface and 47°F at a depth of approximately 8 m; on July 29, 1969, it was 60° to 63°F at the surface. Effluent circulating coolant water from Yankee is approximately 15°F warmer than influent water. The water is very soft (i.e., low calcium plus magnesium content), according to the analytical data in Table 5.1.

Yankee is on the southern shore of Sherman Reservoir, as shown in Figure 5.2. The locations of the intake and outlet for circulating coolant water and of nearby sampling points are shown in Figure 5.2, and in greater detail in Figure 5.3. Note the proximity of the Yankee Station water outlet to the Sherman Station water intake.

5.2 Tritium in Water

5.2.1 *Sampling and analysis.* Water was collected to measure tritium concentrations beyond the point of release during and after the discharge of reactor-plant waste solution (see Section 4.1.3) into effluent circulating coolant water. Samples were obtained at the locations and times listed in Table 5.2. Water was collected in 50-ml portions at the water surface and, in some instances, 2.5 m below the surface (see Note 2 to Table 5.2). All of the samples at the south end of Sherman Reservoir were collected while the waste solution was being released. The Sherman hydroelectric power station was operating during the entire period, and a distinct pattern of water flow from the Yankee outlet to the Sherman water intake was visible.

The water samples were prepared for tritium analysis by distilling at least 10 ml of water to separate tritium from nonvolatile radionuclides. The distilled water was then mixed with scintillating solution to measure the tritium in a liquid-scintillation counter. The energy-response settings of the counter were adjusted to optimize detection of the low-energy beta particles of ^{3}H. For routine analysis, the minimum detectable concentration was 2 pCi/ml. Some samples were counted with an improved detection limit of 0.2 pCi/ml in a modified liquid-scintillation apparatus.† Results at the higher concentrations were confirmed by analyzing several samples with both detectors.

5.2.2 *Results and discussion.* The ^{3}H concentration at the circulating coolant water outlet was 79 pCi/ml (Table 4.2). Values from the traverses in front of the coolant-water outlet–samples 22 A to E and 23 A to E in Table 5.3 -- conform to the observed flow pattern in that tritium concentrations were relatively high near the water intake for

* 1 mile = 1.61 km; 1 cubic foot per second (cfs) = 28.3 liter/sec.

† We thank R. Lieberman, SERHL, EPA, for these analyses.

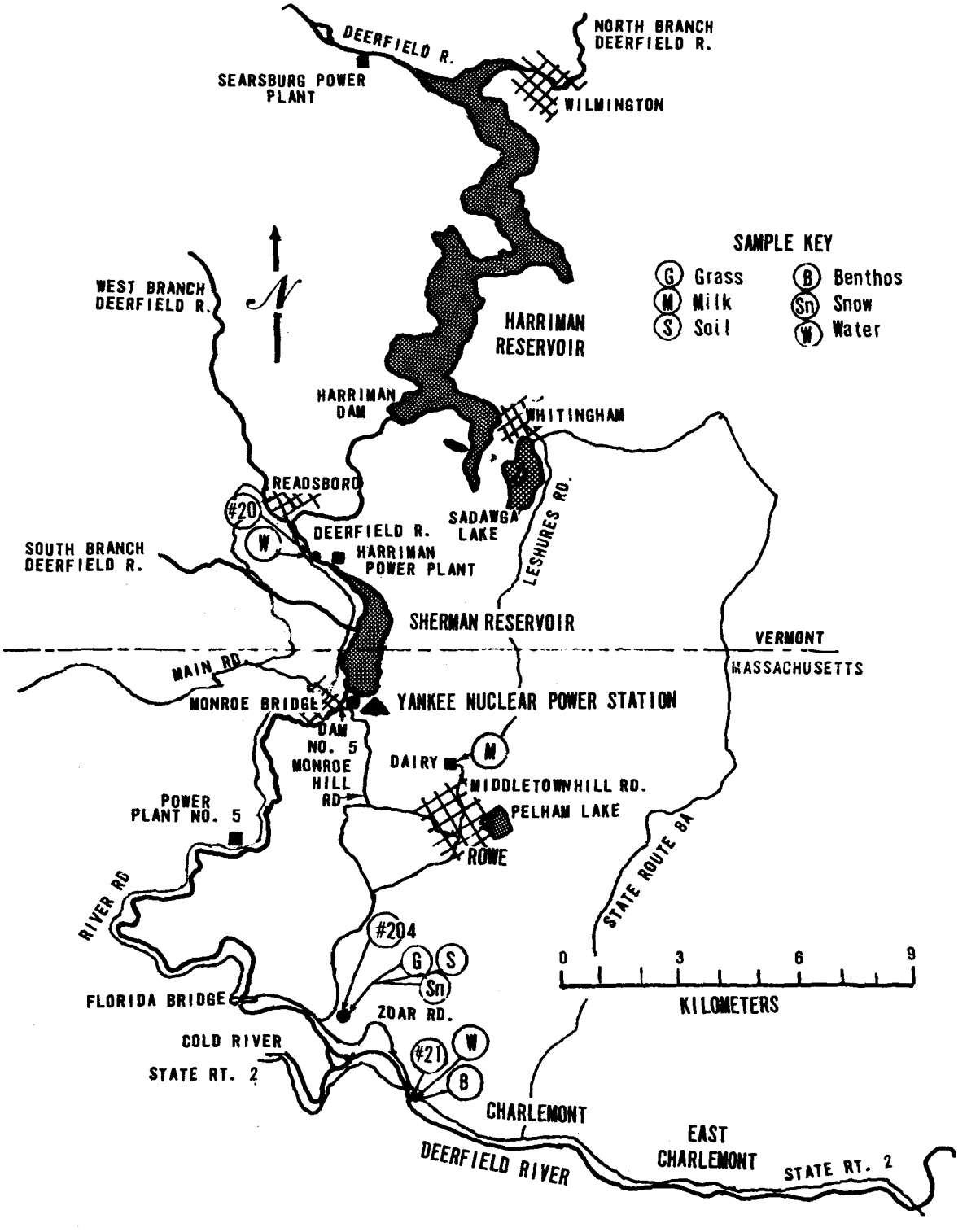

Figure 5.1. Deerfield River Near Yankee Nuclear Power Station.

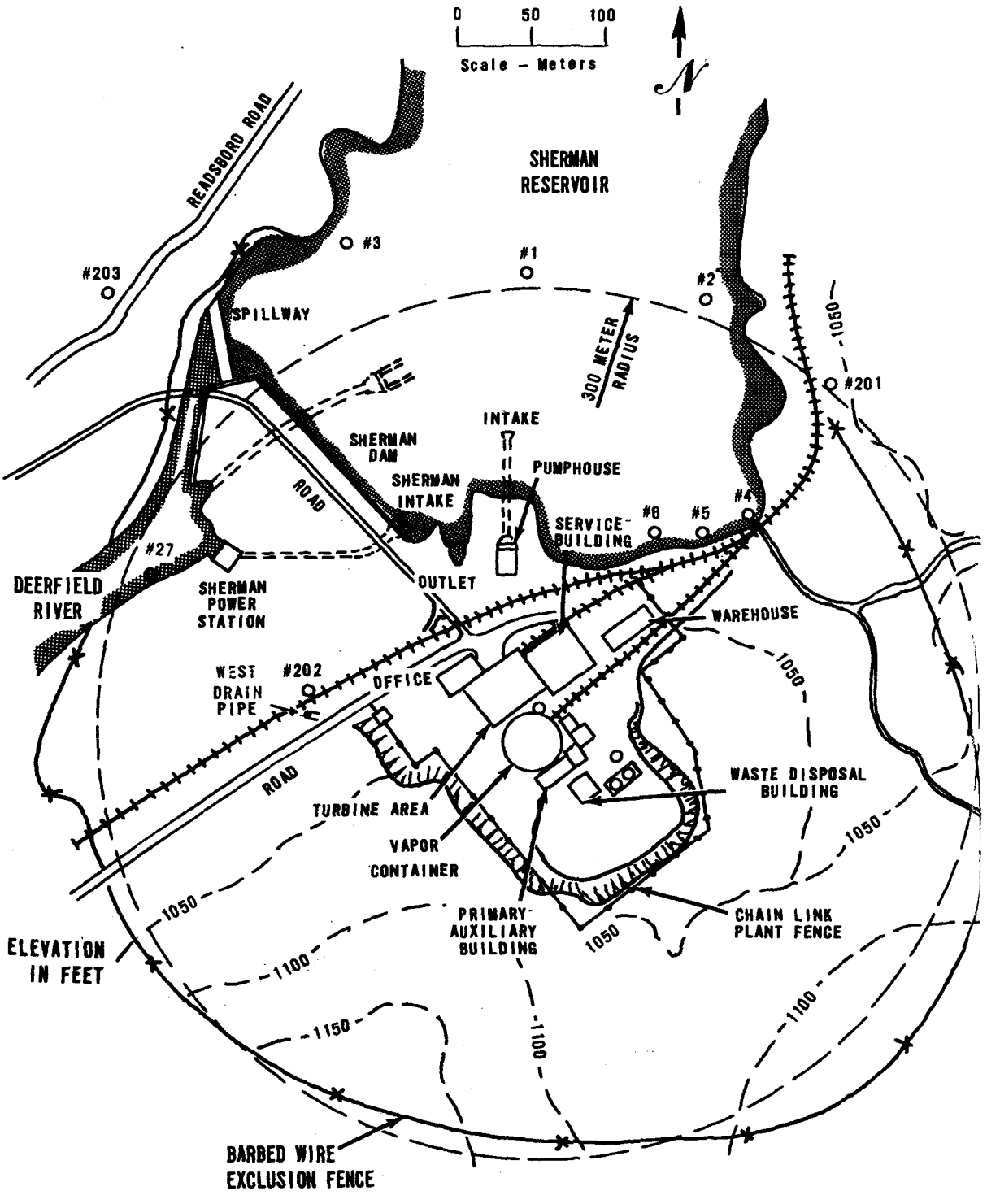

Figure 5.2. Yankee Nuclear Power Station. Note: Elevations refer to New England Power Co. datum; add 106 ft to obtain USGS elevation above mean sea level.

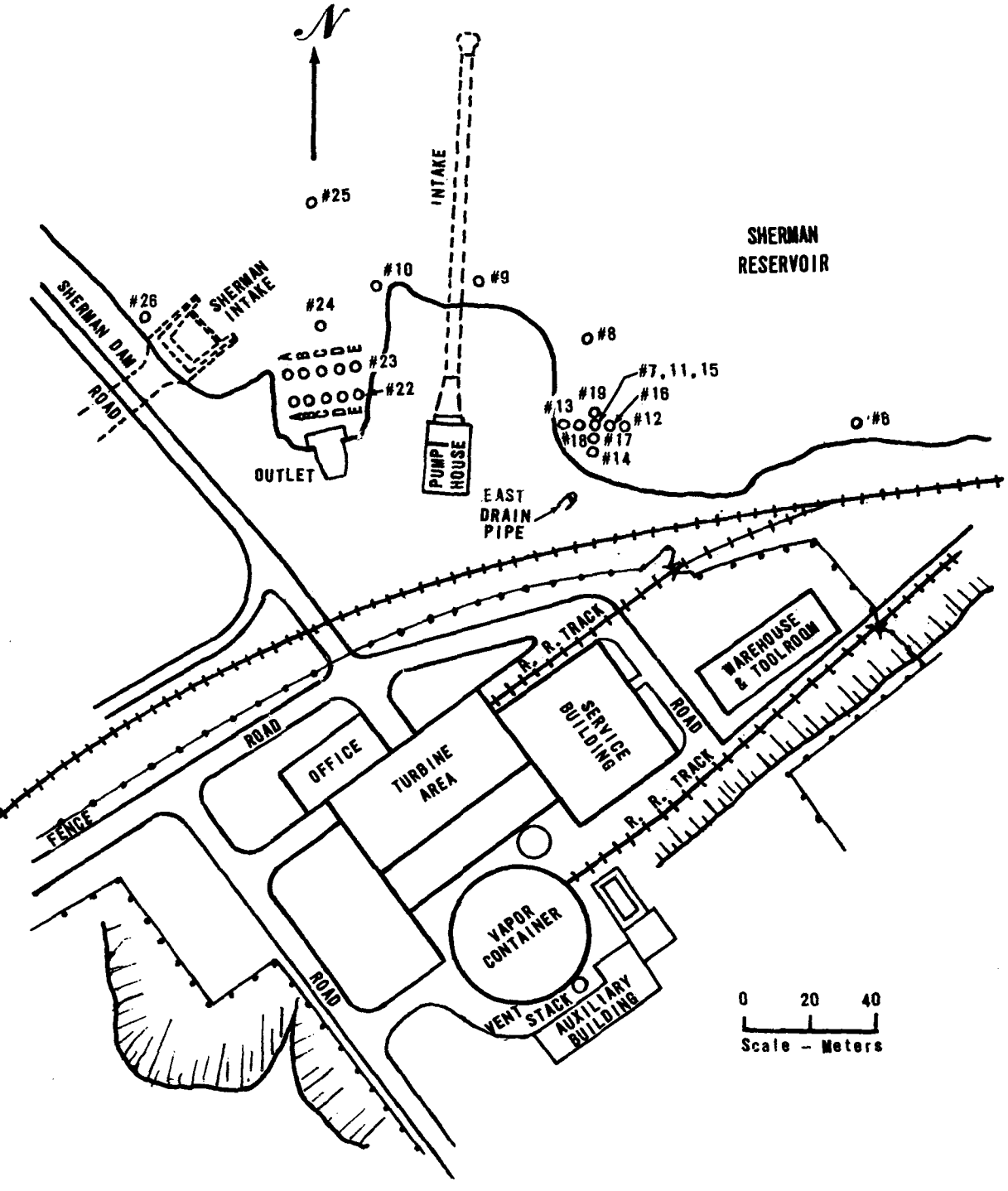

Figure 5.3. Yankee Nuclear Power Station Detailed Plan.

Table 5.1

Concentration of Stable Substances in Water from Deerfield River

Substance	Concentration, μ g/liter
Sodium	3,500
Magnesium	2,900
Potassium	900
Calcium	700
Iron	140
Aluminum	80
Boron	57
Manganese	55
Zinc	36
Nickel	21
Barium	17
Strontium	6
Copper	5
Arsenic	<13
Beryllium	<0.03
Cadmium	<3
Chromium	<1
Cobalt	<3
Lead	<5
Molybdenum	<5
Phosphorus	<13
Silver	<0.3
Vanadium	<5

Notes:

1. Sample was collected at location #27, below outflow of Sherman Station, on June 3, 1969 at 1345.
2. We thank Robert Kroner, Water Quality Office, EPA, Cincinnati, Ohio, for this analysis.
3. Concentrations were measured by emission-spectrographic analysis, except that sodium, magnesium, potassium, and calcium were by atomic absorption spectrometry.

Table 5.2

Tritium Sampling Points

#	Location	Collection date and time, 1969
1	Sherman Reservoir near 300-m station perimeter	June 3, 1200
20	north of Harriman Station (backgrou- d)	June 4, 1000
21	Deerfield River west of Charlemont (DRM 27)	June 4, 1600
22 A	Sherman Reservoir, 8 m north of outlet weir at west shore	June 3, 1200-1210
B	3 m from west shore	
C	at centerline	
D	3 m from east shore	
E	at east shore	
23 C (2)	16 m north of outlet weir at centerline	June 3, 1300
23 A	16 m north of outlet weir at west shore	June 3, 1215-1225
B	3 m from west shore	
C	at centerline	
D	3 m from east shore	
E	at east shore	
26	northwest of Sherman Station intake	June 3, 1230
27	#5 Reservoir below Sherman Station outlet (DRM 40)	June 3, 1345

Notes:

1. numbers indicate locations shown on Figures 5.1, 5.2 and 5.3.
2. all samples were collected at surface; in addition, samples were collected at 2.5-m depth for #1, 22, 23 and 26.

Table 5.3

Tritium Concentration in Sherman Reservoir and Deerfield River

#	Depth	Concentration, pCi/ml				
		Sherman Reservoir, near weir				
		A	B	C	D	E
22	surface	72	65	65	62	4
	2.5-m depth	69	58	60	12	< 2
23 (2)	surface			16		
23	surface	41	42	1.5	0.6	< 2
	2.5-m depth	18	32	16	< 2	3
26	surface	4				
	2.5m depth	< 2				
		Sherman Reservoir, near 300-m perimeter				
1	surface	< 2				
	2.5-m depth	< 2				
		Sherman Reservoir, background				
20	surface	0.4 ± 0.2				
		Deerfield River, below Sherman Station				
27	surface	27				
21	surface	2.4 ± 0.4				

Notes:

1. Sampling points are described in Table 5.2. See Figure 5.1 for locations #20 to 21; Figure 5.2 for locations #1 and 27; and Figure 5.3 for locations # 22, 23, and 26.
2. ± values are 2 σ counting error; < values are 3 σ counting error.
3. 1 pCi/ml = 1 x 10^{-6} μCi/ml
4. values at locations #20, 21, 23 C and 23 D are by R. Lieberman, SERHL, EPA.

Sherman Station and low on the opposite (east) side (see Figure 5.3). No tritium above background was found at the 1,000-ft (300-m) station perimeter in Sherman Reservoir, approximately 600 ft north of the outlet weir. The background concentration of 0.4 pCi/ml at the north end of Sherman Reservoir is similar to background values between 0.1 and 1.4 pCi/ml measured in U.S. surface water between January and June, 1969. (10)

The tritium concentration on the east bank of No. 5 Reservoir, just below the Sherman plant outlet, was 27 pCi/ml. This value was approximately one-third of the concentration at discharge, and similar to the average concentration before intake by Sherman Station (No. 23 A). The coolant water flow rate of 310 cfs (140,000 gal/min) and the average Sherman Station flow rate of 728 cfs (on June 3) indicate that effluent circulating coolant water from Yankee was diluted more than two-fold before reaching No. 5 Reservoir.

The ^3H concentration of 2.4 pCi/ml in the sample collected on the east bank of the Deerfield River near Charlemont at DRM 27 (location No. 21), approximately 24 hours after termination of the release, was 2.0 pCi/ml higher than the background concentration. The peak tritium concentration had undoubtedly passed location No. 21 several hours before sample collection: if the ^3H were completely mixed with water at estimated volumes of 1 x 10^7 $ft.^3$ in No. 5 Reservoir and 1 x 10^7 ft^3 in the Deerfield River between Dam No. 5 and location No. 21, (11) the time of flow from Sherman Station to location No. 21 is 2 x 10^7 $ft^3 \div 993$ ft^3/sec = 2 x 10^4 sec (i. e., 6 hours). In addition, the tritium in water may have been diluted by approximately a factor of two between Sherman Station and location No. 21.

The Yankee contractor for environmental surveillance reported similar ^3H concentrations in several samples collected in the Sherman Reservoir and immediately downstream during 1967 and 1968; in 1969, most of the samples contained no detectable ^3H: (1)

Location	Tritium Concentration, pCi/ml *
Mohawk Park (DRM 27)	4.1 ± 0.8 (Nov. 8, 1967)
Harriman Station (background)	< 2 (19 samples, 1969)
Hydroelectric Station #5	< 2 (18 samples, 1969)
	5 ± 0.8 (April 30, 1969)
Sherman Reservoir	< 2 (19 samples, 1969)

*Sample of Aug. 30, 1969 not included because of apparent contamination; DRM 27 is not a routine sampling station, and the sample was collected only as result of Abnormal Occurrence 67-11 at Yankee. (12)

Thus, tritium in radioactive liquid wastes discharged at Yankee can be used as a tracer to determine dispersion near the point of release and dilution in the Deerfield River. The dispersion would be different from the observed pattern when Sherman Station does not operate, so that the water is retained in Sherman Reservoir or released at the dam. The short-term concentrations of ^{3}H at the point of discharge and beyond were below 3 percent of the limiting annual average of 3,000 pCi/ml (3×10^{-3} μCi/ml) given in 10 CFR 20. Because the water is not ingested by humans, there is no direct radiation exposure to humans by this route.

5.3 Other Radionuclides in Water

5.3.1 *Unfiltered samples.* Water samples (3.5 liters) were collected at locations No. 20, 21, and 27 at the same time as the tritium samples, and also at the Yankee outlet weir on June 3, 1969, at 1000, before liquid waste was released from the Test Tank. The samples were acidified with 10 ml concentrated HCl, evaporated to 45 ml, and analyzed with a NaI(Tl) gamma-ray spectrometer. Thirty ml of each concentrated solution were then analyzed radiochemically for ^{89}Sr, ^{90}Sr, and ^{137}Cs, and 15 ml were evaporated to dryness and counted with a low-background G-M detector for gross beta activity.

The average gross beta activity of the four samples in Table 5.4 and the two in Table 4.2 was $2.3 \pm 0.2 \times 10^{-9}$ μCi/ml, and the average ^{90}Sr content, $1.1 \pm 0.3 \times 10^{-9}$ μCi/ml; no individual sample had significantly higher values than the averages, hence the ^{90}Sr in water is attributed to fallout from atmospheric nuclear weapon tests, and the gross beta activity, to fallout plus naturally occurring radionuclides. No ^{89}Sr ($< 2 \times 10^{-9}$ μCi/ml) or ^{137}Cs ($< 5 \times 10^{-10}$ μCi/ml) was detected by radiochemical analysis, and no radionuclides (generally $< 2 \times 10^{-9}$ μCi/ml) were found by gamma-ray spectrometry. These results are consistent with the calculated discharges in Table 4.2.

The gross beta activity measured in Sherman Reservoir and the Deerfield River is within the ranges of the most recent published data by the MDPH and Yankee's contractor for environmental surveillance, but is considerably below maximum values reported by the latter:

Location	MDPH (2) May-Nov., 1968	Yankee contractor Jan.-Dec., 1969(1)
Harriman Station	2-6 pCi/liter*	< 4.5-377 pCi/liter
Sherman Reservoir	---	< 4.5-21
Sherman Dam Sluiceway	2-8	---
Station #5	1-6	< 4.5-12
Monroe Bridge	1-5	---

* 1 pCi/liter = 1×10^{-9} μCi/ml.

The highest concentration reported by Yankee's contractor -- 377 pCi/liter at Harriman Station on Oct. 31, 1969 – was found by the contractor to be due to dissolved ^{60}Co;(1) the source of this radionuclide, at a location upstream from Yankee, is unknown, but laboratory contamination may be a possibility. The gross alpha activity during 1969,

Table 5.4

Gross Beta Activity and ^{90}Sr Concentration in Water from Sherman Reservoir and Deerfield River, pCi/liter

#	Sample	Gross beta	^{90}Sr
–	Yankee outlet, no waste discharged	2.4 ± 0.5	1.1 ± 0.5
20	Sherman Reservoir water (background)	2.5 ± 0.5	1.0 ± 0.5
27	#5 Reservoir, DRM 40	2.2 ± 0.5	0.7 ± 0.5
21	Deerfield River, DRM 27	1.9 ± 0.5	1.1 ± 0.5

Notes:

1. pCi/liter = 1×10^{-9} μ Ci/ml; ± values are 2 σ counting error.
2. See Table 5.2 for sampling locations and times; water at outlet was sampled on June 3, 1969 at 1000.
3. Values are based on 1 liter unfiltered water for gross beta and 2 liters unfiltered water for ^{90}Sr.

measured by the contractor, (1) was <2.3 pCi/liter except for one or two values near the minimum detectable level at all three sampling locations. The most recent radioactivity concentrations reported for raw surface water in the general area by the Federal Water Pollution Control Administration are for the Connecticut River:

gross beta activity (Wilder, Vt., Dec. 1968)[13]: 4 pCi/liter
^{90}Sr (Northfield, Mass., July-Sept. 1967)[14]: 1.1

5.3.2 *Suspended solids.* An 11.4-liter sample of water, collected at location 23 C(2) (see Table 5.2) during release of Test Tank waste by Yankee, was immediately passed through a membrane filter (8-μ pore diameter) to separate suspended solids for radiometric analysis. The filter was counted for 1000 minutes with a NaI(Tl) gamma-ray spectrometer, and was then ashed, weighed, and analyzed chemically for radiostrontium and radiocesium content.

Three macroplankton samples were collected from Sherman Reservoir on July 29, 1969, by towing a 10-cm-dia. plankton net at a depth of 1.5 m behind a slowly moving boat. The samples were obtained in front of the Yankee outlet, in the bay east of the Yankee pump house, and just upstream from the outlet at Harriman Station, to provide a background value. The volume of sampled water was estimated to be between 250 and 500 liters in each collection. Because of heavy rains, the Reservoir was approximately 2 m higher than during the June field trip, and the water was muddy. Very little plankton was observed. The plankton samples were separated on Whatman No. 41 filter paper from the 50 - 100 ml of water in which they were suspended. They were then ashed and analyzed in the same way as the filter sample described above.

The sample collected on the membrane filter, which appeared to be mostly silt, contained a small amount of ^{90}Sr but no detectable ^{137}Cs (see Table 5.5). The samples collected with the plankton net contained some silt -- especially the heaviest sample -- and showed ^{137}Cs but no ^{90}Sr. The concentration of the two radionuclides per liter of water, based on the values in Table 5.5, were:

Sample	^{90}Sr, pCi/liter	^{137}Cs, pCi/liter
filter-Yankee outlet	0.12	< 0.05
net-Yankee outlet	< 0.0005	0.004
Yankee bay	< 0.0008	0.002
Harriman Station	< 0.0005	0.004

The filtered sample had approximately 10 percent of the ^{90}Sr concentration in unfiltered water (see Table 5.4), while the other samples had less than 0.1 percent. The ^{137}Cs concentration in the samples collected with the plankton net was approximately 7 percent of the ^{137}Cs concentration in Reservoir water (see Table 4.2). No ^{89}Sr (<1 pCi/sample) or long-lived photon-emitting radionuclides (generally <5 pCi/sample) were detected in these samples. The ^{90}Sr and ^{137}Cs in these samples are attributed to fallout.

Table 5.5
Gross Beta Activity and Concentrations of ^{90}Sr and ^{137}Cs in Suspended Solids from Surface Water in Sherman Reservoir

Sample	Water volume, liter	Ash wt., mg	Radionuclide content, pCi/sample		
			Gross beta	^{90}Sr	^{137}Cs
June 3, 1969, membrane filter					
near Yankee outlet	11.4	20.4	NA	1.4 ± 0.3	< 0.5
July 19, 1969, plankton net					
near Yankee outlet	~400	60.1	4.0	< 0.2	1.4 ± 0.5
bay east of pump house	~400	12.7	1.4	< 0.3	0.8 ± 0.4
near Harriman Station outlet	~400	233.	9.0	< 0.2	1.6 ± 0.4

Notes:
1. NA - not analyzed.
2. ± values are 2 σ error of counting; < values are 3 σ error of counting.

5.4 Radionuclides in Vegetation

5.4.1 *Sampling and analysis.* Dead leaves that were barely submerged at the edge of Sherman Reservoir near location No. 2 (see Figure 5.2) were collected on June 2, 1969. Common water moss, Fontinalis sp., * was collected on June 3, 1969 from rocks at a depth of 2.5 m near the shore at location No. 23 E (in front of Yankee outlet weir -- see Figure 5.3). The samples were weighed while wet, after drying at 100°C, and again after ashing at 400°C. Both samples contained silt.

The ashed samples were analyzed with a Ge(Li) detector plus 1600-channel analyzer and with a NaI(Tl) detector plus 200-channel analyzer to identify and quantify photon-emitting radionuclides. Spectra obtained with the Ge(Li) detector are shown in Figures 5.4, 5.5, and 5.6. Radiochemical analysis was performed to measure ^{90}Sr, and to confirm the gamma-spectral identification of ^{137}Cs and ^{106}Ru. In addition, stable calcium and strontium were measured by atomic absorption spectroscopy, and the silica content was determined by gravimetric analysis.

5.4.2 *Results and discussion.* Longer-lived fission and activation products were detected in the two samples at the concentrations given in Table 5.6. The ^{60}Co in both samples and ^{54}Mn and ^{58}Co in moss are attributable to Yankee; all other radionuclides are

Table 5.6
Radionuclides in Water Moss and Dead Leaves from Sherman Reservoir, pCi/g ash weight

Radionuclide	Water moss	Dead leaves
^{54}Mn	26	2
^{58}Co	~ 4	NM
^{60}Co	13	4
^{90}Sr	4	3
^{95}Zr	12	< 4
^{95}Nb	18	NM
^{103}Ru	~12	NM
^{106}Ru	28	4
^{125}Sb	1	2
^{137}Cs	2	6
^{141}Ce	~ 9	NM
^{144}Ce	58	10
^{40}K	180	27
^{226}Ra	15	3
^{232}Th	~70	~6
^{7}Be	~38	NM
Sr^{++} (mg/g ash)	0.14	0.15
Ca^{++} (mg/g ash)	13.7	12.2
SiO_2 (mg/g ash)	124	278
Ash wt/wet wt, %	7.1	6.7
Dried wt/wet wt, %	14.5	15.5

Notes:
1. Water moss was collected on June 3, 1969, from rock near weir (location 23 E) at 2.5-m depth; dead leaves were collected on June 2, 1969, at water-line on shore of Sherman Reservoir (near location 2).
2. < values are 3σ counting error.
3. Concentrations pertain to collection time.
4. NM - not measured.
5. The following radionuclides were not detectable: ^{51}Cr (<15 pCi/g); ^{59}Fe (<5 pCi/g); ^{89}Sr and ^{110m}Ag (<1 pCi/g).

* We thank M. C. Palmer, Environmental Protection Agency, Cincinnati, for identifying the moss.

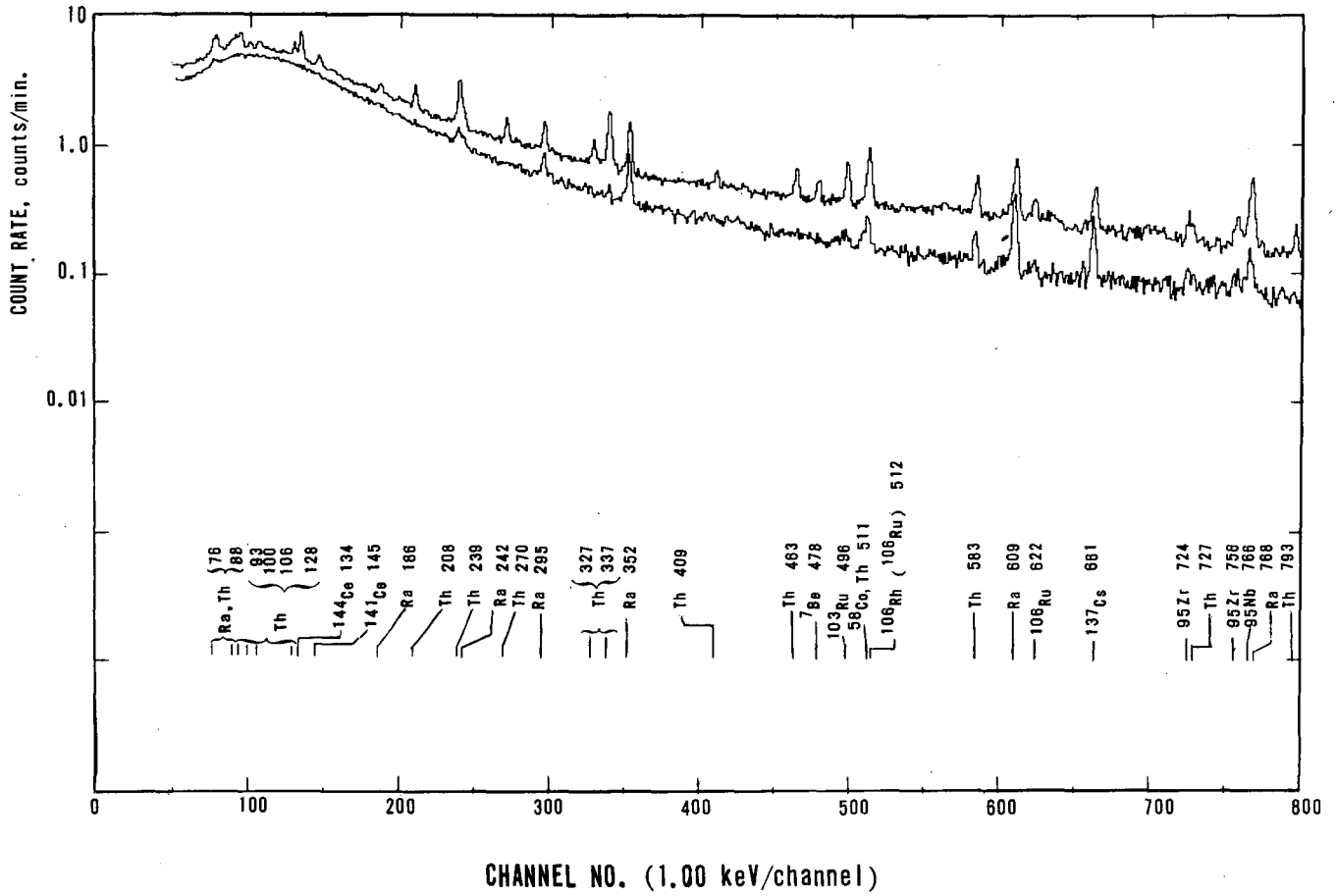

Figure 5.4. Gamma-ray Spectrum of Water Moss, 0 – 800 keV.

Detector: *Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal*

Sample : *18 g (35 cc) ash, collected June 3, 1969, in Sherman Reservoir near outlet for Yankee cooling water.*

Counts : *(upper curve) July 9–10, 1969 (1,000 minutes, background not subtracted); (lower curve) counter background; Ra and Th refer to ^{226}Ra and ^{232}Th plus progeny.*

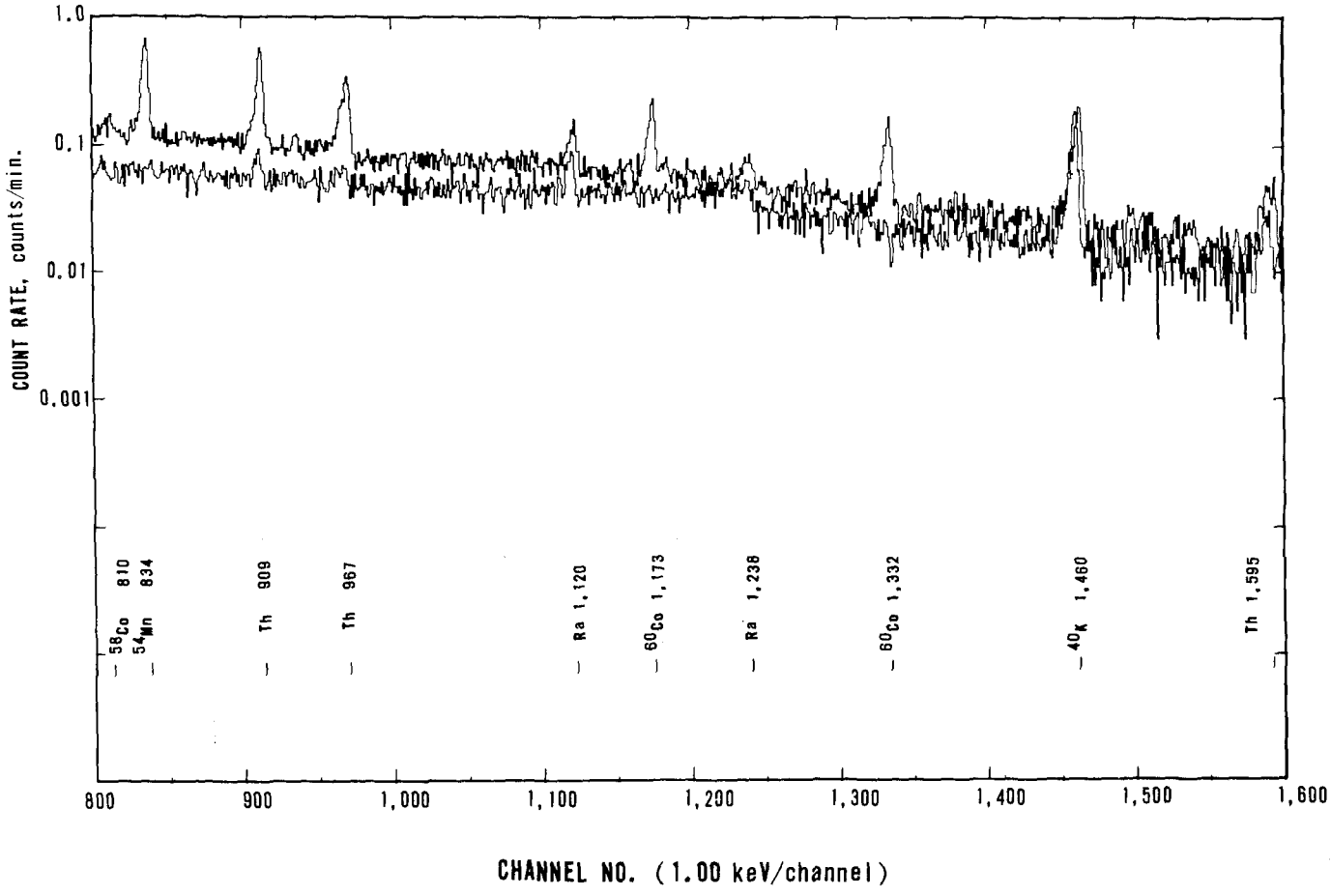

Figure 5.5. Gamma-ray Spectrum of Water Moss, 800 – 1,600 keV.

Detector: Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal

Sample : 18 g (35 cc) ash, collected June 3, 1969, in Sherman Reservoir near outlet for Yankee cooling water.

Counts : (upper curve) July 9–10, 1969 (1,000 minutes, background not subtracted); (lower curve) counter background; Ra and Th refer to ^{226}Ra and ^{232}Th plus progeny.

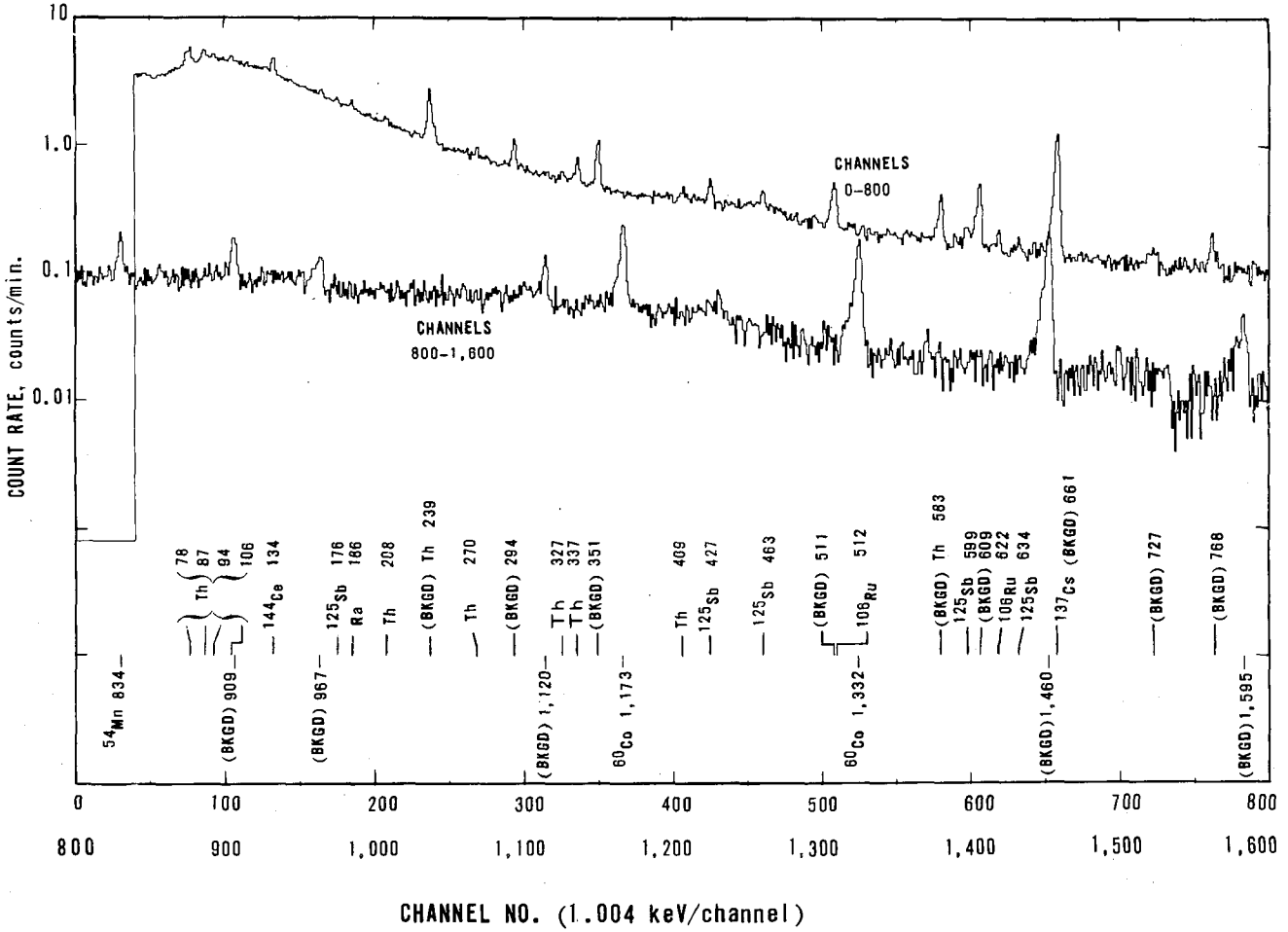

Figure 5.6. Gamma-ray Spectrum of Dead Leaves from Sherman Reservoir.

Detector: *Ge(Li), 10.4 cm^2 x 11 mm, trapezoidal*

Sample : *210 g (450 cc) ash, collected June 2, 1969, at east shore near 300-m perimeter.*

Count : *Nov. 12-13, 1969 (1,000 min.); Ra, Th, and Bkgd refer to ^{226}Ra plus progeny, ^{232}Th plus progeny, and counter background (see Figures 5.4 and 5.5), respectively.*

probably from fallout (see Table 6.2 for radionuclide content of vegetation samples collected on land) or occur naturally. The radionuclides may be in both the organic material and the accompanying silt (see Section 5.6 for the radionuclide content of sand, silt and clay in sediment samples). Because of its proximity to the outlet, the water moss would be expected to collect radionuclides discharged in circulating coolant water. The dead leaves were collected within 200 m of the east yard drain, and may have retained radionuclides from effluents at that drain.

In terms of wet weight, the concentrations of ^{54}Mn, ^{58}Co, and ^{60}Co in the moss and leaves range from 1×10^{-7} to 2×10^{-6} μCi/g (see Table 5.7). Accumulation factors for these radionuclides--defined as concentration in the media divided by the concentration in water--can not be calculated because neither the average radionuclide concentrations in water near the media nor the exposure periods of the media are known. The accumulation factors of ^{90}Sr and ^{137}Cs from fallout and stable strontium and calcium from Sherman Reservoir water are approximately 2000 in moss, as shown in Table 5.7.

Thus, despite the extremely low concentrations of radionuclides discharged by Yankee, some of these radionuclides could be detected in organic material in Sherman Reservoir, at concentrations considerably higher than in the water (see Table 4.2, last column). The moss was seen only near the discharge weir, and may be confined to that area because the water is colder everywhere else; dead leaves are found at many locations near the edge of Sherman Reservoir. Concentration of radionuclides in these media does not appear to have any consequence as a health hazard to humans through consumption, external radiation, or return of radionuclides to water after concentration. In future studies, it would be of interest to analyze these media again, both at the indicated sites and at background locations, to check attribution of the noted radionuclides to Yankee, and to examine the use of these media as convenient indicators of discharged radionuclides.

5.5 Radionuclides in Fish

5.5.1 *Collection and analysis.* Fish were collected on June 18, 1969, from both the Sherman and Harriman Reservoirs by the electro-shocking method.† As shown in Figure 5.1, the two reservoirs are well separated, hence movement of fish between them is unlikely and the fish from Harriman Reservoir can serve as the background sample.

The collected fish are listed in Table 5.8. The fish from each reservoir were combined in three categories according to their feeding habits: bottom feeders, insect eaters and predators. Catfish, a bottom feeder, were analyzed separately since this type of fish was available from both reservoirs. Also listed are the numbers of fish, total wet weight, and age as determined by annular scale marks. Ages of the crappie and catfish are unknown.

Table 5.7

Radionuclide Concentration in Water Moss and Dead Leaves

	Water moss		Dead leaves
Substance	**Amount per wet wt.**	**Accumulation factor***	**Amount per wet wt.**
^{54}Mn (μCi/g)	1.8×10^{-6}	---	1.4×10^{-7}
^{58}Co	3×10^{-7}	---	---
^{60}Co	9×10^{-7}	---	3×10^{-7}
^{90}Sr	3×10^{-7}	9×10^{2}	2×10^{-7}
^{137}Cs	1.4×10^{-7}	3×10^{3}	4×10^{-7}
Sr^{++} (μg/g)	9.9	1.6×10^{3}	10.0
Ca^{++} (mg/g)	0.97	1.4×10^{3}	0.82

*Calculated by dividing values in preceding column by concentration in water. Concentrations in water for ^{90}Sr and ^{137}Cs are average values from analysis of ion-exchange resin in Table 4.2; for Sr^{++} and Ca^{++}, values are from Table 5.1.

† We thank Colton H. Bridges and associates, Bureau of Wildlife Research and Management, Division of Fisheries and Game, State of Massachusetts, for collecting these samples and providing data on fish ages.

Samples were frozen immediately after collection. For analysis, the fish were thawed, weighed, and dissected into the following tissues that were expected to concentrate the radionuclides of interest:

muscle	- ^{134}Cs and ^{137}Cs analysis
kidney + liver	- ^{55}Fe, ^{58}Co and ^{60}Co analysis
bone	- ^{89}Sr and ^{90}Sr analysis

No analyses were performed for ^{131}I in the thyroid because of the lapse of time between sample collection and analysis.

Liver plus kidney were analyzed directly by gamma-ray spectrometry with a NaI(Tl) detector, and also with a NaI(Tl)-gamma-ray coincidence/anticoincidence spectrometer system. The iron fraction was separated, and analyzed for ^{55}Fe with an x-ray proportional detector, and for stable iron with an atomic absorption spectrometer.

Bone was ashed at 600°C, and strontium was then separated chemically. Radiostrontium was measured by counting total strontium and ^{90}Y. Stable strontium and calcium were determined by atomic

Table 5.8

Fish Collected in Sherman and Harriman Reservoirs

Reservoir	Category	Type	Total weight, kg (number)	Average age, yr (range)
Sherman	Bottom Feeder	White sucker	7.2 (11)	4.7 (2-8)
		Catfish, bull head	1.9 (17)	---
	Insect Eater	Rock bass	0.55 (16)	3.4 (2-7)
		Golden shiner	0.30 (3)	5.2 (2-7)
		Crappie	0.20 (4)	---
		Yellow perch	0.65 (19)	6.2 (2-9)
	Predator	Small mouth bass	1.3 (3)	4.8 (2-7)
Harriman	Bottom Feeder	Common sucker	3.9 (10)	3.6 (2-5)
		Catfish, bull head	0.65 (10)	---
	Insect Eater	Rock bass	1.3 (11)	5.8 (3-7)
		Yellow perch	1.10 (9)	4.7 (4-6)
		Lake trout	0.60 (1)	4
		Brown trout	0.50 (1)	3
	Predator	Chain pickerel	1.8 (2)	8.5 (8-9)

Table 5.9

Radionuclide (pCi/kg)[a] and Stable Ion (g/kg)[a] Concentration in Fish Tissue

	Bone				Muscle					
Category	^{90}Sr	Ca	Sr	Ash/wet weight	^{22}Na	^{137}Cs	K	Ca	Sr	Ash/wet weight
Sherman Reservoir										
Bottom Feeder-sucker	2230	32	0.059	0.10	3.1	250	3.42	0.59	0.0011	0.016
								0.35[b]	0.00064[b]	0.009[b]
Bottom Feeder-catfish	3510	36	0.091	0.11	3.1	120	2.82	0.86	0.0010	0.014
Insect Eater	3070	40	0.072	0.12	2.0	110	3.56	2.45	0.0019	0.013
Predator	2950	41	0.068	0.12	3.0	650	4.42	1.15	0.0011	0.018
Harriman Reservoir										
Bottom Feeder-sucker	2370	38	0.072	0.12	1.9	170	3.09	0.89	0.0021	0.012
Bottom Feeder-catfish	3530	36	0.091	0.11	NM	210	3.62	0.72	0.0020	0.026
Insect Eater	2320	32	0.058	0.11	0.5	520	3.60	0.96	0.0010	0.027
Predator	2790	32	0.043	0.10	1.2	460	4.12	1.09	0.0005	0.014

[a]All kg values are wet weights.

[b]Bone was removed very thoroughly.

Notes:

1. ± values (2 σ counting errors) are: ^{90}Sr, 90 pCi/kg; ^{22}Na, 0.3 pCi/kg; and ^{137}Cs, 10 pCi/kg.
2. NM - not measured.

absorption spectroscopy.

Muscle was ashed at 400°C and then analyzed by gamma-ray spectrometry. Cesium-137 and ^{40}K in muscle were determined by gamma-ray spectrometry, and the potassium content was calculated from the ^{40}K measurement. A gamma ray at 0.51 MeV energy was observed by coincidence/anticoincidence spectrometry and the emitting radionuclide was identified as ^{22}Na by its photon spectrum and by chemical separation. Stable strontium and calcium were also measured in these samples. To evaluate the contribution of incompletely separated bone to the radiostrontium content in muscle, an additional sample--muscle of sucker from the Sherman Reservoir--was prepared for stable strontium and calcium analysis with special care to remove all bone.

5.5.2 *Results and discussion.* The ^{90}Sr and ^{137}Cs concentrations in fish were not consistently higher in Sherman Reservoir than in Harriman Reservoir (see Table 5.9), hence these radionuclides in all of the fish are attributed to fallout. The average ^{90}Sr concentration in bone for all fish was 2840 pCi/kg wet weight, 42 pCi/mg of strontium, and 79 pCi/g of calcium. The average ^{137}Cs concentration in muscle for all fish (adjusted for the number of fish in each category), was 235 pCi/kg wet weight, and 67 pCi/g of potassium. These concentrations fall within the range of previously reported values. (20-22) The average observed ratio for radiostrontium [$OR_{bone/water} = (Sr/Ca)_{bone} \div (Sr/Ca)_{water}$] was 0.17 ± 0.05 (2σ) based on a ^{90}Sr concentration of 0.32 pCi/liter of water (Table 4.2) and a calcium concentration of 0.7 mg/liter of water (Table 5.1). The average $OR_{bone/water}$ for stable strontium was 0.22 ± 0.06 (2σ). Published $OR_{bone/water}$ values are between 0.1 and 0.7. (15-18) Because the calcium content of the water is very low, the principal source of calcium retained by the fish may be food rather than water, (19) however, and use of the OR bone/water may be inappropriate.

In terms of the accumulation factor (AF)--the ratio of concentration per weight of fish to concentration in water (in Tables 4.2 and 5.1), the values for ^{90}Sr, strontium, calcium, and ^{137}Cs are as follows:

$$AF_{^{90}Sr} = 7.9 \times 10^3 \qquad AF_{Ca} = 5.2 \times 10^4$$
$$AF_{Sr} = 1.2 \times 10^4 \qquad AF_{^{137}Cs} = 4.7 \times 10^3$$

Reported accumulation factors in fish range from 200 to 8,500 for ^{90}Sr, (18,23) from 50 to 100,000 for strontium, (16,23,24) from 600 to 100,000 for calcium, (17) and from 122 to 15,000 for ^{137}Cs. (20-22)

Some of the differences in ^{90}Sr and ^{137}Cs concentrations among species and locations indicate the necessity for carefully matching test samples and background samples. For example, the higher ^{137}Cs concentration in predator fish than in most other categories, which has been observed previously, appears to result from these fish eating other fish.(22) The higher ^{137}Cs concentration in the nominal insect eaters from Harriman Reservoir may be due to consumption of other fish by the relatively large rock bass and perch in this category. These fish in Sherman Reservoir were lighter (see Table 5.8), hence probably true insect eaters. The higher concentrations of ^{90}Sr and strontium in catfish from both reservoirs should also be noted.

The ^{90}Sr concentration in muscle was not measured directly, but was estimated by multiplying the ^{90}Sr/strontium ratio in bone by the stable strontium concentration in muscle (i.e., assuming that the ratio of ^{90}Sr to strontium was the same in bone and muscle). To correct for strontium in muscle from some fine bones that were not readily removed by the routine dissection procedures, the measured strontium concentrations in muscle were multiplied by 0.6, based on a strontium analysis of muscle from which bone had been thoroughly removed (see footnote b to Table 5.9). After this correction for thorough separation of bone from muscle had been applied, the average concentration of strontium in muscle was computed to be 0.011 of that in bone. By comparison, an average concentration ratio of 0.024 was calculated from date for white crappie in the Clinch River. (25) At the ratio of 0.011, an average ^{90}Sr concentration of 2840 pCi/kg in fish bone would correspond to 32 pCi/kg in muscle.

Concentrations of ^{22}Na in muscle ranged from 0.5 to 1.9 pCi/kg wet weight in Harriman Reservoir and from 2.0 to 3.1 pCi/kg in Sherman Reservoir. The difference may be attributable to Yankee, although no ^{22}Na was detected in any liquid samples from the station. The mean concentration in all fish--2.1 ± 0.4 pCi/kg--is one-half of the value reported for bass collected in the Columbia River in 1964, (26) and similar to the values reported in the muscle of salmon collected near the coast of the state of Washington in 1967. (27)

No ^{89}Sr (<60 pCi/kg wet weight) was detected in fish bone, and no longer-lived photon-emitting radionuclides except ^{137}Cs, ^{22}Na, and naturally

occurring ^{40}K were found in any sample. The minimum detectable level for ^{134}Cs in muscle was 2 pCi/kg, and for radiocobalt in kidney plus liver, 15 pCi/kg. The iron content of the kidney plus liver samples ranged from 0.04 to 0.26 g/kg; but no ^{55}Fe (< 400 pCi/kg wet weight, < 10 pCi/mg Fe) was found. Concentrations of ^{55}Fe between 3 and 50 pCi/mg iron have been reported for freshwater fish collected in Finland during 1965. (28)

In summary, the only radionuclide in fish that might be attributable to Yankee was ^{22}Na at a concentration above background of approximately 2 pCi/kg wet weight. Since there is fishing in Sherman Reservoir, however, it appears reasonable to check radionuclide concentrations periodically in the edible portions of food fish.

5.5.3 *Hypothetical radionuclide concentration in fish.* The concentrations of radionuclides in fish exposed to radioactive effluent from Yankee was computed to demonstrate the procedure and indicate possible critical radionuclides:

Radio-nuclide	Annual average concentration in water, * μ Ci/ml	Concentration factor, (29) ml/g	Hypothetical Concentration in fish, + μCi/100 g wet wt.	Percent of intake guide**.
^{3}H	3×10^{-6}	0.9	3×10^{-4}	0.005
^{14}C	4×10^{-11}	4,550	2×10^{-5}	0.001
^{24}Na	(1×10^{-8})	31.7	(3×10^{-5})	(0.05)++
^{32}P	3×10^{-13}	100,000	3×10^{-6}	0.007
^{51}Cr	7×10^{-11}	200	1×10^{-6}	< 0.001
^{54}Mn	4×10^{-11}	25	1×10^{-7}	< 0.001
^{55}Fe	4×10^{-11}	300	1×10^{-6}	< 0.001
^{59}Fe	1×10^{-12}	300	3×10^{-7}	< 0.001
^{58}Co	4×10^{-11}	500	2×10^{-6}	0.001
^{60}Co	7×10^{-11}	500	4×10^{-7}	0.001
^{63}Ni	4×10^{-12}	40	2×10^{-8}	< 0.001
^{64}Cu	(2×10^{-10})	200	(4×10^{-6})	(0.001)
^{90}Sr	3×10^{-13}	40	1×10^{-9}	< 0.001
^{95}Zr	1×10^{-11}	100	1×10^{-7}	< 0.001
^{95}Nb	1×10^{-11}	30,000	3×10^{-5}	0.01
^{99}Mo	(5×10^{-11})	100	(5×10^{-7})	<(0.001)
^{110m}Ag	4×10^{-12}	3,080	1×10^{-6}	0.002
^{124}Sb	7×10^{-12}	40	3×10^{-8}	< 0.001
^{131}I	1×10^{-11}	1	1×10^{-9}	< 0.001
^{133}I	(3×10^{-10})	1	(3×10^{-8})	(0.001)
^{135}I	(3×10^{-10})	1	(3×10^{-8})	(<0.001)
^{137}Cs	7×10^{-13}	1,000	7×10^{-8}	< 0.001

*The estimated annual discharge (Section 4.3.5) divided by the flow of circulating coolant water of 2.8×10^{14} ml/yr.

+The product of the values in columns 2 and 3, multiplied by the estimated intake of 100 g/day.

**The limiting concentrations from Section 4.3.5 multiplied by the water intake of 2,200 ml/day on which the concentration limits are based, (30) except for limits for ^{90}Sr (200 pCi/day) and ^{131}I (80 pCi/day) from Federal Radiation Council guidance. (31)

++Values in parentheses are based on inferred, not measured, concentrations.

This tabulation is based on an average daily intake of 100 g fish (values of 50 g (32) and 100 g (33) have been reported), a tabulation of concentration factors for edible portions of freshwater fish, (29) the radionuclide release estimates in Section 4.3.5, and the assumption that the radionuclides in the edible portions of all consumed fish had reached equilibrium with radionuclide concentrations in circulating coolant water at the point of discharge. Of these, the radionuclide release estimates and many of the concentration factors are quite approximate, and it is

improbable that radioactive equilibrium is attained in all fish.

The total estimated intake of radionuclides by eating fish is, therefore, below 0.1 percent of the intake guide. The dose rates from the listed radionuclides are 0.3 mrem/yr to the gastrointestinal tract (mostly from ^{95}Nb), 0.2 mrem/yr to bone (from ^{32}P), and less than 0.1 mrem/yr to the thyroid and whole body. These values were computed by comparing the hypothetical daily intakes (column 4, above) to the maximum permissible daily occupational drinking-water intakes listed by the NCRP that correspond to 5 rem/yr to the total body, 15 rem/yr to the GI tract, and 30 rem/yr to bone, (30) or directly applying FRC guidance for radiostrontium and radioiodine. (31)

Of the listed radionuclides, ^{3}H, ^{24}Na, and ^{95}Nb would be readily detected in fish muscle at the indicated concentrations, and should be looked for in fish samples from Sherman Reservoir near the Yankee outfall or the Deerfield River below Sherman Dam. In the analyzed samples (which were collected throughout Sherman Reservoir, however), no ^{95}Nb was found ($< 2 \times 10^{-6}$ μCi/100 g wet weight).

The average concentrations given in Section 5.5.2 of ^{22}Na and ^{137}Cs measured in fish muscle, and of ^{90}Sr in muscle inferred from fish bone analyses, correspond to the following annual radiation exposure at a daily fish consumption of 100 g:

Radionuclide	Average concentration in fish, μCi/100 g	Radiation dose, mrem/yr	Critical organ
^{22}Na	2.1×10^{-7}	0.005	GI tract (30)
^{90}Sr	3.2×10^{-6}	2.7	bone (31)
^{137}Cs	2.4×10^{-5}	0.3	whole body (31)

As indicated in Section 5.5.2, the ^{90}Sr and ^{137}Cs are attributed to fallout, but most of the ^{22}Na may be from Yankee.

5.6 Radionuclides in Benthal Samples

5.6.1 *Sampling and on-site measurements.* The MDPH in 1965 found radionuclides from Yankee in at least one of five benthal samples collected in Sherman Reservoir. An effort was therefore made to confirm this observation and to evaluate the extent of the contamination. Three methods of determining radioactivity in benthal samples were compared with respect to sensitivity and ability to define the extent and magnitude of the contamination. The methods were:*

(1) Use of a 10-cm x 10-cm NaI(Tl) detector as submergible probe; gamma-ray spectra were obtained for 4 - to 20-minute periods while the detector rested on the bottom of Sherman Reservoir.

(2) Collection of core samples by a diver; the core sample, 10 cm in diameter and 13.4 cm in depth, was separated into equal upper and lower fractions and analyzed for radionuclide content.

(3) Collection of samples with an Ekman or a Petersen dredge, and analyses for radionuclide content.

The location and number of probe measurements and samples are given in Table 5.10. Samples were collected from a boat by lowering the diver into the water or dropping a dredge to the Reservoir bottom; the probe was lowered from a second boat which contained the multichannel analyzer with associated power supply (motor-generator) and recording system. The probe was positioned on the Reservoir bottom by the diver, who later collected benthal samples by hand at the same location. Measurements were taken and/or samples collected at three locations across the Reservoir near the 300-m perimeter relative to the containment sphere; at 7 points along the south shore of the Reservoir near

*We thank the MDPH, SERHL, and NERHL for making this study possible; especially Cornelius J. O'Leary, MDPH, for providing equipment and guiding the sampling, Edward Crockett, MDPH, for performing the diving, Charles Phillips, SERHL, for providing and operating the underwater probe, and Raymond H. Johnson, Jr., NERHL, for providing sample collection equipment and advising on sampling procedures.

Yankee; at 2 locations in front of the Yankee circulating coolant water outlet; at the north end of the Reservoir to indicate background values; and at one location in the Deerfield River below the Reservoir. Brief probe measurements were obtained in the relatively shallow water along the south shore of the Reservoir until the area of highest radionuclide concentration was identified. In that area, 10 probe measurements and 5 samples were taken to define the extent of the contamination and the response of the probe.

5.6.2 *Description of benthal samples.** Five of the samples were characterized as shown in Table 5.11. (34) In brief, organic carbon was determined by measuring the carbon dioxide formed in ashing the samples, and the weight of organic matter was estimated by multiplying the organic carbon content by 1.72. Particle-size separation was by wet sieving and sedimentation (for clay). Cation-exchange capacity was determined by saturating the sample with sodium acetate. Mineral constituents were identified by x-ray crystallographic analysis of preferred-oriented aggregation specimens prepared on ceramic plates.

The background sample (No. 20) and sample No. 24 are sandy, while samples No. 19 and 25 are loamy

Table 5.10
Benthal Sampling Points

	Approximate location		Number of samples		
#*	Depth, m	Distance from shore, m	Probe measurement	Hand-collected	Dredge-collected+
		Sherman Reservoir near 300-m station perimeter. June 2, 1969			
1	21	140	1	2	2
2	2.5	20	1	2	2
3	2	20	1	1	2
		Sherman Reservoir near south shore, June 3, 1969			
4	2.5	11	1	0	0
5	2.5	10	1	0	0
6	3	6	1	0	0
7	4	9	1	0	0
8	14	15	1	0	0
9	9	9	1	0	0
10	3	6	2	0	0
11	7	9	1	0	0
12	7	12	1	0	0
13	5	6	1	0	0
14	7	9	1	0	0
15	6	9	1	2	1
16	6	10.5	1	2	1
17	7.5	9	1	2	1
18	6	7.5	1	2	1
19	4.5	9	1	2	1
		Sherman Reservoir north of outlet weir, June 3, 1969			
24	12	30**	0	0	1
25	20	60**	0	0	1
		Sherman Reservoir north of Harriman Station, June 4, 1969			
20	1.5	10	1	2	1
		Deerfield River west of Charlemont, June 4, 1969			
21	0.2	0.5	0	1	0

* numbers refer to locations in Figures 5.1, 5.2 and 5.3.

+ samples # 1-3 and 15-20 were collected with an Ekman dredge; # 24 and 25 were collected with a Petersen dredge.

** distances are from Yankee circulating water outlet along centerline.

*We thank Profs. Clyde R. Stahnke and Larry Wilding, Agronomy Dept., Ohio State University, for performing these analyses.

and have much higher fractions of silt, clay, and organic material. Samples No. 1 to 3 and No. 15 to 18 also appear to be loamy, while sample No. 21 is sandy. The loamy samples showed a cation-exchange capacity of approximately 16 milliequivalents per 100 gram (meq/100 g) and the sandy samples, 2 meq/100 g. The two samples that were examined in detail consisted mostly of illite, with some vermiculite, quartz, and kaolinite. Stahnke's estimate of the contribution of the various components to the total cation-exchange capacity of sample No. 19, hand, top, is in agreement with the measured value of 16.8 meq/100 g:

Component	Approximate capacity, meq/100 g	Fraction of total sample	Contribution to total capacity, meq/100 g
organic matter	200	0.050	10
allophane (amorphous constituents)	150	0.44 x 0.071 x 0.95	4.5
illite	40	0.39 x 0.071 x 0.95	1.1
vermiculite	150	0.08 x 0.071 x 0.95	0.8
			16.4

The listed cation-exchange capacities are commonly used values. The contributions to the total capacity are the products of the individual capacities and the sample fraction, obtained from Table 5.11. The contribution by the fractions of quartz and kaolinite in the analyzed samples was considered negligible.

5.6.3 *Analysis.* The spectra obtained with the probe were plotted as shown in Figure 5.7, and the gross count rates of ^{137}Cs and ^{60}Co were obtained at the energy ranges of 0.63 - 0.71 MeV and 1.10 - 1.40 MeV, respectively. The ^{60}Co count rate of each spectrum was the difference between the gross

Table 5.11

Mineralogical Analysis of Benthal Samples*

Number :	19	19	20	24	25
Collected by :	hand	hand	dredge	dredge	dredge
Core fraction :	top	bottom			
Texture	loam	sandy loam	sand	sand	silt loam
	Organic material, % of total dried weight				
Organic carbon	2.92	2.88	0.23	0.46	2.92
Organic matter	5.02	4.95	0.40	0.79	5.02
	Particle size distribution, % of total mineral weight				
Clay (<2 μdia.)	7.1	6.9	2.0	1.2	6.4
Silt (2-50 μdia.)	48.4	43.4	1.1	4.1	52.9
Sand (50-2,000 μdia.)	44.5	49.7	96.9	94.7	40.7
	Cation exchange capacity, meq/100 g of individual fraction				
Total	16.8	14.8	1.83	2.44	16.4
Clay & organic material+	143	129	77	123	148
Clay**	94	71	51	72	99
	Clay mineral, % of total clay++				
Illite (mica)	55 (39)	---	---	---	60 (41)
Vermiculite	10 (8)	---	---	---	15 (8)
Quartz	25 (6)	---	---	---	20 (6)
Chlorite	<5 (<3)	---	---	---	<5 (<3)
Kaolinite	10 (3)	---	---	---	5 (3)

* By C.R. Stahnke and L. Wilding, Agronomy Dept., Ohio State University.

\+ The cation-exchange capacity was assumed to be entirely due to clay (including allophane) and organic material.

** A cation-exchange capacity of 200 meq/100 g was assumed for organic material.

++ Values in parentheses are percentages of total clay that are crystalline; the amorphous constituents were removed with 0.5 M NaOH.

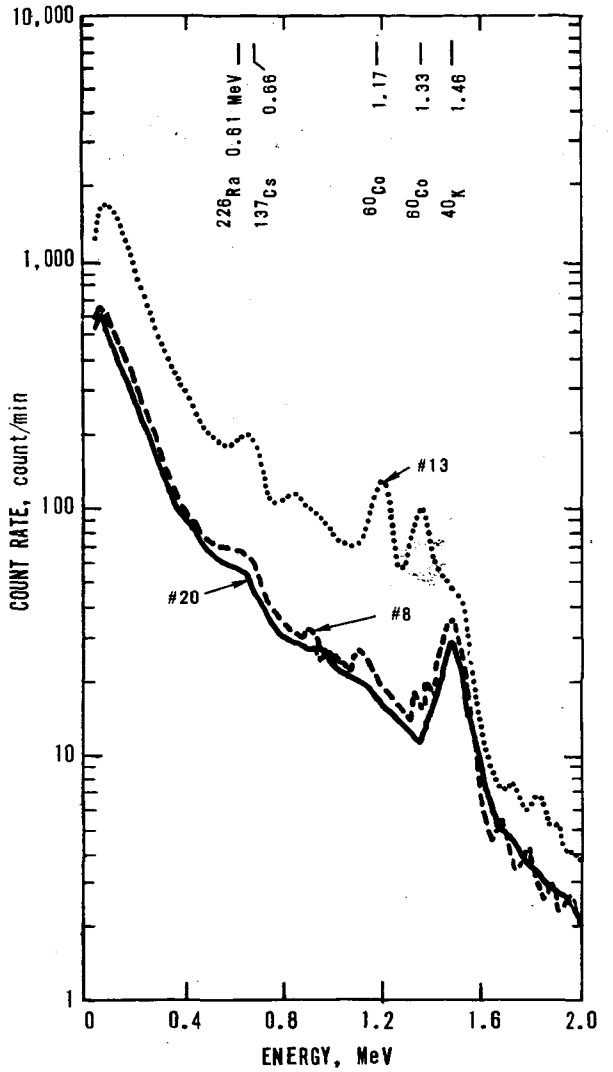

Figure 5.7. Gamma-ray Spectra of Bottom of Sherman Reservoir.

detector: *10 × 10-cm. NaI(Tl) Probe*

location: *see Table 5.10*

counted : *June 3, 1969.*

reading in this energy range and the value at the background location (No. 20). The background location showed gamma rays of naturally occurring ^{40}K, ^{226}Ra plus progeny, and ^{232}Th plus progeny, and also ^{137}Cs from fallout. To calculate the count rate of ^{137}Cs in each spectrum, (1) the background (No. 20) spectrum was subtracted, (2) the low count rate attributed to ^{137}Cs at the background location (50 c/m) was added, and (3) the Compton continuum attributed to ^{60}Co on the basis of the net count rate of ^{60}Co and a typical ^{60}Co spectrum was subtracted. At locations No. 1 and 3, the count rates in the energy region of the ^{137}Cs gamma ray were actually lower than in sample No. 20.

The benthal samples were either placed directly into sample containers or were first separated in the field with a U. S. No. 10 sieve (2-mm-dia. mesh). At the laboratory, the samples were air dried and thoroughly mixed. The samples were analyzed gamma-spectrometrically with a NaI(Tl) detector and 200-channel analyzer as shown in Figure 5.8. Three of the samples were separated with a standard No. 270 sieve (53-μ-dia. mesh) into sand and silt plus clay fractions. They were further separated into silt and clay fractions by extracting the clay into water as a

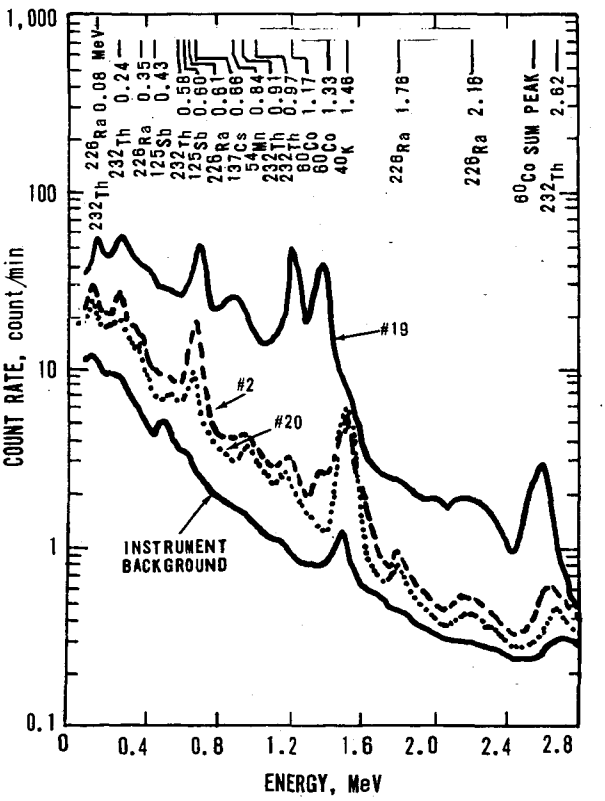

Figure 5.8. Gamma-ray Spectra of Benthal Samples from Sherman Reservoir.

detector: *10 × 10-cm NaI(Tl)*

samples : *#2, 89 g; #19, 101 g; #20, 137 g.*

counts : *#2, June 26, 1969 (200 min)*
#19, Aug. 14, 1969 (100 min)
#20, Nov. 12, 1969 (1000 min).

suspension in 11 successive extractions. Sodium carbonate was added as flocculating agent and the pH was adjusted to a value of 9. (35) These separated samples were also analyzed by gamma-ray spectrometer.

Some of the samples were analyzed in duplicate for ^{90}Sr content by leaching strontium from 10-g portions of the benthal material, separating it chemically, and counting radiostrontium and ^{90}Y with a low-background G-M counter. (36) Sample No. 17 was analyzed radiochemically for antimony to confirm the ^{125}Sb results obtained by gamma-ray spectrometry. Several of the samples were also analyzed with a Ge(Li) detector and 1600-channel spectrometer to identify the photon-emitting radionuclides through precise measurement of characteristic gamma-ray energies (± 1 keV). This was especially necessary for ^{54}Mn and ^{125}Sb, whose

Table 5.12

Concentration of Radionuclides in Benthal Samples from Sherman Reservoir and Deerfield River

#	Sample Collection	Weight/volume of analyzed sample, g/cc	Concentration, pCi/g dried weight							
			^{60}Co	^{137}Cs	^{90}Sr	^{54}Mn	^{125}Sb	^{40}K	^{226}Ra	^{232}Th
1	hand, top	46/100	1.9	3.3	---	0.7	< 0.1	15	0.9	0.8
	hand, lower	85/100	< 0.1	0.7	---	< 0.1	< 0.1	19	1.4	0.8
	dredge, s	350/400	1.2	4.5	0.6	0.3	0.4	16	0.6	1.2
	dredge, u	52/100	0.8	3.4	---	< 0.1	< 0.1	12	0.6	0.7
2	hand, top	89/100	0.6	3.6	---	< 0.1	0.4	18	0.9	0.8
	hand, lower	71/100	0.2	1.8	---	< 0.1	< 0.1	18	1.2	0.6
	dredge, s	300/400	1.0	5.0	0.4	0.3	0.3	19	1.0	0.9
	dredge, u	265/400	1.0	4.3	---	0.2	0.2	17	0.9	0.9
3	hand	640/400	< 0.1	0.2	0.1	< 0.1	0.2	15	0.5	0.6
	dredge, s	144/100	0.3	1.2	---	0.2	0.3	16	0.9	0.7
	dredge, s	122/100	0.5	0.8	---	0.2	0.3	16	0.8	0.4
	dredge, p	142/100	< 0.1	0.3	---	< 0.1	< 0.1	18	1.1	0.8
15	hand, top	416/400	6.0	3.4	---	0.4	0.5	21	0.9	0.9
	hand, lower	107/100	6.0	4.9	---	0.5	0.9	20	1.2	0.7
	dredge, u	118/100	4.6	3.2	0.2	0.3	0.6	20	1.0	0.7
16	hand, top	493/400	0.9	1.4	---	< 0.1	0.3	21	0.7	0.7
	hand, lower	428/400	0.5	1.4	---	< 0.1	0.2	22	0.9	0.6
	dredge, u	500/400	7.0	4.6	---	0.4	0.7	20	1.0	0.8
17	hand, top	396/400	4.2	4.3	---	0.3	0.6	20	0.7	0.8
	hand, lower	412/400	1.5	3.4	---	0.1	0.6	22	0.7	0.9
	dredge, u	417/400	10.6	5.7	0.2	0.5	0.7	23	0.9	1.4
18	hand, top	440/400	0.5	1.4	---	< 0.1	0.2	23	0.8	0.8
	hand, lower	500/400	0.1	1.0	---	< 0.1	0.2	25	0.8	0.8
	dredge, u	384/400	32.0	5.2	0.2	2.0	0.5	18	0.7	1.0
19	hand, top	101/100	20.2	6.4	0.1	1.5	0.8	21	0.8	0.9
	hand, lower	103/100	18.6	6.1	---	0.9	0.9	19	1.5	0.9
	dredge, u	505/400	9.1	3.7	---	0.8	0.5	18	1.0	0.9
20	hand, top	593/400	< 0.1	0.4	---	< 0.1	< 0.1	10	0.5	0.5
	hand, lower	600/400	< 0.1	0.8	---	< 0.1	0.2	14	0.4	0.4
	dredge, u	560/400	< 0.1	0.5	0.1	< 0.1	< 0.1	14	0.5	0.4
21	hand	600/400	< 0.1	0.3	0.1	< 0.1	0.2	11	0.6	0.4
24	dredge, s	674/400	1.8	2.5	0.1	0.4	0.4	14	0.5	0.6
25	dredge, s	350/400	1.6	4.2	0.4	0.2	0.6	22	0.9	1.0

Notes:

1. Sample collection definitions:
   - hand = 10-cm-dia core collected by hand
   - top = 0 cm to 6.7 cm from surface
   - lower = 6.7 cm to 13.4 cm from surface
   - u = unscreened
   - s = passed through US sieve #10
   - p = retained on US sieve #10
2. 2 σ values are approximately ± 0.1 pCi/g for ^{54}Mn, ^{60}Co, ^{125}Sb, and ^{137}Cs; ± 0.02 pCi/g for ^{90}Sr; ± 0.3 pCi/g for ^{226}Ra and ^{232}Th; and ± 1 pCi/g for ^{40}K; < values are 3 σ counting error.
3. 1 pCi/g = 1 x 10^{-6} μCi/g.

gamma rays could not be as clearly identified by the NaI(Tl) spectrometer as those of ^{60}Co and ^{137}Cs.

The concentrations of photon-emitting radionuclides were computed from count rates accumulated in 100- and 300-min periods. Calibration curves had been established with 100- and 400-cc solutions of standardized radionuclides at specific gravity 1.00. At higher specific gravity (1.25 - 1.75), the results were multiplied by the factor 1.1 to correct for the observed lower counting efficiencies. The ^{226}Ra and ^{232}Th values were computed on the assumption that radioactive progeny were in equilibrium.

5.6.4 *Results and discussion of sample analyses.* The ^{90}Sr and gamma-ray spectral results summarized in Table 5.12 show ^{60}Co and ^{137}Cs attributable to Yankee operations at every sample location in the southern end of Sherman Reservoir. The background sample from the north end of Sherman Reservoir (No. 20) contains ^{137}Cs attributed to fallout at concentrations of 0.4 to 0.8 pCi/g, but no ^{60}Co (< 0.1 pCi/g). The radionuclide content of sample No. 21 collected in the Deerfield River at DRM 27 is similar to that of the background sample. The highest concentrations of ^{60}Co and ^{137}Cs are in samples No. 18 and 19, in the small bay east of the pump house. The highest concentration of radionuclides was found at the same location by MDPH in 1965.

The samples collected at the south end of Sherman Reservoir also contained relatively low concentrations of ^{54}Mn, ^{90}Sr, and ^{125}Sb. All three radionuclides occur in fallout, but the background values in sample No. 20 suggest that they are from Yankee if their concentrations are considerably larger than 0.1 pCi/g. Sample No. 20 provides an appropriate background only for samples No. 21 and 24, however, because these three are set apart by their relatively sandy nature, as reflected in their high specific gravity and low concentrations of naturally occurring radionuclides. For all other samples, No. 1-hand-lower (see Table 5.12) may serve as background: its radionuclide concentrations are lowest among these samples, and similar to the concentrations in sample No. 20. The concentration of ^{90}Sr from fallout on land ranged from 0.1 to 1.5 pCi/g soil, and that of ^{54}Mn was approximately 0.1 pCi/g soil (see Section 6.4).

Radionuclide concentrations were generally highest in the dredge samples, intermediate in hand-top samples, and lowest in hand-lower samples. The differences are in most cases not large, and are reversed in a few samples. The largest differences between the hand-top and hand-lower concentrations occur at locations No. 1 and 2, where relatively little radioactivity was found in the lower sample. The largest differences between dredge- and hand-collected samples are at locations No. 16, 17 and 18, where the dredged samples contained approximately an order of magnitude more ^{60}Co. These values suggest that radionuclides attributable to Yankee are dispersed throughout the bottom deposits at the south end of Sherman Reservoir, even below the depth of 6.7 cm, but that concentrations are highest near the surface.

Collection by hand appears preferable in view of the better sample definition as to location and depth than for a dredged sample. In Sherman Reservoir, however, the dredged samples provided the most sensitive indication of radioactivity on the bottom, possibly because they contained mostly the surface of the sediment.

The benthal samples (see Section 5.2.1) collected by the Sanitary Engineering Division, MDPH, on November 2, 1965, were taken at the following locations:(2)

(1) middle of the reservoir, 100 m S of the Vermont state line;

(2) 50 m from the east shore, 300 m S of the Vermont state line;

(3) 50 m from the west shore and 500 m upstream from the dam;

(4) 50 m from the east shore and 500 m northeast of the condenser coolant discharge;

(5) 10 m from the south shore and 100 m east of the dam.

The last of these locations is in the same general area as locations No. 7 and No. 11-19 in the present study. The MDPH samples were dried, and analyzed by NaI(Tl) gamma-ray spectrometry with an 11-isotope matrix at NERHL.

Results were as follows:(2)

MDPH Benthal Samples of Nov. 2, 1965, pCi/g

	Station #5	Station #1-4	Possible origin
^{60}Co	2.8	ND* - 0.1	Yankee
^{137}Cs	6.5	ND - 2.5	fallout + Yankee
^{54}Mn	1.9	ND - 0.4	fallout + Yankee
^{65}Zn	ND	ND - 0.05	not significant
^{106}Ru	6.4	0.2 - 2.4	fallout + Yankee
^{125}Sb	2.0	0.07 - 0.9	fallout + Yankee
^{134}Cs	0.3	ND - 0.1	Yankee
^{144}Ce	7.9	0.7 - 3.7	fallout + Yankee
^{40}K	17	14. - 20.	natural
^{226}Ra	0.9	0.5 - 0.8	natural
^{232}Th	5.1	3.5 - 5.4	natural

*ND: not detected

Compared to the measurements near MDPH station No. 5, values at location No. 15 in this study are similar for ^{60}Co, ^{137}Cs, ^{40}K, and ^{226}Ra, and somewhat lower for ^{54}Mn, ^{125}Sb, and ^{232}Th. The radionuclides ^{106}Ru and ^{144}Ce were detected in 1965, but not in this study, possibly because these radionuclides decayed in the 3.5-year interval between measurements and were not replaced. The radionuclides ^{65}Zn and ^{134}Cs were very low or undetectable in 1965, and undetectable in 1969. Radionuclides measured in 1965 at concentrations above 0.1 pCi/g at MDPH stations No. 1 to 4 are all attributable to fallout or naturally occurring radioactivity, while values of 0.1 pCi/g or less are highly uncertain.

The benthal samples collected by Yankee staff for analysis by their contractor for environmental surveillance at 3 locations in Sherman Reservoir, 2 locations in No. 5 Reservoir just below Sherman Dam, and 9 downstream locations in the Deerfield River between Mohawk Park and Red Mill Dam, all contained the following radionuclides:(1)

Yankee Benthal Samples of Dec. 14-15, 1967, pCi/g

	Sherman Reservoir and #5 Reservoir	Deerfield River
^{54}Mn	0.5 - 1.4	0.2 - 0.6
^{60}Co	0.4 - 1.5	0.1 - 0.4
^{137}Cs	1.7 - 3.4	0.4 - 1.6
^{40}K	6 - 15	5 - 11
gross alpha	3.5 - 4.2	0.8 - 5.2

No ^{234}U, ^{235}U, ^{238}U, or ^{239}Pu was detected. The maximum concentrations of ^{54}Mn, ^{60}Co, and ^{137}Cs were in one sample from No. 5 Reservoir. Concentrations in the samples from the Deerfield River were very low, but showed a general downward trend with increased distance from Yankee.

These concentrations of ^{54}Mn, ^{60}Co, and ^{137}Cs in Sherman Reservoir are within the range of values measured in this study. The data by Yankee's contractor show that the three radionuclides discharged at Yankee were also deposited in No. 5 Reservoir, and possibly farther downstream in the Deerfield River.

5.6.5 *Distribution of radionuclides in benthal samples as function of particle size.* Three samples were separated into sand, silt plus clay, and clay fractions to observe the distribution of radionuclides among particle-size ranges (see Table 5.13). It had been noted in a tracer study with ^{65}Zn that the radionuclide concentration is relatively high in the fine fraction (< 43-μ dia.), and that radionuclide contents in a variety of samples are more readily comparable if the fine fraction is analyzed in each. (37)

The smaller particles generally, but not invariably, contained higher concentrations of the deposited fission and activation products. This trend suggests that samples containing a relatively high fraction of silt plus clay should be selected for more sensitive detection of radionuclides in sediment. For accurate background subtraction, the background samples must contain a similar particle-size distribution as the samples of interest. Radionuclide analysis of the clay fraction did not improve the analytical sensitivity, however, because only a small amount of clay was separated. Separation of silt plus clay from sand in wet media rather than after drying, as in this study, was recommended to fractionate the radionuclides more accurately.(38)

5.6.6 *Results and discussion of probe measurements.* The probe measurements identified ^{137}Cs at the background location (No. 20) at the level of 50 c/m, but did not detect any ^{60}Co, according to Table 5.14. In the southern end of Sherman Reservoir, ^{137}Cs at count rates distinctly above background was found at all locations except No. 1, 3, 4, 8, and 12, and ^{60}Co, at all locations except No. 3. The highest count rates, at locations No. 7, 11, and 13-19, coincided with the highest concentrations of ^{60}Co and ^{137}Cs in benthal samples (see samples No. 15-19 in Table 5.12). The probe data from this area indicate that count rates decrease toward the east, at locations No. 16 and 12, but do not clearly delineate the distribution of the radioactivity. Duplicate measurements at location No. 10 are consistent, but the three different values at locations No. 7, 11, and 15 (which were intended to be at the same spot) suggest the difficulty of locating exactly the same spot (note also the differences in recorded depth in Table 5.10).

The counting efficiencies of the probe for ^{60}Co and ^{137}Cs, given in Table 5.15 in terms of the ratio of probe count rate to benthal concentration, varied considerably among locations. This would be expected from an uneven vertical and horizontal distribution of radionuclides in the sediment. The averages in Table 5.15 may indicate the magnitude of the ratios of count rates to radionuclide concentrations.

The sensitivity of the probe is shown by the low and "less-than" readings in Table 5.14 as compared to

Table 5.13

Radionuclide Distribution in Dredged Benthal Samples as a Function of Particle Size, pCi/g dry weight

Radionuclide	#19 dredged			#24			#25		
	Sand	Silt & Clay	Clay	Sand	Silt & Clay	Clay	Sand	Silt & Clay	Clay
^{60}Co	2.6(12)*	14.9(88)	40(3)	0.8(50)	16.4(50)	27(2)	0.7(20)	1.3(80)	4.5(6)
^{137}Cs	4.5(52)	3.4(48)	10(2)	2.1(86)	7.2(14)	11(0.5)	3.4(31)	3.2(69)	14. (6)
^{90}Sr	NM	NM	NM	NM	NM	NM	NM	0.37	NM
^{54}Mn	0.2(15)	0.9(85)	< 3	0.2(65)	2.3(35)	3(1)	0.2(30)	0.2(70)	< 2
^{125}Sb	0.5(37)	0.7(63)	< 3	0.3(87)	0.9(13)	< 3	0.6(34)	0.5(66)	< 2
^{40}K	18. (41)	21. (59)	< 13	13. (93)	20. (7)	< 10	20.(29)	21. (71)	< 9
^{226}Ra	0.8(35)	1.2(65)	< 13	0.4(89)	1.0(11)	< 10	0.6(22)	0.9(78)	< 11
^{232}Th	0.8(35)	1.2(65)	< 13	0.4(82)	1.8(18)	< 10	0.6(20)	1.0(80)	< 2
Fraction by wt., separated**	0.45	0.55	0.0072	0.95	0.047	0.0010	0.30	0.70	0.013
analysis†	0.44	0.56	0.07	0.95	0.053	0.012	0.41	0.59	0.064
Particle diameter, mm	> 0.053	< 0.053	< 0.002	> 0.053	< 0.053	< 0.002	> 0.053	< 0.053	< 0.002

* Values in parentheses indicate the percent of the radionuclide that is in the indicated fraction.

† See Table 5.11 (values for #19 are taken from top hand sample).

** Sand, as retained on US sieve #270; silt and clay, as passing through US sieve #270; clay diameter as defined.

Note: NM - not measured.

the measured benthal concentrations in Table 5.12. Thus, the values at locations No. 1, 2, 3, and 20 suggest that ^{60}Co and ^{137}Cs at concentrations of 0.5 pCi/g are near the limit of detectability. Manganese-54 and ^{125}Sb in sample No. 19 were not clearly detected at concentrations near 1 pCi/g. The sensitivity of the probe can also be computed from the average ratios in Table 5.15 of approximately 500 and 80 c/min per pCi/g for ^{60}Co and ^{137}Cs, respectively, and the lowest significant count rates of 20 c/min. The resulting sensitivities of 0.04 and 0.25 pCi/g, respectively, appear too optimistic, however, especially for ^{60}Co.

The probe was found to be a highly convenient device for surveying the Reservoir bottom because results are immediately available to indicate the location and distribution of radioactivity. The probe measurements are more qualitative than quantitative, and their practical sensitivity is less by possibly a factor of five than gamma-ray spectral analysis of benthal samples in the laboratory. Hence, probe measurements are most useful if followed by sample collection and analysis.

5.6.7 *Significance of radioactivity in sediment.* The most important aspect of the radioactivity in the benthos was considered to be its detectability at the outfall of a station that discharges approximately one-hundredth as much radioactivity (other than ^{3}H) in liquid effluent as other full-scale commercial nuclear power stations. It is, nevertheless, desirable to consider the distribution, total amount, and health impact of the radionuclides at the bottom of Sherman Reservoir.

Three explanations have been suggested for the location of highest radionuclide concentrations to the east of the pump house peninsula: (1) radioactive material deposited at a location just beyond the turbulent water at the outlet to the west of the pump house; (2) benthal material with particularly high affinity for the radionuclides happens to be at that site; or (3) most of the radionuclides at that location were discharged from the nearby yard drain. No information is available to select the correct explanation. It may be noted that most samples at the site of highest radionuclide concentrations were unusual in that they contained more ^{60}Co than ^{137}Cs. The radionuclides can not be attributed to any single discharge of wastes, according to Yankee records; (12) the MDPH sampling indicates that they had been at the site of highest concentration for at least 3.5 years.

At a very approximate estimate, the amounts of ^{60}Co and ^{137}Cs in sediment at the southern end of Sherman Reservoir are as follows:

area, south end of Sherman Reservoir = 3×10^4 cm x 2×10^4 cm = 6×10^8 cm^2
depth of radioactivity in benthal material = 10 cm
density of benthal material = 1.1 g/cm^3
radionuclide concentration, ^{60}Co and ^{137}Cs = 1 pCi/g, each
total amount = $6 \times 10^8 \times 10 \times 1.1 \times 1 = 7 \times 10^9$ pCi = 7 mCi, each

Table 5.14

Net Count Rate of ^{60}Co and ^{137}Cs with NaI(Tl) Underwater Probe in Sherman Reservoir

Location	Counting time, min	^{60}Co, count/min	^{137}Cs, count/min
at 300-m perimeter			
# 1	20	40 ± 10	20 ± 10
2	10	60 ± 20	170 ± 10
3	10	< 20	< 20
within 300-m perimeter			
4	4	90 ± 30	60 ± 20
5	4	170 ± 30	140 ± 30
6	4	220 ± 30	160 ± 30
7	4	1,650 ± 50	230 ± 40
8	4	40 ± 30	70 ± 20
9	4	70 ± 30	90 ± 30
10	8	490 ± 30	110 ± 20
	4	530 ± 40	110 ± 30
11	4	2,130 ± 50	200 ± 40
12	4	630 ± 40	40 ± 30
13	4	2,220 ± 50	320 ± 50
14	4	1,940 ± 50	180 ± 40
15	10	1,960 ± 30	370 ± 30
16	10	780 ± 30	120 ± 20
17	10	2,270 ± 30	300 ± 30
18	10	1,290 ± 30	130 ± 20
19	10	4,860 ± 50	300 ± 40
background 20	20	< 20	50 ± 10

Notes: 1. See Figure 5.3 for location of #6-19, Figure 5.2 for #1-5, and Figure 5.1 for #20.
2. ± values are 2 σ counting error; < values are 3 σ counting error.
3. #10 was measured twice in succession; #7, 11, and 15 are approximately the same location.

Table 5.15

Ratio of Count Rate by Underwater Probe to Radionuclide Concentration in Benthal Samples

	^{60}Co, c/min / pCi/g			^{137}Cs, c/min / pCi/g		
Location	hand, top	hand, lower	dredge	hand, top	hand, lower	dredge
#1	20	400	40	6	30	5
2	100	300	60	50	100	40
3	--	---	60	--	---	40
15	330	330	430	110	80	120
16	870	1,600	120	90	90	30
17	540	1,500	250	70	90	60
18	2,600	13,000	400	90	140	20
19	240	260	590	40	50	90
20	---	---	---	120	70	120
range	20-2,600	260-13,000	40-590	6-120	30-140	5-120
avg. of #2, 15, 16, 17, 19 and 20:	420	800	290	80	80	80

Note: count rates from Table 5.14 and radionuclide concentrations from Table 5.12 (two dredge values averaged for locations #1 and 2).

In comparison, discharges during the 10 years of operation at the annual ^{60}Co and ^{137}Cs releases estimated in Section 4.3.5 would be approximately 20 mCi ^{60}Co (of which 9 mCi would have decayed) and 2 mCi ^{137}Cs. Both sets of estimates are highly uncertain, but suggest that a considerable portion of the discharged ^{60}Co and ^{137}Cs remained in benthal material.

The radionuclide concentrations in the sediment are too low to result in any detectable direct radiation exposure to humans. The possibility of radionuclides in benthal material entering the food chain through uptake by fish, however, has been suggested. (4) Although, at the observed concentrations, the uptake by fish would be expected to be very low, this potential exposure pathway should be evaluated periodically by comparing radionuclide levels in benthal material and fish.

5.7 References

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6. Radionuclides in the Terrestrial Environment

6.1 Introduction

6.1.1 *Sampling.* Release data by Yankee (see Appendix B.2) and radioactivity measurements in airborne effluents during this study (Section 3.3) suggest that radionuclide concentrations in ground-level air and deposition on ground and vegetation attributable to Yankee were extremely low. Because so little airborne radioactivity is released, few radioactivity measurements are performed on land by the Yankee contractor for environmental surveillance. In 1969, they consisted only of gross alpha and beta activity analyses in soil from 9 locations.(1) The Yankee Hazards Summary Report also mentions the collection of airborne particles (on filters and gummed trays) and hay, but states that "with complete information available on the amount of radioactivity released from the plant, the need for an extensive post-operational survey will be limited".(2)

The following samples and measurements were obtained in the neighborhood of Yankee:

(1) Air was collected in 96-liter Saran-plastic bags at four locations 300 to 500 m NE to E of the Yankee stack to measure radionuclides in ground-level air during the release of gas from the gas surge drum. Air was pumped by hand at the rate of approximately 1 liter/min. The collection technique was satisfactory, but the bags leaked, hence no samples were available for the intended analysis of ^{85}Kr. As indicated in Section 6.2.1, the estimated concentration of ^{85}Kr in the collected air was, in any case, too low for detection. A release rate higher by an order of magnitude than the one described in Section 3.1.3 had been anticipated, but was not attained because of the limited size of the orifice at the discharge into the stack.(3)

(2) One sample of snow was collected from the ground on site, and a background sample was collected at a distance of 8 km.

(3) One set of grass and soil samples was collected at the on-site location (0.2 km west of the stack), two were collected just beyond the 0.3-km station perimeter and one was collected at the background location, 8 km distant (see locations No. 201-204 in Figures 5.1 and 5.2).

(4) Two samples of milk from cows that grazed on a pasture 3.1 km SE of Yankee were collected at the dairy in Rowe (see Figure 5.1). One sample was obtained before gas was released from the surge drum, and the other, one day after the gas release.

(5) Three deer that had died in accidents near Yankee and three that had died similarly at distant locations were compared for radionuclide content.

(6) External radiation exposure was measured with survey meters at the following number of points: 10 on site at Yankee, 5 at the 0.3-km station perimeter, 8 in the immediate environs, and 3 at background locations.

Calculations of expected concentrations of radionuclides from Yankee in the environment are presented in Section 6.2 and Appendices C.1 to C.5 to demonstrate the procedure and indicate the magnitude of radionuclide concentrations that may be attributed to Yankee. The computed concentrations in snow, vegetation, soil, and milk were several orders of magnitude below detectability. Values measured in airborne effluent (Section 3.3) were used as source terms; meteorological data were taken from a summary of short-term measurements on site or from U. S. Weather Bureau data for Albany, N.Y.; and dispersion over a flat terrain was assumed. As a consequence, results of these calculations are gross approximations. They are considered useful guides

for planning environmental surveillance, however, as long as radionuclide releases are so low that calculated radiation doses are far below AEC limits.

Sample analyses and measurement results are described in detail in Sections 6.3 to 6.7. The detected radionuclides are believed to have been deposited as fallout from atmospheric nuclear tests, or to occur naturally. Their concentration varied so much among samples, however, that careful sample selection and numerous samples are needed to determine with assurance whether any of these radionuclides should be attributed to effluents from the station. The external radiation exposure rate above background was 1 to 3 microroentgen per hour (μR/hr) at the Yankee exclusion perimeter, and was estimated to be 0.7 μR/hr at the nearest habitation and 0.3 μR/hr at the town of Monroe Bridge. This radiation was attributed to gamma rays from stored radioactive wastes at the station.

6.1.2 *Environment of Yankee.* The plant lies in the deep narrow valley of the Deerfield River in the Berkshire Mountains of northwest Massachusetts. The elevation of the plant is approximately 350 m (1150 ft); within 1.5 km to the east, south, and west, the mountains rise to elevations between 550 and 640 m (1800 and 2100 ft). The slopes of the mountains are wooded, and there are few open spaces or roads in this area within a 3-km radius. Sherman Reservoir is immediately to the north of the Yankee Plant (see Figures 5.1 and 5.2).

Populated areas within a 3-km radius include the town of Monroe Bridge (1.2 km SW, pop. 200 in 1960), a few houses on Main Road in Monroe township approximately 2 km west, part of Rowe township (4.3 km SE, pop. 230), and a few houses above the valley in southern Vermont (the Vermont border is 1.2 km north of the station). The Sherman hydroelectric station is immediately west of Yankee, Harriman hydroelectric station is 2.6 km to the north, and the Readsboro Road carries traffic along the west bank of Sherman Reservoir, 0.4 km to the northwest. Towns at slightly greater distances from Yankee include Charlemont and Florida in Massachusetts and Readsboro and Whitingham in Vermont. Nearby cities are North Adams and Greenfield, Mass.; the Albany-Troy area, 60 km west, is the nearest large population center.

There appears to be no farming within 3 km of the plant. The dairy at which milk was collected apparently is the only one in Rowe. A herd of three cows was seen in Whitingham township, and a few cows were reputed to be in Monroe township. The only edible crops from the immediate area are said to be apples and maple syrup.(3) The only nearby industry is a glassine paper plant in Monroe Bridge, which uses part of the water that is retained by No. 5 Dam, just below Sherman Dam.

6.1.3 *Meteorology and Climatology.* An aerovane and an anemometer mounted at the station indicate that winds in the valley are predominantly along the axis of the valley, but that appreciable turbulence occurs.(2) Under unstable conditions, the air within the valley would be expected to mix with the air above the ridges and to flow in the direction of the wind at these higher elevations. Under stable conditions, the air in the valley would be isolated from the general airflow unless the airflow is along the axis of the valley, but dispersion is expected to be increased by the air turbulence at the plant site.(2) Structures near the stack, at or just below stack height, enhance dispersion.

6.2 Estimation of Radioactivity Concentrations

6.2.1 *Dispersion of ^{85}Kr in air.* Calculation of the dispersion of radioactive gas from the stack can only be approximate in view of the complex terrain and air turbulence near the stack. An approximate value of the normalized dispersion at ground level on the plume center-line, $X\bar{u}/Q$ (in m^{-2}), was derived from curves of dispersion *vs.* distance as a function of stack height and stability categories.(4) Values were taken from Figures 3-5B and 3-5C in this reference for the 46-m height of the Yankee stack and a distance of 300 m between stack and sampling points. Other sample-collection information and the calculated results are given in Appendix C.1.

The concentration of ^{85}Kr in air at ground level, X (in pCi/m^3), was computed from the graphic values of $X\bar{u}/Q$, values in Appendix C.1 of the measured release rate, Q (in pCi/sec), and the mean wind speeds, $\bar{u}$ (in m/sec). The calculated concentrations (7 and 3 pCi/m^3) are lower than the ^{85}Kr background (approximately 1½ pCi/m^3) in air.(5) Actual concentrations from Yankee may be even lower because of greater air turbulence than was considered in the computation. The computed concentrations could have been detected in several cubic meters of

air, but not in the 96-liter volumes that were collected.

6.2.2 *Accumulation of ^{90}Sr in snow.* The sampling locations had been covered with snow for several months prior to sampling on April 1, 1969. At Albany, rainfall totalled 0.3 cm between March 26 and 31;[6] this is believed to have fallen as snow in the mountains where Yankee is located. To calculate the radionuclide content in the 2-cm-deep snow samples at and near Yankee, washout in the recently precipitated snow was added to dry deposition during the period in which this snow had been on the surface.

Deposition by washout, W (in pCi/m^2), was computed by the following equation, derived from equation 5.64 of Slade:[7]

$$W = \frac{Q'_o L T \exp(-L x / \bar{u})}{\Theta \bar{u} x} \qquad (6.1)$$

where Q'_o : virtual release rate at stack, pCi/sec

L : washout coefficient, sec^{-1}

T : duration of washout, sec

Θ : sector width, radians

$\bar{u}$: wind speed at release height, m/sec

x : distance from stack to sampling point, m

Dry deposition, D (in pCi/m^2), was computed by integrating equation 5.44 of Slade[8] with respect to the cross-wind direction and then distributing deposition across the appropriate 20° sector:

$$D = \frac{(2/\pi)^{1/2}}{\Theta} \cdot \frac{v_d Q'_x T}{x \sigma_z \bar{u}} \cdot \exp(-h/2\sigma_z^2) \qquad (6.2)$$

where:

v_d : deposition velocity, m/sec

Q'_x : depletion-corrected release rate at point of interest, pCi/sec

T : total duration of deposition, sec

Θ : sector width, radians

x : distance to point of interest, meters

σ_z : standard deviation of vertical concentration distribution, meters

$\bar{u}$: release height wind speed, m/sec

h : effective release height, meters

The results of the calculation are shown in Appendix C.3. The washout of ^{90}Sr from the atmosphere was computed for 2 cm of snow that was assumed to have fallen during a 34-hour period with midpoint on March 29, 1969. It was also assumed that the wind blew from the stack to both of the sampling points during the entire snowfall. Dry deposition was summed for the period March 29-31, at the average wind frequency to the sector shown in Appendix C.2. The source term was used without correction for depletion. Values of v_d and L for ^{90}Sr were taken to be 3×10^{-3} m/sec and 1×10^{-5} sec^{-1}, respectively.[9]

6.2.3 *Accumulation of ^{90}Sr in vegetation.* Deposition under neutral and unstable atmospheric stability and during precipitation was computed with equations 6.1 and 6.2 for five sampling locations (see Appendix C.4). Deposition parameters on which these values are based are listed in Appendix C.2. Atmospheric stability and wind data in Appendix C.2 are from instruments at Sherman Dam (for locations No. 201 to 203) and on the hillside (for location No. 204 and the dairy farm). They were obtained in April, May, and June of 1959.[2] The data for the valley are probably representative despite the brevity of the record because air flow patterns within a deep valley are highly recurrent. Deposition was summed for April and May, 1969, because the samples were collected on June 3, and it was assumed that the growing season consisted of these two months. Rainfall data are from the April and May 1969 summaries for Albany.[6] To convert deposition per square meter to concentration per gram ash, the average grass density was taken to be 0.33 kg dry weight per square meter,[10] and the ash/dry weight ratio, 0.07,[11] for an overall ratio of 23 g ash/m^2. A half-time for radiostrontium in vegetation of 14 days,[12] taken from the mid-point of the 2-month period, was used to correct for removal of the radionuclide from the grass before sampling.

6.2.4 *Accumulation of ^{90}Sr on soil.* The deposition calculations described in Sections 6.2.2 and 6.2.3 were used to compute average annual accumulation of ^{90}Sr in soil, as shown in Appendix C.5. The average of annual precipitation values in 1968 and 1969 at Albany[6] was used, deposition values were corrected for decay according to the 28-year half life of ^{90}Sr, and the parameters affecting deposition calculations that are given in Appendix C.2 were applied. It was assumed that one-half of the deposited activity remained in the soil, and that one-half of the activity in the soil was in the 2-cm-deep soil-layer collected for analysis.

6.2.5 *Iodine-131 in cows' milk.* The concentration

of ^{131}I in milk from cows at the dairy 3.1 km from Yankee was estimated by combining calculations of dry deposition and transfer from grass to milk. It was assumed that ^{131}I was released from the gas surge drum at concentration Q'_O (in pCi/sec) for the 6.75 hour period following 1500 on June 3, 1969, that the wind blew continuously throughout that period from the Yankee stack toward the pasture, and that atmospheric stability was one-third neutral and two-thirds unstable. According to equation 6.2, the deposition parameters in Appendix C.2, and a deposition velocity for ^{131}I of 1×10^{-2} m/sec,(8) the total deposition was $1.04 \times 10^{-4} Q'_O$.

The ^{131}I concentration in milk, M, was computed by multiplying the deposited amount of activity by the effective daily grazing area(10) and the ratio of concentration in milk 1.25 days after initial ingestion of ^{131}I to the average daily intake of ^{131}I. (13) The latter was taken from curve C in Figure 14.2 of reference 13. Thus,

$$M = 1.04 \times 10^{-4} Q'_O \text{ pCi/m}^2 \times 45 \text{ m}^2\text{/day} \times 2.9 \times 10^{-3} \text{ day/liter} = 1.4 \times 10^{-5} Q'_O \text{ pCi/liter} \quad (6.3)$$

For $Q'_O < 50$ pCi/sec, the minimum detectable release rate (see Table 3.6), the computed concentration is $< 7 \times 10^{-4}$ pCi/liter. Even if ^{131}I were discharged continuously at the same rate, its concentration in milk would be higher by only an order of magnitude, far below detectable levels. Concentrations of ^{89}Sr, ^{90}Sr, and ^{137}Cs in milk would be similarly far below detectable levels because their release rates were less than 50 pCi/sec (see Section 3.3.5) and their transfer from cows' feed to milk are within a factor of five of the ^{131}I transfer.

6.3 Radionuclides in Snow

Two snow samples were collected on April 1, 1969; locations and amounts of sample are given in Table 6.1. The snow was melted and passed through 0.45-μ-dia. membrane filters. For both samples, 7.6 liters of filtrate were evaporated to 35 ml. The membrane filters and the 35-ml samples were analyzed by gamma-ray spectrometry with NaI(Tl) detectors and 200-channel analyzers. The membrane filter from the on-site sample was also analyzed with the Ge(Li) detector plus 1,600-channel spectrometer. Aliquots of the four samples were then analyzed radiochemically for radiostrontium and radiocesium.

The radionuclide content in on-site sample No. 202 is so similar to that in background sample No.

Table 6.1

Radionuclides in snow, April 1, 1969

	Concentration, pCi/liter			
	#202, 0.2 km W		#204, 8 km S	
Radionuclide	Soluble	Insoluble	Soluble	Insoluble
^{3}H	700 ± 200	---	500 ± 200	---
^{54}Mn	< 2	< 1	< 2	< 1
^{60}Co	< 3	< 1	< 3	< 2
^{89}Sr	< 0.2	< 0.1	< 0.2	< 0.2
^{90}Sr	1.1	0.2	0.8	0.6
^{95}Zr	< 2	4	< 2	8
^{95}Nb	< 2	16	< 2	18
^{103}Ru	NM	3	NM	NM
^{106}Ru	< 8	< 2	< 8	15
^{131}I	< 2	< 1	< 2	< 1
^{137}Cs	2	3	1.3	4
^{141}Ce	NM	13	NM	12
^{144}Ce	< 10	26	< 10	39

Notes:

1. The sample at location #202 consisted of 16.6 l of water, and was taken from the top 2 cm of snow in a 3-m^2 area; the sample at location #204 consisted of 7.6 l of water, and was taken from the top 2 cm of snow in a 2.3-m^2 area.

2. ± values are 2 σ counting error; < values are 3 σ counting error; NM - not measured.

204 (see Table 6.1) that the entire radioactivity is attributed to fallout from atmospheric nuclear tests. The computed ^{90}Sr concentrations from Yankee in Appendix C.3 are two orders of magnitude lower than the measured concentrations in the on-site sample. To detect particulate radionuclides that are released from the Yankee stack at similar rates, are deposited to approximately the same degree as ^{90}Sr, and moreover do not occur in fallout, it would appear necessary to collect 500-fold larger samples.

6.4 Radionuclides in Vegetation and Soil

Four samples were collected on June 4, 1969, at the locations shown in Table 6.2. Several kilograms of vegetation were obtained by cutting grass and weeds in an area of approximately 10 m^2. Soil samples of approximately 500 cc were taken from the top 2 cm at the same locations after removing the covering vegetation.

The vegetation samples were dried at 110°C in cloth bags and then ashed at 500°C. The dried weights could not be measured because the samples accidentally ignited during drying. The ashed samples were analyzed gamma-spectrometrically in 400-cc volumes with NaI(Tl) and Ge(Li) detectors plus multichannel analyzers. Aliquots were then analyzed radiochemically for strontium, cesium, ruthenium, and antimony. Soil samples were dried at 110°C and then analyzed with gamma-ray spectrometers, and radiostrontium was determined in aliquots with the leaching procedure referred to in Section 5.6.3.

The radionuclides measured in the vegetation and soil samples are listed in Tables 6.2 and 6.3, respectively. The radionuclides usually found in fallout were observed. The much lower concentrations in some of the nearby samples (No. 201, 202, and 203) than in the background sample (No. 204) suggest considerable variability in deposition or accumulation of these radionuclides. To determine if the higher concentrations of some radionuclides in nearby vegetation--^{90}Sr (No. 201), ^{106}Ru (No. 202), and ^{137}Cs (No. 201)--could possibly be attributable to Yankee would require analysis of several samples at each location to establish standard deviation values. The wide differences of radionuclide concentrations in vegetation are also indicated by the ^{90}Sr and ^{137}Cs contents of six deer rumen, which ranged from 11 to 51 pCi/g ash and from 9 to 86 pCi/g ash, respectively (converted from pCi/kg wet weight in Section 6.6.2).

Table 6.2
Radionuclide (pCi/g ash) and Stable Ion (mg/g ash) Concentration in Vegetation, June 4, 1969

Substance	#201 0.3 km NE	#202 0.2 km W	#203 0.4 km NW	#204 8 km S
^{54}Mn	< 2	< 2	< 2	< 2
^{89}Sr	< 5	< 5	< 5	< 5
^{90}Sr	64 ± 3	20 ± 1	19 ± 1	57 ± 3
$^{95}Zr + ^{95}Nb$	34 ± 1	35 ± 1	52 ± 2	36 ± 2
^{106}Ru	< 8	38 ± 5	< 8	< 8
^{125}Sb	< 2	< 2	< 2	< 2
^{137}Cs	13 ± 1	3 ± 1	5 ± 1	6 ± 1
^{144}Ce	18 ± 2	NM	38 ± 4	NM
calcium	56	36	33	39
strontium	0.40	0.20	0.30	0.27
potassium	550	230	250	590
silica	NM	103	NM	18

Notes:
1. $\pm$ values are 2 σ counting error; $<$ values are 3 σ counting error; NM - not measured.
2. see locations in Figures 5.1 and 5.2.

Table 6.3
Radionuclide (pCi/g dried) and Stable Ion (mg/g dried) Concentration in Soil, June 4, 1969

Substance	#201 0.3 km NE	#202 0.2 km W	#203 0.4 km NW	#204 8 km S
^{90}Sr	0.51 ± 0.05	0.49 ± 0.05	0.14 ± 0.02	1.54 ± 0.05
^{95}Zr	0.13 ± 0.02	0.66 ± 0.03	0.40 ± 0.06	NM
^{137}Cs	1.5 ± 0.1	1.4 ± 0.1	3.5 ± 0.2	5.4 ± 0.3
^{144}Ce	1.4 ± 0.5	2.0 ± 0.5	1.0 ± 0.5	1.3 ± 0.5
calcium	2.0	2.3	1.7	1.1
strontium	0.042	0.054	0.053	0.060
potassium	11.4	11.5	17.9	8.7

Note: see footnotes to Table 6.2.

The samples also contained naturally occurring ^{40}K, U plus progeny, and Th plus progeny. No ^{89}Sr (< 0.2 pCi/g) or photon-emitting radionuclides other than listed (generally < 1 pCi/g) were found in soil.

Computed concentrations of ^{90}Sr in grass from deposition of airborne particles released by Yankee are lower than measured values by four orders of magnitude, according to Appendix C.4. Strontium-90 concentrations in soil computed for long-term deposition of airborne particles from Yankee (Appendix C.5) are also lower than measured values by four orders of magnitude.

6.5 Radionuclides in Milk

Two four-liter samples of raw milk were collected at the dairy in Rowe, one at the morning milking on June 3, 1969, and the other at the evening milking on June 4. A 3.5-liter sample of milk was analyzed with a NaI(Tl) detector plus multichannel analyzer for photon-emitting radionuclides, and a l-liter aliquot was analyzed radiochemically for radiostrontium*. Analyses were by the procedure for routinely collected milk-network samples, but the counting periods were longer to improve precision of measurement.

The radionuclides were at essentially the same concentrations in both samples, as shown in Table 6.4. Average radionuclide concentrations in pasteurized milk at nearby cities during June 1969 were:(14)

Radionuclide	Albany, N. Y.	Boston, Mass.	Hartford, Conn.
^{90}Sr	8 pCi/l	11 pCi/l	8 pCi/l
^{131}I	<10	<10	<10
^{137}Cs	<20	19	13

Table 6.4
Radionuclide Concentration in Milk, pCi/liter

Radionuclide	June 3, 1969 morning	June 4, 1969 evening
^{89}Sr	6 ± 2	7 ± 2
^{90}Sr	17 ± 1	13 ± 1
^{131}I	<3	<3
^{137}Cs	48 ± 2	53 ± 2
^{140}Ba	<3	<3

Notes:
1. Gas was released from surge drum at Yankee Nuclear Power Station on June 3, 1969, at 1500-2145.
2. Milk is from a dairy at Rowe, see Figure 5.1.
3. Analysis was by NERHL, PHS.
4. ± values are 2 σ counting error; < values are 3 σ counting error.

* We thank NERHL, PHS for analyzing these samples.

Strontium-89 concentrations between 5 and 11 pCi/liter were reported at 15 stations throughout the country, but no ^{89}Sr was detected at these 3 cities. The ^{90}Sr and ^{137}Cs concentrations in the milk at Rowe are higher than at the cited cities, but less than the highest average values in the U. S. of 31 and 126 pCi/l, respectively, during the month. No ^{131}I was detected at any U.S. milk station.(14)

At the measured or minimum detectable release rates at Yankee, the concentration of ^{131}I in milk was computed to be four orders of magnitude lower than could be measured, and the other radionuclides would be similarly undetectable (see Section 6.2.5). No noticeable differences can, therefore, be expected in the two sets of values. Even if the radionuclide releases at these rates were continuous (i.e., from sources at Yankee other than the surge drum), the resulting levels could not be detected in the milk (see Section 6.2.5). Hence, the measured concentration of ^{89}Sr, ^{90}Sr, and ^{137}Cs are attributed to fallout.

6.6 Radionuclides in Deer

6.6.1 *Sampling and analysis.* To begin evaluation of the radionuclide content in wildlife, six deer were collected, three within 3 km of Yankee, and three from more distant sites.* The deer are described in Table 6.5. Four had been killed in automobile accidents, deer D-1 was killed at a fence, and D-6 was found dead from undetermined causes. Samples of bone (femur) and muscle, and the entire rumen content from each deer, were preserved in plastic bags on dry ice. Muscle and rumen-content samples were ashed at 400°C and analyzed for photon-emitting radionuclides with 10- x 10-cm NaI(Tl) detector and with the coincidence/anticoincidence NaI(Tl) system. Bone samples were ashed at 600°C. All samples were analyzed for radiostrontium. Bone and rumen-content samples were analyzed with an atomic-absorption spectrometer for stable calcium and strontium.

6.6.2 *Results and discussion.* The stable ion and radionuclide concentrations measured in the deer samples are listed in Table 6.6. All concentrations refer to wet weight, but ash weight/wet weight ratios are given so that concentrations relative to ash weight may be calculated.

The average concentrations of ^{90}Sr and ^{137}Cs in deer collected near Yankee are, with one exception, higher than for deer obtained at a distance; average ^{22}Na concentrations, on the other hand, were lower in deer near the station, as shown below:

sample	radionuclide	nearby deer, pCi/kg	background deer, pCi/kg
bone	^{90}Sr	10,100 ± 1,300	8,700 ± 2,100
muscle	^{22}Na	3 ± 1	7 ± 3
	^{90}Sr	6 ± 3	6 ± 3
	^{137}Cs	1,270 ± 910	570 ± 420
rumen	^{22}Na	11 ± 8	16 ± 12
	^{90}Sr	650 ± 240	330 ± 150
	^{137}Cs	960 ± 650	450 ± 410

Table 6.5

Description of Sampled Deer

Deer No.	Location*	Distance from Yankee, km	Date of death, 1969	Age year	Age month	Sex
D-1	Charlemont	12	April 16	1	10	M
D-2	Ashfield	26	February	1	6	ND**
D-3	Buckland	29	June	6	10	F
D-4	Rowe	3	April 25	2	11	F
D-5	Rowe	3	Jan. 12	5	0	F+
D-6	Rowe	1	April 2	2	10	M

*all are southeast of Yankee

**ND - not determined

+pregnant

* We thank Colton H. Bridges, Bureau of Wildlife Research and Management, Division of Fisheries and Game, State of Massachusetts, for collecting the deer and determining their ages.

The differences are in no case significant because of the relatively large standard deviations, suggesting that all of the measured radioactivity was from fallout.

Because of the large differences in radionuclide concentrations among samples, collection of more samples appears necessary to determine adequate mean values and standard deviations. This problem was also encountered with vegetation and soil samples (see Section 6.4), but is especially serious in animals because their radionuclide contents are affected by so many variables--e.g., environment, location, season, age, food supply, and individual differences.

Even the highest ^{137}Cs concentration in muscle is not unusually high compared to deer muscle collected in areas distant from nuclear power stations.(15,16) Jenkins and Fendley reported numerous cases in which levels of ^{137}Cs in the muscle of Whitetail deer from the southeastern United States, collected during winter and early spring, approach 150,000 pCi/kg.(15) The ^{137}Cs levels in both rumen and muscle in this study are higher than those in four deer obtained in June 1969 from the vicinity of Dresden, Illinois.(17)

The rumen contained mostly grasses and leaves. If these samples can be considered typical, the accumulation factor (AF), in pCi ^{137}Cs/kg muscle per pCi ^{137}Cs/kg rumen content, ranges from 0.64 to 3.23, with an average AF value of 1.5 ± 0.4 (1 σ). These values are similar to those reported previously.(15-17)

The observation of ^{22}Na in the muscle and rumen of deer parallels that in fish muscle (see Table 5.9). The AF for ^{22}Na from rumen to muscle is 0.5 ± 0.2.

The mean ^{90}Sr concentration in bone was 9,400 ± 3,100 pCi/kg, 95 ± 11 pCi/g calcium, and 132 ± 40 pCi/mg strontium. The mean concentration in bone is approximately 4 times higher than in deer collected in Illinois,(17) but is similar to concentrations reported for deer from South Carolina,(16) Colorado,(18) and California.(19,20) The average AF values from diet to bone for the six deer, assuming rumen content to be a typical diet, are as follows for ^{90}Sr, strontium and calcium:

Table 6.6

Radionuclide (pCi/kg)* and Stable Ion (g/kg)* Concentration in Deer Samples

Sample Type	Nuclide	Background samples			Samples from Vicinity of Yankee		
		D-1	D-2	D-3	D-4	D-5	D-6
Bone	^{90}Sr	12,500 ± 340	10,000 ± 260	3,650 ± 140	10,600 ± 320	11,000 ± 260	8,600 ± 220
	Sr	0.090	0.060	0.069	0.078	0.081	0.053
	Ca	106	100	87	117	102	82
	ash wt./ wet wt.	0.28	0.26	0.23	0.31	0.26	0.22
Muscle	^{22}Na	7.1 ± 1.1	10.5 ± 0.5	3.1 ± 0.6	3.4 ± 0.4	1.9 ± 0.3	Not anal.
	^{90}Sr	5.3 ± 1.9	9.9 ± 2.1	4.4 ± 1.4	4.4 ± 1.4	10.2 ± 1.8	4.5 ± 1.9
	^{137}Cs	990 ± 40	550 ± 20	160 ± 10	1,470 ± 60	2,060 ± 90	270 ± 10
	K	3.77	3.39	3.31	3.76	2.94	2.24
	ash wt./ wet wt.	0.013	0.015	0.010	0.012	0.009	0.011
Rumen Content	^{22}Na	10.5 ± 0.7	30.3 ± 1.3	8.3 ± 0.7	4.9 ± 0.3	8.9 ± 0.6	20.0 ± 0.1
	^{90}Sr	339 ± 9	480 ± 11	171 ± 6	370 ± 14	760 ± 20	810 ± 20
	^{137}Cs	920 ± 40	170 ± 10	250 ± 10	1,440 ± 60	1,210 ± 50	210 ± 10
	K	4.23	3.68	4.89	5.29	3.07	2.26
	Sr	0.0027	0.0043	0.0019	0.0030	0.0029	0.0086
	Ca	0.33	1.65	1.09	0.44	0.53	2.06
	ash wt./ wet wt.	0.014	0.019	0.016	0.017	0.014	0.016

*Kg wet weight

Note: ± values are 2 σ counting error.

$AF_{^{90}Sr} = 23$
$AF_{Sr} = 24$
$AF_{Ca} = 160$

The average observed ratio from diet to bone for strontium relative to calcium ($OR_{bone/diet}$) is 0.20 ± 0.05 for both ^{90}Sr and stable strontium. The AF and $OR_{bone/diet}$ for ^{90}Sr agree with those previously reported for deer collected in Illinois, (17) but the $OR_{bone/diet}$ and AF from diet to bone are slightly smaller than for Alaskan caribou (0.31 and 37, respectively).(21)

The mean ^{90}Sr concentration of 6 pCi/kg deer meat is one-tenth of that reported for Alaskan caribou or reindeer meat, (22) but is 6 times that in meat taken to be a typical component of New York City diets during Jan.-March 1969. (23) The average ^{90}Sr concentration in deer muscle was approximately 1/1500 of the concentration in bone, a much smaller ratio than that reported for Alaskan caribou.(21) This ratio undoubtedly varies because muscle reflects recent dietary intake of ^{90}Sr more directly than does bone. The average AF for ^{90}Sr from rumen to muscle is 0.016; in 25 Alaskan caribou and reindeer, the AF ranged from 0.004 to 0.21, with an average of 0.036.(24,25)

Strontium-89 was not detected in rumen content, muscle, or bone. Minimum detectable levels at the 3-sigma confidence limit were 20 pCi/kg wet weight in rumen content and muscle, and 400 pCi/kg wet weight in bone. No ^{134}Cs was detected in deer muscle at the minimum detectable concentration of 2 pCi/kg. The fission products ^{106}Ru (in deer D-2 and D-3) and ^{95}Zr (traces) were identified by their characteristic gamma rays in rumen contents, as were naturally occurring ^{40}K, ^{226}Ra plus progeny, and ^{232}Th plus progeny.

6.6.3 *Hypothetical radiation dose from eating deer meat.* The radiation dose a person might receive from eating deer meat was estimated from Federal Radiation Council values, according to which 170 mrem/year is equivalent to a daily intake of 200 pCi ^{90}Sr. (26) At the average ^{90}Sr concentration of 6 pCi/kg deer meat and an annual consumption of 79 kg meat (0.22 kg/day)(23) applied entirely to deer meat, the radiation dose to bone marrow is 1.1 mrem/year. The average ^{137}Cs concentration of 920 pCi/kg muscle was 150 times more than that of ^{90}Sr and the limit for ^{137}Cs is 150 times higher, hence the radiation dose from ^{137}Cs to the whole body is also 1.1 mrem/yr. The additional radiation dose to the whole body from ^{22}Na at an average concentration of 5 pCi/kg is negligible (0.002 mrem/yr). These doses are believed to be entirely from radionuclides in fallout, but provide an upper limit if some of the radioactivity in deer were attributed to Yankee.

6.7 External Radiation

6.7.1 *Detection instruments.* Radiation exposure rates were measured with cylindrical NaI(Tl) gamma-ray detectors (5-cm diameter x 5-cm length) connected to portable count-rate meters.(27) The instruments had been calibrated by comparing their count rates in the natural radiation background at Cincinnati with measurements by a muscle-equivalent ionization chamber and Shonka electrometer.(28)* Radiation levels during calibration ranged from 5 μR/hr over water in a lake to 19 μR/hr over granite. The count rate, C (in count/min), of the survey instruments varied linearly with the radiation exposure rate, R (in μR/hr), of the ionization chamber; a typical calibration curve had the equation:

$$R = 7.0 \times 10^{-4} C + 3.3 \qquad (6.4)$$

Radiation exposure rates at the measurement locations near Yankee were computed by applying these calibration curves to the observed count rates.

Despite the dependence of the counting efficiency of the detectors on the energy distribution of the gamma-ray flux, the calibration curves were applicable in a variety of natural radiation backgrounds. In numerous measurements, the standard error of the survey meters was ± 0.35 μR/hr, and the exposure values computed from the readings were within 4 percent of the values measured with the ionization chamber during 95 percent of the time. (27) Similar calibration curves could also be applied to readings within or beneath mixtures of noble-gas fission products that were emitted from the stack of a boiling water reactor. (17)

6.7.2 *Measurements.* The 26 radiation measurement locations listed in Table 6.7 were selected for the following reasons:

(1) points O and P were considered to be sufficiently distant from Yankee but similar in natural radiation to yield

*We thank Richard Stoms, PHS, Cincinnati, for making the two sets of instruments available.

terrestrial background values for comparing with and subtracting from exposure rates near the station; point M yielded the background value over water;

(2) eight points, 0.37 to 1.1 km distant from the center of the station, provide values for computing potential radiation exposure of persons in the environment;

(3) five points indicate the radiation exposure at the 0.3-km exclusion perimeter of the station; and

(4) ten points on site were intended to aid in identifying the source of external radiation from Yankee and to check off-site exposure values by extrapolating from these higher, more accurately measured values.

The first and third sets of measurements were taken while the station was operating at full power; the second set was obtained during refueling, when the reactor was not operating and neither short-lived nor stored radioactive gases were being discharged.

Table 6.7

External Radiation Exposure Rate Measurements near Yankee

Location		Exposure Rate,[c] μR/hr		
Point[a]	Distance[b]	June 4, 1970	Nov. 18, 1970	Feb. 8, 1971
A	0.30 km NE	10.4 ± 0.0[d]	12.2 ± 0.2	
B	0.29 km NE		10.5 ± 0.7	
C	0.30 km NNE		9.3 ± 0.4	
D	0.40 km NNW		7.0 ± 0.6	
E	0.40 km NW		9.5 ± 0.2	5.9 ± 0.1
F	0.37 km NW	7.5 ± 0.1	9.2 ± 0.1	
G	0.39 km WNW		9.3 ± 0.1	7.1 ± 0.0
H	0.25 km NW		9.5 ± 0.2	7.2 ± 0.0
I	0.18 km W		13.4 ± 0.2	
J	0.14 km W	11.0 ± 0.0	14.5 ± 0.1	9.4 ± 0.1
K	0.14 km W	11.5 ± 0.1	14.3 ± 0.1	8.7 ± 0.1
L	0.30 km WSW		8.8 ± 0.1	6.0 ± 0.0
M	2.0 km N		6.5 ± 0.2	
N	1.1 km SW		9.2 ± 0.1	
O	8 km S	7.5 ± 0.0	8.5 ± 0.1	5.7 ± 0.1
P	17 km SE	7.4 ± 0.1	8.4 ± 0.1	5.6 ± 0.1
Q	0.12 km NW			14.0 ± 0.1
R	0.18 km NW			9.2 ± 0.0
S	0.31 km NW			5.7 ± 0.1
T	0.16 km W			12.4 ± 0.0
U	0.21 km W			8.5 ± 0.1
V	0.30 km WSW			6.8 ± 0.1
W	0.22 km WSW			6.9 ± 0.1
X	0.39 km WNW			6.7 ± 0.0
Y	0.44 km W			5.7 ± 0.1
Z	1.1 km SW			6.0 ± 0.0

a. All points are shown in Fig. 6.1 except those more than 0.4 km distant from Yankee: point M is in Sherman Reservoir; N, near west end of Dam #5 (Fig. 5.1); O, at location #204 (see Fig. 5.1); P, in East Charlemont; Y, on Readsboro Rd; and Z, near the Monroe Bridge school.

b. Distances are from the center of the exclusion area shown in Fig. 6.1.

c. Measurements at points B, C, D, and M were taken 1 m above water level in Sherman Reservoir; all others were obtained 1 m above ground.

d. Exposure rates are averages of 2 to 10 measurements; ± values are 1/2 of the range for 2 measurements or 2 σ values for more than 2 measurements.

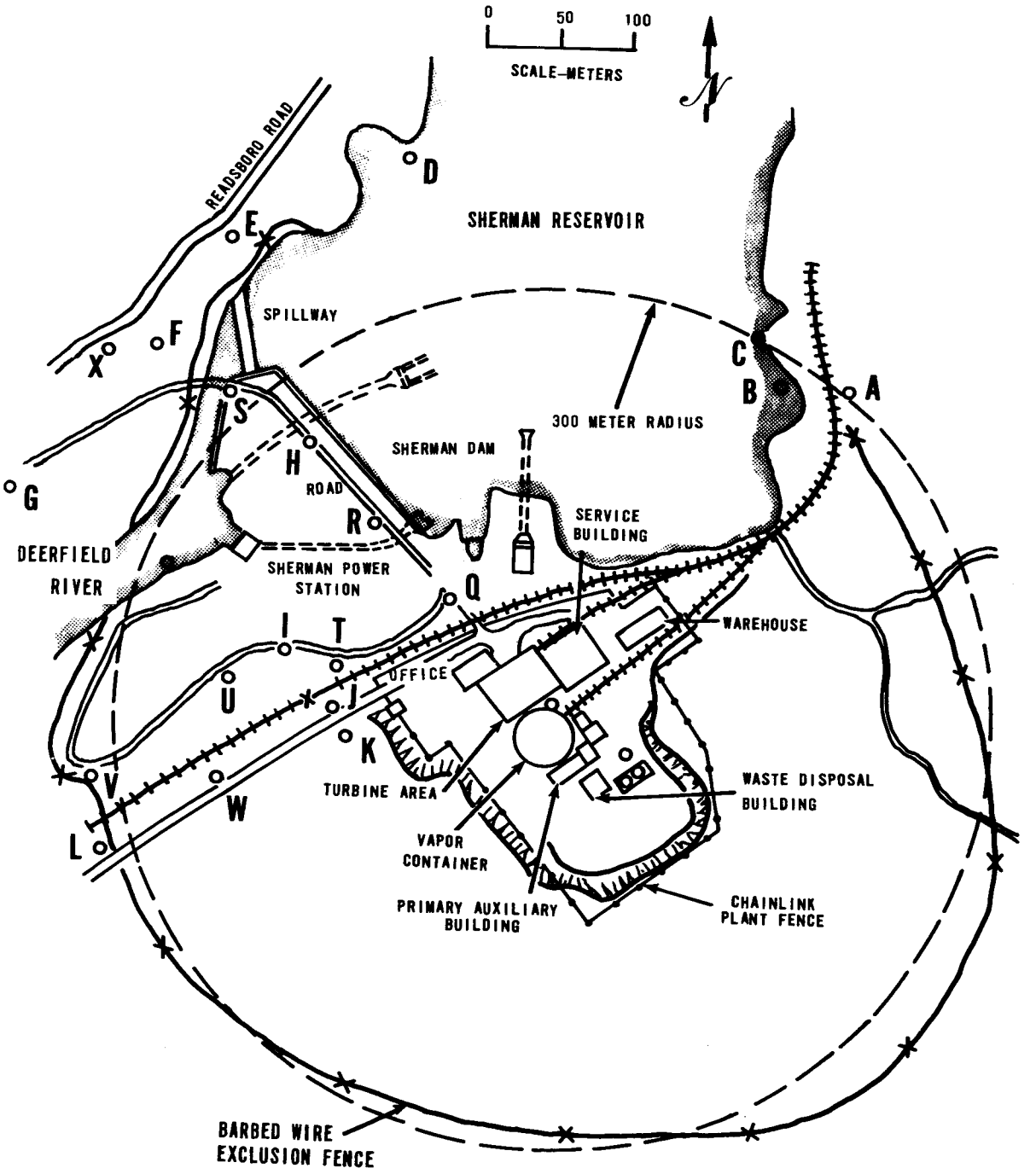

Figure 6.1. Locations of Radiation Exposure Measurements with Survey Meters.

For these measurements, detectors were held 1 m above the surface of the ground or water. The count rates were between 3,400 and 16,000 count/min. On land, locations over grassy terrain were selected to minimize variations in the background; however, a rock fill near point G and cuts in hillsides near points K and Z may have increased the background radiation at these locations. The snow cover to a depth of 0.3 to 1 m on Feb. 8, 1971, undoubtedly lowered background values at all locations. The lower background over water also shows the effect of shielding material (i.e., the water) between the detector and soil or rock.

To observe the effect of distance and direction from the center of the station on the external radiation field, the on-site measuring points were located along several radii toward northeast (NE), northwest (NW), west (W), and west-southwest (WSW), as shown in Figure 6.1. In the opposite directions steep and unpopulated hillsides adjoin the station.

6.7.3 *Results and discussion.* The gross radiation exposure rates in Table 6.7 (which include the natural radiation background) range from 5.7 to 14.5 μR/hr at the station and its immediate environs. The terrestrial background radiation values at locations 0 and P agreed within 0.1 μR/hr, but the average value was different during each of the three measurement periods.

All radiation exposure rates except three at or near Yankee were above the background values obtained on the same days. These higher values are attributed to direct radiation from radioactive waste stored at the station. This explanation was supported qualitatively by (a) the general decrease in exposure rates with distance from the station, (b) the lower values where there was shielding by buildings or topographical features (see Fig. 6.1), and (c) no change in radiation exposures during reactor shut-down or changes in wind directions. For these reasons, the higher radiation exposure rates were not attributed to higher natural radiation background, deposition of radionuclides from the station on the ground, radionuclides in the plume of airborne radioactive effluents from the station, or direct radiation from the Yankee reactor.

Because the net radiation exposure rates beyond the station were so low and the radiation backgrounds could not be measured directly at these points, an attempt was made to check these exposure rates by extrapolating from the higher values measured on site. The resulting sets of values compare as follows:

Location		Exposure rate, μR/hr	
Point	Distance	Measurements minus background	Extrapolation of on-site measurements
A	0.30 km NE	3.3	---
C	0.30 km NNE	2.8	---
	NE perimeter	average 3.0 ± 0.3	---

L	0.30 km WSW	0.3	1.3
V	0.30 km WSW	1.1	1.3
	WSW perimeter	average 1.0 ± 0.5 (1σ)	
S	0.31 km NW	0.0	1.2
	NW perimeter	average 0.6 ± 0.6	
D	0.40 km NNW	0.5	0.8
E	0.40 km NW	0.6	0.8
F	0.37 km NW	0.4	0.9
G	0.39 km WNW	1.1	0.8
X	0.39 km WNW	1.0	0.8
Y	0.44 km W	0.0	0.6
	Readsboro Road	average 0.7 ± 0.3 (1σ)	
N	1.1 km SW	0.7	0.1
Z	1.1 km SW	0.3	0.1
	Monroe Bridge	average 0.3 ± 0.3 (1σ)	

Exposure rates in the first data column were obtained by subtracting background from the gross values in Table 6.7 and then averaging the net values at each location. The natural radiation background was 7.5 μR/hr on June 4, 1970, 8.5 μR/hr over land and 6.5 μR/hr over water on Nov. 18, 1970, and 5.7 μR/hr on Feb. 8, 1971.

Values in the last column were computed by the equation:

$$R = 0.12\ D^{-2} \qquad (6.5)$$

where R is the radiation exposure rate in μR/hr and D is the distance in km from the center of the exclusion area. The constant in this inverse-square relation between radiation exposure and distance from the source was obtained from the net radiation exposures at points Q and R (i.e., the values in Table 6.7 minus 5.7 μR/hr). Radiation exposures along a radius through these two points were intermediate to one through points T, I, and G (where radiation exposures were higher by a factor of 1.4), and through points J, K, V, and W (where radiation exposures were lower by 1.4). No effort was made to extrapolate in the NE direction because directly measured exposure rates at the exclusion perimeter were significantly above background.

The extrapolated exposure rates are generally consistent with the directly measured values along Readsboro Road, where the largest number of measurements are available for comparison, but differ considerably at some other locations. On the whole, the two sets of values suggest that external radiation exposure rates were 3 μR/hr or less at points on the exclusion perimeter, and approximately 0.7 μR/hr along Readsboro Road. The average radiation exposure rate at the town of Monroe Bridge was 0.3 μR/hr, but the value is so low as to be very uncertain.

It would be of interest to obtain more accurate data on the external radiation exposure in the environs of the station, and to check on possible errors in the presented data due to counter calibration, background subtraction, or periodic variations in the intensity of the radioactive waste. Long-term exposure measurements with sensitive detectors such as thermoluminescent dosimeters appear feasible at locations A and C. At other off-site locations, it would be difficult to distinguish between radiation from the station and the natural background by long-term measurements, but it may be possible to do so by instantaneous measurements at selected locations. On the other hand, it may be simpler to reduce off-site exposure rates to natural background levels by improved shielding of stored wastes.

6.7.4 *Estimated external radiation exposure to persons in the environs.* The average instantaneous exposure rates listed in Section 6.7.3, multiplied by 8,760 hours per year, yield a radiation exposure value at the nearest habitation (location E in Figure 6.1) due to Yankee of 6 ± 3 milliroentgen per year (mR/yr), and 3 ± 3 mR/yr near the center of the town of Monroe Bridge. These values are subject to the uncertainties discussed in Section 6.7.3 and have not been corrected for shielding by house walls and time spent by persons at other locations. In comparison, the natural radiation background averaged 64 mR/yr, and its variation was greater than the radiation exposure attributed to Yankee. Exposures of travelers on Readsboro Road, fishermen on the southern end of Sherman Reservoir, and those who approached Yankee from the SSW or NE directions would undoubtedly have occurred during only a small fraction of the year, and accordingly been relatively low. The set of measurements suggests that distance and shielding by the terrain reduced radiation exposure from radioactive wastes stored at Yankee effectively to zero at distances of 2 km or more.

6.8 References

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7. Summary and Conclusions

7.1 Radionuclides in Yankee Effluents

Almost the entire radioactive content of effluents from Yankee consisted of ^{3}H, in accord with the station's operating reports. The other radionuclides discharged to the environment were mostly noble gas fission products in airborne effluents and activation products in liquid effluents. As points of interest, ^{14}C was found in relatively low amounts in both gaseous and liquid wastes, and no ^{131}I was detected in either gaseous or airborne particulate form. Radionuclides other than ^{3}H were discharged only in small amounts from the reactor system because liquid wastes were treated by evaporation and gaseous wastes, by storage. A considerable fraction of the radioactive effluent was contributed by the direct release of airborne and liquid wastes at the secondary coolant system during leakage from the main into the secondary system at the steam generators.

The information on effluent radioactivity that is summarized below and presented in detail in Sections 3 and 4 was obtained in several sampling periods between October 1968 and February 1971. Annual releases were estimated for individual radionuclides by averaging measured concentrations in samples collected at the same points and multiplying these averages by the volumes released annually, as reported by the station. The annual release estimates are presented to indicate magnitudes and permit comparison with other data, but are not based on sufficient samples to be considered accurate records of annual releases. It is expected that detailed and continuous isotopic discharge data will, in the future, be obtained by station operators in response to recent AEC regulations.

The estimated amounts of radioactive gases discharged annually through the stack from four sources at the station were as follows:

Radionuclides in Gaseous Effluent, Ci/yr

	Main coolant sampling	Air ejector	Surge drum	Vapor container
^{3}H	5×10^{-4}	4×10^{-3}	1×10^{-2}	13
^{14}C	2×10^{-3}	1×10^{-2}	1×10^{-1}	2×10^{-1}
^{41}Ar	4×10^{-1}	$<1 \times 10^{-1}$	NA*	NA
^{85m}Kr	2×10^{-2}	NA	NA	NA
^{85}Kr	6×10^{-4}	$<4 \times 10^{-4}$	1×10^{-2}	3
^{87}Kr	2×10^{-2}	NA	NA	NA
^{88}Kr	3×10^{-2}	NA	NA	NA
^{133m}Xe	2×10^{-3}	$<6 \times 10^{-2}$	$<3 \times 10^{-4}$	NA
^{133}Xe	1×10^{-1}	4×10^{-2}	8×10^{-3}	ND
^{135}Xe	7×10^{-2}	1×10^{-1}	NA	NA

*NA = not analyzed; ND = not detected.

Main coolant sampling usually occurs once daily and results in the release of a small volume of gas from the main coolant system. The air ejector at the main condenser in the secondary coolant system continuously discharges noncondensable gases, including radioactive ones that have leaked from the main coolant system. The surge drum collects hydrogen-bearing waste gases from the main coolant and is usually discharged once yearly. Vapor container air includes radioactive gases that leaked from the main coolant system during reactor operation, and emanated during refueling; it is discharged several times yearly when the vapor

container is opened for inspection, repairs, or refueling.

Most of the gaseous radioactivity was released while ventilating the vapor container. Gaseous radioactive effluents from the third and fourth sources consist mostly of longer-lived radionuclides, while the radionuclides from the first and second sources were relatively short-lived. In addition to the measured radionuclides, it was estimated that approximately 3 Ci of short-lived noble gas fission products (^{89}Kr, ^{135m}Xe, ^{137}Xe, and ^{138}Xe) were released annually at the first and second sources. Particulate radionuclides were at very low concentrations; the total based on analyses of filters in ventilating-air and incinerator stack effluents was less than 1×10^{-3} Ci/yr. No ^{131}I ($< 3 \times 10^{-4}$ Ci/yr) in gaseous or particulate form was detected.

The estimated total annual releases of 13 Ci ^{3}H and 4 Ci of all other radionuclides are consistent with releases reported by Yankee for 1969 of 9.19 Ci ^{3}H and 4.13 Ci gross beta-gamma activity. These amounts are considerably below the most restrictive annual release limit of 4.5×10^{3} Ci for individual radionuclides.

The estimated amounts of radionuclides discharged annually into effluent circulating coolant water were as follows:

Reactor plant wastes are treated by evaporation and then discharged in batches; secondary plant effluents are mostly steam-generator blowdown and leakage water, discharged directly and without delay. Some radionuclides were also in effluent yard-drain water.

Tritium was at highest concentration in both wastes. The most prevalent radionuclides after ^{3}H, at far lower concentrations, were the activation products ^{14}C, ^{51}Cr, ^{54}Mn, ^{55}Fe, and ^{58}Co. Shorter-lived radionuclides (half life $<$ 8 days) than those measured could also be in secondary plant effluent; ^{24}Na, for example, was estimated to be released at the rate of 3 Ci/yr. In yard-drain water, if observed concentrations and flow rates were typical, annual releases were of the magnitude of 0.1 Ci ^{3}H, 5×10^{-4} Ci ^{60}Co, and lower for other radionuclides.

The sum of ^{3}H releases--800 Ci/yr--agrees with the value of 1,048 Ci/yr in 1969 reported by Yankee, but the sum of all other radionuclides--0.08 Ci/yr--is higher than the reported gross beta-gamma activity of 0.019 Ci/yr. The AEC limit of 82 Ci/yr for discharging the most hazardous radionuclides listed above--^{90}Sr and ^{131}I--is many orders of magnitude higher than the indicated releases of all radionuclides except ^{3}H. Tritium releases approach the limits most closely; these are 84,000 Ci/yr according to AEC regulations and 3,650 Ci/yr for the station according

Radionuclides in Liquid Effluent, Ci/yr

	Reactor plant	Secondary plant
^{3}H	6×10^{2}	2×10^{2}
^{14}C	1×10^{-2}	1×10^{-3}
^{32}P	8×10^{-5}	ND*
^{51}Cr	ND	2×10^{-2}
^{54}Mn	2×10^{-3}	9×10^{-3}
^{55}Fe	1×10^{-2}	2×10^{-4}
^{59}Fe	6×10^{-4}	4×10^{-3}
^{58}Co	4×10^{-4}	1×10^{-2}
^{60}Co	2×10^{-4}	3×10^{-3}
^{63}Ni	ND	1×10^{-3}
^{90}Sr	3×10^{-5}	6×10^{-5}
^{95}Zr	ND	4×10^{-3}
^{95}Nb	ND	3×10^{-3}
^{110m}Ag	3×10^{-4}	8×10^{-4}
^{124}Sb	8×10^{-5}	2×10^{-3}
^{131}I	2×10^{-4}	4×10^{-3}
^{137}Cs	1×10^{-4}	1×10^{-4}

*ND = not detected.

to the Massachusetts Department of Public Health.

Thus, all radionuclides were released in quantities well below AEC limits. These releases at Yankee of radionuclides other than ^{3}H are approximately two orders of magnitude lower than in liquids and gases at other commercially operated full-scale PWR nuclear power stations. Tritium releases appear to be typical of the power level at PWR stations with stainless steel fuel cladding.

7.2 Radionuclides in the Environment of Yankee

Radionuclides from Yankee were found in the aquatic environment at low concentrations (see Section 5), but not at all in the terrestrial environment (Section 6). That these radionuclides could be detected in sediment and aquatic vegetation at Yankee, despite the relatively low radioactivity level in its liquid effluent, suggests that they can be found at most other nuclear power stations.

At the point of discharge of circulating coolant water into the Sherman Reservoir, ^{3}H was at a concentration of 79 pCi/ml during release of reactor-plant liquid waste. The measured concentration agreed with the value computed from the measured concentration in the waste and the 4,700-fold dilution by circulating coolant water. At the same time, ^{3}H was also measured downstream from the outfall at considerably lower concentrations. Other radionuclides in the waste could not be detected at the point of discharge because their concentrations were too low.

Benthal material (sediment) from Sherman Reservoir within approximately 200 m of the outfall of circulating coolant water has accumulated the following radionuclides from Yankee liquid wastes:

Radionuclides in Sediment, pCi/g dry wt.

	highest concentration	background
5.26-yr ^{60}Co	32	<0.1
28.5 -yr ^{90}Sr	0.6	0.1
30 -yr ^{137}Cs	6	0.7
313 -d ^{54}Mn	2	<0.1
2.77-yr ^{125}Sb	0.9	0.2

The sediment was estimated to contain approximately 7 mCi each of ^{60}Co and ^{137}Cs, probably a considerable fraction of the total release of the two radionuclides during the 10-year life of the station.

Water moss on rock at the outfall and dead leaves submerged in water at the nearby shore of Sherman Reservoir also contained radionuclides attributed to Yankee. Radionuclide concentrations were higher by four or five orders of magnitude than estimated concentrations in water. The values in the one sample of each that was collected were as follows:

Radionuclides in Aquatic Vegetation, pCi/g wet wt.

	water moss	dead leaves
313 -d ^{54}Mn	1.8	0.1
71.3 -d ^{58}Co	0.3	not detected
5.26-yr ^{60}Co	0.9	0.3

In fish from Sherman Reservoir, the average ^{22}Na concentration in muscle ranged from 2.0 to 3.1 pCi/kg wet weight among four sampling categories, compared to averages of 0.5 to 1.9 pCi/kg in background samples. The difference in ^{22}Na concentration may be due to waste discharges by Yankee, although fish with higher ^{22}Na concentrations than in Sherman Reservoir fish have been found elsewhere.

No radioactivity attributed to Yankee could be observed in suspended solids, including plankton, from Sherman Reservoir. These samples were of relatively small volume, however, because the water was low in suspended solids.

No radioactivity attributed to Yankee was found in the following terrestrial samples:

- snow within the station perimeter at Yankee
- vegetation and soil just beyond the Yankee perimeter
- milk from a dairy at Rowe
- deer that had died accidentally within 3 km of Yankee.

Computations based on measured effluent concentrations and a simple model of dispersion in air indicated that radionuclide concentrations in air and on the ground near Yankee were so low that they could not be detected with the available sample volumes and analytical procedures.

External radiation measurements with survey meters yielded an exposure rate above background of 1 to 3 μR/hr at the 0.3-km perimeter at Yankee, 0.7 $\pm$ 0.3 μ R/hr at the nearest habitation (0.4 km distant on the west side of Sherman Reservoir), and 0.3 $\pm$ 0.3 μR/hr at Monroe Bridge (1.1 km distant). The natural

radiation background at somewhat greater distances ranged from 5.7 to 8.5 μR/hr, depending on the time of year. The radiation flux above background is believed to have been gamma rays from radioactive waste stored at Yankee (Section 6.7).

On the basis of these measurements in the environment, the radiation exposure from Yankee to persons living approximately 1 km distant was 3 $\pm$ 3 mR/yr due to direct radiation. The natural background radiation in the area is approximately 64 mR/yr. Radiation exposure from this source to persons living at greater distances would be essentially zero because of the terrain and distance. The radiation dose from Yankee to avid fishermen and fish eaters through ingesting fish caught at the southern end of Sherman Reservoir was 0.3 mrem/yr as inferred from effluent radioactivity data, and was considerably less on the basis of direct radionuclide analyses of fish muscle. The radiation dose from stack effluent was estimated to be 0.4 mrem/yr at the Yankee exclusion boundary. Thus, operation of the Yankee nuclear power station under the observed conditions had an extremely small impact on the radiation dose in the environment. The direct exposure rate was so far below the natural radiation background that it could not be measured with certainty, while inferred radiation doses by two other pathways were each only a fraction of one mrem/yr. No other exposure pathway was observed.

7.3 Monitoring Procedures

The following techniques, in addition to those reported earlier in the study at Dresden, were demonstrated:

(1) measurement of radionuclides that emit only low-energy beta particles, specifically ^{14}C and ^{63}Ni;

(2) measurement of total ^{3}H in air, as distinguished from ^{3}H as water vapor;

(3) measurement of radionuclides in aquatic vegetation;

(4) use of a NaI(Tl) detector plus multichannel analyzer as survey instrument for detecting photon-emitting radionuclides in the benthos, and comparison of survey data with measured concentrations in silt;

(5) comparison of benthal sample collection by hand (diver) and by dredge.

(6) measurement of radiation exposure at low levels from gamma rays emitted at the station.

7.4 Recommendations

The fundamental recommendation for radiological surveillance programs by nuclear power stations, based on observations in this study and the one at the Dresden I BWR, is that all radioactive effluents be analyzed to obtain in detail their radionuclide content. After the radioactive constituents have been identified, analyses can be limited to the radionuclides at highest abundance and of greatest health significance. Once the pattern of radionuclide discharges has been observed, the frequency of analysis can also be reduced. Significant changes in station operation or the radionuclide content of effluents require at least a brief return to more detailed analyses. These radionuclide discharge data provide the basis for estimating population exposure, planning environmental surveillance, and treating wastes at the station. Such data will, in the future, be available from the stations in response to recent AEC regulations.

At a nuclear power station such as Yankee, where few pathways for population exposure exist because of the remote location and very low amounts of discharged radionuclides (except ^{3}H in liquids), a small-scale surveillance program will provide sufficient information if effluent radioactivity is rigorously monitored. The following environmental (offsite) measurements can be suggested:

(1) external radiation exposure measured continuously at off-site locations of potential personal exposure;

(2) radionuclide analyses of fish caught in the southern end of Sherman Reservoir and immediately below Sherman Reservoir, at times when fishermen are active. Radiochemical analysis of edible portions for ^{3}H and ^{14}C content and gamma-ray spectral analysis of large amounts of the same sample are of particular interest.

(3) occasional analyses of other foods from the immediate vicinity of Yankee, including wild life, milk, fruit and vegetables (if any), and maple syrup;

(4) occasional measurements of the radionuclide content of benthal samples

and aquatic vegetation for comparison with radionuclide concentrations in fish, to determine whether the radionuclides deposited in the sediment enter the food chain.

The program of environmental surveillance must be evaluated periodically to consider modifications in response to changes in effluent radioactivity, new patterns of population distribution and environmental use, and increased knowledge of the behavior of radionuclides in the environment.

At nuclear power stations that discharge more radioactivity than Yankee, more extensive environmental surveillance will usually be found desirable. In addition, studies to relate concentrations of radionuclides along critical environmental pathways for human radiation exposure to the release rates of these radionuclides will often be useful. The radionuclide acts as tracer to quantify transfer coefficients from station to man, providing a better and more pertinent basis for calculating exposures at the site than most published values. Such studies may need to be performed only once. At Yankee, the only radionuclide that appears feasible as such a tracer is ^{3}H in water.

The EPA research program of which this study is a part is being continued through field studies at the newer and larger nuclear power stations. In total, these field studies should indicate the degree to which release data are generally applicable, the influence of the environment and station size, design, and operating practices on human radiation exposure, and the need for studying specific environmental pathways for radionuclides.

Appendix A

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Appendix B.1

Main Coolant Data from Yankee Nuclear Power Station Monthly Operating Reports*

Month	Average Power, MWe	Average** Boron Concentration, ppm	Average Tritium Concentration, μCi/ml	Comments
June 1967+	176	586	2.92	
July	73	477	~1.	Maintenance shutdown 7/8-7/25
August	173	375	1.36	
September	174	235	2.40	
October	170	125	1.56	
November	175	4	1.30	Dilution for boron removal 11/3
December	171	0	1.82	
January 1968	156	0	1.67	
February	141	0	1.42	
March	94	0	1.22	
April	0	~2800	0.08	Refueling shutdown 3/23-5/1
May	175	<1500	<3.61	
June	179	1103	3.81	
July	165	978	4.47	
August	177	853	4.86	
September	163	721	4.36	
October	182	585	4.21	
November	143	445	2.07	Maintenance shutdown 11/8-11/15
December	143	317	2.25	
January 1969	179	187	2.24	
February	177	61	1.20	Dilution for boron removal 2/18
March	181	0	1.29	
April	143	9	~0.9	Maintenance shutdown 4/11-4/16
May	156	0	0.531	Primary-Secondary leakage
June	130	0	0.429	Primary-Secondary leakage
July	115	0	0.364	
August	4	2893	0.040	Refueling shutdown 8/2-9/25
September	34	2417	2.20	
October	173	1101	3.05	
November	185	979	3.93	
December	185	870	3.61	
January 1970	185	722	2.57	
February	175	610	1.93	
March	127	538	1.63	Maintenance shutdown 3/21-3/29
April	183	395	1.16	
May	167	305	1.80	Primary-Secondary leakage
June	169	183	1.29	Primary-Secondary leakage
July	177	2	0.83	Dilution for boron removal 7/1
August	116	~330	1.15	Maintenance shutdown 8/21-8/31 Primary-Secondary leakage
September	158	1	0.30	
October	112	0.	n.r.++	
November	3	2760	0.03	Refueling shutdown 10/24-11/30
December	181	1525	1.37	
January 1971	184	1270	3.12	
February	176	1110	3.92	

*Data reviewed and corrected by Yankee staff.

**Calculated weighted average from reported concentrations for given portions of the month, or as a mean where only maximum and minimum values were reported.

+Tritium concentration in main coolant only reported sporadically before June, 1967.

++n.r. - not reported

Appendix B.2

Radioactive Waste Discharge Data from Yankee Nuclear Power Station Monthly Operating Reports

	Liquid Releases						Gaseous Releases	
	Waste Disposal			Secondary Plant				
Month	Volume, 10^5 l	Gross β-γ, μCi	Tritium*, Ci	Volume, 10^5 l	Gross β-γ, μCi	Tritium, Ci	β-γ, mCi	Tritium, Ci **
November 1966	2.5	370	32	4.2	< 5	2.5	495	0.3
December	7.4	22,410	194	6.4	35.9	30.5	134	n.r.+
January 1967	8.8	5,000	219	10.5	48	6.7	196	n.r.
February	10.1	19,460	65	8.1	52	14.7	148	n.r.
March	7.5	11,360	53	6.0	32	9.9	902	8.97
April	10.8	786	78	6.5	10.9	2.3	166	0.06
May	9.8	2,330	132	9.5	99	16.4	150	n.r.
June	7.3	5,150	180	11.1	47	45.9	421	n.r.
July	4.9	2,380	44	6.6	28	29.5	90	6.05
August	7.0	574	94	9.0	7.5	0.3	17	n.r.
September	11.5	3,010	126	11.2	11.6	0.2	78	n.r.
October	8.2	4,420	51	8.6	6.9	0.1	111	n.r.
November	5.4	195	225	7.2	6.3	0.1	7	n.r.
December	6.1	112	195	7.0	10.2	0.2	26	n.r.
January 1968	1.1	53	34	14.8	15.1	0.4	14	n.r.
February	0.7	31	27	11.0	5.0	0.3	20	0.10
March	2.1	73	96	8.2	5.1	0.3	33	4.09
April	3.8	164	100	12.7	7,640	0.1	n.r.	1.90
May	2.4	104	57	10.7	31	0.2	20	n.r.
June	1.2	17	42	7.1	4	0.3	14	n.r.
July	1.1	27	66	10.6	6	0.9	28	0.05
August	1.2	11	126	8.9	5	0.9	99	n.r.
September	0.6	5	48	10.9	17	1.2	76	n.r.
October	1.2	18	150	11.8	4	3.4	84	n.r.
November	2.3	53	229	11.4	11	3.8	107	1.91
December	2.2	74	184	8.7	3	0.7	168	n.r.
January 1969	1.9	35	98	12.2	9.9	1.7	147	n.r.
February	1.2	19	101	10.4	90	1.4	158	0.16
March	2.1	87	191	19.7	478	16.6	162	0.06
April	3.4	168	205	16.0	458	10.5	358	4.96
May	2.0	61	88	13.3	6,000	22.0	817	n.r.
June	1.6	59	28	18.0	4,760	17.2	445	0.13
July	2.2	153	26	9.4	1,150	6.3	311	4.5×10^{-4}
August	4.5	124	45	2.1	450	2.8	24	2.23
September	3.1	60	30	4.7	130	0.7	135	1.39
October	2.1	36	76	15.5	466	12.8	523	1.16
November	0.9	29	75	7.8	1,440	35.3	263	0.01
December	0.8	55	86	8.8	2,900	48.1	790	2.9×10^{-4}

*Monthly reports of tritium liquid waste discharge began in March, 1965.

**Comments on gaseous releases of tritium:

Reported as "gaseous waste releases" in March, 1967, and as "gaseous releases" in February, March, April, June, August, November, and December, 1969; January, February, March, June, August, September and October, 1970.

Reported as "a vapor, from the vapor container" in November, 1966; March and July, 1967; March, April, July, and November, 1968; April, July, August, September, and October, 1969; March, August, October, and November, 1970.

Reported as "an inadvertent gas release" in April, 1967, and February, 1968.

A 62-mCi release during main steam line safety valve test was reported in June, 1969.

n.r. - not reported

Notes: 1. Core lifetimes:

Core VI - November 8, 1966 - March 23, 1968

Core VII - May 1, 1968 - August 2, 1969

Core VIII - September 5, 1969 - October 24, 1970

2. Data reviewed and corrected by Yankee staff.

Radioactive Waste Discharge Data from Yankee Nuclear Power Station Monthly Operating Reports (cont'd).

January 1970	1.4	25	143	14.9	4,390	46.6	1,462	1.9×10^{-5}
February	2.2	34	96	15.8	4,240	44.7	2,054	4.5×10^{-5}
March	2.8	46	86	17.4	10,590	40.8	1,731	3.79
April	3.4	60	239	16.2	265	3.0	714	n.r.
May	1.0	15	47	12.4	1,590	8.5	1,628	n.r.
June	2.5	41	126	14.2	3,790	18.3	1,549	0.08
July	4.0	65	174	9.9	2,758	33.8	4,659	n.r.
August	3.0	44	90	11.5	4,432	73.8	1,743	3.94
September	4.5	161	103	15.1	261	4.1	304	0.03
October	2.5	87	48	12.6	316	5.1	145	0.54
November	2.0	59	14	3.5	382	0.5	25	0.62
December	1.5	55	47	15.0	125	0.3	439	n.r.
January 1971	2.3	56	146	15.3	35	0.5	392	n.r.
February	1.7	25	145	14.8	191	1.1	508	n.r.

Appendix B.3

Estimated Generation Rate of Fission Products in Fuel at 600 MWt Power

Product	Fission yield, Y*	Decay constant, λ, sec^{-1}	Generation rate, μCi/sec	Accumulation in 2 years, μCi
^{3}H	9.5×10^{-5} +	1.78×10^{-9}	8.5×10^{1}	5.9×10^{9}
^{85}Kr	2.9×10^{-3}	2.05×10^{-9}	3.0×10^{3}	1.8×10^{11}
^{85m}Kr	1.3×10^{-2}	4.37×10^{-5}	2.9×10^{8}	6.5×10^{12}
^{89}Sr	4.5×10^{-2}	1.57×10^{-7}	3.6×10^{6}	2.7×10^{13}
^{90}Sr	5.9×10^{-2}	7.82×10^{-10}	2.3×10^{4}	1.7×10^{12}
^{91}Sr	5.8×10^{-2}	1.98×10^{-5}	5.8×10^{8}	3.4×10^{13}
^{95}Zr	6.3×10^{-2}	1.23×10^{-7}	3.9×10^{6}	3.7×10^{13}
^{95}Nb	6.3×10^{-2}	2.29×10^{-7}	8.5×10^{6}**	8.1×10^{13}
^{99}Mo	6.1×10^{-2}	2.90×10^{-6}	8.9×10^{7}	3.6×10^{13}
^{131}I	2.9×10^{-2}	9.96×10^{-7}	1.5×10^{7}	1.7×10^{13}
^{133}I	6.5×10^{-2}	9.21×10^{-6}	3.0×10^{8}	3.8×10^{13}
^{135}I	6.0×10^{-2}	2.87×10^{-5}	8.6×10^{8}	3.5×10^{13}
^{133}Xe	6.6×10^{-2}	1.52×10^{-6}	5.0×10^{7}	3.3×10^{13}
^{133m}Xe	1.6×10^{-3}	3.5×10^{-6}	2.8×10^{6}	8.0×10^{11}
^{135}Xe	6.3×10^{-2}	2.11×10^{-5}	6.7×10^{8}	3.2×10^{13}
^{137}Cs	5.9×10^{-2}	7.30×10^{-10}	2.2×10^{4}	1.5×10^{12}
^{140}Ba	6.6×10^{-2}	6.26×10^{-7}	2.1×10^{7}	3.9×10^{13}

* Harley, N., I. Fisenne, L. D. Y. Ong, and J. Harley, "Fission Yield and Fission Product Decay" in AEC Rept. HASL 164 (1965), p. 251; Russell, I. J. and R. V. Griffith, "The Production of ^{109}Cd and ^{113m}Cd in a Space Nuclear Explosion" in AEC Rept. HASL 142 (1964) p. 306.

+Albenesius, E. L. and R. S. Ondrejein, "Nuclear Fission Produces Tritium", Nucleonics *18* (9), 199 (1960).

**Equilibrium with longer-lived parent is assumed.

Notes:

1. Generation rate = thermal power $\times \frac{\text{fission rate}}{\text{MWt}} \times Y \times \lambda$

$$= 600 \text{ MWt} \times \frac{3.1 \times 10^{16} \text{ fission/sec}}{\text{MWt}} \times Y \times \lambda \times \frac{\mu\text{Ci}}{3.7 \times 10^{4} \text{ dis/sec}}$$

2. Accumulation = thermal power $\times \frac{\text{fission rate}}{\text{MWt}} \times Y \times (1-e^{-\lambda t})$

Appendix B.4

Estimated Turnover Rate of Ionic Fission Products in Main Coolant Water Based on Concentration Measurements, and Ratio of Turnover Rate to Generation Rate

Fission Product	λ decay + λ turnover, sec^{-1}	Turnover rate, μ Ci/sec	Turnover rate/ Generation rate
^{89}Sr	3.0×10^{-5}	1.3×10^{-2}	3.6×10^{-9}
^{90}Sr	3.0×10^{-5}	3.8×10^{-4}	1.6×10^{-8}
^{91}Sr	5.0×10^{-5}	3.1×10^{-1}	5.3×10^{-10}
^{95}Zr	3.0×10^{-5}	8.6×10^{-2}	2.2×10^{-8}
^{95}Nb	3.0×10^{-5}	9.6×10^{-2}	1.1×10^{-8}
^{99}Mo	3.3×10^{-5}	2.3×10^{-1}	2.6×10^{-9}
^{131}I	3.1×10^{-5}	1.0×10^{-1}	6.7×10^{-9}
^{133}I	3.9×10^{-5}	1.6	4.0×10^{-9}
^{135}I	5.9×10^{-5}	3.4	4.0×10^{-9}
^{137}Cs	3.0×10^{-5}	3.8×10^{-4}	1.7×10^{-8}
^{140}Ba	3.1×10^{-5}	2.4×10^{-2}	1.1×10^{-9}
Avg.			8.2×10^{-9}

Notes:

1. $\lambda_{\text{turnover}} = \frac{113 \text{ kg/min}}{64{,}000 \text{ kg}} \times \frac{\text{min}}{60 \text{ sec}} = 3.0 \times 10^{-5} \text{ sec}^{-1}$
2. Turnover rate = Concentration x coolant amount x ($\lambda_{\text{decay}} + \lambda$ turnover)
   = Concentration in μ Ci/g x 6.4×10^{7} g x ($\lambda_{\text{decay}} + 3.0 \times 10^{-5}$) sec^{-1}
3. Concentrations from Table 2.1 for sample of Oct. 4, 1968.
4. Generation rate and λ_{decay} from Appendix B.3.

Appendix B.5

Estimated Turnover Rate of Longer-lived Ionic Activation Products in Main Coolant Water Based on Concentration Measurements

Activation product	Turnover rate, μCi/sec
^{32}P	1.9×10^{-2}
^{51}Cr	1.6
^{54}Mn	1.0
^{55}Fe	1.9×10^{-1}
^{59}Fe	3.6×10^{-1}
^{57}Co	$\sim 1 \times 10^{-3}$
^{58}Co	6.5×10^{-1}
^{60}Co	1.7×10^{-1}
^{110m}Ag	3.6×10^{-2}
^{124}Sb	3.8×10^{-2}
^{181}Hf	$\sim 2 \times 10^{-2}$
^{182}Ta	$\sim 1 \times 10^{-1}$
^{185}W	1.9×10^{-2}

Notes:

1. Concentrations fromTable 2.1 for sample of Oct. 4, 1968.
2. $\lambda_{\text{turnover}} = 3.0 \times 10^{-5}$ sec.$^{-1}$; $\lambda_{\text{decay}} \leq 5 \times 10^{-7} sec^{-1}$ for all listed radionuclides.
3. See footnote 2 to Appendix B.4 for calculation of turnover rate.

Appendix C.1

Test Conditions and Calculations for Sampling Yankee Stack Effluent in Environment, June 3, 1969

Period	1	2
Hours (EDT)	1600-1700	1700-1800
Mean temperature (°F)	73	72
Mean wind speed, m/sec	4.0	5.2
Solar radiation	moderate	moderate
Stability category (Pasquill-Gifford)	B	C
Normalized concentration, m^{-2}	7×10^{-5}	4×10^{-5}
^{85}Kr Release rate (Table 3.6), pCi/sec	4.2×10^{5}	4.2×10^{5}
Computed ^{85}Kr concentration, pCi/m^3	7	3

Appendix C.2

Radionuclide Deposition Parameters

Location	#201	#202	#203	#204	Dairy farm
Distance, m	260	230	450	8000	3100
Azimuth, deg.	046	270	336	180	135
Wind azimuth, deg.	226	090	156	0	315
Mean wind frequency in 20° sector					
unstable	0.060	0.012	0.008	0.012	0.034
neutral	0.045	0.010	0.003	0.019	0.018
Mean wind speed, m/sec					
unstable	3.6	2.2	2.2	4.4	4.9
neutral	3.6	3.1	1.8	4.0	4.0
Standard deviation [a], m					
unstable (A)	35	35	90	1000[b]	1000[b]
neutral (C)	16	16	30	160[b]	160[b]

[a] Letters A and C refer to Pasquill-Gifford Stability Class.
[b] Estimated

Appendix C.3

Computed Accumulation of ^{90}Sr in Snow during March 1969

Location	#202	#204
Deposition, pCi/m^2		
Snow	$2.45 \times 10^{-3} Q_o'$	$7.03 \times 10^{-5} Q_o'$
Dry, unstable	$0.52 \times 10^{-3} Q_o'$	$0.005 \times 10^{-5} Q_o'$
Dry, neutral	$0.03 \times 10^{-3} Q_o'$	$0.001 \times 10^{-5} Q_o'$
Total	$3.0 \times 10^{-3} Q_o'$	$7.0 \times 10^{-5} Q_o'$
Area sampled, m^2	3.0	2.3
Sampled activity, pCi	$9.0 \times 10^{-3} Q_o'$	$16.1 \times 10^{-5} Q_o'$
Volume of melted snow, l	16.6	7.6
Concentration[a], pCi/l	$5.4 \times 10^{-4} Q_o'$	$2.1 \times 10^{-5} Q_o'$
	4.3×10^{-3}	1.7×10^{-4}

[a] Value based on average $Q_o' = 8$ pCi/sec in Section 3.3.5.

Appendix C.4

Computed ^{90}Sr Accumulation in Grass, April-May 1969

Location	#201	#202	#203	#204	Dairy farm
Deposition, pCi/m^2					
Dry, unstable	$2.8 \times 10^{-2} Q_o'$	$1.06 \times 10^{-2} Q_o'$	$2.82 \times 10^{-3} Q_o'$	$0.12 \times 10^{-4} Q_o'$	$1.78 \times 10^{-4} Q_o'$
Dry, neutral	$0.21 \times 10^{-2} Q_o'$	$0.06 \times 10^{-2} Q_o'$	$3.81 \times 10^{-3} Q_o'$	$1.27 \times 10^{-4} Q_o'$	$3.11 \times 10^{-4} Q_o'$
Precipitation, April	$0.05 \times 10^{-2} Q_o'$	$0.02 \times 10^{-2} Q_o'$	$0.06 \times 10^{-3} Q_o'$	$0.04 \times 10^{-4} Q_o'$	$0.17 \times 10^{-4} Q_o'$
Precipitation, May	$0.03 \times 10^{-2} Q_o'$	$0.01 \times 10^{-2} Q_o'$	$0.03 \times 10^{-3} Q_o'$	$0.02 \times 10^{-4} Q_o'$	$0.08 \times 10^{-4} Q_o'$
Total	$3.2 \times 10^{-2} Q_o'$	$1.2 \times 10^{-2} Q_o'$	$6.7 \times 10^{-3} Q_o'$	$1.5 \times 10^{-4} Q_o'$	$5.1 \times 10^{-4} Q_o'$
Remaining at sampling time,[a] pCi/m^2	$6.1 \times 10^{-3} Q_o'$	$2.3 \times 10^{-3} Q_o'$	$1.3 \times 10^{-3} Q_o'$	$2.9 \times 10^{-5} Q_o'$	$9.7 \times 10^{-5} Q_o'$
In grass ash,[b] pCi/g	$2.7 \times 10^{-4} Q_o'$	$1.0 \times 10^{-4} Q_o'$	$5.7 \times 10^{-5} Q_o'$	$1.3 \times 10^{-6} Q_o'$	$4.2 \times 10^{-6} Q_o'$
	2.2×10^{-3}	8.0×10^{-4}	4.6×10^{-4}	1.0×10^{-5}	3.4×10^{-5}

[a]Based on 14-day environmental half life from mid-time of period (May 1) to collection date (June 3), the decay factor is 0.19

[b]1 m^2 of vegetation was assumed to yield 23 g ash; Q_o' = 8 pCi/sec in Section 3.3.5

Note: There were 90 hr of precipitation in April and 44 hr in May.

Appendix C.5

Computed Long-term ^{90}Sr Accumulation in Soil

Location	#201	#202	#203	#204
Annual Deposition, pCi/m^2				
Dry, unstable	$0.175 Q_o'$	$0.065 Q_o'$	$0.0172 Q_o'$	$0.000067 Q_o'$
Dry, neutral	$0.013 Q_o'$	$0.004 Q_o'$	$0.0089 Q_o'$	$0.000774 Q_o'$
Precipitation[a]	$0.004 Q_o'$	$0.001 Q_o'$	$0.0005 Q_o'$	$0.000037 Q_o'$
Total	$0.19 Q_o'$	$0.070 Q_o'$	$0.027 Q_o'$	$0.00088 Q_o'$
8-yr accumulation,[b] pCi/m^2	$1.4 Q_o'$	$0.51 Q_o'$	$0.19 Q_o'$	$0.0064 Q_o'$
In top 2-cm layer,[c] pCi/m^2	$0.35 Q_o'$	$0.13 Q_o'$	$0.048 Q_o'$	$0.0016 Q_o'$
In soil,[d] pCi/g	$1.2 \times 10^{-5} Q_o'$	$4.3 \times 10^{-6} Q_o'$	$1.6 \times 10^{-6} Q_o'$	$5.3 \times 10^{-8} Q_o'$
	9.6×10^{-5}	3.4×10^{-5}	1.3×10^{-5}	4.2×10^{-7}

[a]Based on precipitation of 754 hr/yr during 1968 and 1969.

[b]Corrected for decay of ^{90}Sr.

[c]One-fourth of the accumulation, assuming that one-half is removed from the soil and one-half is below the top 2 cm.

[d]For dry density of 1.5 g/cm^3 and 2-cm sampling depth, 1 m^2 surface area corresponds to 3×10^4 gms soil; value based on average Q_o' = 8 pCi/sec in Section 3.3.5.

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A radiological surveillance study was undertaken at the Yankee Nuclear Power Station to make available information for calculating population radiation exposures at routinely operating commercial PWR stations and to demonstrate effective monitoring procedures. Radionuclide concentrations and external radiation were measured in the immediate environment of the station. At the same time, the radionuclide contents of liquids and gases at the station and of effluents at points of discharge were measured, and levels of environmental radioactivity were estimated from these values.

The radioactivity in effluents at Yankee consisted mostly of ^{3}H, in amounts typical of PWR stations that use fuel clad in stainless steel. The amounts of other radionuclides discharged to the environment from the reactor plant were very small, apparently because of effective containment of fission products other than ^{3}H within the fuel elements and treatment of wastes by storage (for radioactive decay) and evaporation. A considerable fraction of the effluent radioactivity was discharged at the secondary coolant system because these effluents are released without treatment.

In the environment, radionuclides from Yankee were found only in the aquatic environment, at low concentrations. The detected radionuclides do not appear to constitute significant direct radiation exposure to the population; and radiation doses inferred from radionuclide measurements in liquid and gaseous wastes were less than 1 mrem/year through all pathways that were considered. Measurements of external radiation exposure in the environment suggested that a small increment above the natural radiation background was due to gamma rays emitted by wastes stored at Yankee.

KEY WORDS:

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