RADIOLOGICAL EMISSIONS AND ENVIRONMENTAL MONITORING FOR BROOKHAVEN NATIONAL LABORATORY, 1947 - 1961

C.B. Meinhold and A.F. Meinhold
Edited by P. D. Bond

FINAL
May 2001

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NATIONAL LABORATORY

Brookhaven Science Associates
Brookhaven National Laboratory
Upton, New York 11973-5000

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ACKNOWLEDGEMENTS

This work involved an extensive review and analysis of historical data dating back to the early history of the laboratory. Many people helped to track down and interpret these records. The authors are particularly indebted to Victor Cassella for his assistance in retrieving and interpreting historical meteorological data. We thank Mr. Henry Hauptman, Mr. Gerard Shepherd, Ms. Maria Beckman, Ms. Anne Battinger, Ms. Louisa Morrison, Mr. Manny Lilimpakis, and Dr. Casper Sun for their help. Ms. Grace Webster and Ms. Gina Bernard supported the preparation of this final report. A number of employees reviewed drafts of this report and made extremely helpful comments and suggestions. This work was supported by Ms. Gail Penny, DOE/BHG EM Program Manager.

We would also like to thank several retired employees who shared their knowledge and experience at the Brookhaven Graphite Research Reactor in the early years of its operation. Mr. Leigh Phillips, Mr. Jack Phillips, Mr. Ernest Guettinger and Mr. Robert Wayte attended a workshop held at BNL and provided much useful information on its operation and on the ruptures of the fuel cartridges.

The authors would like to dedicate this report to Andrew P. Hull. His untimely death robbed us of a trusted friend and thoughtful scientist. The quality of the report is diminished by his absence, as are we.

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EXECUTIVE SUMMARY

Introduction

Brookhaven National Laboratory (BNL) has monitored its releases to the environment since its inception in 1947. From 1962 to 1966 and from 1971 to the present, annual reports were published that recorded the emissions and releases to the environment from Laboratory operations. In 1998, a report was written to summarize the environmental data for the years 1967 to 1970. One of the purposes of the current report is to complete BNL's environmental history by covering the period from 1948 through 1961. The activities in 1947 were primarily organizational and there is no information on the use of radiation at the Laboratory before 1948. An additional objective of this report is to provide environmental data to the Agency for Toxic Substances and Disease Registry (ATSDR). The report does not provide an estimate of the doses associated with BNL operations.

The report is comprised of two parts. The first part is a summary of emissions, releases, and environmental monitoring information including a discussion of the uncertainties in these data. Part two contains the detailed information on the approach taken to estimate the releases from the fuel cartridge failures at the Brookhaven Graphite Research Reactor (BGRR). A series of appendices present more detailed information on these events in tabular form.

The approach in this report is to be reasonable, conservative, (pessimistic), and transparent in estimating releases from fuel cartridge ruptures. Clearly, reactor stack monitoring records and more extensive records would have greatly improved this effort, but in accordance with Atomic Energy Commission (AEC) Appendix 0230 Annex C-9, many of the detailed records from this time were not retained.

The major facilities in operation during the period of this report were:

- The Brookhaven Graphite Research Reactor (BGRR) (1950 -1969)
- The Hot Laboratory (1951-present)
- The Cosmotron (1953 -1956)
- The Brookhaven Medical Research Reactor (BMRR) (1959 - 2000)
- The Alternating Gradient Synchrotron (AGS) (1960 - present)

These facilities all contributed to airborne and liquid effluent releases.

Routine Airborne Releases

Routine air emissions associated with the operation of BNL from 1948 through 1961 were dominated by those from the Brookhaven Graphite Research Reactor (BGRR). Normal operation of the BGRR resulted in the production of

radionuclides, of which argon-41 (1.8 hour half-life) was the single largest emission from the Laboratory. Other sources of routine radionuclide emissions to air included the Brookhaven Medical Research Reactor (BMRR), the Hot Laboratory, the Cosmotron and the Alternating Gradient Synchrotron. Table E-1 summarizes routine airborne emissions from the BGRR.

Table E-1. Annual routine emissions of argon-41, iodine-131 and carbon-14 and particulates from the BGRR, 1950 through 1961.

Year	BGRR			
	Argon-41 (Ci)	Carbon-14 (Ci)	Iodine-131 (Ci)	Particulates (Ci)
1950	3.5×10^{4}	1.6	8.0×10^{-4}	1.7
1951	1.2×10^{6}	56	1.4×10^{-1}	62
1952	1.5×10^{6}	69	1.8×10^{-1}	76
1953	1.6×10^{6}	74	1.9×10^{-1}	82
1954	1.7×10^{6}	77	2.0×10^{-1}	85
1955	1.7×10^{6}	79	2.0×10^{-1}	87
1956	1.6×10^{6}	74	1.9×10^{-1}	81
1957	1.4×10^{6}	63	1.6×10^{-1}	69
1958	3.9×10^{6}	180	2.0	280
1959	5.1×10^{6}	230	2.7	370
1960	5.2×10^{6}	240	2.7	380
1961	5.6×10^{6}	250	2.9	400

External radiation levels, including background as influenced by world-wide fallout from atmospheric weapons testing and the increments attributable to BNL operations, were monitored at stations located in the central area of the site, at the site's perimeter, and off site. With few exceptions, the only measurable increase in external radiation above background attributed to routine BNL operations at most of the monitoring stations was caused by the radioactive argon-41 component of the BGRR's effluent cooling-air.

Data from three environmental monitoring stations for 1959, 1960 and 1961 and from the Environmental Monitoring Reports for 1962, 1963, 1964 and 1965 were used to estimate the average external exposure in the central portion of the site and at the site perimeter per million Curies (Ci) of argon-41 emitted from the BGRR. The relationship between the annual amount of argon-41 released by the BGRR and the amount of external radiation resulting from these releases measured at the perimeter monitoring stations did not vary significantly over the years.

From 1951 to 1957 when the BGRR operated with the natural uranium fuel, the maximum external exposure from the argon-41 at the site boundary ranged from 10 to 14 mR/year. During 1958 to 1961 when enriched fuel was used, the maximum external exposure from the argon-41 at the site boundary from the

argon-41 ranged from 34 to 46 mR/year. For the central area of the site, in 1951-1957 the maximum external exposure was 56 mR/year, and in 1958-1961 it was 220 mR/year.

Fallout from U.S. and U.S.S.R. nuclear weapons tests obscured the contributions of any routine airborne particulate gross beta activity from BNL operations as measured at the field stations. However, in 1966, as part of a detailed analysis of particulate activity released from the stack, gamma-emitting radionuclides in air samples from the stack were analyzed daily. That study showed that greater than 99% of the gross beta particulate activity resulted from short-lived (less than a few hours half-life) radionuclides.

The 1963 Environmental Monitoring Report (Hull, 1964) noted that in 1963, 2.4 Ci of iodine-131 were released with an average concentration of 7×10^{-10} μCi/cc (microcuries per cubic centimeter). Meteorological evaluations indicated that the concentrations at the perimeter stations were much less than 2×10^{-15} μCi/cc. Since the iodine-131 routinely emitted in 1958, 1959, 1960 and 1961 ranged from 2.0 to 2.9 Ci, iodine-131 concentrations at the perimeter stations would have been about 2×10^{-15} μCi/cc. This is five orders of magnitude below the permissible level. For 1950-1957 the routine releases of iodine-131 were much lower.

Fuel Cartridge Ruptures

During the early years of operation of the BGRR, fuel failures occurred which resulted in radioactive materials being released to the air stream that cooled the reactor. While the air was filtered before it was released from the stack, some emissions to the environment occurred as a result of these events. There were 28 reported ruptures of BGRR fuel during the period 1952-1957. These all occurred with the natural uranium fuel. There was one rupture of a uranium oxide (U_3O_8) sample that was being irradiated for the radioiodine-production program.

Releases of noble gases, particulates and radioiodines have been estimated for each fuel cartridge rupture. This was done using radionuclide inventories and release fractions developed for the Oak Ridge X-10 reactor, combined with probe readings and other qualitative information available for each rupture. These estimates are uncertain, and the assumptions used in deriving them were chosen to result in reasonable overestimates. As indicated in the body of the report, it is very likely that the estimates of emission releases could be overestimated by a factor of 5 or more.

Estimated releases of particulates for individual fuel ruptures ranged from 0.2 to 130 Ci. Noble gas release estimates ranged from 33 to 440 Ci, and iodine releases for individual ruptures ranged from 63 to 500 Ci (Table E-2 and Figure E-1).

The estimated annual noble gas release associated with the BGRR ruptures was highest in 1954. The noble gas release from the BGRR ruptures can be seen to have been very much smaller than the routine releases of argon-41.

Table E-2. Estimates of stack releases for fuel-cartridge ruptures in the BGRR.

Rupture Number	Rupture Date	Particulates (Ci)	Iodines (Ci)	Noble gases (Ci)
1	1/2/52	0.3	63	55
2	3/11/52	1.7	63	55
3	3/12/52	1.7	63	55
4	5/6/52	2.0	63	55
5	12/2/52	3.3	63	55
6	4/17/53	0.7	63	55
7	4/22/53	0.2	63	55
8	5/10/53	1.1	127	33
9	7/23/53	7.7	190	160
10	8/24/53	17	320	270
11	11/8/53	0.3	63	55
12	11/16/53	1.6	63	55
13	1/17/ 54	17	130	110
14	1/22/54	2.9	63	55
15	2/21/54	25	190	160
16	4/11/54	20	500	440
17	6/21/54	0.9	63	55
18	7/6/54	17	320	270
19	6/19/55	1.1	63	55
20	10/12/55	3.9	130	110
21	5/1/56	7.0	190	160
22	5/2/56	130	320	280
23	10/19/56	4.9	130	110
24	11/14/56	3.5	130	110
25	1/15/57	13	250	220
26	3/1/57	0.3	63	55
27	7/23/57	0.8	63	55
28	10/3/57	6.2	130	110
29*	10/28/57	3.0	13	9.1

* not a fuel element, uranium-oxide slug for iodine production.

With regard to measurement of the particulates, a major confounding factor in assessing the levels of airborne radionuclides associated with rupture events was the fallout from weapons tests that took place during this period. Although particulate air samplers were in place at the environmental monitoring stations, the only available data are those from two rain and settled dust collectors. The largest estimated particulate releases from BGRR rupture events in 1954 were in February, while the largest value in rain and settled dust was in September. Thus, the level of particulates in the rain and settled dust samples and the estimated releases from each rupture are not correlated.

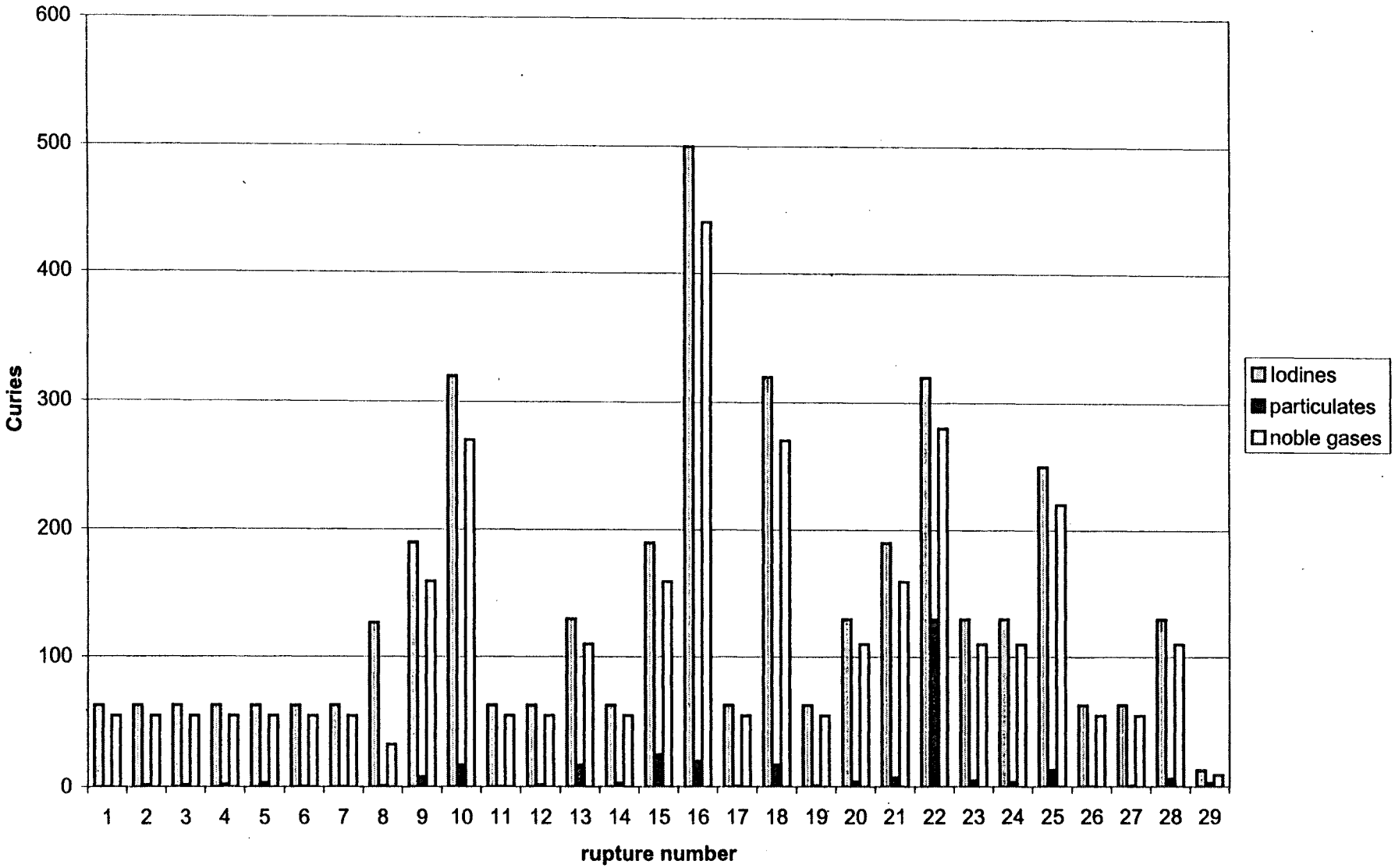

Figure E-1 Estimated releases of radioiodines, particulates and noble gases for each rupture of fuel in the BGRR

Estimates of the exposures to particulates were obtained from our postulated releases combined with a standard meteorological transport dispersion model. The particulate deposition values derived in this manner ranged from 32 pCi/m^2 (1.2 Bq/m^2) to 9.2 x 10^6 pCi/m^2 (3.4 x 10^5 Bq/m^2).

For the radioiodines the deposition ranged from 1.6 x 10^4 pCi/m^2 (5.8 x 10^2 Bq/m^2) to 7 x 10^6 pCi/m^2 (2.6 x 10^5 Bq/m^2) for I-131 and from 3.8 x 10^4 pCi/m^2 (1.4 x 10^3 Bq/m^2) to 1.5 x 10^7 pCi/m^2 (5.7 x 10^5 Bq/m^2) for I-133.

Liquid Releases

Most of the radionuclide contaminated liquid effluents at BNL originated from the BGRR, the Hot Laboratory complex, and from decontamination and hot-laundry operations. Smaller contributions came from the Medical, Chemistry and Physics facilities.

Liquid effluents containing small amounts of radionuclides were discharged to the sewage system. The effluent passed through an Imhoff Tank, where most of the solids were removed, was discharged to a large sand-filter from which it was collected by an underlying tile field, was chlorinated, and then discharged to the headwaters of the Peconic River.

Table E-3 shows gross beta, strontium-90 and cesium-137 activities at the points of discharge from the Imhoff Tank and the Chlorination Plant and concentrations at the Site Boundary. From 1951 through 1961, the estimated annual average concentration of gross beta at the point of discharge from the Imhoft Tank to the sand filter beds ranged from 310 to 780 pCi/l, a range of about 10% to 26% of the applicable radiation protection guide of 3,000 pCi/l. The values at the site boundary ranged from 25 to 260 pCi/l.

Table E-3. Average concentrations (pCi/l) of gross beta, strontium-90 (Sr-90) and cesium-137 (Cs-137) activities at the discharge from the Sewage Treatment Plant Imhoff Tank, Chlorination Plant and the Site Boundary.

Year	Imhoff Tank			Chlorination Plant			Site Boundary		
	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs-137 (pCi/l)	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs-137 (pCi/l)	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs137 (pCi/l)
1951	570	57	280	100	10	51	88	9	44
1952	330	33	170	99	10	50	45	4	22
1953	310	30	150	110	11	55	25	3	13
1954	430	43	220	100	10	50	47	5	24
1955	450	45	230	170	17	86	52	5	26
1956	320	32	160	120	12	59	26	3	13
1957	370	37	190	160	16	82	94	9	47
1958	530	53	270	200	20	100	34	3	17
1959	780	78	390	260	26	130	65	7	33
1960	650	65	330	310	31	150	260	26	130
1961	430	43	210	300	30	150	170	17	85

Beginning in 1960, downstream surface water grab samples were taken along the Peconic River and other nearby surface waters. The yearly average gross-beta concentrations at these downstream and control locations are given in Table E-4 for 1960 and 1961.

Table E-4. Yearly averages for 1960 and 1961 gross beta activities in surface water samples, pCi/l.

Sample Locations	1960	1961
On-Site		
M*	255	170
Peconic River		
A (Schultz Road)	20	34
B (Wading River-Manorville Road)	13	19
D (Calverton)	11	17

*estimated from river flow and monthly total releases at the Chlorination Plant.

The largest yearly average gross beta concentration at the sampled stations was an estimated 255 pCi/l at the on-site perimeter station M in 1960. Off site, the highest average concentration was 34 pCi/l.

Releases to Groundwater

Releases from cesspools and the former landfills were not documented during the early history of the laboratory. Sources of groundwater contamination during this period included potential losses of material from the on-site sewer pipes, an accidental contamination of a monitoring well with waste material and losses to groundwater through the sand filter beds at the sewage treatment plant.

An Inter-Agency agreement (IAG) is in effect among the United States Department of Energy, the United States Environmental Protection Agency and the New York State Department of Environmental Conservation to address groundwater contamination. Areas of Concern (AOCs) being addressed under the IAG include both active facilities such as the Sewage Treatment Plant and inactive facilities such as the former landfills, cesspools, and radioactive waste storage tanks.

Uncertainties

This report summarizes emissions, release estimates and environmental monitoring information from the early years of BNL's operation. A number of gaps in the available information were filled based on assumptions and analyses of the data that were available. In all cases, the intent was to make assumptions that were reasonable, but would overestimate the magnitude of the release.

The highest degree of uncertainty is associated with the estimates of the releases from the fuel ruptures. The approach taken was to use qualitative descriptions of the events as well as any quantitative data that were available, and to apply professional judgment, since much of the information was based on incomplete data in the reports generated 50 years ago. The data and information used in developing the release estimates are given in the report and in detailed attachments. This allows the reader to develop alternative estimates if desired.

Major assumptions made in developing release estimates included those of estimates of the inventories in individual fuel slugs, estimates of the fraction of the inventory released from a single slug; the fraction released to the stack, and the number of slugs that were involved in each rupture. In fact, as shown in the report, there are indications that the release estimates could be overestimated by a factor of 5 or more. This report assumes a release fraction of 80% for radioiodines. However, it is likely that although 80% of the iodines may have been released from the fuel slugs, a significant fraction of this material would have adhered to the surface of the extensive cooling air duct work, including filters, and the stack.

Application of the Data Given in this Report

The data presented here are meant primarily to complete the sequence of BNL's reporting of environmental monitoring data. It will also be made available to the ATSDR in developing a public health assessment for BNL. It should be noted that while the data in this report form the basis for a screening assessment of dose, they are not sufficient to form the basis of a quantitative dose reconstruction.

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Half Lives of Radioisotopes discussed in report

Radionuclide	Symbol	Half-life
Argon-41	Ar	1.8h
Barium-140	Ba	12.74y
Bromine-82	Br	35.3h
Carbon-14	C	5730y
Cerium-144	Ce	284.3d
Cobalt-60	Co	5.27y
Iodine-129	I	1.5×10^{7}y
Iodine-131	I	8.04d
Iodine-133	I	20.8h
Krypton-85	Kr	10.7y
Lanthanum-140	La	40.27h
Nitrogen-13	N	9.97min
Niobiun-95	Nb	35.15d
Oxygen-15	O	122.sec
Phosphorus-32	P	14.29d
Ruthenium-103	Rh	39.28d
Ruthenium-106	Rh	368.2d
Strontium-89	Sr	50.5d
Strontium-90	Sr	29.1y
Uranium-234	U	2.45×10^{5}y
Uranium-235	U	7.03×10^{8}y
Uranium-238	U	4.468×10^{9}y
Xenon-133	Xe	5.24d

Acronyms

AGS	Alternating Gradient Synchrotron
AEC	Atomic Energy Commission
AMSL	Above Mean Sea Level
AOC	Area of Concern
ATSDR	Agency for Toxic Substances and Disease Registry
BNL	Brookhaven National Laboratory
BGRR	Brookhaven Graphite Research Reactor
BMRR	Brookhaven Medical Research Reactor
Bq	Becquerel, 1 decay per second
Ci	Curie, 3.7×10^{10} decays per second
d/m	Distintegrations per minute
DOE	United States Department of Energy
ERDA	Energy Research and Development Administration
FRC	Federal Radiation Council
HEPA	High Efficiency Particulate Air
HFBR	High Flux Beam Reactor
HP&S	Health Physics and Safety
IAG	Inter-Agency Agreement
ICRP	International Commission on Radiological Protection
MW	Mega-Watt, 1 million Watts
MPC	Maximum Permissible Concentration
NBS	National Bureau of Standards
ORNL	Oak Ridge National Laboratory
R	Roentgen, a measure of exposure
rem	A measure of absorbed dose
X-10	Oak Ridge Graphite Reactor

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PART I

Emissions and Environmental Monitoring

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PART I - EMISSIONS AND ENVIRONMENTAL MONITORING

1. INTRODUCTION

Brookhaven National Laboratory (BNL) was established in 1947 on the former site of the Army's Camp Upton located in central Long Island, New York. From the beginning, BNL monitored the environment on and around the Laboratory site to assess the contributions of its operations to radiation in the environment.

Early monitoring focused on radiation, and data were reported in various internal documents and in papers presented at scientific meetings. Monitoring data were also summarized in the Laboratory's annual progress reports, which were submitted to the U.S. Atomic Energy Commission (AEC). The AEC was a predecessor to the U.S. Department of Energy (DOE), which funds BNL today.

In 1960, the AEC instituted a program for publicly reporting radioactivity data collected around major AEC installations. As part of that program, BNL began issuing reports to major Long Island newspapers on radiation levels measured in the environment.

In 1963, BNL pioneered the development of an annual environmental monitoring report; the first was for calendar year 1962 (Hull, 1963). Reports were also prepared for calendar years 1963-1966 (Hull, 1964; Hull, 1966; Hull, 1967; Hull and Gilmartin, 1969).

These reports were not required by the AEC, and therefore were completed when time and staff resources were available. As a result, BNL did not compile annual reports for the years 1967 through 1970, although environmental monitoring continued.

In 1971, for the first time, these reports became a contractual obligation for all AEC facilities. Thus, BNL has prepared a site monitoring report every year from 1971 onward.

In 1998, a report was issued summarizing the environmental data collected for 1967, 1968, 1969, and 1970 thereby filling the gap in BNL's series of environmental monitoring reports beginning in 1962 (Meinhold and Hull, 1998).

One of the purposes of this report is to complete BNL's environmental reporting history by covering the period from 1948 through 1961. An additional objective is to provide environmental data to the Agency for Toxic Substances and Disease Registry (ATSDR).

This report presents monitoring and emissions data for the early years of the Laboratory's history. These data reflect measurements for 1948-1961 of concentrations and amounts of airborne radioactivity, radioactivity released to the Peconic River, and external radiation levels in the vicinity of BNL.

Information in this document comes primarily from monthly Health Physics and Safety summary reports, Reactor Operations Monthly Reports, and BGRR operating logbooks. Other internal memoranda, published reports and papers were also used. Since this information was generated 50 years ago, interpretation is always somewhat uncertain.

The number of significant figures reported from original sources is maintained. Calculations and data presented in Appendices use three or more significant figures but in light of many uncertainties, the data are rounded off to two significant figures in Tables and in the text.

2. BACKGROUND

2.1 BNL Site

Brookhaven National Laboratory is a scientific research center situated in Suffolk County on Long Island, about 70 miles east of New York City. Figure 1 shows its location and the surrounding communities. From 1948 to 1961, the largest nearby populations were the shoreline communities. The land area within ten miles of BNL was mostly forested or under cultivation, with a reported population of 26,400 in 1948 (BNL, 1948a).

Figure 2 shows the BNL site and the principal sources of airborne radionuclide emissions for 1948-1961. Most of the airborne radioactive effluents at BNL originated from the BGRR with smaller contributions from the Hot Laboratory, the Cosmotron, the AGS, and the BMRR. Direct radiation in the environment originated from sources in the Biology fields, with smaller contributions from the accelerators. Environmental monitoring stations recorded airborne emissions from the Laboratory as well as the fallout from atmospheric weapons testing. The principal monitoring stations are shown in Figure 2.

Liquid effluents were collected in the sanitary system and treated at the Sewage Treatment Plant (STP), monitored, and discharged to the Peconic River (Figure 2).

Figure 3 shows the annual wind distribution observed by BNL's meteorology Group between 1960 and 1973. Nagle (1975) gives the monthly and annual wind roses for 1949 -1951 and 1960 -1973. The wind distribution patterns are similar on an annual and seasonal basis. BNL can be characterized as a well-ventilated site. Prevailing winds are from the southwest during the summer, from the northwest during the winter, and about equally from these two directions during the spring and fall.

From 1947-1967 the active area of the site was about 3,600 acres. As can be seen in Figure 1, there was an additional undisturbed tract of land north of Route 25. In 1968 property was acquired that extended the site to about 5,300 acres. Most of the site is wooded, except for a central area of less than 1,000 acres.

The terrain is gently rolling, with elevations varying between 40 and 120 feet above sea level. The land lies on the west rim of the shallow Peconic River watershed, with the river itself rising in marshy areas in the north and east sections of the site. The Peconic River both recharges to, and receives water from, the groundwater aquifer depending on the hydrological conditions. During droughts, the river water typically recharges to groundwater, while with normal to above-normal precipitation, the river receives water from the aquifer.

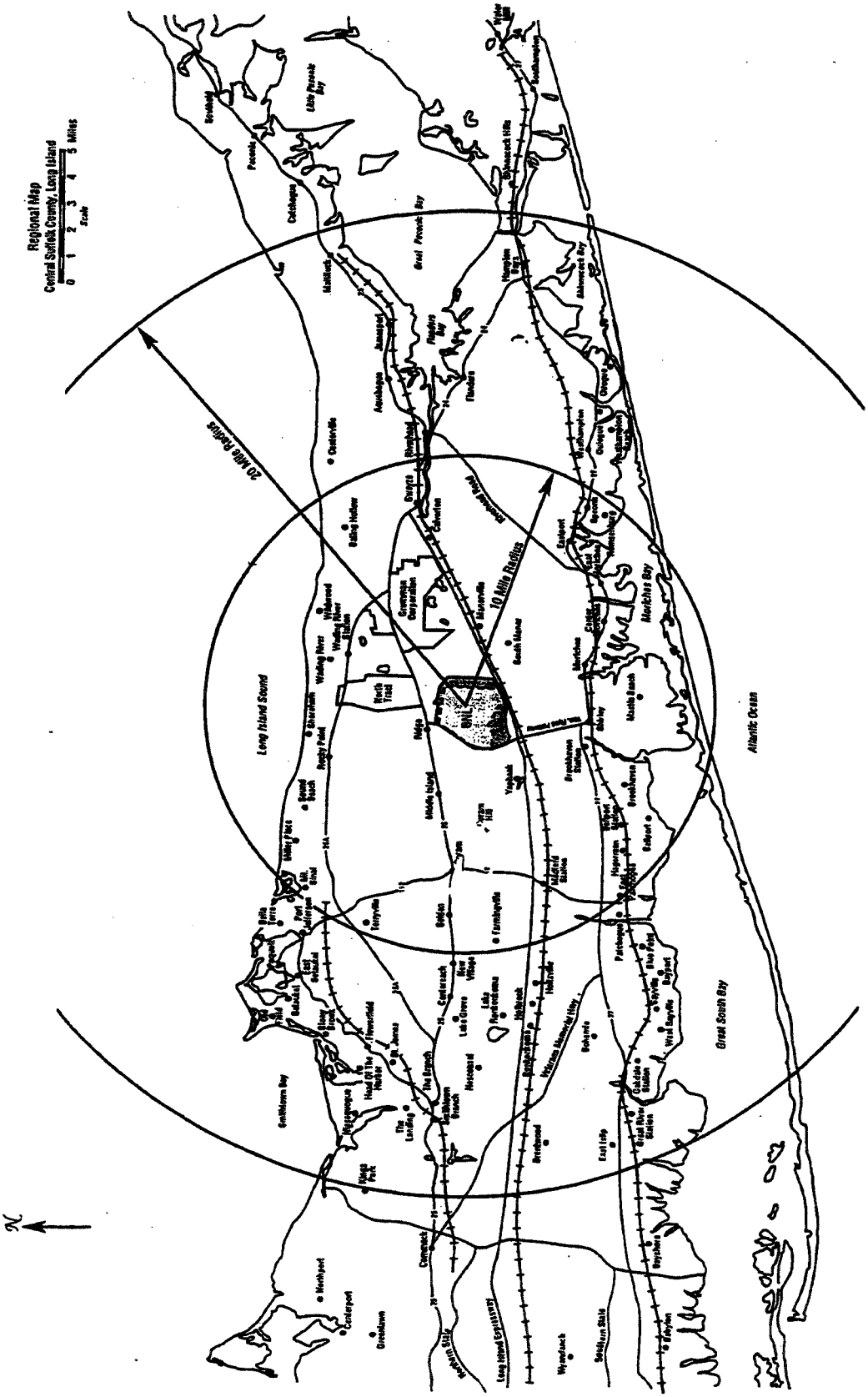


Figure 1. Central Suffolk County, showing the area around Brookhaven National Laboratory.

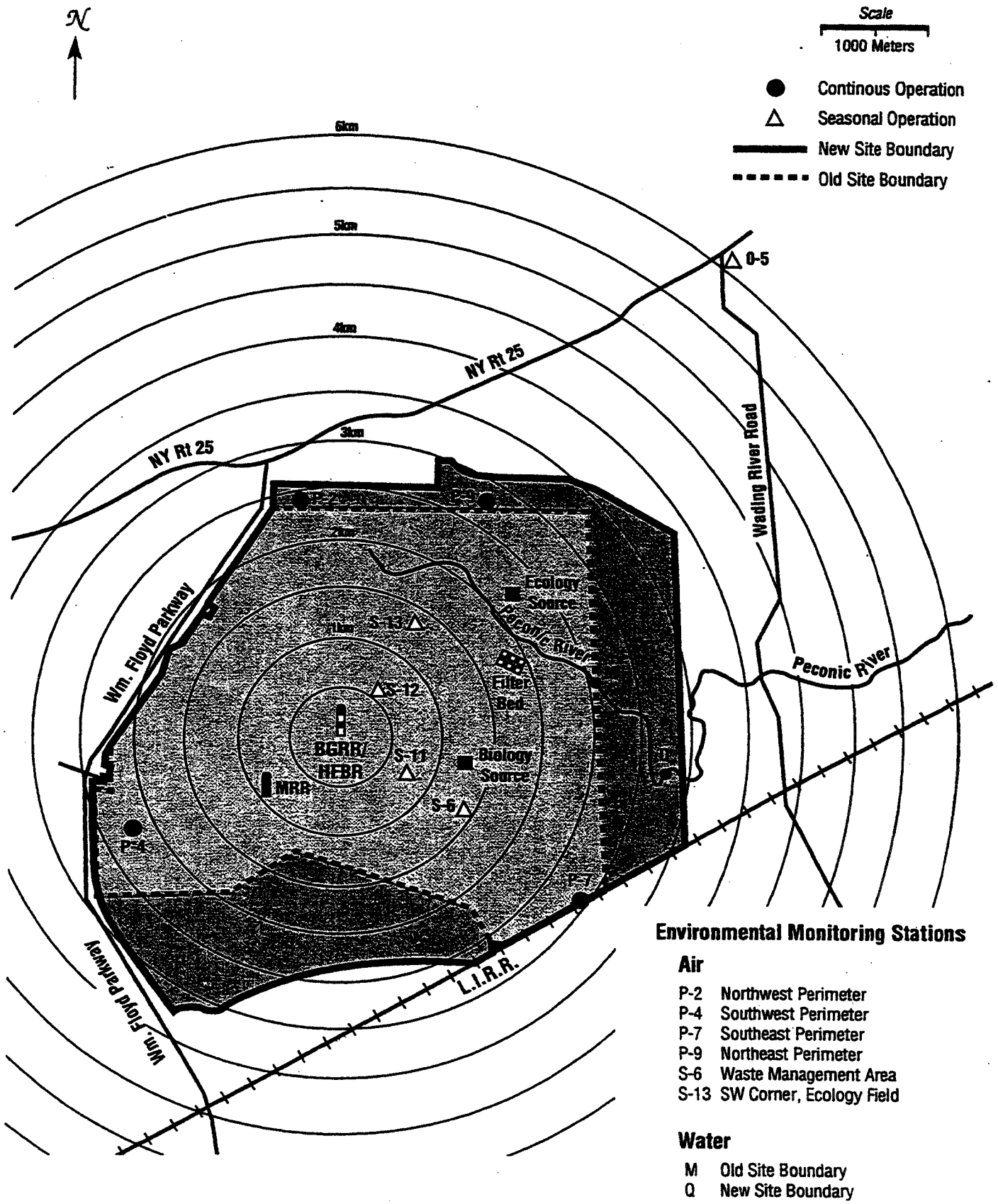
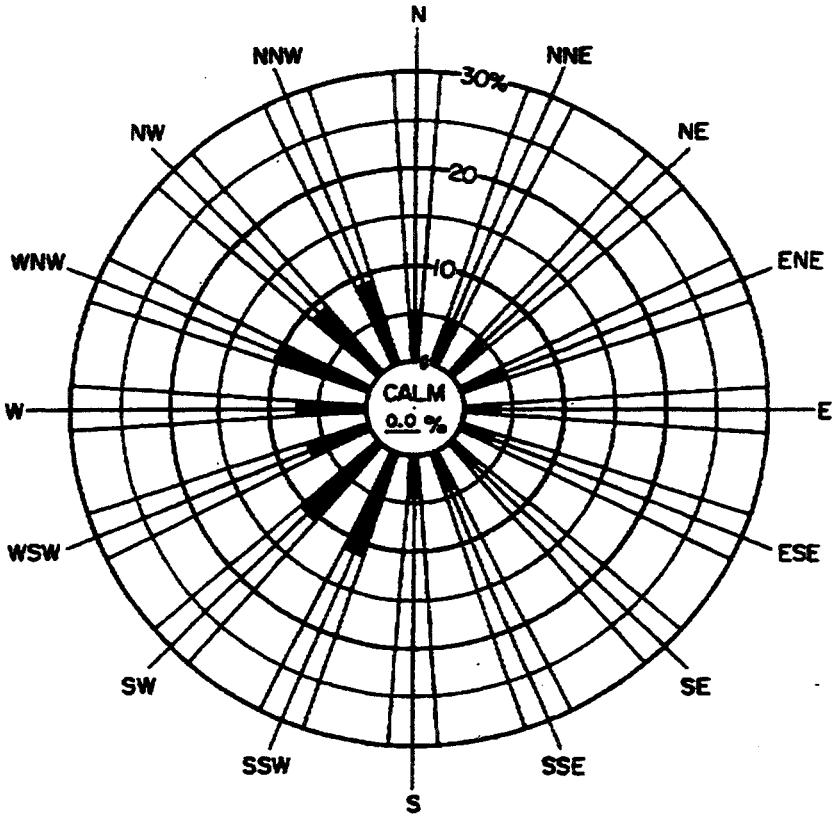


Figure 2. Principal laboratory sources of, and monitoring stations for, environmental radiation. (The site was expanded in 1968, hence the New and Old Site Boundary. The High Flux Beam Reactor (HFBR) came on-line in 1965, and shared the stack with the BGRR).

STATION: BROOKHAVEN NATIONAL LABORATORY
HEIGHT: 355 ft.
PERIOD: January-December, 1960-73



Notes:

1. The arrowheads formed by the wedges indicate the direction toward which the wind blows with the greatest frequency. The predominant wind directions were toward the north-northeast and east-southeast.
2. Each concentric circle represents a 5% frequency. For example, the wind blew toward the NNE approximately 12% of the time.

Figure 3. Annual wind distribution, 1960-73.

Studies of Long Island hydrology and geology in the vicinity of the Laboratory indicate that the uppermost Pleistocene deposits (referred to as the Upper Glacial Aquifer) which are between 31-61 m thick, are generally composed of highly permeable glacial sands and gravels (Warren *et al.*, 1968).

In general, groundwater in the northeast and northwest sections of the site flows towards the Peconic River. In the western portion of the site, groundwater flow tends to be towards the south, while along the southern and southeastern sections of the site, the flow tends to be towards the south to southeast. Figure 4 depicts the typical groundwater configuration for the BNL site.

2.2 Major Scientific Facilities and Release Points

The following were among the major scientific facilities operated at BNL during the period covered by this report:

- Brookhaven Graphite Research Reactor (BGRR)
- Brookhaven Medical Research Reactor (BMRR)
- Cosmotron
- Alternating Gradient Synchrotron (AGS)
- Hot Laboratory

Prior to the operation of the major scientific facilities a wide variety of activities in the Biology, Chemistry, Physics, and Medical Departments involved the use of radioactive material.

The BGRR came on-line in 1950 and operated until 1969. It was the first reactor built in the United States solely to provide neutrons for research, and its size and design allowed it to support a large number of research programs simultaneously.

The BMRR is the first nuclear reactor built exclusively for medical and biological research, and came on-line on March 15, 1959 and operated until October 2000.

The AGS and the Cosmotron were used to study the elementary building blocks of matter and the laws and forces that govern them. Using these accelerators, many new particles were discovered and studied. The Cosmotron operated from 1953 to 1966. The AGS achieved full energy in 1960 and is still in use.

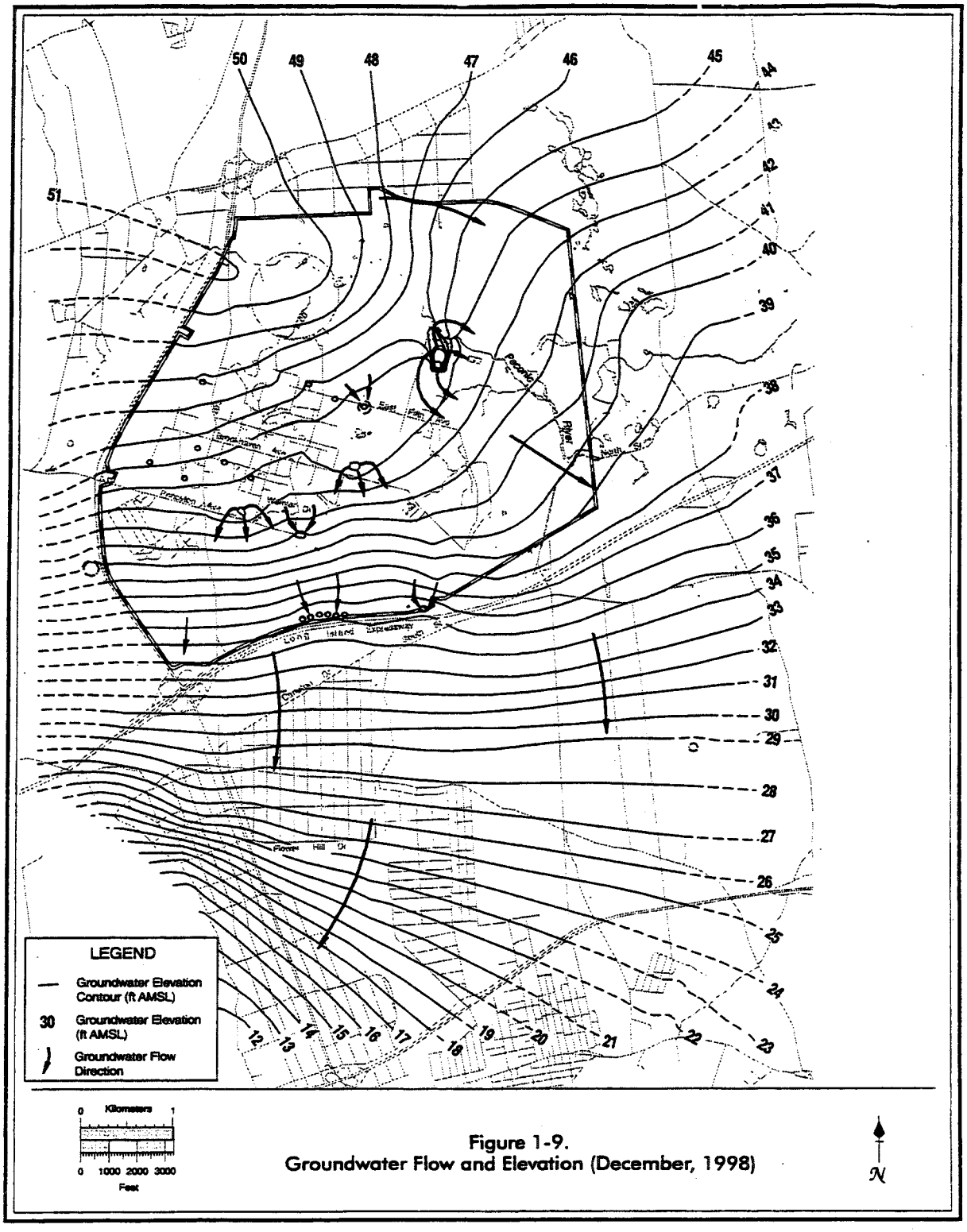


Figure 4. Typical water table contour map for the BNL site (BNL, 1999).

Other programs involving irradiations and the use of radionuclides for scientific investigations were carried out at other BNL facilities, including the Medical Research Center, the Biology Department (including two multi-curie field-irradiation sources), the Chemistry Department, and the Department of Nuclear Engineering. The latter included the Hot Laboratory, where special-purpose radioisotopes were produced and processed for on- and off-site use.

Routine air emissions associated with the operation of Brookhaven National Laboratory from 1948 through 1961 were dominated by those from the BGRR. Other sources of routine radionuclide emissions to air included the BMRR, the Hot Laboratory, the Cosmotron and the AGS. There were also episodic releases of radionuclides, such as those associated with fuel cartridge ruptures at the BGRR. Direct radiation in the environment originated from the Biology Fields sources, with smaller contributions from the accelerators.

The BGRR also contributed to BNL's liquid radioactive effluents. Additional liquid effluent originated from the Hot Laboratory complex, and from decontamination and hot-laundry operations. Smaller contributions came from Medical, Physics, and Chemistry facilities.

Liquids in the sanitary waste system passed through a settling (Imhoff) tank, which removed most of the solids. The effluent flowed onto sand-filter beds, from which most of it was directed by an underlying tile field, chlorinated, and discharged into a small stream which forms one of the headwaters of the Peconic River. Small amounts of radionuclides were released in this effluent to the Peconic River and to the groundwater underlying the sand-filter beds.

Other potential sources of groundwater contamination during this period included leakage of material from the on-site sewer pipes, and contamination originating from the landfill used to dispose of low-level radioactive material. There were also small spills of radioactive material on the ground, which may have contaminated the groundwater. There is one documented accidental disposal of radioactive material into the water table in July 1960 (see additional discussion in Section 6).

2.3 Environmental Monitoring Program

An environmental monitoring program was developed at BNL before the BGRR came on-line. In preparation for the operation of the BGRR, a chain of 15 monitoring stations was established located at distances out to 10 miles in various directions from the center of the site. These stations were equipped with pressurized ionization-chamber-type monitors for gamma radiation, several Geiger-Mueller counters and a moving-tape-type of dust monitor for monitoring beta (β) and alpha (α) emitting radionuclides. These stations were developed by the Laboratory's Instrumentation Division in 1948 when little equipment was

available commercially. A meteorological group was formed and a 420-foot-high tower was constructed and equipped with instrumentation for determining wind speed and direction and temperature as a function of height. The environmental monitoring program at BNL also was active in monitoring the fallout in rain and settled dust associated with world-wide nuclear weapons tests.

Depending on the level of radioactivity, liquid radioactive waste was either collected or combined with the sanitary-waste generated at BNL and discharged to the sewage treatment plant. The BGRR-Hot Laboratory complex had a separate liquid waste system so that releases to the general sanitary waste system could be monitored and controlled.

There were hold-up tanks at other facilities. Depending on the concentration of radioactivity, the contents of these tanks were either released to the sanitary waste system or transferred to the Hot Laboratory. When the radionuclide concentrations were too high for release to the sanitary system the liquid was sent to the evaporation plant which separated the solids from the liquids. The solids were treated as radioactive waste and the condensate was monitored and released to the sanitary system.

Samples were collected at the point where the liquid effluent was discharged to the sand-filter beds and just before its discharge to the stream near the Peconic River headwaters. Samples were analyzed daily for gross activity. Beginning in 1960, downstream surface water grab samples were taken along the Peconic River and other nearby surface waters.

To understand the potential impact of BNL's operations on the local environment, a geohydrological study was undertaken by the United States Geological Survey (USGS) during the period 1950-1952. Groundwater monitoring and an extensive study of the groundwater movement at BNL was included in these studies (Warren *et al.,* 1968). No routine groundwater monitoring was done following the USGS study, with the exception of wells installed in the Hazardous Waste Management Area in 1960 related to an accidental injection (Section 6).

2.4 Historical Records

Historical data were reviewed to gather information on the air emissions from BNL from 1948-1961. The data were collected from sources such as facility operating records, monthly reports, off-normal incident reports, reactor health-physics logbooks, and reactor control room logbooks.

Many of the materials reviewed in this report were related to reactor operations which, at that time, were all marked "Secret" or "Classified". These materials were declassified in the 1950s.

In accordance with AEC Appendix 0230 Annex C-9, many of the detailed records

from this time were not retained. Such items as Daily Operations Reports, log sheets, and recorder charts had retention periods of 1-6 years.

The records used to develop the data in this report are described in the applicable sections of the report.

2.5 Radiation Protection Limits

Radiation Protection Limits applicable at Brookhaven National Laboratory are discussed below and summarized in Table 1.

Table 1. Radiation protection limits (1947-1961).

Year	Occupational	Public Dose Limit
1947	50 mrem/day	3.5 mrem/week
1949	300 mrem/week	
1953		30 mrem/week
1957	(age-18) x 5 rem 3 rem/13 weeks	500 mrem/year
1960		5 rem/30 years (170 mrem/year)
Release Concentration to Sand Filter Beds		
1949 – 1961		Gross activity: 3×10^{-12} Ci/cc (3,000 pCi/l)
MPC* in Air		
1949 – 1961		Iodine-131: 3×10^{-10} µCi/cc

*MPC: maximum permissible concentration

External

The external radiation protection limits for 1948-1961 took the form of agreements between the AEC and BNL. There was no independent federal agency responsible for establishing limits at this time. In 1947 a letter from AEC's W.E. Kelly to BNL Director P.M. Morse established, on a temporary basis, a maximum permissible radiation dose for the public of 3.5 mrem in any 7 days. This was 1% of the maximum dose permitted for workers (Occupational Limit) of 50 mrem/day, i.e. 0.50 mrem/day that results in 7 days/week x .50 mrem/day = 3.5 mrem/week.

Over the next several years the Occupational Limit became 300 mrem/week as the emphasis changed from controlling daily exposure to weekly exposure. In 1953, the AEC and BNL agreed to raise the limit for members of the public to 10% of the occupational limit, or 30 mrem/week.

In 1957 the AEC adopted the 1956 Recommendations of the International Commission on Radiological Protection (ICRP 1957; 1958) which introduced an occupational limit equivalent to 5000 mrem/yr over a working lifetime. Ten

percent of 5000 mrem/yr or 500 mrem/yr then became the limit for members of the public. This remained the limit for 1957-1961, although BNL applied a 170 mrem/yr in late 1960 as recommended by the Federal Radiation Council (Federal Radiation Council, 1960).

Internal

For emitters that concentrate in the body such as iodine-131, the AEC and BNL applied the recommendations of the National Committee on Radiation Protection (NBS, 1953). These recommendations were radionuclide-specific.

For example, in that document the tabulated values of the "maximum permissible" concentration of iodine-131 in air was 3×10^{-9} $\mu Ci/cm^3$ (micro Curies per cubic centimeter). The AEC reduced that concentration by a factor of 10 for the public. It was modified in 1959 (NBS, 1959) to 9×10^{-9} $\mu Ci/cm^3$.

Liquid Releases

Early in the history of the laboratory AEC and BNL agreed to apply a limit of 3×10^{-6} $\mu Ci/cc$ (3,000 pCi/l gross beta activity) at the point of discharge to the sand-filter beds, with the further stipulation that the total activity discharged per year should not exceed 1.5 Curies.

3. THE BROOKHAVEN GRAPHITE RESEARCH REACTOR

Routine air emissions from the operation of Brookhaven National Laboratory from 1948 through 1961 were dominated by those from the BGRR. There were also episodic releases of radionuclides from the BGRR associated with fuel-cartridge ruptures. These airborne emissions are estimated in Section 4, along with emissions from other smaller sources. Because of the unique nature of the BGRR and its importance to the environmental history of the laboratory, this section describes the reactor and its operations in some detail.

The BGRR (Figure 5) came on-line in 1950 and operated until late 1969. The reactor core consisted of fuel cartridges loaded in horizontal fuel channels passing through a cube of graphite 7.6 m on a side. The graphite acted as the neutron moderator and reflector. Figure 6 shows the loading face of the reactor.

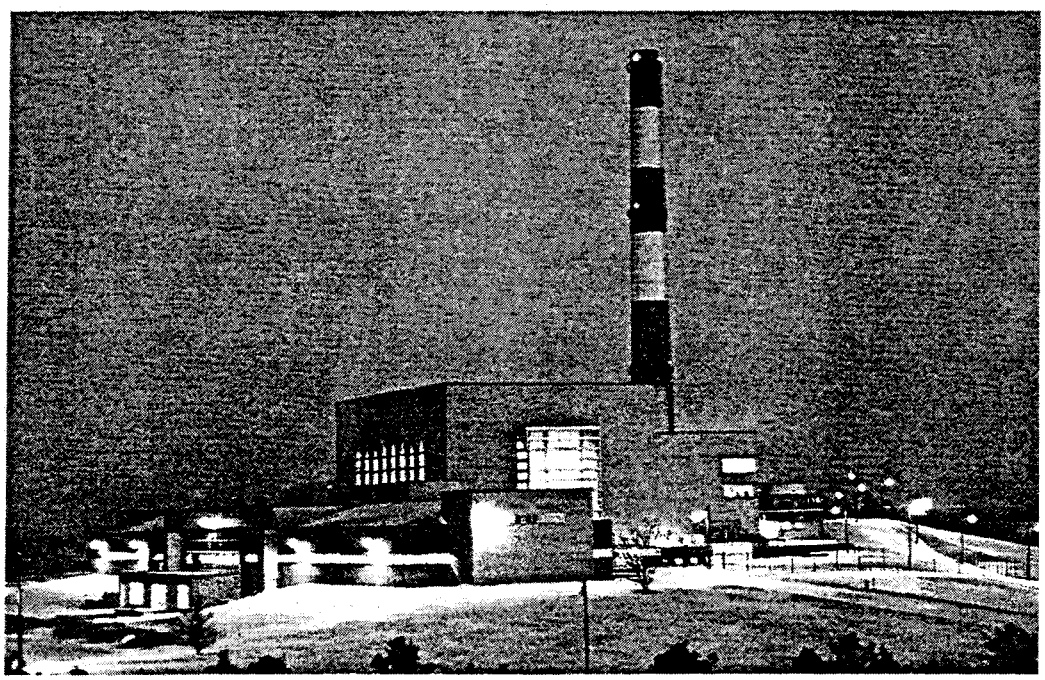


Figure 5. The Brookhaven Graphite Research Reactor.

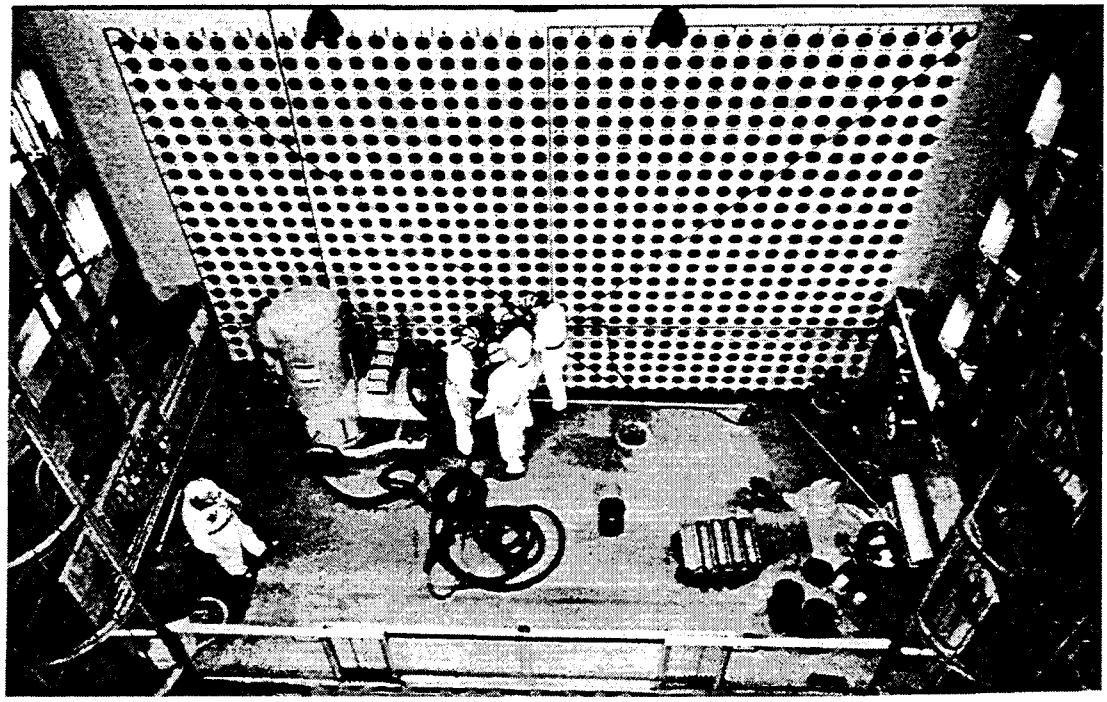


Figure 6. Loading face of the BGRR.

The core was divided into halves (north and south) by a 7 cm-wide vertical gap in the center of the graphite cube. The cooling air was drawn into the reactor at the rate of 7600 m^3/minute. It passed into the central gap and flowed bi-directionally through the north and south halves of the core into the plenum chambers. It then entered the north and south air ducts, passed through exit air filters, heat-exchangers and a venturi into the fan house where the two ducts join. The fans blew the filtered air up a 300-foot stack. The base of the stack was about 50 feet above the immediate surroundings. The exit air filters were designed to be 95% efficient for total particulates and 89% efficient for 3-4 micron particles. As the filter loading increased, its efficiency rose to about 99% (BNL, 1948b; Foelix and Hull, 1963). Figure 7 is a diagram of the BGRR cooling air system.

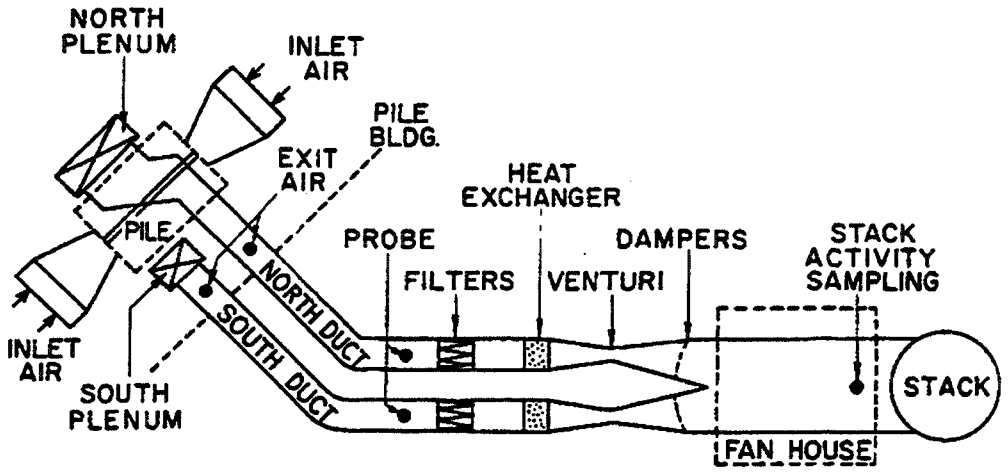


Figure 7. BGRR cooling-air system (from Foelix and Hull, 1963).

From 1950 through 1957/58 the BGRR was fueled with natural uranium. Several hundred 11-foot finned aluminum cartridges that each held 33 fuel slugs (Figures 8, 9) fueled the reactor. The fuel slugs were 1 x 4 inch machined right circular cylinders of natural uranium. The cartridges were placed in the channels, normally one in the south channel and one in the north channel.

The cartridges were connected to a helium system, which served two purposes. One was to provide an inert atmosphere to suppress oxidation of the uranium, and the other was to permit the detection of the loss of cartridge integrity by observing pressure changes in the cartridges. This helium system was developed in an attempt to overcome some of the difficulties experienced at the Oak Ridge Clinton X-10 Reactor, which began operations in 1948, used identical fuel slugs.

Evidence of helium leaks alone were not a particular cause for concern, but helium losses alerted the operating staff to a potential for a cartridge failure. When elevated levels of airborne radioactivity were detected in the cooling air, the failed cartridge was termed a rupture and was removed ("discharged") from the reactor core. There were 28 reported ruptured fuel cartridges and one experimental sample rupture in the period 1952 through 1957. They are summarized in Section 4 and discussed in detail in Part II.

In 1957/58 the aluminum cartridges with the natural-uranium-metal slugs were replaced with fuel elements which consisted of three curved plates containing an alloy of aluminum and enriched uranium, clad on all surfaces by 0.5 mm of aluminum (Figure 10).

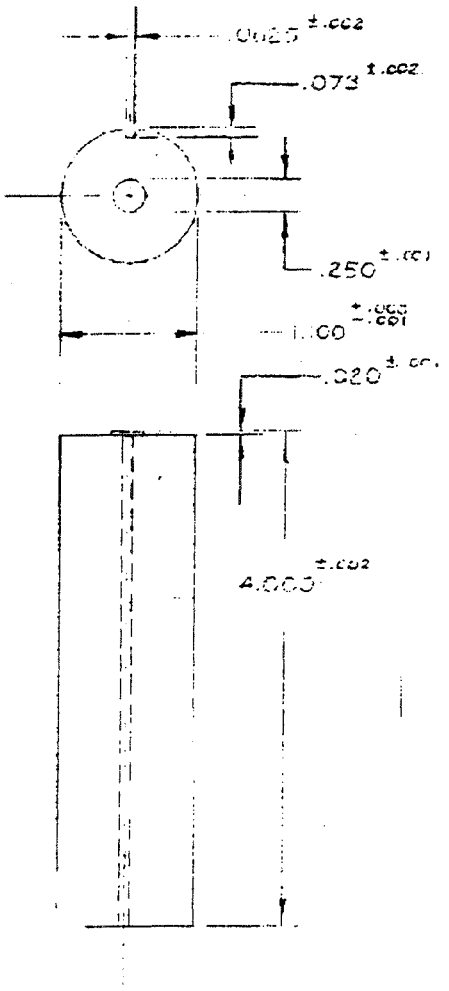


Figure 8. Natural uranium fuel slug (from BNL, 1948a)

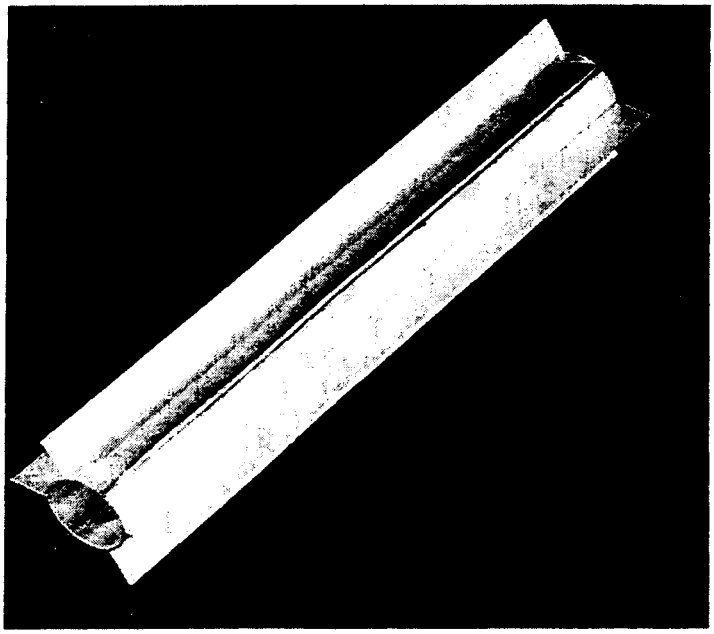


Figure 9. Natural uranium-fuel cartridges (from BNL, 1948a).

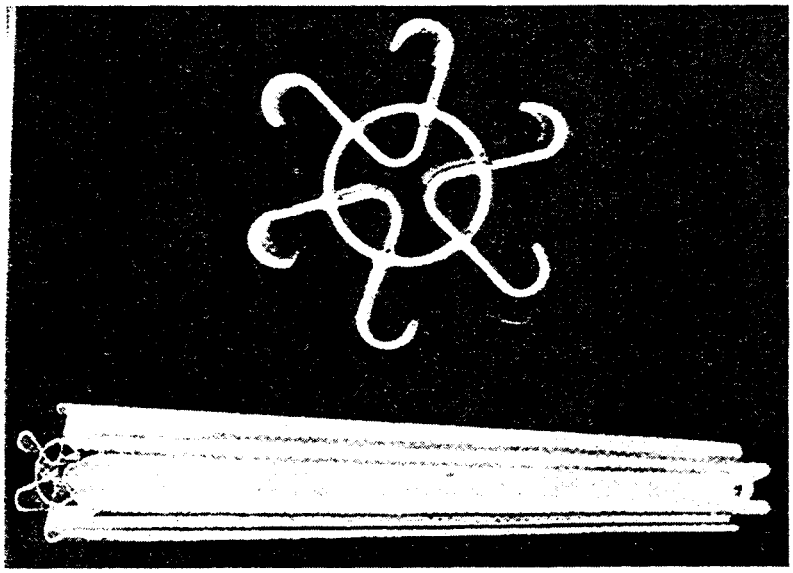


Figure 10. Enriched fuel element (from BNL, 1956).

This replacement was done over a period of 14 months. It began in January, 1957, when 35 channels of enriched fuel were loaded into the central core region (BNL, 1958). In October 1957, the loading was gradually increased to 157 channels. The reactor was shut down from March 20 through April 13, 1958 to remove all of the remaining natural-uranium fuel from the reactor and to complete the loading with enriched fuel.

The new fuel met several objectives, including a higher neutron flux for the experimental program, and improved airflow, which reduced cooling costs. Most significantly, these new fuel elements were inherently less subject to damage of the fuel, referred to (in the enriched fuel) as a cladding failure. There were no reported failures of this new fuel during the period covered by this report (through 1961). The new fuel did result in an increase in the routine releases of argon-41 because of the higher neutron flux. Minor surface contamination of the aluminum-alloy cladding with uranium which occurred during manufacture, resulted in some routine releases of fission products to the cooling air (Table E-1). This type of surface contamination is not unique to the BGRR fuel.

4. AIRBORNE EMISSIONS

Routine emissions from the operation of Brookhaven National Laboratory from 1948 through 1961 were dominated by those from the Brookhaven Graphite Research Reactor. Other sources of routine radionuclide emissions to air included the Hot Laboratory, the Brookhaven Medical Research Reactor, the Cosmotron, and the Alternating Gradient Synchrotron. There were also episodic releases of radionuclides, such as those associated with fuel-cartridge ruptures at the BGRR.

4.1 Emissions from the Brookhaven Graphite Research Reactor

4.1.1 Routine Emissions, Natural-Uranium Fuel

As cooling air passed through the reactor core, certain components of the air became radioactive. The most important radionuclide was argon-41, resulting from the neutron activation of argon-40, which makes up 0.94% of air by volume. This generation of argon-41 is related to the neutron flux which, for a given fuel loading, is related to the power of the reactor. Much smaller amounts of carbon-14 were also produced by neutron activation of the cooling air. Even smaller quantities of other activation products would have been created.

Occasionally fine particles from automobile exhaust, smoke, and salt spray from coastal storms would enter the cooling air stream through the intake filters and become radioactive and were seen as temporary increases in the stack air monitors. These radionuclides had short half-lives and were unlikely to have resulted in measurable releases to the environment.

For the natural uranium used from 1950 to 1957, data given in BNL (1951) provide a value of 6,600 Ci/day of argon-41 with the BGRR operating at 30 MW (220 Ci/MW-day). This value was used to estimate the argon-41 emitted monthly from the BGRR for 1950 through 1957, based on the power level of the reactor in MW-day (Figure 11, Attachment I-A). Table 2 summarizes the annual routine releases of argon-41 from the BGRR. The monthly power levels for the BGRR were abstracted from Reactor Operations Monthly Reports for 1950-1957.

Carbon-14 emissions also were a function of the power of the reactor for a given fuel loading. A report prepared by the Reactor Safeguard Committee (BNL, 1948b) gives a value of 0.28 Ci/day at 30 MW for the BGRR (ratio of 0.01 Ci/MW-day). Attachment I-A gives the estimated monthly carbon-14 emissions. A summary of the annual releases of carbon-14 from the BGRR is given in Table 2.

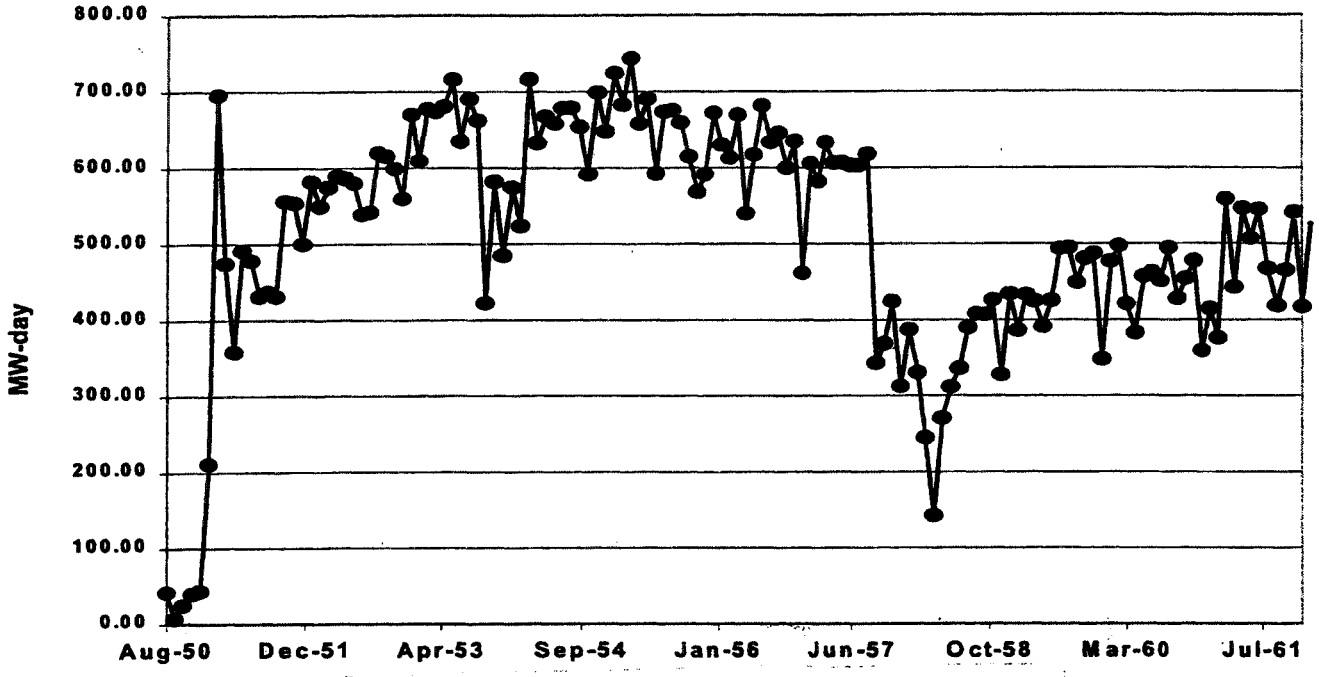


Figure 11. Power history of the BGRR, 1950-1961.

Table 2. Annual routine emissions of argon-41, carbon-14, iodine-131, and particulates from the BGRR, 1950 through 1961.

Year	Argon-41 (Ci)	Carbon-14 (Ci)	Iodine-131 (Ci)	Particulates (Ci)
1950	3.5×10^{4}	1.6	8.0×10^{-4}	1.7
1951	1.2×10^{6}	56	1.4×10^{-1}	62
1952	1.5×10^{6}	69	1.8×10^{-1}	76
1953	1.6×10^{6}	74	1.9×10^{-1}	82
1954	1.7×10^{6}	77	2.0×10^{-1}	85
1955	1.7×10^{6}	79	2.0×10^{-1}	87
1956	1.6×10^{6}	74	1.9×10^{-1}	81
1957	1.4×10^{6}	63	1.6×10^{-1}	69
1958	3.9×10^{6}	180	2.0	280
1959	5.1×10^{6}	230	2.7	370
1960	5.2×10^{6}	240	2.7	380
1961	5.6×10^{6}	250	2.9	400

The natural-uranium fuel slugs were contained in a sealed aluminum cartridge with helium gas flowing around them. The potential routine release of importance from these slugs would have been fission products that might have been swept out of the cartridges by the helium system. Every month, the Reactor Operations Monthly Reports state that no fission products were associated with the helium leaks, but the detection limit is not given.

It seems reasonable to make some estimate of the radioiodine and noble gases that might have been released when helium leaked out of the system. A reasonable estimate can be made using the following assumptions.

- Inventory of fission products was the same for natural and enriched fuel.
- Release characteristics of the fission products were similar.
- Enriched fuel resulted in a release of 0.51 mCi of I-131/MW-day.
- 5% of the channels holding the natural uranium fuel had He leaks.
- Therefore 5% of 0.51 mCi/MW-day of I-131 was released from the natural uranium fuel.

The first assumption is plausible, since although the flux was greater with the enriched fuel, the number of fuel channels was smaller and the power level of the reactor was lower. The second assumption is that the release characteristics of the fission products with natural-uranium slugs were similar to that of the enriched-fuel elements. Uranium-235 produces the fission products. Small amounts of it existed as a contaminant on the aluminum cladding of the enriched uranium-aluminum alloy fuel elements. The unclad natural fuel also had small amounts of uranium-235 on its surface because the fuel is only 0.72% uranium-235.

The remaining assumptions focus on the amount of iodine-131 that could have been released during normal operations. Section 4.1.2 of this report provides the reference for the estimate of 0.51 mCi/MW-day of iodine-131 released as a function of the power of the reactor for the enriched-fuel loading. This would be the release level of iodine-131 from the natural-uranium loading if there were no cartridges containing the fuel or if all of the channels leaked enough to sweep out all of the iodine-131. However, the data indicate that for 1951 through 1957 only 5% of the channels had helium leaks. For the six months the reactor operated in 1950, less than 1% of the channels leaked. Using this information, monthly and annual routine releases of iodine-131 were estimated; they are given in Table 2.

The 1966 Environmental Monitoring Report discussed the results of a detailed study that explored the relationship between measured releases from the BGRR stack and the environmental airborne concentrations. Aside from argon-41, iodine-131 was the most important radionuclide that would contribute to a potential dose that was discharged to the atmosphere from the BGRR. Bromine-82 and iodine-133 were released in somewhat larger concentrations. However, calculations indicate that due to their shorter half-life and other dosimetric properties, the resulting doses from iodine-133 would have been about half that of iodine-131, and the doses from bromine-82 less than 20% of that resulting from iodine-131 exposure.

These analyses do not include estimates of the krypton-85 nor iodine-129 released. These radionuclides are released in much smaller quantities. For example, the daily production of iodine-131 was about 2 Ci/day, while that of krypton-85 was about 0.4 mCi/day, and iodine-129 is an order of magnitude less (see radionuclide release estimates for Rupture Number 8 in Part II, Attachment II-A).

Small amounts of krypton-85 would have continued to be emitted from the non-ruptured fuel elements even after they were removed from the reactor and stored in the spent fuel canal.

Small amounts of particulates were also released during normal operations. In the Health Physics and Safety Summary of February, 1954, F.P. Cowan notes that "During normal operation of the pile this monitor indicates release of filterable activity at the rate of about 0.14 mCi/min." This is equivalent to 0.2 Ci/day. The average monthly power level for 1950-1957 was 555 MW-day, or 18.5 MW-day for each day of operation. These values were used to derive a ratio of 0.011 Ci of particulates per MW-day for the natural uranium fuel (Table 2).

4.1.2 Routine Emissions, Enriched-Uranium Fuel

While the BGRR was running with enriched-uranium fuel, the routine emissions of argon-41, carbon-14 and iodine-131 were greater than they were when running with natural uranium. Routine releases of fission products associated with uranium on the surface of the enriched fuel were estimated based on measurements made in 1963.

There are several sources for the relationship between power and argon-41 released from the BGRR when it was running with enriched fuel, and some variations in the estimates. This analysis used a value of 977 Ci/MW-day, derived from measurements published in the BNL Environmental Monitoring Report for 1965 (Hull, 1967).

Routine carbon-14 emissions from the BGRR when enriched fuel was used were also related to the power of the reactor. No carbon-14 emissions data were found, so a ratio was derived using the amount of carbon-14 released per MW-day when natural-uranium fuel was used in the reactor as given in Section 4.1.1, and the ratio of argon-41 released per MW-day for natural- and enriched- fuel. This value is 0.044 Ci/MW-day.

Routine iodine-131 and particulate releases from the enriched-uranium fuel occurred during normal operations due to trace amounts of uranium that became trapped on the aluminum cladding during manufacture. These routine emissions can also be related to the power of the reactor, although other factors affected the iodine-131 and particulate concentrations and the relationship is not as clear as it is for argon-41 and carbon-14. Foelix and Hull (1963) report that on average 0.51 mCi/MW-day of iodine-131 was released in 1962. A value of 0.07 Ci/MW-day of particulates was derived from data in the 1965 Environmental Monitoring Report (Hull, 1967).

Table IA-2 in Attachment I-A gives the monthly power history of the reactor from January of 1958 through December 1961. This analysis assumed the enriched fuel relationship for argon release to power level beginning in January 1958, although the fuel change was not complete until April 1958. This table also gives the calculated monthly emissions of argon-41, carbon-14 and iodine-131 from the BGRR. Table 2 in Section 4.1.1 summarizes the annual routine releases of argon-41, carbon-14, and iodine-131 from the BGRR.

As indicated in Section 4.1.1, the 1966 Environmental Monitoring Report reported that bromine-82 and iodine-133 were released in somewhat larger concentrations than iodine-131, however, the resulting doses would have been smaller.

4.1.3 Significance of BGRR Routine Emissions 1950-1961

External radiation levels, including natural background as influenced by fallout from above ground nuclear weapons tests and the increments attributable to BNL operations, were monitored at fixed monitoring stations located on-site, at the site's perimeter, and off site. The data collected at these stations are discussed in more detail in Section 5.

The measurable increase in external radiation levels above background attributed to BNL operations at most of the monitoring stations was caused by the radioactive argon-41 component of the BGRR's cooling-air effluent.

The relationship between the annual amount of argon-41 released by the BGRR and the amount of external radiation measured at the perimeter monitoring stations did not vary significantly over the years. Data from perimeter monitoring stations for 1959, 1960 and 1961 and from the Environmental Monitoring Reports

for 1962, 1963, 1964 and 1965 were used to estimate the average external exposure per million Curies of argon-41 emitted from the BGRR. This is discussed in detail in Section 5.

From 1951 to 1957 when the BGRR operated with the natural uranium fuel, the maximum external exposure from the argon-41 at the site boundary ranged from 10 to 14 mR/year. During 1958 to 1961 when enriched fuel was used, the maximum external exposure from the argon-41 at P-9 (the site boundary at the time) ranged from 34 to 46 mR/year.

Fallout from nuclear weapons tests obscured the contributions of any airborne particulate gross beta activity from routine BNL operations as measured at the field stations. However, in 1966, as part of a detailed analysis of particulate activity released from the stack, radionuclides in air samples from the stack were analyzed daily. That study, reported in detail in the 1966 Environmental Monitoring Report (Hull and Gilmartin, 1969) showed that the gross beta particulate activity resulted primarily from short-lived radioactivity. For example, in 1966, the particulate activity discharged from the BGRR and the High Flux Beam Reactor (HFBR) (HFBR operations began in 1965) stack was 323 Ci. The daily analysis showed that less than 0.2 Ci (about 0.06%) of this total was due to intermediate and long-lived radionuclides, i.e. those with half-lives of several hours to many days.

The Environmental Monitoring Report (Hull, 1964) noted that in 1963, 2.4 Ci of iodine-131 were released with an average concentration of 7×10^{-10} μCi/cc. Meteorological evaluations indicated that the concentrations at the perimeter stations were much less than 2×10^{-15} μCi/cc. Since the iodine-131 emitted in 1958, 1959, 1960 and 1961 ranged from 2.0 to 2.9 Ci with an average of 2.6 Ci, the iodine-131 concentrations at the perimeter stations would be about 2×10^{-15} μCi/cc. This is five orders of magnitude below the permissible level given in Table 1. For 1950-1957 the routine releases of iodine-131 were much lower.

4.1.4 Emissions Associated with Ruptures of the Fuel Cartridges

There were 28 reported ruptures of BGRR fuel during the period covered by this report (Table 3). These all occurred with the natural uranium fuel used until 1957-58. The severity of the rupture and the quantity of radioactive material released were related to how quickly the rupture was detected and the reactor shut down, and how long the fuel cartridge was in the reactor before it ruptured. There was one rupture of a uranium oxide (U_3O_8) sample that was being irradiated for the iodine-production program, which is designated Rupture 29 in this report.

Part II of this report details the approach used in these evaluations. Table 3 gives estimates of the radionuclide releases associated with each rupture.

These estimates have a high degree of uncertainty because the measurements were very rough, and the extent to which the stack monitoring data for a particular rupture are relevant to other ruptures is also uncertain. The analysis of Rupture 29, where the entire sample is assumed to have been released, suggests that particulate releases may be overestimated by about a factor of 5 or more (see Attachment II-A).

The estimated noble gas release associated with the BGRR ruptures was highest in 1954 with a value of 1100 Ci of xenon-133. During that year, the routine release of the noble gas argon-41 was about 170 million Curies. Although the half-life of argon-41 (1.83 hours) is shorter than that of xenon-133 (5.2 days), this difference is unimportant for exposure out to tens of miles from the reactor stack. At longer distances there would have been some decay of the argon but the ratio is so large that even if we compare 1/2 of the argon-41 to all of the xenon-133, we find 1100 Ci Xe-133/85,000,000 Ci Ar-41 or for each curie of xenon-133 in the environment there would have been about 7500 Curies of argon-41. In addition, the routine hourly discharge of argon-41 in 1959 was about 650 Ci which is larger than the largest noble gas release for any rupture of 440 Ci.

The overall result of this analysis is that the xenon-133 release from the BGRR ruptures is insignificant compared to the routine releases of Ar-41 and to standards in effect at the time.

With regard to the particulates, a major confounding factor in assessing the levels of airborne radionuclides associated with rupture events was the fallout from weapons tests by the United States and the U.S.S.R. that took place during this period (Cowan and Steimers, 1958; Weiss, 1954). These are discussed in more detail in Section 5. Although particulate air samplers were in place at the environmental monitoring stations, with rare exceptions the only available data are from two rain and settled dust collectors.

Fortunately, detailed meteorological data for this time period are available, so rough screening level calculations could be made, utilizing the Lawrence Livermore National Laboratory HOTSPOT computer code 3.1. This code was developed to assess potential downwind air concentrations and deposition values associated with accidental releases. This makes it particularly useful for application to the rupture releases, which were both episodic and relatively brief.

Using this code, the maximum particulate and iodine concentrations were calculated for each rupture event using the release data given in Table 3, the historical meteorological data and the HOTSPOT gaussian plume transport model.

Table 3. Estimates of stack releases for fuel-cartridge ruptures in the BGRR.

Rupture Number	Rupture Date	Particulates (Ci)	Iodines (Ci)	Noble gases (Ci)
1	1/2/52	0.3	63	55
2	3/11/52	1.7	63	55
3	3/12/52	1.7	63	55
4	5/6/52	2.0	63	55
5	12/2/52	3.3	63	55
6	4/17/53	0.7	63	55
7	4/22/53	0.2	63	55
8	5/10/53	1.1	127	33
9	7/23/53	7.7	190	160
10	8/24/53	17	320	270
11	11/8/53	0.3	63	55
12	11/16/53	1.6	63	55
13	1/17/ 54	17	130	110
14	1/22/54	2.9	63	55
15	2/21/54	25	190	160
16	4/11/54	20	500	440
17	6/21/54	0.9	63	55
18	7/6/54	17	320	270
19	6/19/55	1.1	63	55
20	10/12/55	3.9	130	110
21	5/1/56	7.0	190	160
22	5/2/56	130	320	280
23	10/19/56	4.9	130	110
24	11/14/56	3.5	130	110
25	1/15/57	13	250	220
26	3/1/57	0.3	63	55
27	7/23/57	0.8	63	55
28	10/3/57	6.2	130	110
29*	10/28/57	3.0	13	9.1

*Not a fuel element, uranium-oxide slug for iodine production.

Values of wind speed and direction and the distance to the point of the maximum iodine deposition for each rupture using this approach are given in Part II, Attachment II-A. The deposition values range from 1.2 Bq/m^2 to 3.4×10^5 Bq/m^2 (32pCi/m^2 to 9.2μCi/m^2) for particulates, from 5.8×10^2 Bq/m^2 to 2.6×10^5 Bq/m^2 (15.6nCi/m^2 to 7μCi/m^2) for I-131 and from 1.4×10^3 Bq/m^2 to 5.7×10^5 Bq/m^2 (37nCi/m^2 to 15μCi/m^2) for I-133.

4.2 Emissions from the Brookhaven Medical Research Reactor

The Brookhaven Medical Research Reactor (BMRR) was the first nuclear reactor built exclusively for medical and biological research. It came on-line on March 15, 1959 and operated until October 2000. The BMRR was nominally a 3-MW light-water reactor, capable of operating for short periods at power levels up to 5 MW. The BMRR is housed in a 60-foot diameter steel-and-concrete structure adjacent to BNL's Medical Research Center.

The reactor core was contained in a cylindrical tank, two feet in diameter in the central region. The core was supported upright within the tank by a grid plate. The grid plate had space for up to 32 elements and four control rods. Critical configurations had been established with as few as 17 elements (BNL, 1960).

The fuel in the BMRR was enriched uranium, homogeneously dispersed in a uranium-aluminum alloy. The reactor used curved-plate fuel-elements made of a uranium-aluminum alloy containing 12% by weight fully enriched uranium. The BMRR had a peak flux of 5×10^{13} neutrons/cm^2/sec (ERDA, 1977).

The core was moderated and cooled by the forced circulation of light water. The water flowed upward, primarily to allow the short-lived isotopes (produced by neutron interaction with the oxygen in the water) to decay in the expanded upper shielded portion of the reactor vessel. This system eliminated the need for massive shielding of external parts of the coolant piping. The closed-cycle primary coolant underwent heat exchange with a once-through secondary water system. The reactor core was contained within an aluminum vessel, which was surrounded by an air-cooled graphite reflector and a concrete shield (Figure 12).

Air from the interior of the confinement building was used to cool the neutron reflector surrounding the core of the reactor vessel. When air was drawn through the reflector, it was exposed to a neutron flux that causes the argon component of the air to become radioactive argon-41. This generation of argon-41 was related to the neutron flux which, for a given fuel loading, was related to the power of the reactor. After passing through the reflector, the air was routed through a roughing filter, a high efficiency particulate air (HEPA) filter to remove particulates, and a charcoal filter to remove radioiodines. The air was then exhausted to a 150-foot (46-meter) stack adjacent to the confinement building.

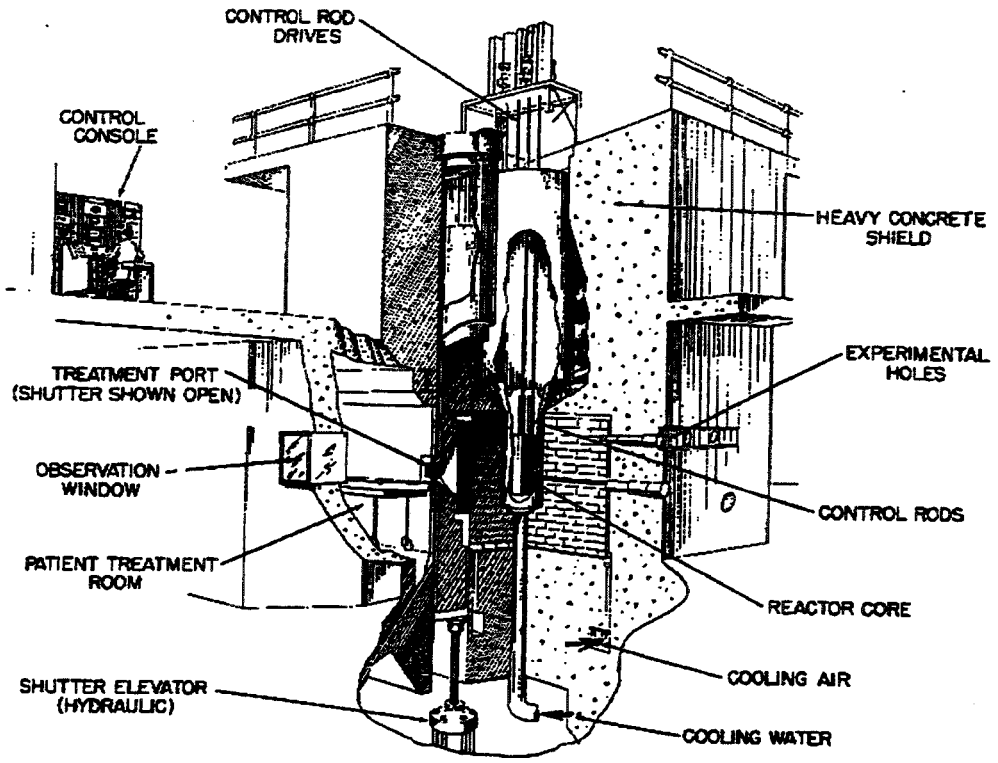


Figure 12. Cutaway view of the BMRR (from ERDA, 1977).

Fuel assemblies from different sources, with some variations in design have been used in the BMRR. These assemblies are described in BNL 1960. The power history of the BMRR, in MW-hours for each month of operation (Figure 13), was taken from the BMRR logbooks. Because of the basic design of the reactor and the low power level at which it operated, the levels of argon-41 released from the BMRR stack were orders of magnitude smaller than the releases from the BGRR.

No original records of direct measurements of the argon-41 released from the BMRR stack are available for the time period of interest, but several documents give values that can be used to relate the release of argon to the power level of the reactor. In 1997 direct measurements of argon-41 released from the BMRR were made. From these measurements an average value of 2.11 Ci/MW-hr was derived.

Argon-41 emissions were calculated for the BMRR using this Ci/MW-hour value. Monthly emissions are given in Attachment I-A. Annual emissions are shown in Table 4. The unusually high values for the summer of 1960 were related to extensive reactor operations required for the dosimetric evaluation of the patient treatment facility.

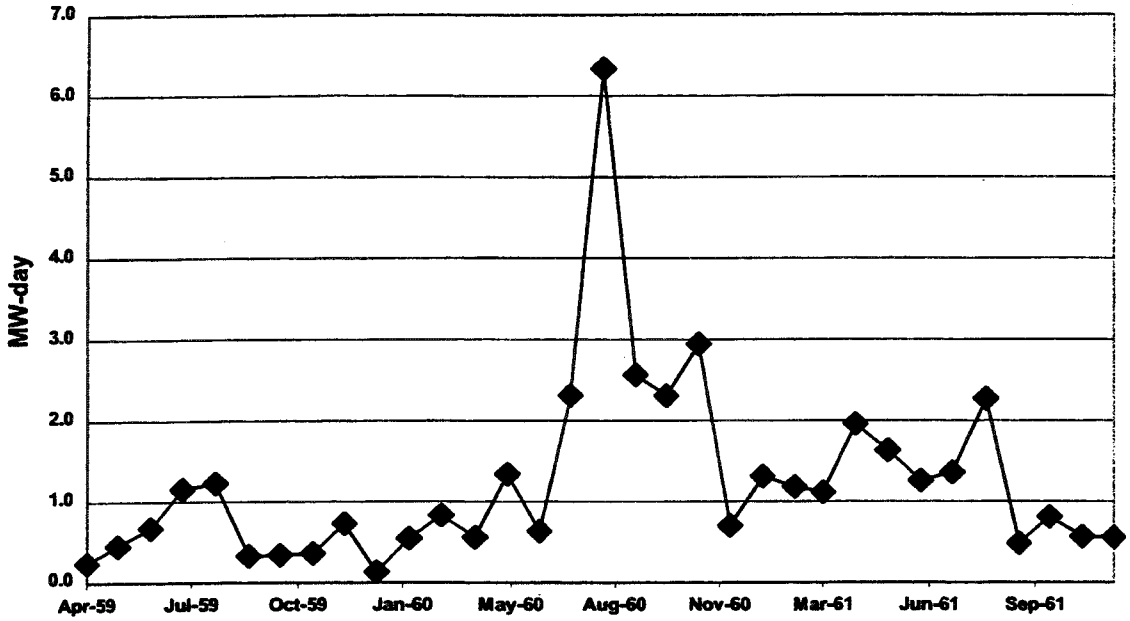


Figure 13. Power history of the BMRR, 1959-1961.

Table 4. Argon-41 and carbon-14 released from the BMRR.

Year	Annual Total Argon-41 (Ci)	Annual Total Carbon-14 (Ci)
1959	280	4.7×10^{-3}
1960	1100	1.8×10^{-2}
1961	740	1.2×10^{-2}

Carbon-14 emissions can also be related to the power level of the reactor, and the ratio of carbon-14 to argon-41 in the BMRR emissions has been calculated to be 1.7×10^{-5}. Carbon-14 emissions were calculated using this value and are given in Table 4 and Attachment I-A.

These emissions of argon-41 and carbon-14 are orders of magnitude smaller than routine releases from the BGRR, and could not have been detected at the site boundary.

In recent years, BNL has measured and reported emissions of additional radionuclides released in the BMRR's effluent (Table 5, Lee *et al.*, 1999). These radionuclides are of negligible dosimetric importance because of their short half-life and extremely low total activity. A rough estimate for 1959, 1960 and 1961 could have been made using the ratios of argon-41 levels between 1997 and these years. However, in view of the extremely low levels of emissions and the large uncertainty in this procedure, this was not done.

Table 5. Routine airborne radionuclide releases from the BMRR in 1997 (from Lee et al., 1999)

Nuclide	Half-life	Ci Released
Ar-41	1.8 h	2.22×10^{3}
Al-26	7.2×10^{5} y	1.21×10^{-8}
As-76	26 h	4.70×10^{-4}
Ba-128	2.4 d	1.86×10^{-4}
Ba-140	12.8 d	1.56×10^{-4}
Br-82	35 h	8.61×10^{-3}
Ce-141	32 d	1.82×10^{-7}
Ce-144	284 d	1.42×10^{-6}
Co-60	5.2 y	2.65×10^{-6}
Fe-59	44 d	3.85×10^{-6}
Hg-203	46 d	5.90×10^{-5}
I-124	4.2 d	1.89×10^{-5}
I-131	8 d	3.33×10^{-5}
I-133	21 h	3.63×10^{-4}
La-140	40 h	8.25×10^{-4}
Mo-99	66 h	1.54×10^{-7}
Na-24	15 h	2.30×10^{-4}
Sb-122	2.7 d	4.81×10^{-7}
Sc-46	84 d	2.15×10^{-8}
Se-75	119 d	2.04×10^{-7}
Sr-91	9.5 h	3.32×10^{-4}
Tc-99m	6.0 h	5.86×10^{-5}
Ti-44	47 y	1.29×10^{-4}
Xe-133	5.2 d	1.02×10^{-4}
Xe-135	9.1 h	1.07×10^{-3}
Zn-65	244 d	1.95×10^{-5}
Zn-69m	13.7 h	1.44×10^{-6}

4.3 Other Sources of Air Emissions

A review of Health Physics and Safety Summary reports and other documents indicates that the only important sources of radionuclide air releases during this period, besides the BGRR and the BMRR, were the Cosmotron, AGS, and the Hot Laboratory.

4.3.1 Cosmotron and Alternating Gradient Synchrotron

The Cosmotron and the AGS were used to study the elementary building blocks of matter and the laws and forces that govern them. Using the Cosmotron and the AGS, many new particles have been discovered and studied. The Cosmotron came on-line in 1953 and was operated until 1966. The AGS achieved full energy in 1960 and is still in operation.

Some radioactive gases were created by activation of the ventilating air within the tunnel and experimental areas of the Cosmotron and AGS. These were released at ground level, but, since they are short-lived (carbon-11, half-life 20.5 min; nitrogen-13, half-life 10.0 min; oxygen-15, 2.1 min), they did not create a measurable radiation level beyond the immediately adjacent on-site area.

4.3.2 Hot Laboratory

The Hot Laboratory officially opened January 15, 1951. It consisted of a central laboratory, a fan house, a radioactive-liquid waste tank farm, and a liquid radioactive-waste concentration plant (ERDA, 1977).

The original purpose of the central facility was to provide appropriately shielded areas for research with large amounts of radioactive material. The "hot" area of the Hot Laboratory included five hot cells, three chemical-processing hot cells, and three high-level hot cells for handling and processing of radioactivity in gaseous, liquid, or solid form. The cells were maintained at negative pressure with respect to their surroundings to minimize the possibility of radioactive releases to the building. Each has individual exhaust air-filters as well as a backup filter preceding discharge to the BGRR stack.

The iodine production program at the Hot Laboratory and an accidental release of uranium hexafluoride in 1957 deserve mention.

Iodine Production Program

A separate exhaust-air system was installed for the process hot cells originally set up for acid-dissolution of irradiated uranium samples to recover radioiodines. It includes a NaOH scrubber and backup charcoal filter, and was ducted to a stainless steel pipe within the BGRR stack (ERDA, 1977). Cowan and Gemmell (1960) also refer to filters and scrubbers at the Hot Lab.

From March of 1952 to June of 1960 there was a program of iodine-131 production at the Hot Laboratory. The detailed data for rupture 29 given in Part II (a production sample for this program) provides information on the radionuclide inventory of these samples.

A rough estimate of the radionuclide emissions from this process can be made If it is assumed that (1) all the production samples were 225 g of U_3O_8, equivalent to 209 g of uranium-235 or 17% of the inventory in a single fuel slug, (2) they were in the BGRR for 30 days, with 1 run each month (3) since the objective of the effort was to collect the iodine-131, only 10% of radiodines but 100% of the noble gases were released to the BGRR stack. This assumption of 10% is an overestimate because of redundancies in back up systems. For each of these years, the approximate emission amounts were the following:

- Iodine-131: 5 Ci
- Iodine-133: 10 Ci
- Iodine-129: 5×10^{-9} Ci
- Krypton-85: 3×10^{-2} Ci
- Xenon-133: 100 Ci.

Uranium Hexafluoride Release

In 1957 there was an accidental release of uranium hexafluoride from the Hot Laboratory. The following quotation was taken from a letter from R.W. Powell to F.L. Horn dated September 10, 1957:

> "A quantity of uranium hexafluoride was vaporized into the air and vented to the atmosphere when an explosive reaction ruptured the fluoride volatility pilot plant at the Hot Laboratory on May 15, 1957. A total of 29 unirradiated natural uranium slugs had been used for dissolution studies between January 22, 1957 and May 15.

A mass balance estimate of the amount of material lost in this event gave 14.5 kg, and an AEC investigation provided a value of 22.28 kg.

The Health Physics Summary for May 1957 made the following statement:

> "As a result of the explosion in the semiworks area, low levels of uranium contamination were spread through the cold area of the Hot Laboratory and somewhat higher levels were spread through the hot area. Smears counted about 40 d/m of alpha in the cold area. Average smears in the hot area counted 400 d/m of alpha and 1500 d/m of β/γ".

It seems prudent to assume that 50% of the uranium was released, primarily

because of the extensive contamination of the laboratory building, and because uranium hexafluoride is highly reactive. Under that assumption, 0.005 Ci of uranium-238, and 0.005 Ci of uranium-234 would have been released. The uranium-235 activity would have been much less. If 100% were released, the activity would be a factor of 2 higher.

4.3.3 Other Facilities

During this period BNL had a variety of hoods and ventilating systems for experiments with tracers or low level radioactivity, but these sources released only small amounts of material (Cowan and Gemmell, 1960).

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5. AREA MONITORING STATIONS

The measurements and assessments of radioactivity in air and water and levels of external radiation exposure in the vicinity of BNL include those associated with naturally occurring radioactive elements and cosmic radiation, those resulting from fallout from atmospheric nuclear weapons tests, and those attributable to BNL's operations. External radiation levels and airborne particulate radionuclides were measured at area monitoring stations located in the central area of the BNL site and at the site boundary (Figure 3). The releases from the BGRR and other facilities were controlled by the Radiation Protection Limits in effect during this period.

5.1 External Radiation

External radiation levels, including background as influenced by fallout and the increments attributable to BNL operations, were monitored at fixed monitoring stations located in the central portion of the site, at the site's perimeter, and off site. Most of the monitors remain in their original locations, although some were added over the period covered by this report.

Background at a given station was determined from the radiation level prevailing when no obvious BNL contributions were detectable at the station. The potential error in making this determination was minimized by reference to meteorological data (to establish the direction of the reactor's air effluent plume) and to the log-book indications of the gamma -field sources. These background measurements are approximately 30% higher than the values obtained today because most weapons fallout radionuclides with short and intermediate half-lives have decayed. Furthermore, the detectors used during this time contained small quantities of naturally occurring radionuclides in the insulating material of the ion chamber.

The measurable increase in external radiation above background attributed to BNL operations at most of the monitoring stations was caused by the radioactive argon-41 component of the BGRR's cooling-air effluent.

Late in 1961, a 10,000 Ci cesium-137 gamma-source was installed in the ecology forest in the northeast area of the BNL site, about 800 meters equidistant from the north and east boundaries (Hull, 1963; Figure 3). The source was used to irradiate an otherwise undisturbed wooded area for ecological studies. The cesium-137 source produced a measurable dose rate at stations E-9 (later P-9) and S-13, both located northeast of the BGRR stack.

Each station's equipment included an ion chamber and dynamic capacitor electrometer assembly (Kuper and Chase, 1950). These units could accurately measure less than 10 μR/hr, and detect changes of the order of 1 μR/hr. During

the early years of laboratory operation, a set of rooftop Geiger-Muller tubes were placed at the stations. This equipment is described in detail in the BNL Area Survey Manual (Weiss, 1955).

When the reactor was first put into operation, radiation levels due to argon-41 at the edge of the site averaged over a week, were restricted to no more than 1% of the limit prescribed for occupational exposure (50 mR/day) (Cowan and Gemmell, 1960). Therefore 50 mR/day x 7 days x 0.01 = 3.5 mR/week became the public limit. When necessary, reactor operations were curtailed to ensure that exposures at the edge of the site did not exceed 3.5 mR/week.

Data analyzed monthly for the perimeter monitoring stations (P-2, P-4, P-7, P-9) and the monitoring stations located in the central portion of the site (P-10, P-11 and P-12) are available only for 1959, 1960, and 1961 (Attachment IA, Tables IA-6, IA-7, IA-8). Raw data for most of the earlier years may be available but are not presented here due to the limited scope of the project. Because of the strong correlation between the argon-41 released from the BGRR and the readings at the monitoring stations, identification and analysis of these data was not considered necessary to estimate the Ar-41 exposure.

Data from P-7, P-8 and P-9 for 1959, 1960 and 1961 and from the Environmental Monitoring Reports for 1962, 1963, 1964 and 1965 were used to estimate the maximum average external exposure in the central portion of the site and at the site perimeter per million Curies of argon-41 emitted from the BGRR. These relationships are derived in Table IA-5 in Attachment A and were used to generate the data in Table 6, which gives the estimated and measured radiation levels at the environmental monitoring stations associated with argon-41 from the reactor stack.

From 1951 to 1957 when the BGRR operated with the natural uranium fuel, the maximum external exposure from the argon-41 at the site boundary ranged from 10 to 14 mR/year. During 1958 to 1961 when enriched fuel was used, the maximum external exposure from the argon-41 at P-9 from the argon-41 ranged from 34 to 46 mR/year. For the central area of the site, in 1951-1957 the maximum external exposure was 56 mR/year, and in 1958-1961 it was 220 mR/year.

Table 6. Annual argon-41 released from the BGRR and measured (in bold) and estimated (see text) external exposure levels at area monitoring stations.

	Argon-41[1] (Ci)	Boundary Stations[2] (mR/year)		Central Area Stations[3] (mR/year)		Background[4] (mR/year)
		Average	Maximum	Average	Maximum	Average
1950	3.5×10^4	0.2	0.3	0.8	1.2	85
1951	1.2×10^6	5.9	10	28	40	90
1952	1.5×10^6	7.4	13	35	50	92
1953	1.6×10^6	7.8	14	37	53	96
1954	1.7×10^6	8.3	14	39	56	99
1955	1.7×10^6	8.3	14	39	56	100
1956	1.6×10^6	7.8	14	37	53	100
1957	1.4×10^6	6.9	12	32	46	110
1958	3.9×10^6	20	34	90	130	130
1959	5.1×10^6	**26**	**46**	120	170	140
1960	5.2×10^5	**25**	**39**	**140**[5]	**220**[5]	100
1961	5.6×10^6	**30**	**37**	**120**	**170**	97

[1] Total Ci of argon-41 released from the BGRR
[2] P-2,P-4,P-7,P-9
[3] P-10, P-11, P-12
[4] average background measurements available for P-7 and P-9 includes weapons fallout and instrument contamination
[5] July through December only

5.2 Estimating Particulates Releases

A major confounding factor in assessing the environmental levels of airborne radionuclides resulting from BNL operations was the fallout from weapons tests by the United States and the U.S.S.R. that took place during this period (Cowan and Steimers, 1958; Weiss, 1954). The highest daily deposition of weapons test fallout on the Brookhaven site was 300,000 disintegrations per minute, per square meter (dis/min/m^2) measured in March 1955 (Health Physics Summary March, 1955). In units used below, this value is approximately 350 millicuries per square mile (mCi/mi^2) Although particulate air samplers were in place at the environmental monitoring stations, the only available data are from two rain and settled dust collectors. Tables 7 and 8 show monthly activities measured in rain and settled dust from 1953 to 1961 given in units of milliCuries per square mile (mCi/mi^2). While natural radioactivity levels were quite consistent, the levels associated with atmospheric tests were highly variable, as were releases associated with BGRR rupture events.

The largest estimated particulate release from BGRR rupture events in 1954 was in February, while the largest value in rain and settled dust was in September. The largest estimated particulate release in 1955 from the BGRR ruptures was in October while the largest value in rain and settled dust was in September. In 1956 the largest estimated particulate release from BGRR rupture events was in

May while the highest value in rain and settled dust was in September. For 1957 the highest estimated particulate release associated with the BNL ruptures was in January while the highest value in rain and settled dust was again in September. The lack of correlation between the rain and settled dust results and the rupture events demonstrate that these measurements are of little help in estimating the releases associated with the ruptures.

Although there were particulate monitors at the base of the stack, these measurements were not found.

As a result, in this report, an approach to estimate downwind air concentrations and ground deposition of radionuclides (both particulates and iodines) used the Lawrence Livermore National Laboratory HOTSPOT code.

5.3 Radionuclides in Milk

In response to concerns about fallout from multi-national weapons tests, BNL was requested to analyze milk samples from local farms. In 1961, two sets of milk samples were obtained from farms in Center Moriches, Yaphank, and Islip and analyzed for iodine-131. All six samples showed iodine-131 activity of about 25 pCi/l, which agrees with levels in milk found by other laboratories and confirming the source of the iodine to be weapons fallout (Health Physics and Safety Summary for December 1961).

Table 7. Radionuclide activity in rain and settled dust collections, 1953-1957, mCi/mi^2 and relationship to weapons tests and rupture events.

	1953			1954			1955		
Month	Activity[1] (mCi/mi^2)	Tests[2]	Ruptures[3] (Ci of particulates)	Activity[1] (mCi/mi^2)	Tests[2]	Ruptures[3] (Ci of particulates)	Activity[1] (mCi/mi^2)	Tests[2]	Ruptures[3] (Ci of particulates)
Jan	na			38.4		R1: (2); R2: (2.9)	4.9		
Feb	na			7.2	P	R (25)	57.4	N (2)	
Mar	na	N (3)		106	P		1210	N (6)	
Apr	na	N (4)	R1: (0.7); R2: (0.2)	87.9	P (2)	R: (20)	641	N (3)	
May	na	N (3)	R: (1.1)	31.0	P (2)		346	N (2); P	
Jun	na	N (1)		21.7	P	R: (0.9)	36.5	P (8)	R: (1.1)
Jul	na		R: (7.7)	17.2		R: (17)	9.7	U	
Aug	na	U (2)	R: (17)	48.8			22.4	U (2)	
Sep	14.8	U (3)		468	U (2)		8.4	U	
Oct	10.1	P		180	U (8)		12.5	U (2)	R: (3.9)
Nov	1.5		R1:(0.3); R2(1.6)	276			13.8	U (2)	
Dec	11.3			50.4			12.0		
Total				1332.6			2374.6		

na: data not available before September 1953.

[1] Activity measured in rain and settled dust on the BNL site.

[2] Reported Nuclear Detonations, locations, and number: N=U.S. Nevada Test Site; P= U.S. Pacific Test Site; U=U.S.S.R.; A= U.S. Atlantic Test Site (Taken from UNSCEAR 2000 "Report to the General Assembly with the Scientific Annexes.")

[3] R denotes a BGRR fuel cartridge rupture, amount released is in parentheses.

Table 7 (cont.). Radionuclide activity in rain and settled dust collections, 1953-1957, mCi/mi^2 and relationship to weapons tests and rupture events.

Month	1956			1957		
	Activity[1] (mCi/mi^2)	Tests[2]	Ruptures[3] (Ci of particulates)	Activity[1] (mCi/mi^2)	Tests[2]	Ruptures[3] (Ci of particulates)
Jan	82.2			348	U	R
Feb	103	U		36.6	U	
Mar	105	U (2)		58.5	U	R
Apr	105	U		285	U	
May	125	P (5)	R1: (7.0); R2: (130)	112	N; P	
Jun	32.6	P		38.8	N; P	
Jul	212	P (5)		262	N	R
Aug	62.0	U (2)		331	N; U	
Sep	427	U (2)		726	N; P; U	
Oct	49.1	P,U	R: (4.9)	112	N; P; U	R1: (6.2); R2: (3.0)
Nov	32.0		R: (3.5)	44.3	P; U	
Dec	105.0	U		97.4	N; U	
Total	1439.9			2138.4		

na: data not available before September 1953.

[1] Activity measured in rain and settled dust on the BNL site.

[2] Reported Nuclear Detonations, locations, and number: N=U.S. Nevada Test Site; P= U.S. Pacific Test Site; U=U.S.S.R.; A= U.S. Atlantic Test Site. (Taken from UNSCEAR 2000 "Report to the General Assembly with the Scientific Annexes.")

[3] R denotes a BGRR fuel cartridge rupture, amount released is in parentheses.

Table 8. Radionuclide activity in rain and settled dust collections, 1958-1961, mCi/mi^2 and relationship to weapons tests[1,2].

Month	1958	1959	1960	1961
January	102 (U, 2)	208	8.1	2.3
February	18.4 (U, 3)	232	270	3.5
March	144 (U, 8)	535	3.8	9.2
April	435 (P)	311	9.8	275
May	246 (P, 3)	85.4	10.4	19.2
June	101 (P, 4)	152	10.6	5.0
July	245 (P, 3)	19.2	5.2	5.8
August	197 (N; A; P)	14.5	2.0	9.2
September	59.6 (N, 2; A; U, 2)	2.6	15.6	100 (U, 26)
October	1109 (N, 13; U; 17)	6.5	11.2	180 (U, 22)
November	356 (U, 2)	4.9	2.4	416 (U, 10)
December	224	3.9	2.0	372
Total	3237.0	1575.0	351.1	1397.2

[1] Reported Nuclear Detonations, locations, and number: N=U.S. Nevada Test Site; P= U.S. Pacific Test Site; U=U.S.S.R.; A= U.S. Atlantic Test Site. (Taken from UNSCEAR 2000 "Report to the General Assembly with the Scientific Annexes.")

[2] There were no BGRR fuel cartridge ruptures from 1958-1961.

6. LIQUID EMISSIONS AND MONITORING

6.1 Liquid Waste System

Beginning in 1948, low-level liquid radioactive wastes at BNL were disposed of by release into a conventional sanitary waste treatment facility. Even before the BGRR came on-line, the Chemistry, Physics and Biology Departments used radionuclides that contributed to small concentrations of radionuclides in the sewage effluent.

Liquids in the sanitary waste system passed through an Imhoff Tank, which removed most of the solids. The effluent flowed onto sand-filter beds, from which most of it was directed by an underlying tile field, chlorinated, and discharged into a small stream which forms one of the headwaters of the Peconic River.

Figure 14 is a schematic illustration of the sewage plant as it existed during the period covered by this report. The monitoring arrangements included continuous proportional samplers at the discharge from the Imhoff Tank and at the point where the liquid effluent was discharged to the stream.

Total beta activity discharged to the sand-filter beds and released from the Chlorination Plant was measured. Routine analyses of the liquid effluent for specific radionuclides were not performed before 1961, but occasional analysis indicated that most of the activity in the effluent consisted of long-lived fission-products in which cesium-137 predominated.

Periodic measurements of the sewage effluent began late in 1948. These early samples were simple grab samples. A formal analysis and reporting system was established in 1951 when proportional samplers were developed that automatically took samples at the stations on a periodic basis.

Thus routine liquid emissions data became available in 1951. Measured values of gross beta concentration and activity discharged to the filter beds (Imhoff Tank), discharge to the river (Chlorination Plant) and computed values at the original site boundary (station M) are given for each year beginning in 1951 in Tables IB-1 through IB-33 in Attachment I-B. These data were abstracted from monthly Health Physics and Safety summaries.

An approximation of the strontium-90 and cesium-137 concentrations were estimated using a relationship between the total beta concentration and of these two radionuclides derived from data published in the Environmental Monitoring Reports for 1962, 1963, 1966 and 1968. The derived relationship is 50% of gross beta for cesium-137 and 10% of gross beta for strontium-90.

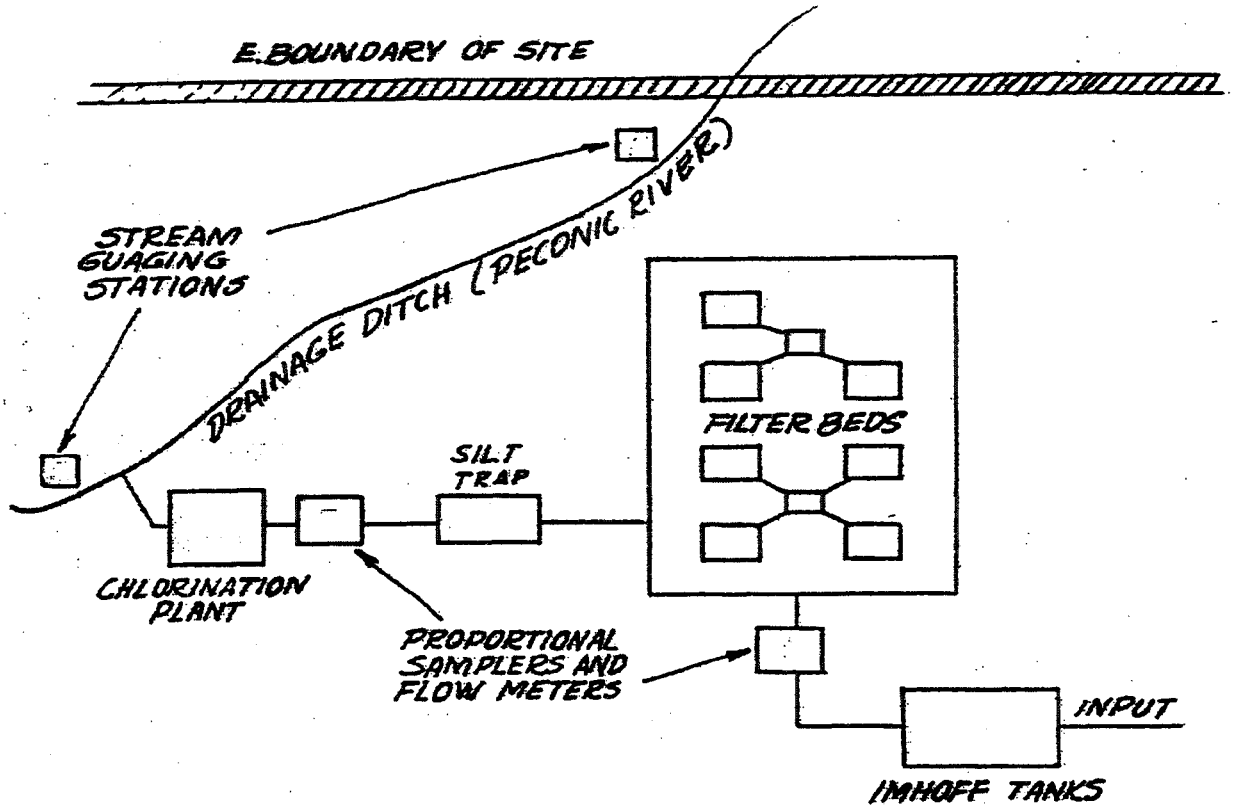

Figure 14. Sewage Treatment Plant, as it was from 1950-1961.

Discharges into the Peconic River for 1951-1961 were higher than in later years (Meinhold and Hull, 1998). These higher levels of gross beta and cesium-137 resulted from waste associated with the storage and handling of the irradiated natural-uranium slugs used to fuel the BGRR from 1950-1958. This stored fuel was not entirely removed from the reactor canal until 1962.

1948 - 1950

During this period routine grab sampling on an hourly schedule was used to estimate liquid releases since on-line proportional samplers were not yet available. The data reported in the monthly Health Physics Summary Reports are given below for this time period. The reported sensitivity for routine analysis was given as 10^{-12} to 10^{-13} Ci/cc.

1948

Sewage and water samples were taken routinely, starting late in 1948. In most cases, no measurable activity was detected (11/1948).

1949

In 1949, low levels of activity were detected in the sewage effluent.

March: Concentrations of the order of 3×10^{-14} Ci/cc were detected at the exit of the Imhoff Tank on several occasions. A maximum concentration of 2.3×10^{-15} Ci/cc was detected in the Peconic River below the point of discharge.

April: Activity levels at the exit of the Imhoff Tank were high enough to be detectable for most daily samples tested, ranging from a barely detectable 10^{-14} Ci/cc, to 6×10^{-13} Ci/cc. This peak concentration was due to radiophosphorous used in the Biology Department.

May: Daily samples of the Imhoff tank effluent showed background or barely detectable activities throughout the whole month.

June: Effluent from the Imhoff tank was at background except for a few days when levels of about 10^{-14} Ci/cc were measured.

July: Sewage effluent was at, or near, background for the entire month.

August: Sewage effluent showed only background activity except for two days when traces of activity were measured.

September: No detectable activity was noted in the laboratory sewage effluent for the month.

October: There was no detectable activity in the sewage effluent for the month, except on a few days when barely detectable levels of 10^{-14} Ci/cc were noted.

November: All water and sewage samples showed background levels, except for the sewage sample of November 8, which had a concentration of about 10^{-12} Ci/cc. This was due to a cobalt-60 spill at Building T-527.

December: No data were found for December.

1950

January: Sewage effluent was at background except for three days when the activity was about 3×10^{-14} Ci/cc. This was caused by discharging of the contents of the radiochemistry laboratory's holdup tank.

February: All sewage samples read background except on February 2, which was 10^{-13} Ci/cc of beta-gamma. This was caused by the discharge of the Chemistry holdup tank.

March: The sewage effluent activity was at background except on one day when it contained of 3×10^{-14} Ci/cc.

April: The sewage effluent showed no detectable activity in April.

May: The sewage effluent was at background except on two dates when activity of about 10^{-13} Ci/cc was detected. This was due to the emptying of the Chemistry holdup tank.

June: The sewage effluent was at background during June.

July: Sewage activities greater than the minimum detectable amount of about 10^{-14} Ci/cc were noted in eight consecutive days. Values ranged from 10^{-13} to 2×10^{-14} beta-gamma activity, with no alpha activity.

August: On August 10, the Laboratory began processing the daily Imhoff tank effluent without using nitric acid. By this method a detectable concentration was obtained every working day for the rest of the month. The concentration for the non-acid samples varied from 2×10^{-14} to 4×10^{-12} Ci/cc, with an average of 5×10^{-13} Ci/cc. Average concentrations with nitric acid appeared to be about 1/10 as great. Decay studies indicated that the activity was primarily iodine-131. If the above average was representative, the Laboratory discharged about 700 μCi of iodine-131 per day, since the daily flow amounts to about 375,000 gallons. It was believed that the iodine was from the Medical Department's Iodine Therapy Program.

September: Low levels of about 5×10^{-14} Ci/cc were detected in the Imhoff Tank's effluent on most days during September, with isolated values of 2×10^{-13} and 5×10^{-13}. Only small differences were noted in samples processed with and without nitric acid, suggesting that the proportion of iodine present had decreased markedly compared with August.

October: The Imhoff Tank's effluent had a maximum activity of 5×10^{-13} Ci/cc.

November: The average flow rate at the Imhoff tank was 1.6×10^{6} liters/day and at the Chlorination Plant, it was 7×10^{5} liters/day. Activities in the 10^{-13} to 10^{-14} Ci/cc range were noted at the Imhoff Tank 18 days out of the month.

December: Sewage samples at the Imhoff Tank showed an activity less than the minimum detectable concentration of 10^{-14} Ci/cc about half the time, and values ranging from 10^{-13} Ci/cc to 10^{-14} Ci/cc the rest of the time. Activity at the Chlorination Plant was non-detectable except on two days when very low or possibly spurious values of about 3×10^{-14} Ci/cc were obtained.

1951 - 1961

By 1951, proportional samplers were in place, and monthly data were consistently reported for flows and concentrations at the Imhoff Tank and the Chlorination Plant. The proportional samplers included a provision for actuating a sampler with each 2,000 gallons of flow in combination with a positive-action battery-operated sampler. Table 9 summarizes data for the years 1951-1961. Detailed tables with monthly data are given in Attachment I-B.

Table 9. Average concentrations (pCi/l) of gross beta, strontium-90 (Sr-90) and cesium-137 (Cs-137) at the Sewage Treatment Plant Imhoff Tank, Chlorination Plant, and the site boundary.

Year	Imhoff Tank			Chlorination Plant			Site Boundary		
	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs-137 (pCi/l)	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs-137 (pCi/l)	Gross Beta (pCi/l)	Sr-90 (pCi/l)	Cs137 (pCi/l)
1951	570	57	280	100	10	51	88	9	44
1952	330	33	170	99	10	50	45	4	22
1953	310	30	150	110	11	55	25	3	13
1954	430	43	220	100	10	50	47	5	24
1955	450	45	230	170	17	86	52	5	26
1956	320	32	160	120	12	59	26	3	13
1957	370	37	190	160	16	82	94	9	47
1958	530	53	270	200	20	100	34	3	17
1959	780	78	390	260	26	130	65	7	33
1960	650	65	330	310	31	150	260	26	130
1961	430	43	210	300	30	150	170	17	85

The descriptions given below were taken from Health Physics Summary reports.

1951

In 1951, the total waste-water flow measured at the Imhoff Tank was >3 x 10^8 liters (no data were found for January and February). About 76% of the liquid effluent discharged onto the sand-filter beds at the Sewage Treatment Plant was directed to the Peconic River, as measured at the Chlorination Plant.

The average concentration of gross-beta emitters in the effluent discharged onto the sand-filter beds was 566 pCi/l (Table IB-1). The average concentration of beta emitters in the waters released to the Peconic River at the Chlorination Plant was 102 pCi/l (Table IB-2) which was 3.4% of the applicable radiation protection guide of 3,000 pCi/l. The change in the concentration of beta emitters through the sand-filter beds was primarily due to their holdup in the top few inches of the beds.

At the site's perimeter (original site perimeter, station M), the yearly average concentration of gross beta emitters was estimated to be about 88 pCi/l (Table IB-3).

The unusually high values for May were primarily due to the release of iodine-131, particularly in the urine of patients receiving therapeutic doses (Health Physics Summary, May 1951). The unusually high values for September were the result of a release of 20 mCi from the Hot Laboratory on September 11, 1951.

1952

In 1952, of the total waste-water flow discharged onto the sand-filter beds of 3.6 x 10^8 liters, 74% was directed to the Peconic River. The average concentration of gross-beta emitters in this discharge to the sand-filter beds was 333 pCi/l (Table IB-4) which was 11% of the limit of 3000 pCi/l.

The average concentration of gross beta emitters in the waters released to the Peconic River at the Chlorination Plant was 99 pCi/l (Table IB-5).

At the site perimeter station M, the total flow was 6.2 x 10^8 l. The concentration of gross-beta emitters at station M was estimated to be 45 pCi/l (Table IB-6).

1953

The total BNL waste-water flow discharged onto the sand-filter beds in 1953 was 4.1 x 10^8 liters. About 78% of the volume of the liquid effluent was directed to the Peconic River at the Chlorination Plant. The average concentration of gross-beta emitters in the effluents released to the sand-filter beds was 305 pCi/l (Table IB-7), which was 10% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in the water released to the Peconic River at the Chlorination Plant was 111 pCi/l (Table IB-8).

At the site perimeter station M, the total flow was 1.36 x 10^9 liters, and the concentration of gross-beta emitters was estimated to be 25 pCi/l (Table IB-9).

1954

In 1954, the total wastewater flow at the Imhoff Tank was 3.9 x 10^8 liters, approximately 85% of which was directed to the Peconic River. The concentration of gross-beta emitters in the wastewater was about 430 pCi/l (Table IB-10), which was 14.3% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in the water released to the Peconic River at the Chlorination Plant was 100 pCi/l (Table IB-11).

At the site perimeter station M, the total flow was 7.6 x 10^8 liters, and the concentration of gross-beta emitters was estimated to be 47 pCi/l (Table IB-12).

1955

In 1955, the total waste-water flow measured at the Imhoff Tank was 4.8 x 10^8 liters. About 87% of the liquid effluent discharged onto the sand-filter beds was directed to the Peconic River as measured at the Chlorination Plant.

The average concentration of gross-beta emitters in the effluent discharged onto the sand-filter beds was 453 pCi/l (Table IB-13), which was 15% of the limit of 3000 pCi/l. The average concentration of beta emitters in the waters released to the Peconic River at the Chlorination Plant was 171 pCi/l (Table IB-14).

At the site's perimeter (station M) the yearly average concentration of gross-beta emitters was estimated to be about 52 pCi/l (Table IB-15).

1956

In 1956, 84% of the total waste-water flow of 5.2 x 10^8 liters discharged onto the sand-filter beds was directed to the Peconic River. The average concentration of gross-beta emitters in the effluent discharged onto the sand-filter beds was 320 pCi/l (Table IB-16), which was 10.6% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in the waters released to the Peconic River at the Chlorination Plant was 118 pCi/l (Table IB-17).

At the site perimeter station M, the total flow was 2.1 x 10^9 liters. The concentration of the gross-beta emitters was estimated to be 26 pCi/l (Table IB-18).

1957

In 1957, the total wastewater flow at the Imhoff Tank was about 6.0 x 10^8 liters; about 73% of it was directed to the Peconic River. The average concentration of gross-beta emitters in the effluent discharged to the sand-filter beds was 372 pCi/ (Table IB-19), which was 12.4% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in water released to the Peconic River at the Chlorination Plant was 164 pCi/l (Table IB-20).

At the perimeter station M, the average concentration of gross-beta emitters was 76.7 pCi/l (Table IB-21).

1958

The total waste-water flow discharged onto the sand-filter beds in 1958 was 6.1 x 10^8 liters, of which about 84% was directed to the Peconic River. The average concentration of gross-beta emitters in the effluent was 531 pCi/l (Table IB-22), which was 17.7% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in water released to the Peconic River at the Chlorination Plant was 204 pCi/l (Table IB-23).

At perimeter station M, the average concentration of gross-beta emitters was 34 pCi/l (Table IB-24).

1959

In 1959, the total waste-water flow at the Imhoff Tank was about 7.6 x 10^8 liters. About 71% of the total volume of the liquid effluent discharged onto the sand-filter beds at the Sewage Treatment Plant was directed to the Peconic River. The average concentration of gross-beta emitters in the effluent discharged to the sand-filter beds was 776 pCi/l (Table IB-25), which was 25.9% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in water released to the Peconic River at the Chlorination Plant was 262 pCi/l (Table IB-26).

At perimeter station M, the average concentration of gross-beta emitters was 65 pCi/l (Table IB-27).

The unusually high values for June and July were "traced to the evaporation plant where a number of defective tubes were permitting active waste to leak across into the condensate" (Health Physics Summary, July 1959). The unusually high values in November and December were attributed to the "pumping of D waste, accumulating in a sump at the hot lab into the sewer rather than into a D tank as required by established procedures" (Health Physics Summary, November 1959).

1960

In 1960, the total waste-water flow at the Imhoff Tank was about 6.4 x 10^8 liters. About 91% of the total volume of the liquid effluent discharged onto the sand-filter beds was directed to the Peconic River. Its average concentration of gross - beta emitters was 649 pCi/l (Table IB-28), which was 21.6% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in water released to the Peconic River at the Chlorination Plant was 309 pCi/l (Table IB-29).

At the perimeter station M, the average concentration of gross-beta emitters was 255 pCi/l (Table IB-30).

The source of the unusually high value for November was not clearly identified. It was, however, identified as being mostly cesium-137.

1961

In 1961, the total wastewater flow at the Imhoff Tank was about 7.6 x 10^8 liters, and about 86% of the total volume of the liquid effluent was directed to the Peconic River. The average concentration of gross-beta emitters in the effluent discharged to the sand-filter beds was 426 pCi/l (Table IB-31), which was 14.2% of the limit of 3000 pCi/l.

The average concentration of gross-beta emitters in water released to the Peconic River at the Chlorination Plant was 300 pCi/l (Table IB-32).

At the perimeter station M, the average concentration of gross-beta emitters was 170 pCi/l (Table IB-33).

6.2 Peconic River Grab Samples

In 1960, surveillance started of the downstream river system into which the treated effluent was discharged. Monthly grab water samples were obtained at on- and off-site locations along the upper tributary of the Peconic River, into which BNL routinely discharged low-level radioactive wastes. Reference grab-samples also were obtained from other nearby streams and bodies of water outside the Laboratory's drainage area. Samples of sediment, vegetation, and animals were not taken in the years covered by this report. The sampling locations were as follows.

Peconic River, proceeding downstream (within the Laboratory's drainage area)

A Peconic River at Schultz Road, 15,900 ft. downstream
B Peconic River at Wading River-Manorville Road, 23,100 ft. downstream
D Peconic River at Calverton, 46,700 ft. downstream

Controls (not within the Laboratory's drainage area)

E Peconic River, upstream from BNL effluent outfall
F Peconic River, north tributary (independent of BNL drainage)
G Carmans River at Middle Island
H Carmans River, outfall of Yaphank Lake
I Artist's Lake (maintained by water table, no surface flow)
J Lake Panamoka (maintained by water table, no surface flow)

The individual yearly average gross-beta concentrations at these downstream and control locations are given in Table 10 for 1960 and 1961.

Table 10. Yearly averages for 1960 and 1961 gross beta in surface water samples, pCi/l (from EM report 1965).

Sample Locations	1960	1961
On-Site		
M*	255	170
Peconic River		
A	20	34
B	13	19
D	11	17
Control Locations		
E	8	17
F	5	10
G	7	6
H	9	9
I	13	14
J	6	16

*estimated from river flow and monthly total releases at the Chlorinating Plant.

1960

The largest yearly average gross-beta concentration at the sampled stations in 1960 was an estimated 255 pCi/l at the onsite perimeter station M. Off site, the

highest measured concentration of 20 pCi/l was at station A, the downstream location closest to the BNL site boundary. Gross-beta concentrations decrease with distance from the laboratory. The concentrations at control locations ranged from 5 to 13 pCi/l.

1961

The largest yearly average gross beta concentration at the sampled stations in 1961 was an estimated 170 pCi/l at the onsite perimeter station M. Off site, the highest measured concentration was at station A, the downstream location closest to the BNL site boundary, where the average gross-beta concentrations were 34 pCi/l. Gross-beta concentrations decrease with distance from the laboratory. Control-location concentrations ranged from 6 to 17 pCi/l.

6.3 Losses to Groundwater

Historical operations at BNL have resulted in soil, sediment and groundwater contamination. An Inter-Agency agreement is in effect among the United States Department of Energy, the United States Environmental Protection Agency and the New York State Department of Environmental Conservation. This IAG requires that environmental impacts associated with past activities at BNL are thoroughly investigated and appropriate response actions formulated, assessed, and implemented.

Areas of Concern (AOCs) being addressed under the IAG include both active facilities such as the Sewage Treatment Plant and inactive facilities such as the former landfills, cesspools, and radioactive waste storage tanks.

Characterizing the extent of contamination in soil, sediment, and groundwater associated with past practices at BNL is the responsibility of the Environmental Restoration Division. Lengthy assessments have been and are being published that document the extent of contamination and DOE's proposed response actions. A historical site review was conducted in 1993 to identify potential sources of contamination. This information is available at local libraries in the Administrative Record for the site and are not discussed here.

The following section discusses releases to groundwater that were known and documented during the early years of the laboratory history. Releases from cesspools and the former landfills had not been documented.

Sand-Filter Beds

Of the total volume of waste-water discharged onto the sand-filter beds, approximately 70-90% was recovered at the Chlorination Plant. The percentage varied considerably over a year. The balance is assumed to have percolated into the groundwater underlying the filter bed. Some of this groundwater then moved

into the Peconic River, depending on the time of year, the height of the water table, and of the stream.

Less than 25% of the fission product activity reaching the sand-filter beds passed immediately through the beds and into the Peconic River. Hull (1970) analyzed the radionuclide retention of the sand-filter beds. Table 11 gives the fractions of input activity retained. This retention includes loss to ground water and radioactive decay. The material retained by the sand-filter beds was periodically removed and disposed of in the sanitary landfill.

Table 11. Gross beta activity at the sand-filter beds (from Hull, 1970).

Year	Influent (mCi)	Effluent (mCi)	Yearly Fraction Retained
1951	160.5	21.5	0.87
1952	116.6	27.9	0.76
1953	132.9	35.8	0.73
1954	182.1	48.5	0.73
1955	223.8	75.0	0.67
1956	170.0	55.0	0.68
1957	300.8	105.1	0.65
1958	325.1	106.0	0.67
1959	586.6	169.5	0.71
1960	542.9	177.8	0.67
1961	384.4	219.1	0.43

Sewer Lines

Many of the sewer pipes that transport liquid effluents from the main laboratory complex to the sewage-treatment plant were inherited from the days when the site was a large Army camp. It is reasonable to assume that they lost significant amounts of effluent to the surrounding soil. Hull (1970) reviewed eighteen releases of one millicurie or more into the sewage collection system for which the amounts discharged were reported. Almost all of these releases consisted of mixed fission products. Hull concluded that there was considerable hold-up and/or loss of activity in the conduits leading to the sewage-treatment plant, as well as in the plant itself. Only 34% of the amounts of mixed fission products reported as having been released into the system passed onto the sand-filter beds.

Contamination Incidents

In November 1960, it was discovered that the Biology Department's 16 Curie cobalt-60 source was leaking. It was determined that a total of 8 mCi was in the water inside the lead source-container. Soil samples, taken from beneath the drainage holes in the outer container, showed activity of 20,000 disintegrations per minute per gram (d/m/g) near the holes. The activity decreased very quickly

to 500 d/m per gram at a depth of 21 inches, and there was no activity detected at a depth of 4 feet. The total activity in the soil was estimated to be about 10 mCi, based on these data and those from some additional laterally located samples. Because of the known retention of the radionuclides by the soil, it was concluded that no more than a few mCi could have escaped to the groundwater. A test-well was driven in the most favorable direction as chosen by the USGS for detecting activity. Water samples were taken before, during, and after the well was pumped. There was no detectable activity.

Groundwater downgradient of a Waste Management area well was accidentally contaminated in 1960. The data on this incident are contained in previously reported documentation (SAIC, 1992). This incident occurred when a technician thought he was putting evaporator concentrate slurry into an underground storage tank. In fact, he put the concentrate waste down a monitoring well. The resulting contamination has been addressed in the BNL restoration program.

7. UNCERTAINTIES AND USE OF THIS REPORT

7.1 Summary Of Uncertainties

This report presents emissions, release estimates and environmental monitoring information from the early years of BNL's operation. A number of gaps in the available information generated 50 years ago were filled based on assumptions and analyses of the data that were available. In all cases, the intent was to use conservative, (pessimistic), but reasonable assumptions.

Each set of release estimates and monitoring data presented in this report carries its own uncertainties and assumptions. These are discussed in the following sections. The highest degree of uncertainty is associated with the estimates of the releases from the fuel ruptures. They are discussed here, and in the detailed analysis of the rupture events which is presented in Part II.

7.1.1 Airborne Emissions

BGRR Routine Emissions

Routine emissions of radionuclides from the BGRR are inherently uncertain because they were not measured directly. Emissions given in this report (Section 4) were based on documented relationships between the power level of the reactor and releases of argon-41 and carbon-14. Power levels were available in the monthly Reactor Operations Summary Reports. Since the argon-41 releases were measured in 1965 (Hull, 1967), these relationships provide very good estimates of the carbon-14 and argon-41 releases from the BGRR for the period 1957-1961. For the earlier period (1950-1956) the argon-41 release was based on design-based calculations. As a result the earlier argon-41 release estimates are good, but less certain than the later values.

The routine release of iodine-131 for the period 1957 to 1961 is based on a series of measurements made in 1963. The source of iodine-131 was fissioning of uranium-235 trapped on the surface of the aluminum cladding. This surface contamination occurred during the manufacture of the enriched fuel elements. This report used a proportional relationship between the power level of the reactor and iodine-131 releases. The major uncertainty in this assumption is the expectation that the amount of uranium surface contamination of the fuel elements was consistent. As a result the values are more uncertain than the estimates of argon-41 and carbon-14, but still provide fairly good estimates of the iodine-131 releases during this period.

Iodine-131 releases for the early years of reactor operation when the reactor was using natural uranium fuel are even more uncertain. The Reactor Operations Monthly Reports reported that there were no fission products released during

normal operation. However, it seems likely that some radioiodine and noble gases were released via the helium system. The approach used in this analysis is based on the release associated with enriched fuel so the uncertainty in that estimate is also embedded in the estimate for the natural uranium fuel. In addition, the mechanics of transport through the helium system would have reduced the radioiodine concentrations. The largest uncertainty is associated with the assumption that the uranium-235 on the surface of the natural uranium fuel is similar to that on the cladding of the enriched fuel. Taken together it appears that the routine release of iodine from the natural uranium fuel is highly uncertain, although small.

Very small amounts of krypton-85, and iodine-129 would also have been expected in the BGRR effluent.

BGRR Rupture Release Estimates

The estimates of the releases during the 29 ruptures are the most uncertain values given in this report. The approach taken was to use qualitative descriptions of the events as well as any quantitative data that were available, and to apply professional judgement in making reasonable but conservative assumptions. Data and information used in developing the release estimates are given in the report (Part II) and in detailed attachments. This allows the reader to follow the logic used in making subjective assumptions and to develop alternative estimates if desired.

Major assumptions made in developing release estimates included those related to estimates of the inventories in individual slugs, in estimating the fraction of the inventory released from a single slug, and in estimating the number of slugs that were lost in each rupture.

Inventories of important radionuclides were developed by modifying the estimates developed for the Oak Ridge X-10 reactor based on the time the fuel channel was in the reactor and the higher BGRR flux. There is some uncertainty in this estimate because it does not consider the location of each cartridge in the reactor, however, the flux distribution across the reactor was reasonably uniform while the reactor was running with natural uranium fuel (BNL, 1958).

The available information on the plutonium inventory included estimates of 0.4 g/slug (from the X-10 inventory), 0.57 and 0.59 g/slug (Reactor Operations Monthly Reports) and 0.8 g/slug ("highest group average", Shepherd, 1997). The selection of 1 g/slug used in this report is based on the expectation that the fuel cartridges with the longer operating histories are most likely to rupture and also to have the larger plutonium inventories. The 1 g/slug assumption is a high estimate, but is likely to be within a factor of 2 of the actual value.

Release fractions for noble gases and iodines were based on the assumptions

used in the assessment of the Oak Ridge X-10 reactor (100% noble gases, 80% iodines). It was assumed that the BGRR filter and extended ductwork did not remove any additional noble gas or radioiodine activity. This may overestimate the quantity released but provides a reasonable assumption for noble gases. Assuming that 80% of the iodines in a slug were released from the stack could overestimate the amount released by a factor of 10.

As discussed in detail in Part II, the number of slugs lost in a rupture, and the percent of particulates released from a single slug have a high degree of uncertainty because the measurements were very rough, and the extent to which the stack monitoring data for a particular rupture are relevant to other ruptures is also uncertain. The analysis of Rupture 29, where the entire sample is assumed to have been released suggests that particulate releases may be overestimated by about a factor of 5 or more.

Estimates for the non-generic ruptures are more uncertain than for the generic rupture. Several ruptures (13, 16, and 18) were difficult to analyze because of conflicting or missing information.

BMRR Routine Release Estimates

Estimates of routine releases of argon-41 from the BMRR were derived from a direct relationship with power level from data collected in 1997. These emissions were not directly measured, but the relationship between power and argon-41 should provide a reasonable estimate. Other radionuclides are released in the BMRR effluent, but these have extremely low activities and short-half lives and were not estimated.

Other Sources of Air Emissions

Emissions from the Hot Laboratory were estimated for the iodine production program and from a release of uranium hexafluoride that occurred in 1957. There are large uncertainties in the releases associated with these situations. Both the iodine and uranium releases are upper estimates. There were probably other small releases for radionuclides to the air during the time covered by his report, related to work being done at the Hot Laboratory, Medical, Chemistry and Biology. The significant releases were noted in the Health Physics Summary Reports and are reflected in this report. There are no other records of airborne releases from other facilities.

7.1.2 Air, Precipitation and Settled Dust Monitoring

External radiation levels and airborne particulate radionuclides were measured at area monitoring stations located on the BNL site and at the site boundary (Section 5). For measurements of external exposure, background at a given station, excluding radon and its decay products, was determined from the

radiation level prevailing when no obvious BNL contributions were detectable at the station. The potential error in making this determination was minimized by reference to meteorological data (to establish the direction of the reactor's air effluent plume) and to the logbook indications of the gamma -field and ecology forest sources.

A major confounding factor in assessing the environmental impacts of airborne radionuclides was the fallout from weapons tests. Furthermore, the detectors used during this time contained small quantities of naturally occurring radionuclides in the insulating material of the ion chamber that contributed to an over-estimate of exposures.

Monthly-analyzed data for the perimeter monitoring stations P-2, P-4, and P-7 are available only for 1959, 1960, and 1961. Estimates for the other years and for Station P-9 were derived using the relationship between argon-41 releases at the BGRR and measurements made at the monitoring stations when both emissions and boundary exposures were measured. These values are uncertain because they were not measured directly, but are felt to represent reasonable estimates because the relationship between the annual amount of argon-41 released by the BGRR and the amount of external radiation measured at the perimeter monitoring stations did not vary significantly over the years.

The major uncertainty in particulate monitoring is due to the lack of data from the particulate air sampling program. The rain and settled dust collections provide a reasonable overview of the level of air contaminants, but the relationship to individual ruptures is quite uncertain.

7.1.3 Liquid Emissions

Liquid Waste System

Data describing water flow rates and concentrations of gross beta activity were abstracted from monthly summary reports (Section 6). Monthly averages of the total beta activity concentrations were available at the Imhoff Tank and at the Chlorination Plant. To estimate concentrations at the site boundary, the total activity discharged at the Imhoff tank was calculated, and then assumed to be present in the water that passed station M. This is an uncertain estimate because the flows were affected by rainfall that occurred during the month, and not all of the discharged activity would actually have left the site boundary.

In the early history of the Laboratory individual radionuclides were not usually analyzed for and reported separately. To support a comparison with data collected in later years, concentrations of cesium-137 and strontium-90 were estimated as a percentage of total beta activity, based on data reported for 1966 and 1968. This relationship probably overestimates the concentrations of these radionuclides for the early years of the Laboratory's history, because strontium-

90 and cesium-137 concentrations in the effluent would have been lower when the evaporator and tank farm were not in use and thus fission product waste was not procured for disposal.

Flow measurements were reported at the Imhoff Tanks, the Chlorination Plant and perimeter station M. These flows are important because they were used to estimate the total activity

Measurements in Surface Water and Milk

Data for surface water samples in the Peconic River and other control locations were found only for 1960 and 1961. Data for concentrations of iodine-131 in milk were reported only in 1961. These samples were obtained and analyzed in response to a large deposition of weapons fallout.

Losses to Groundwater

Losses to groundwater are only qualitatively estimated for the sewage lines. It is known that releases to groundwater occurred through leaks in the sewage lines, discharges to the sand-filter beds at the sewage treatment plant, disposal of materials on landfills, and documented spills. Data describing these releases are limited in the early years of the Laboratory. As mentioned earlier these groundwater impacts have been identified and are being addressed under BNL's Environmental Restoration Program.

7.2. Application of the Data Given in this Report

The data presented here are meant primarily to fill the gap in the sequence of BNL's reporting of environmental monitoring data. It will also be made available to the ATSDR in developing a public health assessment for BNL. This report does not address the estimation of doses associated with BNL operations. However, the data presented here are of sufficient quality to form the basis of a screening assessment of on- and off-site doses. Transport modeling and conservative assumptions can be used to estimate doses to a hypothetical individual living at the site boundary during the early years of the site's operation. This screening level analysis is the kind of assessment that is provided in BNL's annual site environmental monitoring reports.

The data are not sufficient to form the basis of a quantitative dose reconstruction. The screening assessment could be used to determine the need for such a time and resource intensive effort. If a dose reconstruction effort were to be completed at BNL, this report would provide a starting point for the development of definitive exposure estimates.

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ATTACHMENT I-A

POWER LEVELS AND ROUTINE EMISSIONS FROM THE BGRR AND BMRR

Table IA-1. Monthly power levels and estimated routine argon-41, iodine-131, particulates, and carbon-14 emissions for the BGRR when natural-uranium fuel was used (8/50 through 12/57).

Year-Month		Days/ month	Power for month (MW-day)	Power for month (MW)	Argon-41 (Ci)	Iodine-131 (Ci)	Particulates (Ci)	Carbon-14 (Ci)
1950	Aug	31	42.29	1.36	9.30E+03	2.16E-04	0.47	0.42
	Sep	30	7.38	0.25	1.62E+03	3.76E-05	0.08	0.07
	Oct	31	24.62	0.79	5.42E+03	1.26E-04	0.27	0.25
	Nov	30	39.26	1.31	8.64E+03	2.00E-04	0.43	0.39
	Dec	31	43.34	1.40	9.54E+03	2.21E-04	0.48	0.43
1951	Jan	31	211.34	6.82	4.65E+04	5.39E-03	2.32	2.11
	Feb	28	695.42	24.8	1.53E+05	1.77E-02	7.65	6.95
	Mar	31	474.58	15.3	1.04E+05	1.21E-02	5.22	4.75
	Apr	30	358.26	11.9	7.88E+04	9.14E-03	3.94	3.58
	May	31	491.36	15.9	1.08E+05	1.25E-02	5.40	4.91
	Jun	30	478.45	16.0	1.05E+05	1.22E-02	5.26	4.78
	Jul	31	430.92	13.9	9.48E+04	1.10E-02	4.74	4.31
	Aug	31	436.92	14.1	9.61E+04	1.11E-02	4.81	4.37
	Sep	30	431.00	14.4	9.48E+04	1.10E-02	4.74	4.31
	Oct	31	556.21	17.9	1.22E+05	1.42E-02	6.12	5.56
	Nov	30	553.95	18.5	1.22E+05	1.41E-02	6.09	5.54
	Dec	31	499.93	16.1	1.10E+05	1.27E-02	5.50	5.00
1952	Jan	31	581.82	18.8	1.28E+05	1.48E-02	6.40	5.82
	Feb	29	549.39	18.9	1.21E+05	1.40E-02	6.04	5.49
	Mar	31	575.19	18.6	1.27E+05	1.47E-02	6.33	5.75
	Apr	30	589.65	19.7	1.30E+05	1.50E-02	6.49	5.90
	May	31	586.26	18.9	1.29E+05	1.49E-02	6.45	5.86
	Jun	30	580.11	19.3	1.28E+05	1.48E-02	6.38	5.80
	Jul	31	538.95	17.4	1.19E+05	1.37E-02	5.93	5.39
	Aug	31	542.14	17.5	1.19E+05	1.38E-02	5.96	5.42
	Sep	30	619.58	20.7	1.36E+05	1.58E-02	6.82	6.20
	Oct	31	615.37	19.9	1.35E+05	1.57E-02	6.77	6.15
	Nov	30	598.89	20.0	1.32E+05	1.53E-02	6.59	5.99
	Dec	31	559.49	18.1	1.23E+05	1.43E-02	6.15	5.59
1953	Jan	31	669.72	21.6	1.47E+05	1.71E-02	7.37	6.70
	Feb	28	609.01	21.8	1.34E+05	1.55E-02	6.70	6.09
	Mar	31	677.51	21.9	1.49E+05	1.73E-02	7.45	6.78
	Apr	30	673.44	22.5	1.48E+05	1.72E-02	7.41	6.73
	May	31	680.60	22.0	1.50E+05	1.74E-02	7.49	6.81
	Jun	30	634.19	21.1	1.40E+05	1.62E-02	6.98	6.34
	Jul	31	634.19	20.5	1.40E+05	1.62E-02	6.98	6.34
	Aug	31	690.49	22.3	1.52E+05	1.76E-02	7.60	6.90
	Sep	30	662.32	22.1	1.46E+05	1.69E-02	7.29	6.62
	Oct	31	422.23	13.6	9.29E+04	1.08E-02	4.64	4.22
	Nov	30	581.56	19.4	1.28E+05	1.48E-02	6.40	5.82
	Dec	31	484.53	15.6	1.07E+05	1.24E-02	5.33	4.85

Table IA-1. (cont.)

Year-Month-		Days/ month	Power for month (MW-day)	Power for month (MW)	Argon-41 (Ci)	Iodine-131 (Ci)	Particulates (Ci)	Carbon-14 (Ci)
1954	Jan	31	574.57	18.5	1.26E+05	1.47E-02	6.32	5.75
	Feb	28	523.69	18.7	1.15E+05	1.34E-02	5.76	5.24
	Mar	31	716.57	23.1	1.58E+05	1.83E-02	7.88	7.17
	Apr	30	632.88	21.1	1.39E+05	1.61E-02	6.96	6.33
	May	31	667.29	21.5	1.47E+05	1.70E-02	7.34	6.67
	Jun	30	658.15	21.9	1.45E+05	1.68E-02	7.24	6.58
	Jul	31	678.71	21.9	1.49E+05	1.73E-02	7.47	6.79
	Aug	31	679.51	21.9	1.49E+05	1.73E-02	7.47	6.80
	Sep	30	654.08	21.8	1.44E+05	1.67E-02	7.19	6.54
	Oct	31	591.88	19.1	1.30E+05	1.51E-02	6.51	5.92
	Nov	30	699.16	23.3	1.54E+05	1.78E-02	7.69	6.99
	Dec	31	647.70	20.9	1.42E+05	1.65E-02	7.12	6.48
1955	Jan	31	724.21	23.4	1.59E+05	1.85E-02	7.97	7.24
	Feb	28	682.84	24.4	1.50E+05	1.74E-02	7.51	6.83
	Mar	31	743.38	24.0	1.64E+05	1.90E-02	8.18	7.43
	Apr	30	657.70	21.9	1.45E+05	1.68E-02	7.23	6.58
	May	31	690.22	22.3	1.52E+05	1.76E-02	7.59	6.90
	Jun	30	592.02	19.7	1.30E+05	1.51E-02	6.51	5.92
	Jul	31	673.22	21.7	1.48E+05	1.72E-02	7.41	6.73
	Aug	31	675.72	21.8	1.49E+05	1.72E-02	7.43	6.76
	Sep	30	659.37	22.0	1.45E+05	1.68E-02	7.25	6.59
	Oct	31	614.54	19.8	1.35E+05	1.57E-02	6.76	6.15
	Nov	30	568.05	18.9	1.25E+05	1.45E-02	6.25	5.68
	Dec	31	591.41	19.1	1.30E+05	1.51E-02	6.51	5.91
1956	Jan	31	672.04	21.7	1.48E+05	1.71E-02	7.39	6.72
	Feb	29	629.89	21.7	1.39E+05	1.61E-02	6.93	6.30
	Mar	31	612.87	19.8	1.35E+05	1.56E-02	6.74	6.13
	Apr	30	669.39	22.3	1.47E+05	1.71E-02	7.36	6.69
	May	31	540.06	17.4	1.19E+05	1.38E-02	5.94	5.40
	Jun	30	617.06	20.6	1.36E+05	1.57E-02	6.79	6.17
	Jul	31	681.83	22.0	1.50E+05	1.74E-02	7.50	6.82
	Aug	31	633.31	20.4	1.39E+05	1.61E-02	6.97	6.33
	Sep	30	645.56	21.5	1.42E+05	1.65E-02	7.10	6.46
	Oct	31	599.90	19.4	1.32E+05	1.53E-02	6.60	6.00
	Nov	30	634.67	21.2	1.40E+05	1.62E-02	6.98	6.35
	Dec	31	461.56	14.9	1.02E+05	1.18E-02	5.08	4.62

Table IA-1. (cont.)

Year-Month	Days/ month	Power for month (MW-day)	Power for month (MW)	Argon-41 (Ci)	Iodine-131 (Ci)	Particulates (Ci)	Carbon-14 (Ci)
1957 Jan	31	605.40	19.5	1.33E+05	1.54E-02	6.66	6.05
Feb	28	582.36	20.8	1.28E+05	1.49E-02	6.41	5.82
Mar	31	633.57	20.4	1.39E+05	1.62E-02	6.97	6.34
Apr	30	606.39	20.2	1.33E+05	1.55E-02	6.67	6.06
May	31	607.18	19.6	1.34E+05	1.55E-02	6.68	6.07
Jun	30	602.59	20.1	1.33E+05	1.54E-02	6.63	6.03
Jul	31	603.33	19.5	1.33E+05	1.54E-02	6.64	6.03
Aug	31	618.17	19.9	1.36E+05	1.58E-02	6.80	6.18
Sep	30	343.13	11.4	7.55E+04	8.75E-03	3.77	3.43
Oct	31	368.49	11.9	8.11E+04	9.40E-03	4.05	3.68
Nov	30	423.97	14.1	9.33E+04	1.08E-02	4.66	4.24
Dec	31	312.62	10.1	6.88E+04	7.97E-03	3.44	3.13

Table IA-2. Monthly power levels and estimated routine argon-41, iodine-131, and carbon-14 emissions for the BGRR when enriched fuel was used (January 1958 through December 1961).

Year-Month	Days/ month	Power for month (MW-day)	Power for month (MW)	Argon-41 (Ci)	Iodine-131 (Ci)	Particulates (Ci)	Carbon-14 (Ci)
1958 Jan	31	387.08	12.5	3.78E+05	1.97E-01	27.10	17.0
Feb	28	330.63	11.8	3.23E+05	1.69E-01	23.14	14.6
Ma	31	245.02	7.90	2.39E+05	1.25E-01	17.15	10.8
Apr	30	142.69	4.76	1.39E+05	7.28E-02	9.99	6.28
May	31	271.15	8.75	2.65E+05	1.38E-01	18.98	11.9
Jun	30	311.79	10.4	3.05E+05	1.59E-01	21.83	13.7
Jul	31	336.26	10.9	3.29E+05	1.71E-01	23.54	14.8
Aug	31	390.06	12.6	3.81E+05	1.99E-01	27.30	17.2
Sep	30	407.86	13.6	3.98E+05	2.08E-01	28.55	18.0
Oct	31	406.72	13.1	3.97E+05	2.07E-01	28.47	17.9
Nov	30	426.02	14.2	4.16E+05	2.17E-01	29.82	18.7
Dec	31	327.84	10.6	3.20E+05	1.67E-01	22.95	14.4
1959 Jan	31	434.19	14.0	4.24E+05	2.21E-01	30.39	19.1
Feb	28	386.23	13.8	3.77E+05	1.97E-01	27.04	17.0
Mar	31	433.43	14.0	4.23E+05	2.21E-01	30.34	19.1
Apr	30	424.42	14.2	4.15E+05	2.16E-01	29.71	18.7
May	31	391.30	12.6	3.82E+05	2.00E-01	27.39	17.2
Jun	30	425.84	14.2	4.16E+05	2.17E-01	29.81	18.7
Jul	31	493.72	15.9	4.82E+05	2.52E-01	34.56	21.7
Aug	31	494.85	16.0	4.83E+05	2.52E-01	34.64	21.8
Sep	30	449.18	15.0	4.39E+05	2.29E-01	31.44	19.8
Oct	31	480.88	15.5	4.70E+05	2.45E-01	33.66	21.2
Nov	30	487.20	16.2	4.76E+05	2.48E-01	34.10	21.4
Dec	31	348.44	11.2	3.40E+05	1.78E-01	24.39	15.3
1960 Jan	31	476.79	15.4	4.66E+05	2.43E-01	33.38	21.0
Feb	29	497.29	17.2	4.86E+05	2.54E-01	34.81	21.9
Mar	31	421.00	13.6	4.11E+05	2.15E-01	29.47	18.5
Apr	30	382.81	12.8	3.74E+05	1.95E-01	26.80	16.8
May	31	457.02	14.7	4.47E+05	2.33E-01	31.99	20.1
Jun	30	462.45	15.4	4.52E+05	2.36E-01	32.37	20.4
Jul	31	450.82	14.5	4.40E+05	2.30E-01	31.56	19.8
Aug	31	494.67	16.0	4.83E+05	2.52E-01	34.63	21.8
Sep	30	428.14	14.3	4.18E+05	2.18E-01	29.97	18.8
Oct	31	454.55	14.7	4.44E+05	2.32E-01	31.82	20.0
Nov	30	477.37	15.9	4.66E+05	2.43E-01	33.42	21.0
Dec	31	359.48	11.6	3.51E+05	1.83E-01	25.16	15.8

Table A-2. (cont.)

Year-Month	Days/ month	Power for month (MW-day)	Power for Month (MW)	Argon-41 (Ci)	Iodine-131 (Ci)	Particulates (Ci)	Carbon-14 (Ci)
1961 Jan	31	414.71	13.4	4.05E+05	2.12E-01	29.0	23.2
Feb	28	376.43	13.4	3.68E+05	1.92E-01	26.4	22.3
Mar	31	559.23	18.0	5.46E+05	2.85E-01	39.2	22.2
Apr	30	442.70	14.8	4.33E+05	2.26E-01	31.0	24.5
May	31	546.60	17.6	5.34E+05	2.79E-01	38.3	22.9
Jun	30	507.18	16.9	4.96E+05	2.59E-01	35.5	20.8
Jul	31	545.19	17.6	5.33E+05	2.78E-01	38.2	20.1
Aug	31	467.28	15.1	4.57E+05	2.38E-01	32.7	23.0
Sep	30	418.57	14.0	4.09E+05	2.13E-01	29.3	17.0
Oct	31	465.48	15.0	4.55E+05	2.37E-01	32.6	20.4
Nov	30	541.93	18.1	5.29E+05	2.76E-01	37.9	19.9
Dec	31	417.07	13.5	4.07E+05	2.13E-01	29.2	17.0

Table IA-3. Power history, argon-41, and carbon-14 released from the BMRR.

Month		MW-hours	Argon-41 (Ci)	Carbon-14 (Ci)
1959	Apr	5.7	12.0	2.0×10^{-4}
	May	10.8	22.7	3.9×10^{-4}
	Jun	16.2	34.2	5.8×10^{-4}
	Jul	27.8	58.6	1.0×10^{-3}
	Aug	29.6	62.4	1.1×10^{-3}
	Sep	7.8	16.5	2.8×10^{-4}
	Oct	8.2	17.3	2.9×10^{-4}
	Nov	8.7	18.3	3.1×10^{-4}
	Dec	17.5	37.0	6.3×10^{-4}
Annual Total			279	4.7×10^{-3}
1960	Jan	3.1	6.5	1.1×10^{-4}
	Feb	13.0	27.4	4.7×10^{-4}
	Mar	19.9	42.0	7.1×10^{-4}
	Apr	13.3	28.1	4.8×10^{-4}
	May	32.2	67.9	1.2×10^{-3}
	Jun	15.1	31.9	5.4×10^{-4}
	Jul	55.5	117	2.0×10^{-3}
	Aug	152	321	5.5×10^{-3}
	Sep	61.6	130	2.2×10^{-3}
	Oct	55.4	117	2.0×10^{-3}
	Nov	70.7	149	2.5×10^{-3}
	Dec	16. 8	35	6.0×10^{-4}
Annual Total			1073	1.8×10^{-2}
1961	Jan	31.5	66.4	1.1×10^{-3}
	Feb	28.4	59.8	1.0×10^{-3}
	Mar	26.8	56.6	9.6×10^{-4}
	Apr	47.0	99.1	1.7×10^{-3}
	May	39.2	82.8	1.4×10^{-3}
	Jun	30.2	63.8	1.1×10^{-3}
	Jul	32.7	69.1	1.2×10^{-3}
	Aug	54.6	115	2.0×10^{-3}
	Sep	11.6	24.5	4.2×10^{-4}
	Oct	19.4	40.9	7.0×10^{-4}
	Nov	13.6	28.7	4.9×10^{-4}
	Dec	13.4	28.3	4.8×10^{-4}
Annual Total			735	1.2×10^{-2}

Table IA-4. Derivation of argon-41 to power ratio for the BMRR, emissions data for 1997.

Month	MW-hour	Argon-41 (Ci)
Jan	125.81	224
Feb	92.92	172
Mar	15.02	28
Apr	166.35	278
May	58.42	109
Jun	116.15	121
Jul	81.32	120
Aug	65.66	275
Sep	65.83	166
Oct	95.25	320
Nov	103.74	189
Dec	61.78	217
Average Annual	87.44	185
	Ratio (Ci/MW-hour)	2.11

Table IA-5. Derivation of relationship between argon-41 release from the BGRR and average external exposures at boundary and on-site area monitoring stations.

Year	Argon-41 (Ci)	On-site Stations		Boundary Stations	
		Average (mR/yr)	Maximum (mR/year)	Average (mR/year)	Maximum (mR/year)
1959	5.1×10^6	NA	NA	24	46
1960	5.2×10^6	136*	222*	25	39
1961	5.6×10^6	121	172	30	37
1962	5.6×10^6	122	161	34	56
1963	5.6×10^6	118	170	20	44
1964	5.3×10^6	134	167	26	51
1965	4.8×10^6	113	152	24	46
Average	5.2×10^6	124	174	26	46
Average mR/year per Million Curies of Argon-41:		23	33	4.9	8.7

* July through December only

Table IA-6. BNL related radiation levels at area monitoring stations in 1959.

		Boundary Stations (mr/week)			Central Area Stations (mr/week)		
Station	**P-2 N. W. Perimeter**	**P-4 S.W. Perimeter**	**P-7 S.E. Perimeter**	**P-9 N.E. Perimeter**	**P-10**	**P-11**	**P-12**
Month							
January	0.12	0.03	0.54	0.48	NA	NA	NA
February	0.32	0.24	0.66	0.68	NA	NA	NA
March	0.54	0.33	0.22	0.44	NA	NA	NA
April	0.69	0.58	0.22	0.98	NA	NA	NA
May	0.59	0.07	0.22	1.51	NA	NA	NA
June	0.48	0.09	0.70	0.64	NA	NA	NA
July	0.66	0.24	0.15	1.83	NA	NA	NA
August	0.61	0.10	0.20	1.74	NA	NA	NA
September	0.65	0.12	0.16	0.76	NA	NA	NA
October	0.29	0.52	0.44	0.80	NA	NA	NA
November	0.34	0.12	0.44	0.52	NA	NA	NA
December	0.21	0.21	0.72	0.32	NA	NA	NA
Total mr/year	23.9	11.4	20.3	46.3	NA	NA	NA

NA: not available

Table IA-7. BNL related radiation levels at area monitoring stations in 1960.

	Boundary Stations (mr/week)				Central Area Stations (mr/week)		
Station	**P-2 N. W. Perimeter**	**P-4 S.W. Perimeter**	**P-7 S.E. Perimeter**	**P-9 N.E. Perimeter**	**P-10**	**P-11**	**P-12**
Month							
January	0.13	0.26	0.65	0.27	NA	NA	NA
February	0.35	0.60	0.56	0.58	NA	NA	NA
March	0.19	0.23	0.86	0.14	NA	NA	NA
April	0.35	0.20	0.37	0.98	NA	NA	NA
May	1.17	0.12	0.49	0.64	NA	NA	NA
June	0.57	0.20	0.36	0.65	NA	NA	NA
July	0.20	0.17	0.30	1.52	0.80	2.34	6.29
August	0.31	0.52	0.32	1.17	1.58	2.44	5.05
September	0.72	0.67	0.23	0.70	2.47	2.27	3.70
October	0.29	0.36	0.51	0.97	1.04	3.15	3.151
November	0.37	0.35	0.35	0.79	1.14	1.97	4.47
December	0.12	0.18	0.73	0.41	0.66	1.53	2.94
Total mr/year	20.8	16.6	25.0	38.5	7.69	13.7	25.60

NA: not available

Table IA-8. BNL related radiation levels at area monitoring stations in 1961.

	Boundary Stations (mr/week)				Central Area Stations (mr/week)		
Month	**P-2 N. W. Perimeter**	**P-4 S.W. Perimeter**	**P-7 S.E. Perimeter**	**P-9 N.E. Perimeter**	**P-10**	**P-11**	**P-12**
January	0.05	0.15	0.73	0.28	0.30	2.31	1.79
February	0.28	0.41	0.58	0.35	1.59	2.64	2.02
March	0.51	0.67	1.29	0.53	2.46	2.89	1.91
April	0.79	0.81	0.39	0.49	1.75	1.89	0.91
May	1.04	0.30	0.64	0.98	1.46	2.66	3.63
June	0.30	0.27	0.41	1.81	0.62	2.06	6.00
July	1.12	0.70	0.15	1.22	2.28	1.83	5.06
August	0.52	0.53	0.31	0.86	1.46	1.69	5.63
September	0.41	1.20	0.36	0.74	1.51	1.98	4.06
October	0.40	0.47	0.58	0.27	1.66	2.23	2.93
November	0.41	0.20	0.89	0.54	1.11	2.46	2.76
December*	0.33	0.39	0.72	0.54*	1.55	2.67	1.95
Total mr/year	26.5	26.5	30.7	37.4	7.75	16.31	26.65

* includes radiation levels associated with the ecology forest

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ATTACHMENT I-B

Liquid Emissions
Detailed Data Tables

Table IB-1. 1951 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	ND	ND	ND	ND	ND	ND	ND
February	ND	200	ND	20	ND	100	ND
March	0.03	300	8.1	30	0.8	150	4.1
April	0.02	250	5.8	25	0.6	125	2.9
May	0.03	2200	65.6	220	6.6	1100	32.8
June	0.04	250	8.8	25	0.9	125	4.4
July	0.04	80	3.3	8	0.3	40	1.7
August	0.04	240	10.3	24	1.0	120	5.1
September	0.03	1600	53.6	160	5.4	800	26.8
October	0.02	220	5.0	22	0.5	110	2.5
November	0.03	210	5.4	21	0.5	105	2.7
December	0.02	300	7.1	30	0.7	150	3.6
Total	0.31		173.1		17.3		86.5
Weighted Average**		566		57		283	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-2. 1951 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	ND	ND	ND	ND	ND	ND	ND
February	ND	10-50	ND	1-5	ND	5-25	ND
March	ND	113	ND	11	ND	57	ND
April	0.02	25	0.4	3	0.0	13	0.2
May	0.02	180	3.3	18	0.3	90	1.6
June	0.02	86	1.8	9	0.2	43	0.9
July	0.03	25	0.7	3	0.1	13	0.3
August	0.02	130	3.2	13	0.3	65	1.6
September	0.02	190	3.4	19	0.3	95	1.7
October	0.02	120	2.2	12	0.2	60	1.1
November	0.02	90	1.9	9	0.2	45	1.0
December	0.02	90	1.8	9	0.2	45	0.9
Total	0.18		18.5		1.9		9.3
Weighted Average**		102		10		51	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-3. 1951 Boundary Station M. Flow, Gross Beta, Strontium-90 and Cesium-137 amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	ND	ND	ND	ND	ND	ND	ND
February	0.07	10-50	3.5	1-5	0.4	5-25	1.8
March	0.03	113	2.9	11	0.3	57	1.4
April	0.03	13	0.4	1	0.0	7	0.2
May	0.02	134	3.3	13	0.3	67	1.6
June	0.02	84	1.8	8	0.2	42	0.9
July	0.02	25	0.7	3	0.1	13	0.3
August	0.02	130	3.2	13	0.3	65	1.6
September	0.01	190	3.4	19	0.3	95	1.7
October	0.01	120	2.2	12	0.2	60	1.1
November	0.02	78	1.9	8	0.2	39	1.0
December	0.02	74	1.8	7	0.2	37	0.9
Total	0.28		24.9		2.5		12.4
Weighted Average***		88		9		44	

ND: no data

* Gross beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed based on an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-4. 1952 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

Month	Flow 10^9 l	Gross Beta Conc. pCi/l	Gross Beta Amount mCi	Strontium-90* Conc. pCi/l	Strontium-90* Amount mCi	Cesium-137* Conc. pCi/l	Cesium-137* Amount mCi
January	0.02	150	3.7	15	0.4	75	1.8
February	0.02	220	5.1	22	0.5	110	2.5
March	0.02	210	5.1	21	0.5	105	2.5
April	0.03	700	17.7	70	1.8	350	8.9
May	0.03	340	11.0	34	1.1	170	5.5
June	0.03	740	23.8	74	2.4	370	11.9
July	0.04	170	6.3	17	0.6	85	3.2
August	0.04	510	19.6	51	2.0	255	9.8
September	0.03	460	14.9	46	1.5	230	7.4
October	0.03	240	6.9	24	0.7	120	3.4
November	0.03	68	1.9	7	0.2	34	1.0
December	0.03	77	2.2	8	0.2	39	1.1
Total	0.36		118.2		11.8		59.1
Weighted Average**		333		33		166	

ND: no data

* Strontium-90 and Cesium-137 reconstructedfrom an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-5. 1952 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

Month	Flow 10^9 l	Gross Beta Conc. pCi/l	Gross Beta Amount mCi	Strontium-90* Conc. pCi/l	Strontium-90* Amount mCi	Cesium-137* Conc. pCi/l	Cesium-137* Amount mCi
January	0.02	50	1.1	5	0.1	25	0.5
February	0.02	70	1.5	7	0.1	35	0.7
March	0.02	80	1.8	8	0.2	40	0.9
April	0.02	140	2.7	14	0.3	70	1.3
May	0.03	100	2.9	10	0.3	50	1.4
June	0.03	150	4.3	15	0.4	75	2.2
July	0.03	60	1.8	6	0.2	30	0.9
August	0.03	190	6.1	19	0.6	95	3.1
September	0.03	110	3.0	11	0.3	55	1.5
October	0.02	80	1.3	8	0.1	40	0.7
November	0.02	36	0.6	4	0.1	18	0.3
December	0.02	45	0.7	5	0.1	23	0.3
Total	0.28		27.8		2.8		13.9
Weighted Average**		99		10		50	

ND: no data

* Strontium-90 and Cesium-137 reconstructedfrom an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-6. 1952 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

Month	Flow 10^9 l	Gross Beta* Conc. pCi/l	Gross Beta* Amount mCi	Strontium-90** Conc. pCi/l	Strontium-90** Amount mCi	Cesium-137** Conc. pCi/l	Cesium-137** Amount mCi
January	0.03	32	1.1	3	0.1	16	0.5
February	0.03	43	1.5	4	0.1	22	0.7
March	0.06	33	1.8	3	0.2	17	0.9
April	0.06	42	2.7	4	0.3	21	1.3
May	0.13	22	2.9	2	0.3	11	1.4
June	0.14	32	4.3	3	0.4	16	2.2
July	0.05	36	1.8	4	0.2	18	0.9
August	0.04	159	6.1	16	0.6	80	3.1
September	0.02	110	3.0	11	0.3	55	1.5
October	0.01	80	1.3	8	0.1	40	0.7
November	0.02	34	0.6	3	0.1	17	0.3
December	0.02	34	0.7	3	0.1	17	0.3
Total	0.62		27.8		2.8		13.9
Weighted Average***		45		4		22	

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-7. 1953 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.03	640	19.7	64	2.0	320	9.8
February	0.03	540	14.5	54	1.4	270	7.2
March	0.03	770	22.4	77	2.2	385	11.2
April	0.03	380	10.5	38	1.1	190	5.3
May	0.03	250	8.0	25	0.8	125	4.0
June	0.03	190	6.6	19	0.7	95	3.3
July	0.04	74	3.0	7	0.3	37	1.5
August	0.04	160	6.4	16	0.6	80	3.2
September	0.04	230	9.0	23	0.9	115	4.5
October	0.04	420	16.1	42	1.6	210	8.0
November	0.03	130	4.5	13	0.5	65	2.3
December	0.04	120	4.4	12	0.4	60	2.2
Total	0.41		125.1		12.5		62.5
Weighted Average**		305		30		152	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-8. 1953 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.02	480	9.1	48	0.9	240	4.6
February	0.02	350	5.7	35	0.6	175	2.8
March	0.02	220	4.9	22	0.5	110	2.4
April	0.02	180	3.8	18	0.4	90	1.9
May	0.02	60	1.5	6	0.1	30	0.7
June	0.03	70	2.1	7	0.2	35	1.1
July	0.04	32	1.2	3	0.1	16	0.6
August	ND	44	ND	4	ND	22	ND
September	0.04	52	2.0	5	0.2	26	1.0
October	0.03	50	1.6	5	0.2	25	0.8
November	0.03	40	1.2	4	0.1	20	0.6
December	0.03	30	1.0	3	0.1	15	0.5
Total	0.31		34.2		3.4		17.1
Weighted Average**		111		11		55	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table B-9. 1953 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.03	265	9.1	27	0.9	133	4.6
February	0.03	172	5.7	17	0.6	86	2.8
March	0.17	29	4.9	3	0.5	15	2.4
April	0.29	13	3.8	1	0.4	7	1.9
May	0.29	5	1.5	1	0.1	3	0.7
June	0.21	10	2.1	1	0.2	5	1.1
July	0.11	12	1.2	1	0.1	6	0.6
August	0.08	ND	ND	ND	ND	ND	ND
September	0.05	43	2.0	4	0.2	21	1.0
October	0.05	36	1.6	4	0.2	18	0.8
November	0.05	26	1.2	3	0.1	13	0.6
December	0.09	11	1.0	1	0.1	6	0.5
Total	1.36		34.2		3.4		17.1
Weighted Average***		25		3		13	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** "Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-10. 1954 Imhoff Tank Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.03	182	5.9	18	0.6	91	3.0
February	0.03	317	9.3	32	0.9	159	4.7
March	0.03	620	20.1	62	2.0	310	10.0
April	0.03	550	18.4	55	1.8	275	9.2
May	0.03	270	9.0	27	0.9	135	4.5
June	0.04	342	12.5	34	1.3	171	6.3
July	0.05	370	16.9	37	1.7	185	8.4
August	0.04	500	22.3	50	2.2	250	11.1
September	0.04	390	13.8	39	1.4	195	6.9
October	0.04	414	15.0	41	1.5	207	7.5
November	0.03	790	24.8	79	2.5	395	12.4
December	0.00	430	0.0	43	0.0	215	0.0
Total	0.39		167.9		16.8		84.0
Weighted Average**		430		43		215	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-11. 1954 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.03	64	1.6	6	0.2	32	0.8
February	0.02	73	1.7	7	0.2	37	0.9
March	0.03	95	2.5	10	0.3	48	1.3
April	0.03	160	4.8	16	0.5	80	2.4
May	0.04	50	1.8	5	0.2	25	0.9
June	0.03	72	2.2	7	0.2	36	1.1
July	0.03	130	4.5	13	0.4	65	2.2
August	0.05	140	6.3	14	0.6	70	3.2
September	0.04	70	3.0	7	0.3	35	1.5
October	0.04	64	2.4	6	0.2	32	1.2
November	0.03	190	5.0	19	0.5	95	2.5
December	0.00	80	0.0	8	0.0	40	0.0
Total	0.36		35.9		3.6		18.0
Weighted Average**		100		10		50	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-12. 1954 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.07	24	1.6	2	0.2	12	0.8
February	0.04	45	1.7	4	0.2	22	0.9
March	0.04	66	2.5	7	0.3	33	1.3
April	0.04	111	4.8	11	0.5	55	2.4
May	0.06	29	1.8	3	0.2	15	0.9
June	0.04	52	2.2	5	0.2	26	1.1
July	0.03	130	4.5	13	0.4	65	2.2
August	0.05	122	6.3	12	0.6	61	3.2
September	0.09	34	3.0	3	0.3	17	1.5
October	0.10	25	2.4	2	0.2	12	1.2
November	0.08	62	5.0	6	0.5	31	2.5
December	0.12	0	0.0	0	0.0	0	0.0
Total	0.76		35.9		3.6		18.0
Weighted Average***		47		5		24	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-13. 1955 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	290	10.2	29	1.0	145	5.1
February	0.03	520	15.8	52	1.6	260	7.9
March	0.03	330	11.5	33	1.2	165	5.8
April	0.04	840	33.0	84	3.3	420	16.5
May	0.04	820	34.7	82	3.5	410	17.4
June	0.04	500	21.9	50	2.2	250	10.9
July	0.05	250	13.3	25	1.3	125	6.7
August	0.05	410	22.3	41	2.2	205	11.1
September	0.04	250	10.6	25	1.1	125	5.3
October	0.04	480	18.5	48	1.8	240	9.2
November	0.03	370	11.8	37	1.2	185	5.9
December	0.03	410	12.9	41	1.3	205	6.5
Total	0.48		216.5		21.7		108.3
Weighted Average**		453		45		227	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-14. 1955 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	90	3.6	9	0.4	45	1.8
February	0.03	150	4.7	15	0.5	75	2.4
March	0.04	110	4.0	11	0.4	55	2.0
April	0.04	230	9.4	23	0.9	115	4.7
May	0.04	310	10.9	31	1.1	155	5.5
June	0.04	260	9.9	26	1.0	130	5.0
July	0.04	200	8.3	20	0.8	100	4.2
August	0.05	190	8.9	19	0.9	95	4.5
September	0.04	140	5.4	14	0.5	70	2.7
October	0.03	140	4.6	14	0.5	70	2.3
November	0.03	110	3.2	11	0.3	55	1.6
December	0.03	90	2.7	9	0.3	45	1.4
Total	0.44		75.7		7.6		37.9
Weighted Average**		171		17		86	

ND: no data

* Strontium-90 and Cesium-137 reconstructedfrom an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-15. 1955 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.21	17	3.6	2	0.4	9	1.8
February	0.15	32	4.7	3	0.5	16	2.4
March	0.14	28	4.0	3	0.4	14	2.0
April	0.14	68	9.4	7	0.9	34	4.7
May	0.12	89	10.9	9	1.1	45	5.5
June	0.08	126	9.9	13	1.0	63	5.0
July	0.04	200	8.3	20	0.8	100	4.2
August	0.06	157	8.9	16	0.9	79	4.5
September	0.05	109	5.4	11	0.5	54	2.7
October	0.07	69	4.6	7	0.5	34	2.3
November	0.21	15	3.2	2	0.3	8	1.6
December	0.20	14	2.7	1	0.3	7	1.4
Total	1.47		75.7		7.6		37.9
Weighted Average***		52		5		26	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-16. 1956 Imhoff Tank. Flow, Gross Beta, Strontium-9,0 and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.03	450	14.8	45	1.5	225	7.4
February	0.03	320	10.5	32	1.0	160	5.2
March	0.04	450	16.1	45	1.6	225	8.0
April	0.03	370	11.9	37	1.2	185	6.0
May	0.04	300	11.5	30	1.2	150	5.8
June	0.04	380	16.7	38	1.7	190	8.4
July	0.05	310	16.6	31	1.7	155	8.3
August	0.07	370	24.8	37	2.5	185	12.4
September	0.06	210	11.7	21	1.2	105	5.8
October	0.04	200	8.9	20	0.9	100	4.4
November	0.04	220	8.9	22	0.9	110	4.4
December	0.04	330	13.7	33	1.4	165	6.8
Total	0.52		166.1		16.6		83.0
Weighted Average**		320		32		160	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-17. 1956 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.03	200	6.1	20	0.6	100	3.0
February	0.03	90	2.9	9	0.3	45	1.4
March	0.04	70	2.5	7	0.3	35	1.3
April	0.03	80	2.8	8	0.3	40	1.4
May	0.04	90	3.3	9	0.3	45	1.6
June	0.04	150	5.3	15	0.5	75	2.6
July	0.04	190	7.4	19	0.7	95	3.7
August	0.05	180	9.0	18	0.9	90	4.5
September	0.05	90	4.2	9	0.4	45	2.1
October	0.04	70	2.7	7	0.3	35	1.3
November	0.03	70	2.4	7	0.2	35	1.2
December	0.04	120	4.4	12	0.4	60	2.2
Total	0.45		52.9		5.3		26.4
Weighted Average**		118		12		59	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-18. 1956 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.15	40	6.1	4	0.6	20	3.0
February	0.20	14	2.9	1	0.3	7	1.4
March	0.27	9	2.5	1	0.3	5	1.3
April	0.31	9	2.8	1	0.3	4	1.4
May	0.30	11	3.3	1	0.3	5	1.6
June	0.24	22	5.3	2	0.5	11	2.6
July	0.17	42	7.4	4	0.7	21	3.7
August	0.15	61	9.0	6	0.9	30	4.5
September	0.09	47	4.2	5	0.4	23	2.1
October	0.06	42	2.7	4	0.3	21	1.3
November	0.06	44	2.4	4	0.2	22	1.2
December	0.06	72	4.4	7	0.4	36	2.2
Total	2.07		52.9		5.3		26.4
Weighted Average***		26		3		13	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-19. 1957 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	400	14.2	40	1.4	200	7.1
February	0.04	310	11.6	31	1.2	155	5.8
March	0.04	910	37.1	91	3.7	455	18.6
April	0.04	690	30.2	69	3.0	345	15.1
May	0.05	440	21.8	44	2.2	220	10.9
June	0.05	300	15.4	30	1.5	150	7.7
July	0.06	290	18.2	29	1.8	145	9.1
August	0.07	310	20.3	31	2.0	155	10.1
September	0.06	300	18.5	30	1.9	150	9.3
October	0.05	280	15.3	28	1.5	140	7.7
November	0.05	240	12.1	24	1.2	120	6.0
December	0.05	180	8.3	18	0.8	90	4.2
Total	0.60		223.0		22.3		111.5
Weighted Average**		372		37		186	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-20. 1957 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.03	140	4.4	14	0.4	70	2.2
February	0.03	90	3.0	9	0.3	45	1.5
March	0.04	290	10.4	29	1.0	145	5.2
April	0.04	380	14.7	38	1.5	190	7.3
May	0.04	150	6.0	15	0.6	75	3.0
June	0.03	180	6.3	18	0.6	90	3.1
July	0.04	120	5.0	12	0.5	60	2.5
August	0.05	140	6.9	14	0.7	70	3.4
September	0.04	170	7.1	17	0.7	85	3.5
October	0.04	130	5.2	13	0.5	65	2.6
November	0.04	120	5.0	12	0.5	60	2.5
December	0.04	70	2.9	7	0.3	35	1.4
Total	0.47		76.7		7.7		38.4
Weighted Average**		164		16		82	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-21. 1957 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.06	71	4.4	7	0.4	36	2.2
February	0.07	44	3.0	4	0.3	22	1.5
March	0.07	146	10.4	15	1.0	73	5.2
April	0.11	139	14.7	14	1.5	70	7.3
May	0.11	56	6.0	6	0.6	28	3.0
June	0.07	86	6.3	9	0.6	43	3.1
July	0.05	97	5.0	10	0.5	48	2.5
August	0.07	104	6.9	10	0.7	52	3.4
September	0.05	130	7.1	13	0.7	65	3.5
October	0.05	104	5.2	10	0.5	52	2.6
November	0.05	97	5.0	10	0.5	48	2.5
December	0.06	48	2.9	5	0.3	24	1.4
Total	0.82		76.7		7.7		38.4
Weighted Average***		94		9		47	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-22. 1958 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.04	1020	45.1	102	4.5	510	22.5
February	0.04	550	20.5	55	2.1	275	10.3
March	0.04	540	23.3	54	2.3	270	11.7
April	0.04	570	25.5	57	2.5	285	12.7
May	0.05	530	25.9	53	2.6	265	12.9
June	0.05	670	35.0	67	3.5	335	17.5
July	0.06	320	19.3	32	1.9	160	9.6
August	0.07	650	43.7	65	4.4	325	21.8
September	0.06	320	17.8	32	1.8	160	8.9
October	0.05	510	28.0	51	2.8	255	14.0
November	0.05	360	19.4	36	1.9	180	9.7
December	0.05	440	22.3	44	2.2	220	11.2
Total	0.61		325.7		32.6		162.8
Weighted Average**		531		53		266	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-23. 1958 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90		Cesium-137	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.04	460	18.8	46	1.9	230	9.4
February	0.04	190	7.0	19	0.7	95	3.5
March	0.04	160	6.8	16	0.7	80	3.4
April	0.05	180	8.2	18	0.8	90	4.1
May	0.05	140	6.4	14	0.6	70	3.2
June	0.04	240	10.3	24	1.0	120	5.1
July	0.05	180	8.4	18	0.8	90	4.2
August	0.05	160	7.8	16	0.8	80	3.9
September	0.04	170	6.5	17	0.7	85	3.3
October	0.04	230	9.8	23	1.0	115	4.9
November	0.05	200	10.5	20	1.1	100	5.3
December	0.05	160	7.2	16	0.7	80	3.6
Total	0.53		107.7		10.8		53.9
Weighted Average		204		20		102	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-24. 1958 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
Month	Flow 10^9 l	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi	Conc. pCi/l	Amount mCi
January	0.07	277	18.8	28	1.9	139	9.4
February	0.11	64	7.0	6	0.7	32	3.5
March	0.31	22	6.8	2	0.7	11	3.4
April	0.35	23	8.2	2	0.8	12	4.1
May	0.45	14	6.4	1	0.6	7	3.2
June	0.40	26	10.3	3	1.0	13	5.1
July	0.32	27	8.4	3	0.8	13	4.2
August	0.24	32	7.8	3	0.8	16	3.9
September	0.18	37	6.5	4	0.7	18	3.3
October	0.20	50	9.8	5	1.0	25	4.9
November	0.27	39	10.5	4	1.1	19	5.3
December	0.28	26	7.2	3	0.7	13	3.6
Total	3.17		107.7		10.8		53.9
Weighted Average***		34		3		17	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed based on an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-25. 1959 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.05	230	11.0	23	1.1	115	5.5
February	0.05	350	17.0	35	1.7	175	8.5
March	0.06	340	19.8	34	2.0	170	9.9
April	0.06	320	17.8	32	1.8	160	8.9
May	0.06	850	53.5	85	5.3	425	26.7
June	0.07	2190	143.2	219	14.3	1095	71.6
July	0.08	1020	77.4	102	7.7	510	38.7
August	0.08	420	35.0	42	3.5	210	17.5
September	0.07	380	27.7	38	2.8	190	13.9
October	0.07	650	44.3	65	4.4	325	22.1
November	0.06	1330	81.3	133	8.1	665	40.6
December	0.06	1040	59.1	104	5.9	520	29.5
Total	0.76		586.9		58.7		293.5
Weighted Average**		776		78		388	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-26. 1959 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	180	7.2	18	0.7	90	3.6
February	0.04	200	8.1	20	0.8	100	4.1
March	0.05	190	8.8	19	0.9	95	4.4
April	0.05	170	8.6	17	0.9	85	4.3
May	0.05	320	15.2	32	1.5	160	7.6
June	0.05	320	15.0	32	1.5	160	7.5
July	0.05	444	23.1	44	2.3	222	11.6
August	0.07	240	18.0	24	1.8	120	9.0
September	0.05	330	15.8	33	1.6	165	7.9
October	0.05	250	11.9	25	1.2	125	5.9
November	0.05	280	15.3	28	1.5	140	7.6
December	0.05	200	10.4	20	1.0	100	5.2
Total	0.60		157.4		15.7		78.7
Weighted Average**		262		26		131	

ND: no data

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-27. 1959 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.24	30	7.2	3	0.7	15	3.6
February	0.19	43	8.1	4	0.8	22	4.1
March	0.25	36	8.8	4	0.9	18	4.4
April	0.32	26	8.6	3	0.9	13	4.3
May	0.32	48	15.2	5	1.5	24	7.6
June	0.26	58	15.0	6	1.5	29	7.5
July	0.27	86	23.1	9	2.3	43	11.6
August	0.23	77	18.0	8	1.8	38	9.0
September	0.15	104	15.8	10	1.6	52	7.9
October	0.11	104	11.9	10	1.2	52	5.9
November	0.03	280	15.3	28	1.5	140	7.6
December	0.05	200	10.4	20	1.0	100	5.2
Total	2.41		157.4		15.7		78.7
Weighted Average***		65		7		33	

ND: no data

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-28. 1960 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.05	400	19.8	40	2.0	200	9.9
February	0.04	618	19.8	62	2.0	309	9.9
March	0.04	560	23.0	56	2.3	280	11.5
April	0.05	960	43.9	96	4.4	480	22.0
May	0.05	561	19.8	56	2.0	281	9.9
June	0.05	903	19.8	90	2.0	451	9.9
July	0.07	868	19.8	87	2.0	434	9.9
August	0.07	544	19.8	54	2.0	272	9.9
September	0.06	720	45.2	72	4.5	360	22.6
October	0.06	733	19.8	73	2.0	366	9.9
November	0.05	2830	141.4	283	14.1	1415	70.7
December	0.05	520	23.9	52	2.4	260	12.0
Total	0.64		416.1		41.6		208.1
Weighted Average**		649		65		325	

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-29. 1960 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.05	230	11.3	23	1.1	115	5.7
February	0.07	265	18.3	26	1.8	132	9.2
March	0.08	220	17.2	22	1.7	110	8.6
April	0.04	230	9.8	23	1.0	115	4.9
May	0.04	292	11.0	29	1.1	146	5.5
June	0.04	317	12.0	32	1.2	159	6.0
July	0.04	449	19.9	45	2.0	224	10.0
August	0.05	264	12.0	26	1.2	132	6.0
September	0.04	350	13.5	35	1.4	175	6.8
October	0.04	252	9.3	25	0.9	126	4.7
November	0.04	570	24.7	57	2.5	285	12.4
December	0.04	370	15.6	37	1.6	185	7.8
Total	0.57		174.7		17.5		87.3
Weighted Average**		309		31		154	

* Strontium-90 and Cesium-137 reconstructedfrom an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-30. 1960 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.05	230	11.3	23	1.1	115	5.7
February	0.05	265	18.3	26	1.8	132	9.2
March	0.08	220	17.2	22	1.7	110	8.6
April	0.10	95	9.8	10	1.0	48	4.9
May	0.10	115	11.0	12	1.1	58	5.5
June	0.08	145	12.0	14	1.2	72	6.0
July	0.05	369	19.9	37	2.0	184	10.0
August	0.04	264	12.0	26	1.2	132	6.0
September	0.03	350	13.5	35	1.4	175	6.8
October	0.04	250	9.3	25	0.9	125	4.7
November	0.05	493	24.7	49	2.5	246	12.4
December	0.02	370	15.6	37	1.6	185	7.8
Total	0.68		174.7		17.5		87.3
Weighted Average***		255		26		128	

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

Table B-31. 1961 Imhoff Tank. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	853	37.0	85	3.7	427	18.5
February	0.05	403	18.5	40	1.9	202	9.3
March	0.05	340	17.9	34	1.8	170	9.0
April	0.06	399	24.1	40	2.4	200	12.0
May	0.07	405	26.4	41	2.6	203	13.2
June	0.07	780	51.8	78	5.2	390	25.9
July	0.07	1730	128.7	173	12.9	865	64.4
August	0.09	330	28.2	33	2.8	165	14.1
September	0.09	160	13.8	16	1.4	80	6.9
October	0.07	229	16.5	23	1.7	115	8.3
November	0.06	120	6.9	12	0.7	60	3.4
December	0.06	209	11.5	21	1.2	105	5.8
Total	0.76		325.7		32.6		162.9
Weighted Average**		426		43		213	

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-32. 1961 Chlorinating Plant. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta		Strontium-90*		Cesium-137*	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.04	440	17.5	44	1.8	220	8.8
February	0.04	470	18.7	47	1.9	235	9.4
March	0.06	350	19.3	35	1.9	175	9.7
April	0.05	376	19.0	38	1.9	188	9.5
May	0.05	428	22.8	43	2.3	214	11.4
June	0.06	320	18.4	32	1.8	160	9.2
July	0.06	430	26.0	43	2.6	215	13.0
August	0.07	350	23.0	35	2.3	175	11.5
September	0.08	260	22.1	26	2.2	130	11.0
October	0.06	158	9.9	16	1.0	79	4.9
November	0.06	150	8.8	15	0.9	75	4.4
December	0.05	204	10.2	20	1.0	102	5.1
Total	0.60		179.3		17.9		89.7
Weighted Average		300		30		150	

* Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

** Weighted average = total amount (mCi)/total flow (10^9l).

Table IB-33. 1961 Boundary Station M. Flow, Gross Beta, Strontium-90, and Cesium-137; amounts and concentrations.

		Gross Beta*		Strontium-90**		Cesium-137**	
	Flow	Conc.	Amount	Conc.	Amount	Conc.	Amount
Month	10^9 l	pCi/l	mCi	pCi/l	mCi	pCi/l	mCi
January	0.02	440	17.5	44	1.8	220	8.8
February	0.02	470	18.7	47	1.9	235	9.4
March	0.12	156	19.3	16	1.9	78	9.7
April	0.15	130	19.0	13	1.9	65	9.5
May	0.18	129	22.8	13	2.3	65	11.4
June	0.19	98	18.4	10	1.8	49	9.2
July	0.11	233	26.0	23	2.6	117	13.0
August	0.13	171	23.0	17	2.3	86	11.5
September	0.10	212	22.1	21	2.2	106	11.0
October	0.10	104	9.9	10	1.0	52	4.9
November	0.08	107	8.8	11	0.9	54	4.4
December	0.07	149	10.2	15	1.0	74	5.1
Total	1.27		215.6		21.6		107.8
Weighted Average***		170		17		85	

* Gross Beta concentration calculated from total amount (mCi) at Chlorinating Plant, divided by the flow at the boundary or the Chlorinating Plant, whichever was greater.

** Strontium-90 and Cesium-137 reconstructed from an average of 1966 and 1968 ratio to Gross Beta (Sr-90=10%, Cs-137=50%).

*** Weighted average = total amount (mCi)/total flow (10^9l).

PART II

Estimation of Releases from Ruptures of the BGRR Fuel Cartridges

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PART II - ESTIMATION OF RELEASES FROM RUPTURES OF BGRR FUEL CARTRIDGES

1. Introduction

Prior to 1963, few quantitative measurements were made of emissions of radionuclides in the BGRR stack effluent. As a result, an alternate approach was developed to estimate releases associated with ruptures of the fuel cartridges. This section describes the approach and data that were used to derive these estimates.

The uranium fuel cartridges were finned aluminum-alloy tubes that were in channels in the graphite cube. Each cartridge consisted of thirty-three slugs, and each channel contained two cartridges (one in the north half of the graphite cube and the other in the south half). When a cartridge ruptured, oxygen came in contact with the hot fuel and some of the oxidized uranium-fuel slugs would be released.

Cartridges were designated by their location in the core of the reactor. For example, Cartridge N C-3-3 was in the northern half of the reactor cube, C denotes that it was in the C quadrant (A,B,C,D), and 3-3 means that it was three channels over and three channels down from the center of the cube.

2. Ruptures of the Fuel Cartridges

There were 28 reported ruptures of BGRR fuel during the period covered by this report (Table 1). All ruptures occurred with the natural-uranium fuel used until 1957-58. There was one rupture (#29) of a uranium oxide (U_3O_8) sample that was being irradiated for the iodine-production program. Several cladding failures occurred with the enriched fuel after the period covered by this report; these are described in Foelix and Hull (1963) and other reports.

Each rupture is described in the Monthly Reactor Operations Reports (Reactor Operations, 1950-1961). For many of these events, the Health Physics Monthly Summary also contains descriptive material. Reports from the BGRR Health Physics Group Leader to the Health Physics Division Manager (L. Gemmell) often contained information on such incidents. Detailed data on dates and other pertinent information were found in the Reactor Operations Logbooks and in the BGRR Health Physics Divisions Logbooks.

Of particular importance in the Reactor Operations Logbooks were notations on north- and south-probe readings. These probes were oil-soaked cloths attached to the end of long rods. The probes were in the center of the exit air ducts, and located between the reactor, and the air filters. Some of the radioactive material released in a rupture would be captured by the cloths. When the probes were withdrawn from the ducts, they were placed in a fixed position relative to an installed radiation-survey instrument. These measurements provided a quantitative albeit rough measure of the relative size of the release from each rupture. The probe

measurements are given in Table II-1 and in Attachment II-A. The probe measurement technique was adopted from that used at the Oak Ridge (X-10) reactor. This technique was used because the fixed monitoring instruments at that time were incapable of detecting rupture events. The first successful real-time monitoring systems for quantitative measurement of fission products was installed in 1962 (Foelix and Hull, 1963).

Table II-1. Summary of fuel cartridge ruptures in the BGRR.

RUPTURE NUMBER	CLASSIFICATION	RUPTURE DATE	NUMBER SLUGS REPORTED LOST*	PROBE READING mR/hr	CHANNEL NUMBER	DAYS IN REACTOR BEFORE RUPTURE
1	Generic	1/2/52	2	40	S C-1-0	468
2	Generic	3/11/52	2	--	S A-1-0	534
3	Generic	3/12/52	1	220	S A-1-1	571
4	Generic	5/6/52	1	250	N C-4-0	577
5	Non-Generic	12/2/52	1	--	S D-0-3	837
6	Generic	4/17/53	1 -2	85	S A 3-1	115
7	Generic	4/22/53	0	31	S B-0-1	102
8	Generic	5/10/53	1	140	N B-2-7	1
9	Generic	7/23/53	0	1000	N C-3-3	1073
10	Non-generic	8/24/53	5-10	--	S A-2-4	1103
11	Generic	11/8/53	0	33	S A-1-3	191
12	Generic	11/16/53	0	202	N C-1-1	1189
13	Non-generic	1/17/ 54	2	2000	S B-5-5	1248
14	Generic	1/22/54	2	375	S D-1-1	336
15	Non-generic	2/21/54	3	>2000	N B-4-5	1285
16	Non-generic	4/11/54	0	>2000	N B-0-1	107
17	Generic	6/21/54	5?	120	S B-3-5	1403
18	Non-generic	7/6/54	5	80	S A-5-2	1366
19	Generic	6/19/55	1	144	N D-8-3	1766
20	Generic	10/12/55	2	500	N D-9-1	1881
21	Generic	5/1/56	3	900	S A-1-3	879
22	Non-Generic	5/2/56	5	>2000	S B-2-5	2076
23	Generic	10/19/56	0	625	S A-3-4	472
24	Generic	11/14/56	0	450	S A-1-1	371
25	Non-generic	1/15/57	19	--	S A-3-3	633
26	Generic	3/1/57	1	39	S B-0-5	83
27	Generic	7/23/57	0	100	N D-10-3	2529
28	Generic	10/3/57	0	800	S B-6-5	1086
29**	--	10/28/57	--	>2000	D-08	30

* Reported slugs lost are from a Memo J.E. Phillips to R.W. Powell, October 15, 1959.
** not a fuel element, uranium oxide sample for iodine production.

There are two additional important sources of information. The first is a report from L.P. Phillips to F.P. Cowan (Phillips, 1957) describing detailed measurements of particulate releases for three of the more important ruptures. This report is of considerable help in judging the release fractions of the slug inventories. The second is a report from M.M. Weiss to file (Weiss, 1954) on an environmental measurement related to one of the ruptures.

The severity of each rupture and the quantity of radioactive material released were related to how quickly the rupture was detected and the reactor shut down, and how long the fuel cartridge had been in the reactor. For this report, each rupture was qualitatively classified as either generic or non-generic based on these criteria and on the comments and descriptions given in available reference material. Non-generic ruptures exhibit one or more of the following characteristics:

- severe oxidation
- difficulty in removing the fuel cartridge
- measured radioactivity in the environment related to the rupture.

Generic ruptures are those ruptures for which the documentation gave no evidence of severe oxidation, there was no difficulty in removing the fuel element from the fuel channel, and there were no observations of excess radioactivity in the environment.

3.0 Fission Product Inventories

In 1993, a Dose Reconstruction for emissions from the Oak Ridge Reservation was published (ChemRisk, 1993). In Appendix E of Volume II-Part B of that report are source-term estimates for ruptures of graphite reactor slugs. This Appendix from ChemRisk (1993) is reproduced in Attachment II-B of this report.

Since the BGRR slugs were identical to those used by Oak Ridge, this appendix is particularly helpful in estimating inventories for the BGRR slugs. Table 2 compares the BGRR and the Oak Ridge X-10 reactor, it is taken from the 1948 Report on the Brookhaven Nuclear Reactor (BNL, 1948).

The data in the Oak Ridge Report permit us to evaluate the radionuclide inventory in the BGRR slugs by a simple modification of the Oak Ridge values to account for the higher neutron flux at the BGRR, and for the different times the slugs were in the reactors. The Oak Ridge analysis also is helpful in providing a list of the radionuclides that are important in environmental releases.

Table II-2. Comparison of Oak Ridge and Brookhaven Reactors (from BNL, 1948).

	X-10	Brookhaven
Pile Size	24 feet	25 feet
Lattice Size	8 inches	8 inches
Uranium Diameter	1.1 inches	1.1 inches
Area of Channel	3.05 inches^2	5.6 inches^2
Vol. Aluminum/Vol. Uranium	0.108	0.2
Gap	None	2.75 inches
Metal Temperature	270°C	350°C
Exit Air Temperature	80°C	200°C
Power Output	4000 KW	25,000 KW
Maximum Flux	10^{12} neutrons/cm^2 sec	5×10^{12} neutrons/cm^2 sec
Air Flow	100,000 cfm	270,000 cfm
Pressure Drop in Reactor	36 mm Hg	103 mm Hg
Pumping Power	1,400 KW	5,600 KW

Even though the slugs used in the BGRR and the X-10 reactor were identical, the fuel cartridges were different, and the BGRR cooling-air stream passed through a filter that the X-10 reactor lacked. While the X-10 analysis can be used to support an estimate of the inventory of radionuclides in the BGRR slugs, because of these differences it cannot support an estimate of the release fraction of the radionuclides lost during a fuel cartridge rupture.

Uranium, plutonium, and various fission products were released from ruptured fuel slugs. The Oak Ridge report noted that fission products likely to have been released from the ruptured slugs include barium-140, cerium-144, cesium-137, iodine-129, iodine-131, iodine-133, lanthanum-140, niobium-95, ruthenium-106, ruthenium-103, strontium-89, strontium-90, zirconium-95, and the fission gases krypton-85 and xenon-133.

Using the data and analyses in the Oak Ridge report, together with the modifications given above, an assessment of the upper range for the BNL BGRR ruptures is developed.

The Oak Ridge rupture used as the reference event occurred in February 1947. This fuel element was in the Oak Ridge reactor for 850 days. The fission-product content of the Oak Ridge slugs was estimated (ChemRisk, 1993) using the following equation:

$A_i = (5 \times 10^{11}\ n/cm^2\text{-sec})(577 \times 10^{-24} cm^2)(N)(Yield_i)\ (1-e^{-\lambda\ t_{irr}})(2.703 \times 10^{-11}\ Ci\text{-sec/atom})$

where:

A_i	= activity of radionuclide i in each fuel slug (Ci)
5×10^{11} n/cm^2-sec	= average Oak Ridge graphite reactor thermal neutron flux
577×10^{-24}cm^2	= fission cross-section for uranium-235
N	= number of uranium-235 atoms per slug
$Yield_i$	= fission yield of radionuclide i for uranium-235
λ	= decay constant of radionuclide i (sec^{-1})
t_{irr}	= irradiation time in reactor (sec)

2.703×10^{-11} Ci-sec/atom = conversion from atoms/sec to curies

Table 3 gives the estimated inventories for the Oak Ridge slug. The decay constants are given in units of days for use in calculations presented in the next section.

Table 3. Estimated inventory of Oak Ridge reactor slug for first event, 1 slug, February 1947 (data from ChemRisk, 1993),

Radionuclide	Type	Inventory (Ci)	λ (days^{-1})
Krypton-85	Noble Gas	6.69×10^{-2}	1.76×10^{-4}
Xenon-133	Noble Gas	10.89	1.31×10^{-1}
Iodine-131	Iodine	4.86	8.61×10^{-2}
Iodine-133	Iodine	10.89	8.19×10^{-1}
Iodine-129	Iodine	1.51×10^{-7}	1.11×10^{-10}
Barium-140	Particulate	10.56	5.42×10^{-2}
Cesium-137	Particulate	0.49	6.32×10^{-5}
Cerium-144	Particulate	8.80	2.44×10^{-3}
Lanthanum-140	Particulate	10.56	4.14×10^{-1}
Niobium-95	Particulate	10.72	1.98×10^{-2}
Ruthenium-103	Particulate	4.86	1.75×10^{-2}
Ruthenium-106	Particulate	0.49	1.88×10^{-3}
Strontium-89	Particulate	8.04	1.31×10^{-2}
Strontium-90	Particulate	0.53	6.85×10^{-5}
Zirconium-95	Particulate	10.72	1.05×10^{-2}
Uranium-235	Particulate	1.79×10^{-5}	NA
Uranium-238	Particulate	3.88×10^{-4}	NA
Plutonium	Particulate	4.82×10^{-3}	NA

NA: the inventory of these elements in the slug is not calculated based on their decay constants.

To estimate the inventory of long-lived radionuclides of each of the slugs involved in BGRR ruptures, it is necessary to adjust the Oak Ridge estimates to account for different times each slug was in the reactor and for the higher average flux of the BGRR. A flux ratio of five was derived based on the maximum reported fluxes for the BGRR (5×10^{12} n/cm^2-sec) and for X-10 (1×10^{12} n/cm^2-sec). This relationship is an approximate one.

To estimate the inventory of activation and fission products in BGRR slugs two equations are needed, depending on the half-life of the radionuclides. For the purposes of this report, long-lived radionuclides are those for which the difference in time in the reactor is important for irradiations on the order of 100 days, and short-lived radionuclides are those for which the time in the reactor is unimportant. Uranium and plutonium-inventories will be analyzed separately as described below.

Long-lived Radionuclides

For long-lived radionuclides the relationship is

$$A_{i(BGRR)} = (A_{i(X\text{-}10)}\ \text{Ci}) \times \frac{(1-e^{-\lambda\, t_{BGRR}})}{(1-e^{-\lambda\, t_{X\text{-}10}})} \times \text{flux ratio}$$

where:

$A_{i(BGRR)}$	= activity of radionuclide i in BGRR slug (Ci)
$A_{i(X\text{-}10)}$	= activity of radionuclide i in Oak Ridge X-10 slug (Ci)
t_{BGRR}	= number of days slug was in the BGRR
$t_{X\text{-}10}$	= number of days X-10 slug was in reactor (850 days)
λ	= the decay constant of radionuclide i (days^{-1})
flux ratio	= ratio of the maximum BGRR neutron flux to the maximum X-10 flux (5)

For example, for BGRR Rupture Number 1, the cesium-137 inventory in each slug in the cartridge would be

$$A_{Cs\text{-}137}\ \text{Ci} = 0.49\ \text{Ci} \times \frac{(1-e^{-(6.3 \times 10\text{-}5 \times 468)})}{(1-e^{-(6.3 \times 10\text{-}5 \times 850)})} \times 5 = 1.4\ \text{Ci}$$

where 0.49 Ci is the X-10 reference inventory of cesium-137.

Short-lived Radionuclides

For short-lived radionuclides, such as iodine-131, iodine-133, xenon-133, zirconium-95, and niobium-95, the difference in time in the reactor is irrelevant for irradiations on the order of 100 days. In these cases, the Oak Ridge estimates need only be multiplied by the flux ratio of 5 to account for the difference in the neutron flux.

$A_{i(BGRR)} = (A_{i(X\text{-}10)}\ Ci) \times \text{flux ratio}$

Again using the BGRR Rupture Number 1 cartridge, the iodine-131 inventory in each slug in the cartridge would be

$A_{Iodine\text{-}131}\ Ci = 4.86\ Ci \times 5 = 24\ Ci$

Where 4.86 Ci is the X-10 reference inventory of iodine-131.

Uranium

For uranium-235, uranium-238, and uranium-234 the inventory depends only on the amount of metal in a slug and the natural percentage of uranium-235. Since the BGRR and the Oak Ridge slugs were identical, the amount of uranium-235 and uranium-238 can be taken directly from the reference X-10 slug. These values are 1.79×10^{-5} Ci for uranium-235 and 3.88×10^{-4} Ci for uranium-238. For this analysis, uranium-234 was included, with a value equal to that of uranium-238, i.e., 3.88×10^{-4} Ci, because they are in equilibrium.

Plutonium

The Oak Ridge analysis used a value of 4.82×10^{-3} Ci in each slug, irrespective of time in the reactor. For BGRR slugs an adjustment for the higher flux is required. Using the Oak Ridge approach, the plutonium activity in a BGRR slug would be:

$A_{Pu(BGRR)} = (A_{Pu(X\text{-}10)}\ Ci) \times \text{flux ratio} = 4.82 \times 10^{-3}\ Ci \times 5$

$= 2.4 \times 10^{-2}\ Ci$

$= .4\ g/slug$

The Operations Monthly Report for January 1954 reports that there are 437 grams of plutonium per ton of uranium, or 0.57 g/slug. In the Operations Monthly Report for October 1957, 19.566 grams per element were reported, which is 0.59 g/slug. Shepherd in his historical overview of the Brookhaven Graphite Research Reactor Fuel Operations Accountability and Reporting gives a value of 0.5 g/slug, averaged over 45,000 slugs, and 0.8 g/slug for the highest group average (Shepherd, 1997). Since many of the ruptured slugs had been in the reactor for more than 1000 days (12 of 29 ruptures), the plutonium inventory would be at the high end of the distribution. For this report 1 g/slug is used which provides a reasonable but conservative estimate. In all of these references, the plutonium inventory is given without reference to the specific isotope.

BGRR Slug Inventory

The results of applying this approach to estimating the radionuclide inventory of the BGRR slugs involved in the ruptures are given in Table 4. The inventory for individual radionuclides in each rupture are given in the Tables in Attachment II-A.

4.0 Fraction of Inventory Released

While the radionuclide inventory in each slug can be assessed with a fair degree of confidence, estimating the fraction released when a cartridge ruptured is more technically challenging and decidedly more uncertain.

4.1 Oak Ridge Screening Estimates

The Oak Ridge analysis (ChemRisk, 1993) used the following values to make screening-level estimates of the fraction of radionuclides released from a slug during a rupture:

- 100% of noble gas inventory
- 80% of the radioiodine inventory
- 10% of the particulate inventory

To quote from the Oak Ridge Study (ChemRisk, 1993):

> For the purposes of screening calculations, 10% of particulate fission products actually present in each slug at the time of rupture were assumed to be released when the uranium oxidized based on professional judgement. 100% and 80% were applied to noble gas and iodine inventories respectively.

In the same appendix (ChemRisk, 1993), the same release fractions are used for the radionuclide inventories associated with the radioactive lanthanum separation operations at Oak Ridge. In particular,

> The noble gas release fraction of 100% is based on the nonreactive nature of xenon and krypton. The release fraction for iodine is based on analyses of iodine release fractions at the Hanford plant performed as part of the Hanford dose reconstruction project. The release fraction for particulate radionuclides is based on measured particulate emissions from RaLa processing at the Idaho Chemical Processing Plant during 1957 compared to the estimated radionuclide inventories in the materials testing reactor (MTR) fuel used as the barium source at that plant.

Table II-4. Fuel-cartridge ruptures and estimated radionuclide inventory contained in one slug.

RUPTURE				INVENTORY IN ONE SLUG		
Rupture Number	Rupture Date	Channel Number	Days In Reactor	Particulates (Ci)	Noble Gases (Ci)	Iodines (Ci)
1	1/2/52	S C-1-0	468	320	55	79
2	3/11/52	S A-1-0	534	320	55	79
3	3/12/52	S A-1-1	571	320	55	79
4	5/6/52	N C-4-0	577	320	55	79
5	12/2/52	S D-0-3	837	330	55	79
6	4/17/53	S A 3-1	115	260	55	79
7	4/22/53	S B-0-1	102	250	55	79
8	5/10/53	N B-2-7	1	23	6.7	32
9	7/23/53	N C-3-3	1073	330	55	79
10	8/24/53	S A-2-4	1103	330	55	79
11	11/8/53	S A-1-3	191	290	55	79
12	11/16/53	N C-1-1	1189	330	55	79
13	1/17/ 54	S B-5-5	1248	340	55	79
14	1/22/54	S D-1-1	336	310	55	79
15	2/21/54	N B-4-5	1285	340	55	79
16	4/11/54	N B-0-1	107	250	54	79
17	6/21/54	S B-3-5	1403	340	55	79
18	7/6/54	S A-5-2	1366	340	55	79
19	6/19/55	N D-8-3	1766	340	55	79
20	10/12/55	N D-9-1	1881	340	55	79
21	5/1/56	S A-1-3	879	330	55	79
22	5/2/56	S B-2-5	2076	340	55	79
23	10/19/56	S A-3-4	472	320	55	79
24	11/14/56	S A-1-1	371	310	55	79
25	1/15/57	S A-3-3	633	320	55	79
26	3/1/57	S B-0-5	83	240	55	79
27	7/23/57	N D-10-3	2529	350	55	79
28	10/3/57	S B-6-5	1086	330	55	79
29*	10/28/57	--	--	160	54	77

* not a fuel element; uranium-oxide slug for iodine production.

4.2 Approach to Determining BGRR Release Fractions

Although the X-10 and BGRR fuel slugs were identical, the fuel cartridges were very different. For example, the X-10 fuel slugs were in individual cans, requiring extensive welding. In an attempt to overcome the difficulties with the X-10 ruptures, the BGRR fuel-slugs were encased in 11-foot aluminum tubes with one weld on each end and a helium atmosphere maintained around the fuel slugs. Although the BGRR ruptures did result in the release of radioactivity, the extent of oxidation which led to the releases varied considerably from rupture to rupture.

Table II-1 gives the reported number of slugs lost. This loss refers to the inability to account for the individual slugs when an aluminum cartridge was cut into pieces in preparation for shipment often months after the cartridge was moved to the canal. This means that, for example, when 0 slugs were noted as lost, all 33 slugs in a cartridge were counted in the canal where the cutting was performed. However, when a slug was missing, it is not known how much of the oxidized slug was dissolved in the canal water. When cutting operations took place, the water often became opaque due to the quantity of material in it. For these reasons, it is inappropriate to rely on the reported "lost" slugs in estimating the release to the stack resulting from the ruptures. This belief was confirmed by retired employees who attended a workshop at BNL in July, 1999 to discuss approaches to estimating releases from the BGRR from fuel-cartridge ruptures.

For each ruptured slug, it is reasonable to assume that all the noble gases were released along with a major fraction of the radioiodines. However for particulates, there was a major difference between the X-10 reactor and the BGRR. The BGRR had additional high efficiency air-filters on the exit air.

When installed, the exit air-filters were described as glass-tex cloth with the following characteristics: 95% to 99% at full load; 89% initial for 3-4 micron particles (BNL, 1948). In a partially loaded condition they were reported to be somewhat more efficient (Foelix and Hull, 1963). An experimental study by Billings (cited in Hull et. al, 1968) supports this assumption. McLintock (cited in Hull et. al, 1968) reported that the efficiency of the exit air filters is about 80% for particulates in the 0.3 micron range.

A study of particle sizes found on vegetation associated with rupture number 22 (Health Physics Summary, June 1956) reported:

> 1) The particles (both active and inactive) removed from the leaves ranged from 0.5 to 4.5 microns in diameter.
>
> 2) The greatest percentage of the particles were about one micron in diameter, and less than three percent were greater than two microns in diameter.
>
> 3) The greatest percentage of activity removed from the leaves was associated with particles of 1 to 2 microns in diameter.

These data lead to the assumption made in the report of a further factor of 10 reduction in the release of particulates from the BGRR slugs. We then assume for each generic slug rupture that:

- 100% of noble gases inventory is released from the stack
- 80% of the radioiodine inventory is released from the stack

- 1% of the particulate inventory is released from the stack

There were some studies performed during several rupture events at the BGRR that provide guidance in estimating the fraction of the particulate radionuclides released to the environment. The most reliable estimates of the quantity of particulates released during rupture events was that done by L. Phillips in 1957 (Phillips, 1957; Attachment II-C). He reported releases for ruptures 21, 22, and 25. The following table is taken from that memo.

Table II-5. Ruptures No. 21, 22, and 25.

Rupture No.	Date Ruptured	Number of Megawatt Days	Curies Released Decay Time After Rupture				
			2-1/4 Hours	63 Hours	4-1/2 Days	13-1/2 Days	115 Days
21	5/1/56	18,573	3.6	0.8	0.6	0.32	-
22	5/2/56	37,888	60	13	10	-	2.5
25	1/15/57	12,882	7.2	1.53	1.2	0.63	-

These data help in two ways. First, Rupture 21 was typical of a "generic" rupture. For Rupture 21, there was a probe measurement of 900 mR/hr given in the reactor operations log book.

For this rupture, Phillips determined that 3.6 Ci were released as of 2.25 hours post-release. Since the vast majority of the particulate radionuclides used in the Oak Ridge analyses have long half-lives compared with 2.25 hours, a correction of about a factor of 2 to reflect decay during this period was applied, ie. a release of about 7 Ci is estimated.

The selection of the correction factor of 2 is based primarily on professional judgement. There is some support for this value in the data reported for Rupture 25 where two sets of measurements were made. Phillips estimated 7.2 Curies were in the environment as of 2.5 hours after the release. Cowan reported a measurement of 25 Curies made one half hour after the rupture (Health Physics and Safety Summary, January 1957). The Phillips's analytical technique excluded the noble gases, radioiodines and spurious background radiation, which was not possible with the air sample results reported by Cowan. Multiplying 7.2 by 2 results in an estimate of 14.4 Curies, which is quite close to the 25 curies, given that this value is clearly an overestimate.

Therefore, 7 Curies is the estimated amount of particulate released during Rupture 21. Since the probe data indicated a value of 900 mR/hr, a quantitative estimate of the quantity of radioactive material released for each generic rupture can be calculated whenever a probe reading exists, i.e., 7 Ci/900 mR/hr, or 7.8 mCi/mR/hr.

This value appears more accurate than warranted, but will be used to calculate the releases.

The BGRR inventory given in Table II-4 indicates a particulate inventory of 330 Ci for each slug involved in Rupture 21. Applying the release fraction of 1%, 3.3 Ci per slug can be expected to be the particulate release. Dividing the estimated release of 7 Ci by 3.3 Ci/slug gives an estimate of the number of slugs involved. For Rupture 21 this is three slugs, since partial values for the number of slugs will be increased to a whole number to obtain a "slug-equivalent value". This provides a reasonable, albeit conservative, approach for all generic ruptures.

4.3 Approach to Generic Ruptures

Generic ruptures are those for which the available reference documentation gave no evidence of severe oxidation, no observations of excess radioactivity in the environment or no difficulty in removing the fuel element.

The particulate releases were based on the probe measurements. For generic ruptures the probe reading times 7.8 mCi/mR provides a reasonable estimate of the particulates released. This was used to estimate the number of "slug equivalents" involved in the rupture, based on an assumption of 1% of the particulate inventory being released from the stack . With an estimate of the number of "slug equivalents" (fractional slug equivalents are increased to the next largest whole number) the iodine- and noble-gas-releases can be estimated (100% of the noble gas inventory and 80% of the radioiodine inventory per slug equivalent).

4.4 Approach to Non-Generic Ruptures

For the non-generic ruptures, more emphasis is placed on the general discussion of the events in the various referenced materials. and estimates of radionuclide releases are developed on a case by case basis. These rupture numbers are 5, 10, 13, 15, 16, 18, 22, and 25. Rupture 29 was a rupture of a sample of uranium oxide used in the Hot Lab's Iodine Production Program, not a fuel cartridge. The documentation for the data and descriptions discussed below can be found in Attachment II-A.

Rupture 5

For Rupture 5, there is a notation in the reactor log books that the south probe reading was off-scale. However, the descriptive material available for this event does not reflect such a serious release. For Rupture 6 there was a probe reading of 80 mR/h, 27 minutes before shut down at 2130 hours on 4/17/53 and a probe reading on 4/18/53 at 0840 of 85 mR/h, some 11 hours later. For Rupture 5, the shutdown occurred at 0805 on 12/2/52 and the next morning the south probe was 16 mR/h. Although deriving a quantitative relationship is not possible, it seems likely that Ruptures 5 and 6 are quite similar and an assumption of one slug ruptured still seems to be appropriate for Rupture 5 together with the generic approach.

Ruptures 10, 13, and 15

Rupture 10 was clearly a significant event, and was detected at the environmental monitoring stations. Although there was difficulty in removing the fuel elements due to high radiation levels, there was no discussion of difficulty due to swelling of the cartridge. Neither was there any evidence of burning. Therefore, it seems appropriate that all five lost "slugs" (see Table II-1) were involved in this rupture event and that the generic approach can be used.

For Rupture 13, there was a stack-release measurement by A. Solari of 6 Ci (see M. Weiss's memo of 1954). If this activity is used in the generic approach, we would expect loss of two slugs. However, we have a probe measurement of 2,000 mR/h which suggests that five slugs were involved in the rupture. It was noted that "...conditions surrounding this event were even more severe than usual and was similar to those experienced on August 24, 1953" (Rupture 10) and "...at several times during this operation flames were noted in the vicinity of the open container." Under such adverse conditions, it seems reasonable to assume that more than 10% of the particulates escaped the slugs. Assuming that 2 slugs each released 25% of their particulates, a stack release of 7 Ci and a probe reading of about 2,200 mR would be expected. Therefore, for Rupture 13, this report uses 2 slugs ruptured with the release of 25% of the particulates, 100% of the noble gases, and 80% of the radioiodines from each of these slugs. The stack release would then be 2.5% of the particulates in 2 slugs, 80% of the iodines and 100% of the noble gases.

For Rupture 15, the reference material given in Attachment II-A indicates a similarity with ruptures 10 and 13. The major difference was the difficulty in removing the ruptured cartridge from the fuel channel. F.P. Cowan notes in the February 1954 Health Physics Summary that the stack monitor indicated about 9 Ci was released. However, the probe measurements were in excess of 2,000 mR/h. It seems reasonable to use the 3 slugs listed in Table II-1 as lost as the number of ruptured slugs, and as we did with Rupture 13, assume that 25% of the particulates were released from the slugs. The stack release would then be 2.5% of the particulates in 3 slugs, 80% of the iodines and 100% of the noble gases.

Rupture 16

Rupture 16 is a problematic event. The probe reading was greater than 2,000 mR/h and some increase in activity was seen in the dust monitor at two of the environmental monitoring stations. There was no reported difficulty with removal and no evidence of burning, so the release fraction of particulates is assumed to be 0.1. The most confusing was the notation in Table II-1 that 0 slugs were lost. In order to explain the probe reading of >2000 mR/hr, it seems reasonable to assume that 8 slugs ruptured and the release fraction given for the generic ruptures is most applicable.

Rupture 18

For Rupture 18, it is difficult to assess the stack release because, although the probe read 80 mR/hr, there was a notation in the Reactor Operations Report that "...white hot sparks and a dull red glow were noted several times while the element was being disturbed. Several slugs were probably oxidized badly during this event." Unlike Ruptures 22 and 23, this cartridge was easily removed. It would seem appropriate, therefore, that the 5 slugs listed as lost in Table II-1 should be accepted as ruptured, and that the generic approach be used to estimate release fractions. Therefore, for this event, we assume 5 slugs were ruptured in the event and that 100% of the noble gases, 80% of iodines and 1% of the particulates were released, i.e., about 17 Ci.

Ruptures 22 and 25

For Ruptures 22 and 25, it would seem appropriate to use Phillips analysis since this analysis uses available stack emissions estimates based on measurements.

Rupture 25 has the characteristics of a generic rupture, but there is no probe data. However, Phillips provides an estimate of 7.2 Ci released. Since these values are given for 2.25 hours post release, this must be multiplied by a factor of 2 as discussed in Section 4.2. Therefore, approximately 14.4 Ci of particulates were released in Rupture 25. The generic approach would suggest that 4 slugs were involved in the rupture.

For Rupture 22, the situation is more complicated. In Phillips' analysis a value of 60 Ci at 2.25 hours is given, which must be increased to 120 Ci for the immediate post release. Based on the generic approach that would suggest about 33 slugs were involved. This is problematic since only 5 slugs were missing in the canal (Table II-1). A more reasonable assumption is that the release fraction from the fuel slugs was greater than the assumed 10% based on the Oak Ridge studies since more than the usual force was needed to remove the cartridge. If 75% of the 5 slugs had been oxidized and released from the fuel cartridge, about 130 Ci would have been released from the stack. Therefore, an assumption of 5 slugs lost and the particulate release fraction from the slugs of 75% is used. It is assumed that 10% of these particulates were released through the filter.

Rupture 29

Rupture 29 was not a fuel-slug rupture, but that of a 225 gram sample of uranium oxide, U_3O_8. To compare this with a slug rupture, the uranium content of the sample is required, i.e., there are 209 grams of uranium in 225 grams of U-308. Two hundred nine grams is about 17% of the amount of uranium in a slug, which contains 1,175 grams of uranium. The Reactor Operations Logbook for October 1957 noted "...that all of its approximately 225 grams of U_3O_8. had been lost." If all of the radionuclides are assumed to be released from the sample (17% of the inventory in 1 slug) and a 10% release of particulates through the filter, about 3.1 Ci would have

been released. Using the generic rupture approach based on rupture 21, a probe reading of 400 mR/h would have been expected. Since the Operations Log Book gave a probe reading of greater than 2 R (2,000 mR/h), about 15 Ci of particulates would have been calculated, when only 2.7 Ci were available for release. Since this rupture involved a release of the entire sample, the iodine release would have been greater than that used in the generic approach. Even with this relatively small addition to the probe reading, the overall result is that the generic approach apparently overestimates the release by about a factor of 5, which seems satisfactory for an approach based on such little information and with such large uncertainties.

4.5 Estimate of Rupture Releases

Release Estimates

The approach described above was applied to the 28 fuel-cartridge ruptures and one sample rupture that occurred in the BGRR. Table II-6 gives the estimates for particulates, iodines, and noble gases released during each rupture. Attachment II-A gives release estimates for individual radionuclides.

Table II-6. Summary of the stack release for fuel-cartridge and sample ruptures in the BGRR.

Rupture Number	Date	Derived "slug equivalents" Lost	Particulates (Ci)	Iodines (Ci)	Noble gases (Ci)
1	1/2/52	1	0.3	63	55
2	3/11/52	1	1.7	63	55
3	3/12/52	1	1.7	63	55
4	5/6/52	1	2.0	63	55
5	12/2/52	1	3.3	63	55
6	4/17/53	1	0.7	63	55
7	4/22/53	1	0.2	63	55
8	5/10/53	5	1.1	127	33
9	7/23/53	3	7.7	190	160
10	8/24/53	5	17	320	270
11	11/8/53	1	0.3	63	55
12	11/16/53	1	1.6	63	55
13	1/17/ 54	2	17	130	110
14	1/22/54	1	2.9	63	55
15	2/21/54	3	25	190	160
16	4/11/54	8	20	500	440
17	6/21/54	1	0.9	63	55
18	7/6/54	5	17	320	270
19	6/19/55	1	1.1	63	55
20	10/12/55	2	3.9	130	110
21	5/1/56	3	7.0	190	160
22	5/2/56	5	130	320	280
23	10/19/56	2	4.9	130	110
24	11/14/56	2	3.5	130	110
25	1/15/57	4	13	250	220
26	3/1/57	1	0.3	63	55
27	7/23/57	1	0.8	63	55
28	10/3/57	2	6.2	130	110
29*	10/28/57	0.17	3.0	13	9.1

* not a fuel-element, uranium-oxide slug for iodine production.

4.6 Uncertainties

The intent of this report is to be both reasonable albeit conservative in the approach to estimating releases from fuel cartridge ruptures. An attempt has been made to be as clear as possible in documenting these estimates. Reactor-stack monitoring records and more extensive probe reading records would have greatly improved this effort. But these records were not retained in accordance with AEC retention schedules. For example, we note BNL-82, July 10, 1964, under Records Inventory and Disposition Schedule, Appendix 0230, Appendix C-9, Industrial Plant Item 57 through 61; "... retention period 1-6 years. The filing units described will be disposed of on July 1964". These disposition records refer to much of the information that would have helped to make more definitive estimates.

It is important that the rupture releases given in this report are understood to be highly uncertain. Not only does the data for rupture 29 indicate a potential overestimate by about a factor of 5, but other aspects of the analyses are equally uncertain. For example, an iodine release fraction of 80% would overestimate the release by about a factor of 10 based on data collected after the incident at the air-cooled Windscale reactor in Great Britain. In addition, it is likely that although 80% of the iodines may have been released from the fuel slugs, a large fraction of this material would have adhered to the surface of the extensive cooling air duct work and the stack. In addition, the probe readings were uncertain both in terms of the precision of the measurement itself and the time-related aspect of probe removal and placement.

Estimates for the non-generic ruptures are even more uncertain than for the generic rupture. Several ruptures were difficult to analyze because of conflicting or missing information. For these ruptures, estimates were derived using the available information, and professional judgment.

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ATTACHMENT II-A

TABLES AND DOCUMENTATION FOR INDIVIDUAL RUPTURE INCIDENTS

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Method to Estimate Emissions from BGRR Fuel Ruptures

Fuel slugs are identical to those in Oak Ridge X-10 Reactor

Assume inventory of radionuclides in fuel slug can be estimated based on X-10 and data from BGRR log books.

Assume X-10 release fractions from a fuel slug
- 10% of particulate inventory
- 80% of radioiodines inventory
- 100% of noble gas inventory

Assume 10% of particulate release (1% of particulate inventory) gets through the filters and is emitted up stack.

Assume probe reading is a measure of particulate release from fuel

Particulate emissions up the stack are calibrated to probe reading from detailed measurements of ruptures 21,22,25

7.8 mCi (emitted)/mR/hr (probe)

Calculate fraction of particulate inventory of a slug that is emitted up stack

Compare particulate emissions to 1% particulate inventory of a fuel slug to calculate effective number of slugs involved and round to next higher integer (slug equivalents)

Take the inventory in the slug equivalents and assume no loss in extensive air ducts for non-particulate radioisotopes so that -
- 80% of radioiodines in slug equivalents are emitted up stack
- 100% of noble gases in slug equivalents are emitted up stack.

For non-generic ruptures additional assumptions are made that are detailed in the discussion of the specific rupture

A Guide to the Rupture Information Tables

The First Table

The Rupture Number is assigned serially by the date of the rupture.

- Class-- Either Generic or Non-generic as described in sections 4.3 and 4.4
- Rupture Date-- Taken from the Reactor Operations Monthly Report
- Rupture time--Taken From the Reactor Operation logbooks
- Channel Number--Gives the location of the ruptured cartridge in the BGRR discussed in the introduction to Part II
- Discharge Day ---- the number of days since the first day of reactor operations when the ruptured fuel cartridge was removed -- taken from the Reactor Operations Monthly Report
- Charge Day---the number of days since the first day of reactor operations when the ruptured cartridge was placed in the reactor
- Number of days in reactor-- the difference between number of days given under "Discharge Day" and "Charge Day"
- Average Power Level over period in Reactor and Total MWD---Average power level was taken from the Reactor Operation logbooks. The total power generated during the time the ruptured cartridge was in the reactor is the product of the average power level and the number of days cartridge was in the reactor
- Probe Measurement--This is discussed in Part II section 2. These values are used to estimate the particulate release as described in Part II section 4.2
- Derived slug equivalent--The number of fuel slugs assumed to have been involved in the rupture The approach is given in Part II sections 4.3 and 4.4
- Wind speed at the ground--this was the wind speed during the time of release. This data was taken from the historical meteorological records
- Wind Direction (towards)---- This was the direction the wind blew during the release. This data was taken from the historical meteorological records
- Calculated Distance to Maximum I -131 Deposition /Activity---Using the stack emission releases as given in the narrative below the tables, a transport computer code HOTSPOT described in section 4.1.4 was used to give an estimate of the distance downwind that the maximum deposition occurred and an estimate of the concentration at that point. When the point of maximum deposition was on the laboratory site the value given is at the site boundary

The Second Table

- The first column is a list of all the radionuclides calculated in the rupture release. This list was taken from the assessment done for the ruptures in the Oak Ridge X-10 Reactor. This is discussed in detail in Part II Section 3.0
- The second column provides the quantity of each of the listed radionuclides for the particular fuel elements before rupture as detailed in Part II Section 3.0
- The third Column gives the number of slug equivalents as derived via the descriptive material under the first table
- The fourth column gives the release fractions. For the noble gases 100% of the inventory of each slug equivalent is assumed to have been released. For the Iodines 80% is assumed to be released. The basis for this is given in Part II Sections 4.1 and 4.2. For the particulates the value is derived primarily from the probe measurements. The basis for this is given in Part II Sections 4.2, 4.3 and 4.4
- The fifth column,"Curies Released " is the estimate of the release from the stack. It is the product of second third and fourth columns

NOTES: These notes provide all the information available for each rupture. It was from this material that profession judgments can be made about the severity of each rupture.

Rupture Number 1

Class	Generic
Rupture Date	1/2/52
Rupture Time	1500
Channel Number	S C-1-0
Discharge Day	503
Charge Day	35
Number of Days in Reactor	468
Average Power Level Over Period in Reactor/Total MWD Day	11.94 MW, 5588 MWD
Probe Measurement	40 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	0.8 m/s; 0.0002 l/s rain
Wind Direction (towards)	SW/SSW
Calculated Distance to Maximum I-131 Deposition / Activity	2.9 km / 9.9E+04 Bq/m^2

(1) The probe measurement was used to estimate that 0.3 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 316 Ci the release fraction is 0.3 Ci / 316 Ci = 0.00095. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.19	1	1	0.19
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.64			54.64
I-129	4.16E-07	1	0.8	3.33E-07
I-131	24.30	1	0.8	1.94E+01
I-133	54.45	1	0.8	4.36E+01
Total Iodines	78.75			6.30E+01
Ba-140	52.80		0.00095	5.02E-02
Ce-144	34.13		0.00095	3.24E-02
Cs-137	1.37		0.00095	1.30E-03
La-140	52.80		0.00095	5.02E-02
Nb-95	53.60		0.00095	5.09E-02
Ru-103	24.29		0.00095	2.31E-02
Ru-106	1.80		0.00095	1.71E-03
Sr-89	40.11		0.00095	3.81E-02
Sr-90	1.48		0.00095	1.40E-03
Zr-95	53.29		0.00095	5.06E-02
U-235	1.79E-05		0.00095	1.70E-08
U-238	3.88E-04		0.00095	3.69E-07
U-234	3.88E-04		0.00095	3.69E-07
Pu	0.06		0.00095	5.82E-05
Total Particulates	315.72			0.30

Notes

(1) "At 1540 the Exit Filter Activity monitor alarmed and at the same time the Stack gas activity monitor indicated a substantial rise. The Exhaust gas probe was immediately removed from the duct and a reading of 40 mR/hr was noted on its felt pad. The reactor was shutdown for discharging C-1-0 South at 1546...

The ruptured unit was removed from its storage tube in the canal and inspected. The ruptured areas were noted to be about 40 inches from the gap end and to be two slits, several inches in length, running parallel to and between fins. Black oxide was in evidence although the swelling was negligible."

(Reactor Operations Monthly Report, January 1952)

(2) "Sy Block outlined the ruptured slug incident which happened on the previous evening. The rod was finally pulled out of the Pile around 1 A.M. and a new one was put in its place and the Pile was back on the air before 5 A.M.

The 30-minute air samples taken on the front porch of T-88 and at the Health Physics office window in Chemistry on the same afternoon of the accident, showed no activity. The air sample taken on the following morning at the Igloo Area, downwind from the Pile stack, showed no activity. Two water collectors were placed at the apartment area and the ball diamond during the night. No appreciable activity was found. The results from the Area Monitoring Stations is not yet available."

(Gemmell to Health Physics Surveyors, January 4, 1952)

Rupture Number 2

Class	Generic
Rupture Date	3/11/52
Rupture Time	0825
Channel Number	S A-1-0
Discharge Day	572
Charge Day	38
Number of Days in Reactor	534
Average Power Level Over Period in Reactor One Total MWD	12.62 MW, 6739 MWD
Probe Measurement	Not available
Derived Slug Equivalent	1
Wind Speed at the Ground	7.3 m/s
Wind Direction (towards)	NW
Calculated Distance to Maximum I-131 Deposition / Activity	2.5 km / 2.7E+03 Bq/m^2

(1) Although there was no probe reading, this rupture was similar in every aspect to Rupture 3. These ruptures were a day apart, had similar operating histories, and were in the same location in the reactor. Therefore, the derived probe measurement is 220 mR/hr.

(2) The derived probe measurement was used to estimate that 1.7 Ci of particulates were emitted from the stack (section 4.3, Part II).

(3) Since the particulate inventory of this slug was 319 Ci, the release fraction is 1.7 Ci / 319 Ci = 0.0053. The specific particulate radionuclide releases are partioned by this release fraction.

(4) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(5) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.22	1	1	0.22
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.67			54.67
I-129	4.74E-07	1	0.8	3.79E-07
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			6.30E+01
Ba-140	52.80		0.0053	2.80E-01
Ce-144	36.54		0.0053	1.94E-01
Cs-137	1.55		0.0053	8.24E-03
La-140	52.80		0.0053	2.80E-01
Nb-95	53.60		0.0053	1.94E-01
Ru-103	24.30		0.0053	1.29E-01
Ru-106	2.45		0.0053	1.30E-02
Sr-89	40.16		0.0053	2.13E-01
Sr-90	1.68		0.0053	8.91E-03
Zr-95	53.45		0.0053	2.83E-01
U-235	1.79E-05		0.0053	9.49E-08
U-238	3.88E-04		0.0053	2.06E-06
U-234	3.88E-04		0.0053	2.06E-06
Pu	6.13E-02		0.0053	3.25E-04
Total Particulates	319.40			1.69

NOTES:

(1) "Following the shutdown period of March 7,8, and 9th, the reactor was started at 0800 on March 10th. It had operated approximately 24 hours when it became necessary to shut down due to the evidence of fission products in the South duct. Prior to the shutdown it had been established that the fuel unit in A-1-O-S was leaking."

(Reactor Operations Monthly Report, March 1952)

(2) "There were three cartridge ruptures followed by the hot job of replacement. Some building contamination resulted. There were numerous contamination incidents connected with removal of apparatus from the pile and various maintenance and operating jobs."

(Health Physics and Safety Summary, March 1952)

Rupture Number 3

Class	Generic
Rupture Date	3/12/52
Rupture Time	0220
Channel Number	S A-1-1
Discharge Day	573
Charge Day	2
Number of Days in Reactor	571
Average Power Level Over Period in Reactor and Total MWD	12.02 MW, 6863 MWD
Probe Measurement	220 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	4.2 m/s
Wind Direction (towards)	ESE
Calculated Distance to Maximum I-131 Deposition / Activity	3.0* km / 4.6E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 1.7 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 320 Ci, the release fraction is 1.7 Ci / 320 Ci = 0.0053. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.23	1	1	0.23
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.68			54.68
I-129	5.07E-07	1	0.8	4.06E-07
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			6.30E+01
Ba-140	52.80		0.0053	2.80E-01
Ce-144	37.73		0.0053	2.00E-01
Cs-137	1.66		0.0053	8.80E-03
La-140	52.80		0.0053	2.80E-01
Nb-95	53.60		0.0053	2.84E-01
Ru-103	24.30		0.0053	1.29E-01
Ru-106	2.02		0.0053	1.07E-02
Sr-89	40.18		0.0053	2.13E-01
Sr-90	1.80		0.0053	9.52E-03
Zr-95	53.50		0.0053	2.84E-01
U-235	1.79E-05		0.0053	9.49E-08
U-238	3.88E-04		0.0053	2.06E-06
U-234	3.88E-04		0.0053	2.06E-06
Pu	6.13E-02		0.0053	3.25E-04
Total Particulates	320.45			1.70

NOTES:

(1) "Shortly after the completion of these operations the pile returned to normal operations. As soon as the repressurization of the Helium was completed (the system is normally repressurized during Medical treatments), the fuel units in channels A-1-1-S and C-11-4-N were found to be leaking at high rates. Again the presence of fission products were noted in the South duct, indicating that the fuel unit in A-1-1-S had ruptured. The reactor was again shut down and the leaking units were removed.

The incidence of three ruptured units since January seemed somewhat ominous. A review of all operating conditions was made and it was noted that rapid temperature cycling of the reactor had been more prevalent recently. With this clue, the operating procedures were revised to minimize the amplitude and decrease the frequency of the temperature cycles."

(Reactor Operations Monthly Report, March 1952)

(2) See note 2 for Rupture #2

(Health Physics and Safety Summary, March 1952)

(3) 3/12/52 0220 – Probes slightly rising
0349 – South Probe 220 mR/hr
Shut down the reactor

(Reactor Operations Logbook)

Rupture Number 4

Class	Generic
Rupture Date	5/6/52
Rupture Time	0900
Channel Number	N C-4-0
Discharge Day	629
Charge Day	52
Number of Days in Reactor	577
Average Power Level Over Period in Reactor and Total MWD	13.26 MW, 7651 MWD
Probe Measurement	250 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	4A (30 minutes) 2.3 m/s 4B (450 minutes) 1.1 m/s
Wind Direction (towards)	4A (30 minutes) NE 4B (450 minutes) SE
Calculated Distance to Maximum I-131 Deposition / Activity	4A (30 minutes) 2.25* km / 6.8E+03 Bq/m^2 4B (450 minutes) 4.40 km / 1.4E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 1.95 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 320 Ci, the release fraction is 1.95 Ci / 320 Ci = 0.0061. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.23	1	1	0.23
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.68			54.68
I-129	5.1E-07	1	0.8	4.10E-07
I-131	24.30	1	0.8	1.94E+01
I-133	54.45	1	0.8	4.36E+01
Total Iodines	78.75			6.30E+01
Ba-140	52.80		0.0061	3.22E-01
Ce-144	37.91		0.0061	2.31E-01
Cs-137	1.68		0.0061	1.02E-02
La-140	52.80		0.0061	3.22E-01
Nb-95	53.60		0.0061	3.27E-01
Ru-103	24.30		0.0061	1.48E-01
Ru-106	2.04		0.0061	1.24E-02
Sr-89	40.18		0.0061	2.45E-01
Sr-90	1.81		0.0061	1.11E-02
Zr-95	53.51		0.0061	3.26E-01
U-235	1.79E-05		0.0061	1.09E-07
U-238	3.88E-04		0.0061	2.37E-06
U-234	3.88E-04		0.0061	2.37E-06
Pu	6.13E-02		0.0061	3.74E-04
Total Particulates	320.69		0.0061	1.95

NOTES:

(1) "On May 6, 1952, the North exit filter activity unit and the North duct probe indicated the presence of fission products. The Stack gas Recorder and Helium System instrumentation indicated normal readings and were checked to be in good working condition. The reactor was shut down and the plenum chambers were probed with a hastily constructed gas sampling instrument in an effort to locate the source of the activity. Since no positive indication of a fission product source was obtained, the reactor was again started and operated normally for eleven hours when the exit filter activity and the duct probes indicated a sizable increase in the fission products present. The reactor was again shut down and the probing operation of the previous date was repeated with some success, in that a high background area was located and narrowed by probing with rods coated with gum tape until it was determined that all of the fission products were coming from Channel C-4-O-N. The fuel element in this Channel was then discharged.

The most feasible explanation of this incident seems to be that the 1/8" Helium tube became 'crimped' after it was tested for continuity.
Since no Helium was supplied to the opening, there was no washing out of radioactive gas from the inside of the fuel unit; which explains the normal stack gas activity and Helium System indications."

(Reactor Operations Monthly Report, May 1952)

(2)

5/6/52	0930	North Probe 250 mR/hr. Reactor Shut down
	1735	Reactor at 19 MW
	2330	North Probe 250 mR/hr.

(Reactor Operations Logbook)

Rupture Number 5

Class	Non-generic
Rupture Date	12/2/52
Rupture Time	0800
Channel Number	S D-0-3
Discharge Day	838
Charge Day	1
Number of Days in Reactor	837
Average Power Level Over Period in Reactor and Total MWD	14.29 MW, 11,961 MWD
Probe Measurement	Not available
Derived Slug Equivalent	1
Wind Speed at the Ground	0.8 m/s
Wind Direction (towards)	SW/WSW
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 3.2E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) For rupture #5, there is a notation in the reactor logbooks that the south probe reading was off-scale. However, the descriptive material available for this event does not reflect such a serious release. Fortunately, for rupture #6 there was a probe reading of 80 mR/hr., 27 minutes prior to shutdown at 2130 hours on 4/17/55 and a probe reading on 4/18/55 at 0840 of 85 mR/hr., some 11 hours later. For rupture #5, the shutdown occurred at 0805 on 12/2/52 and the next morning the south probe was 16 mR/hr. Although a quantitative relationship cannot be derived, it seems likely that ruptures 5 and 6 are quite similar and an assumption of one slug being ruptured seems appropriate for rupture #5, together with the generic approach.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 1 slug is assumed to have been released to the stack. For the particulates, 1% of the inventory of 1 slug is assumed to have been released to the stack. For this rupture the particulate inventory is multiplied by 0.01. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.33	1	1	0.33
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.78			54.78
I-129	7.43E-07	1	0.8	5.95E-07
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.80		0.01	5.28E-01
Ce-144	43.79		0.01	4.38E-01
Cs-137	2.41		0.01	2.41E-02
La-140	52.80		0.01	5.28E-01
Nb-95	53.60		0.01	5.36E-01
Ru-103	24.30		0.01	2.43E-01
Ru-106	2.43		0.01	2.43E-02
Sr-89	40.20		0.01	4.02E-01
Sr-90	2.61		0.01	2.61E-02
Zr-95	53.60		0.01	5.36E-01
U-235	1.79E-05		0.01	1.79E-07
U-238	3.88E-04		0.01	3.88E-06
U-234	3.88E-04		0.01	3.88E-06
Pu	6.13E-02		0.01	6.13E-04
Total Particulates	328.61			3.29

NOTES:

(1) "On December 2, 1952, the fuel unit in the south end of D-O-3 indicated a large leak rate on the helium system. Indications of fission products were noted in the south duct 45 minutes later and the reactor was immediately shutdown for discharging. This was the fifth fuel unit to rupture in the Brookhaven pile in such a way as to allow fission products to escape."

(Reactor Operations Monthly Report, December 1952)

(2) 12/2/52 0805 Probe off scale [>2 R/hr.]
12/3/52 0800 – 16:00 South Probe 16 mR/hr.

(Reactor Operations Logbook)

Rupture Number 6

Class	Generic
Rupture Date	4/17/53
Rupture Time	2100
Channel Number	S A 3-1
Discharge Day	977
Charge Day	862
Number of Days in Reactor	115
Average Power Level Over Period in Reactor and Total MWD	17.32 MW, 1,992 MWD
Probe Measurement	85 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	6A (1 hour) 6.4 m/s 6B (3.5 hour) 1.3 m/s 6C (1 hour) 3.4 m/s; 0.0001 l/s rain
Wind Direction (towards)	6A (1 hour) SE 6B (3.5 hour) WNW 6C (1 hour) S
Calculated Distance to Maximum I-131 Deposition / Activity	6A (1 hour) 2.5* km / 6.4E+02 Bq/m^2 6B (3.5 hour) 4.4* km / 1.1E+0.3 Bq/m^2 6C (1 hour) 2.5* km / 1.1E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 0.66 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 258 Ci, the release fraction is 0.66 Ci / 258 Ci = 0.0026. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	4.83E-02	1	1	4.83E-02
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.50			54.50
I-129	1.02E-07	1	0.8	8.17E-08
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.69		0.0026	1.37E-01
Ce-144	12.20		0.0026	3.17E-02
Cs-137	0.34		0.0026	8.82E-04
La-140	52.80		0.0026	1.37E-01
Nb-95	48.23		0.0026	1.25E-01
Ru-103	21.09		0.0026	5.48E-02
Ru-106	0.60		0.0026	1.56E-03
Sr-89	31.19		0.0026	8.11E-02
Sr-90	0.37		0.0026	9.54E-04
Zr-95	38.48		0.0026	1.00E-01
U-235	1.8E-05		0.0026	4.65E-08
U-238	3.88E-04		0.0026	1.10E-06
U-234	3.88E-04		0.0026	1.10E-06
Pu	6.13E-02		0.0026	1.59E-04
Total Particulates	258.04			0.66

NOTES:

(1) "Considerable difficulty was experienced during the weekend of April 17, due to the apparent failure of the leak detection system in locating a ruptured fuel element. At 2157, on April 17, the probes and exit filter monitor for the south duct indicated that fission products were escaping from a fuel element in the south half of the reactor and the reactor was shut down. Since the helium system had not given the response usually noted when a rupture occurs, several fuel elements known to be leaking were suspected and individual leak rates were determined on these elements. This revealed that the leak rates on elements C-1-O-S and B-2-2-S had substantially increased. These elements were removed as possible ruptures and the pile was again started.

Persistent indications of the escape of fission products continued and the pile was again shut down after operating less than two hours.

The entire south face of the pile was then probed with an air sampling detector designed for this purpose. When this operation was completed, A-2-5-S appeared to be the most likely suspect although the data was far from conclusive. This element was removed and the reactor was again started.

Indications that a leaking fuel unit remained in the reactor persisted after startup. However, it was decided to continue the operation of the reactor for a longer period of time. The size of the rupture was apparently small enough to defy location and surveys of the fans indicated that practically all of the particles were being retained on the filters.

The reactor was shut down nine hours later. Probing was again started using an improved technique. A-3-1-S was found to be the offender and the rupture was visible in the vicinity of the welded outer end." (Reactor Operations Monthly Report, April 1953)

(2) 4/17/53 2130 South Probe 80 mR/hr.
2157 Reactor shut down.
4/18/53 0840 South Probe 85 mR/hr.
(Reactor Operations Logbook)

Rupture Number 7

Class	Generic
Rupture Date	4/22/53
Rupture Time	0700
Channel Number	S B-0-1
Discharge Day	789
Charge Day	687
Number of Days in Reactor	102
Average Power Level Over Period in Reactor and Total MWD	20.90 MW, 2132 MWD
Probe Measurement	31 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	5.1 m/s
Wind Direction (towards)	E/ENE
Calculated Distance to Maximum I-131 Deposition / Activity	3.25* km / 2.8E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 0.24 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 250 Ci, the release fraction is 0.24 Ci / 250 Ci = 0.00095. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	4.29E-02	1	1	4.29E-02
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.49			54.49
I-129	9.06E-08	1	0.8	7.25E-08
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.59		0.00095	5.05E-02
Ce-144	10.98		0.00095	1.05E-02
Cs-137	0.30		0.00095	2.89E-04
La-140	52.80		0.00095	5.07E-02
Nb-95	46.63		0.00095	4.48E-02
Ru-103	20.27		0.00095	1.95E-02
Ru-106	0.54		0.00095	5.17E-04
Sr-89	29.53		0.00095	2.83E-02
Sr-90	0.33		0.00095	3.13E-04
Zr-95	36.15		0.00095	3.47E-02
U-235	1.79E-05		0.00095	1.72E-08
U-238	3.88E-04		0.00095	3.72E-07
U-234	3.88E-04		0.00095	3.72E-07
Pu	6.13E-02		0.00095	5.88E-05
Total Particulates	250.17			2.40E-01

NOTES:

(1) "At about 0700 on April 22, 1953, the helium system, stack air activity, exit filter activity and duct probe instruments all indicated a ruptured element. The leaking unit was located by the helium system immediately and fuel element B-O-1-S was discharged. The reactor was at full power again within four and one-half hours. All elements of the leak detection and location system functioned properly.

Eight fuel elements were removed during the shut down scheduled at the end of the month. In all cases the fuel elements had substantial leaks, but in no case was there any indication of fission products' having escaped."

(Reactor Operations Monthly Report, April 1953)

(2) 4/22/53 0730 South Probe 19.5 mR/hr
0745 South Probe 31mR/hr
0755 Reactor Shut down

(Reactor Operations Logbook)

Rupture Number 8

Class	Generic
Rupture Date	5/10/53
Rupture Time	0030
Channel Number	N B-2-7
Discharge Day	997
Charge Day	996
Number of Days in Reactor	1
Average Power Level Over Period in Reactor and Total MWD	21.95 MW, 21.95 MWD
Probe Measurement	140 mR/hr
Derived Slug Equivalent	5
Wind Speed at the Ground	1 m/s
Wind Direction (towards)	E
Calculated Distance to Maximum I-131 Deposition / Activity	4.4 km / 1.1E+03 Bq/m^2

(1) The probe measurement was used to estimate that 1.1 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 23.5 Ci the release fraction is 1.1 Ci / 23.5 Ci = 0.047. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction is greater than would be expected from 4 slugs, it was assumed in estimating the noble gas and radioiodine releases that there were 5 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 5 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	4.24E-04	5	1	2.1E-03
Xe-133	6.69E	5	1	3.3E+01
Total Noble Gases	6.69			33
I-129	8.88E-10	5	0.8	3.5E-09
I-131	2.00	5	0.8	8.02
I-133	29.79	5	0.8	1.19E+02
Total Iodines	31.79			1.27E+02
Ba-140	2.78		0.047	1.30E-01
Ce-144	1.21E-01		0.047	5.70E-03
Cs-137	2.96E-03		0.047	1.39E-04
La-140	1.79E+01		0.047	8.41E-01
Nb-95	1.06		0.047	4.99E-02
Ru-103	4.24E-01		0.047	1.99E-02
Ru-106	5.81E-03		0.047	2.73E-04
Sr-89	5.19E-01		0.047	2.44E-02
Sr-90	3.20E-03		0.047	1.50E-04
Zr-95	5.86E-01		0.047	2.76E-02
U-235	1.79E-05		0.047	8.41E-07
U-238	3.88E-04		0.047	1.82E-05
U-234	3.88E-04		0.047	1.82E-05
Pu	6.13E-02		0.047	2.88E-03
Total Particulates	23.46		0.047	1.10

NOTES:

(1) "On May 9, 1953, fuel was charged into the North and South ends of Channel B-2-7. Shortly after operation was resumed, the instrumentation indicated that the element in the North end of this channel had been ruptured. A rupture after so short a period of operation had never been experienced and it was necessary to probe the channel to confirm the condition.

The ruptured element was removed and operations were resumed. Since this element experienced only a few hours of operation, it will probably be 'cool' enough to remove from the canal for detailed inspection in the near future."

(Reactor Operations Monthly Report, May 1953)

(2) 5/10/53 0049 Probe read 140 mR/hr.
0055 Reactor shut down

(Reactor Operations Logbooks)

Rupture Number 9

Class	Generic
Rupture Date	7/23/53
Rupture Time	1400
Channel Number	N C-3-3
Discharge Day	1074
Charge Day	1
Number of Days in Reactor	1073
Average Power Level Over Period in Reactor and Total MWD	15.90 MW, 17,061 MWD
Probe Measurement	1000 mR/hr
Derived Slug Equivalent	3
Wind Speed at the Ground	1.5 m/s
Wind Direction (towards)	ESE
Calculated Distance to Maximum I-131 Deposition / Activity	4.4 km / 7.8E+03 Bq/m^2

(1) The probe measurement was used to estimate that 7.8 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 333 Ci the release fraction is 7.8 Ci / 333 Ci = 0.023. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was slightly more than would be expected from 2 slugs, it was assumed in estimating the noble gas and radioiodine releases that there were 3 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 3 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.41	3	1	4
Xe-133	54.45	3	1	163.35
Total Noble Gases	54.86			164.59
I-129	9.53E-07	3	0.8	2.29E-06
I-131	24.30	3	0.8	5.83E+01
I-133	54.45		0.8	1.31E+02
Total Iodines	78.75			1.89E+02
Ba-140	52.80		0.023	1.21
Ce-144	46.73		0.023	1.07
Cs-137	3.07		0.023	7.06E-02
La-140	52.80		0.023	1.21
Nb-95	53.60		0.023	1.23
Ru-103	24.30		0.023	5.59E-01
Ru-106	2.66		0.023	6.12E-02
Sr-89	40.20		0.023	9.25E-01
Sr-90	3.32		0.023	7.64E-02
Zr-95	53.60		0.023	1.23
U-235	1.79E-05		0.023	4.12E-07
U-238	3.88E-04		0.023	8.92E-06
U-234	3.88E-04		0.023	8.92E-06
Pu	6.13E-02		0.023	1.41E-03
Total Particulates	333.15		0.023	7.66

NOTES:

(1) "On July 23, evidence of fission products escaping into the north duct was received from two independent detecting systems, but the helium system indicated no new large leaks. This fuel element in the north end of channel C-1-2 had previously been removed from the helium system because of a leak rate which was interfering with the system's sensitivity. In such cases, the element removed is fed from a separate helium tank until discharged. The leak rate of this element was checked and found to have increased from 125 cc/hr to 250 cc/hr in ten days. This element was then discharged and, operations were resumed. The presence of fission products in the ducts ahead of the filters persisted and the reactor was again shut down. Many hours of probing at low power levels followed with inconclusive results. Finally, oil soaked tapes were hung in front of all suspected channels. This resulted in the location of an area in the vicinity of C-3-1 as being questionable. The elements in the north end of channels C-3-1, C-3-2 and C-3-3 were removed. On the basis of probe readings the rupture is being assigned to the element in channel C-3-3, although the data is not conclusive. During this work, leaks developed in the elements in the north end of channels C-6-4 and C-3-8, due to damage to their helium tubes. These elements were also discharged. Operations were resumed following the discharging of these elements and the presence of fission products in the ducts rapidly diminished."

(Reactor Operations Monthly Report, July 1953)

(2)
7/23/53 1400	North Probe 40 mR/hr
1545	Reactor shut down
7/24/53 0145	North Probe dropped to 500 mR/hr
0200	North Probe 1000 mR/hr
0205	Reactor shut down

(Reactor Operations Logbook)

Rupture Number 10

Class	Non-generic
Rupture Date	8/24/53
Rupture Time	1340
Channel Number	S A-2-4
Discharge Day	1104
Charge Day	1
Number of Days in Reactor	1103
Average Power Level Over Period in Reactor and Total MWD	16.07 MW; 17,725 MWD
Probe Measurement	Not available
Derived Slug Equivalent	5
Wind Speed at the Ground	10A (20 minutes) 1.4 m/s 10B (10 minutes) 4.1 m/s 10C (15 minutes) 2.0 m/s
Wind Direction (towards)	10A (20 minutes) SE 10B (10 minutes) NE 10C (15 minutes) NE
Calculated Distance to Maximum I-131 Deposition / Activity	10A (20 minutes) 2.5* km / 3.0E+04 Bq/m^2 10B (10 minutes) 3.5* km / 7.5E+03 Bq/m^2 10C (15 minutes) 4.4* km / 5.1E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) Rupture #10 was a large rupture, although effects were not seen in the environmental monitoring shacks, and there was no discussion of difficulty due to swelling of the cartridge, nor was there any evidence of burning. It would, therefore, seem appropriate to assume that all five "lost slugs" given in Table 1 were ruptured during this event and that the generic approach is appropriate.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 5 slugs is assumed to have been released to the stack. For the particulates, 1% of the inventory of 5 slugs is assumed to have been released to the stack. For this rupture the particulate inventory is multiplied by 0.05. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.42	5	1	2.12
Xe-133	54.45	5	1	272.25
Total Noble Gases	54.87			274.37
I-129	9.80E-07	5	0.8	3.92E-06
I-131	24.30	5	0.8	9.72E+01
I-133	54.45	5	0.8	2.18E+02
Total Iodines	78.75			3.15E+02
Ba-140	52.80		0.05	2.64
Ce-144	46.99		0.05	2.35
Cs-137	3.15		0.05	1.58E-01
La-140	52.80		0.05	2.64
Nb-95	53.60		0.05	2.68
Ru-103	24.30		0.05	1.22
Ru-106	2.68		0.05	1.34E-01
Sr-89	40.20		0.05	2.01
Sr-90	3.41		0.05	1.71E-01
Zr-95	53.60		0.05	2.68
U-235	1.79E-05		0.05	8.95E-07
U-238	3.88E-04		0.05	1.94E-05
U-234	3.88E-04		0.05	1.94E-05
Pu	6.13E-02		0.05	3.07E-03
Total Particulates	333.61			16.68

NOTES:

(1) "At 1340 hours on August 24 an increase was reported in the South probe reading. Several minutes later, while inspecting the readings on the stack air activity and exit filter activity instruments, an exceedingly rapid rise in activity was noted. The reactor was shut down at 1348.

It was immediately evident that the conditions surrounding the event were abnormal and that a larger than normal amount of uranium oxide and fission products had escaped from a fuel can. The duct wall opposite the exit filters was noted to read about 350 mR at a point where there is normally a background reading.

Probing confirmed that the unit in the south end of channel A-2-4 had ruptured. The removal of this unit was accomplished under difficult conditions due to the unusually high radiation levels from the contamination of the handling tools.

It has been estimated that a fairly large portion of the fuel in this element was converted to uranium oxide. (Probably ten slugs)."

(Reactor Operations Monthly Report, August 1953)

(2) "On the afternoon of August 24, one of the fuel elements in the Pile ruptured, releasing large amounts of activated Uranium dust. The exit air filter monitors collected a sufficiently large amount of this material to trip the building radiation alarm on the east balcony and to raise the background on the SU-3 in the control room by a factor of 10 or so. This increase in general background dropped away very rapidly. Two men immediately investigated by smears and MX-5 in a downwind direction from the stack. This brought negative results, indicating no serious release of particulate activity from the stack. The word from various individuals on site and the records from the area monitoring shacks indicate that an appreciable cloud of radioactive gas was emitted.

Radioactive particulates were detected by the continuous dust monitors at the area monitoring stations and by settled dust collectors. The data has not yet been fully analyzed but the levels observed were not large enough to cause any concern.

The analysis of area monitoring data during the recent slug rupture at the Pile shows that a significant amount of particulate activity was detected with the dust monitors but that there was no appreciable deposition of material at the stations. The peaks noted in the dust monitoring curves were sizable in only two cases. The other peaks might well have been due to residual activity from bomb tests."

(Health Physics and Safety Summary, August 1953)

(3) "Data in regard to fallout of activity caused by the rupture of a fuel cartridge at the Pile on 8/24/53 and that caused by Nevada bomb tests were summarized and compared in a memo to Dr. Fox. It was shown that the fallout due to the slug rupture was negligible compared to that due to the bomb tests."
(Health Physics and Safety Summary, November 1953)

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Rupture Number 11

Class	Generic
Rupture Date	11/8/53
Rupture Time	1130
Channel Number	S A-1-3
Discharge Day	1179
Charge Day	988
Number of Days in Reactor	191
Average Power Level Over Period in Reactor and Total MWD	20.96 MW, 4003 MWD
Probe Measurement	33 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	5.1 m/s
Wind Direction (towards)	NE
Calculated Distance to Maximum I-131 Deposition / Activity	3.5* km / 2.5E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 0.26 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 286 Ci the release fraction is 0.26 Ci / 286 Ci = 0.0009. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	7.97E-02	1	1	7.97E-02
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.53			54.53
I-129	1.70E-07	1	0.8	1.36E-07
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.80		0.0009	4.75E-02
Ce-144	18.60		0.0009	1.67E-02
Cs-137	5.62E-01		0.0009	5.06E-04
La-140	52.80		0.0009	4.75E-02
Nb-95	52.42		0.0009	4.72E-02
Ru-103	24.46		0.0009	2.11E-02
Ru-106	0.93		0.0009	8.38E-04
Sr-89	36.84		0.0009	3.32E-02
Sr-90	6.08E-01		0.0009	5.47E-04
Zr-95	47.05		0.0009	4.23E-02
U-235	1.79E-05		0.0009	1.61E-08
U-238	3.88E-04		0.0009	3.49E-07
U-234	3.88E-04		0.0009	3.49E-07
Pu	6.13E-02		0.0009	5.52E-05
Total Particulates	286.13		0.0009	2.58E-01

NOTES:

(1) "Two ruptured fuel elements were removed during the month. In both cases the detection of the rupture was immediate and insignificant amounts of fission products escaped into the ducts. In one case (A-1-3-S), the location of the offending units was routine and consumed a minimum of time; in the second case (C-1-1-N), some difficulty was experienced.

On the day proceeding the rupture of the element in the South end of the channel A-1-3, a power failure occurred which caused an emergency shutdown of the reactor. Following the shutdown, several large helium leaks were detected on the helium system and 1159 hours on November 8, the rupture was detected by the exhaust duct probes. These events provide some indication that the manner in which the reactor is shut down affects the life of fuel elements. An effort will be made to gather more information on this situation."

(Reactor Operations Monthly Report, November 1953)

(2) 11/8/57 1130 South Probe from 17 mR/hr to 27 mR/hr
1145 South Probe 33
1159 Reactor shut down

(Reactor Operations Logbook)

Rupture Number 12

Class	Generic
Rupture Date	11/16/53
Rupture Time	2110
Channel Number	N C-1-1
Discharge Day	1190
Charge Day	1
Number of Days in Reactor	1189
Average Power Level Over Period in Reactor and Total MWD	16.24 MW, 19,309 MWD
Probe Measurement	202 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	1.5 m/s
Wind Direction (towards)	NE
Calculated Distance to Maximum I-131 Deposition / Activity	4.4 km / 3.1E+03 Bq/m^2

(1) The probe measurement was used to estimate that 1.6 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 335 Ci the release fraction is 1.6 Ci / 335 Ci = 0.0048. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.45	1	1	0.45
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.90			54.90
I-129	1.06E-06	1	0.8	8.45E-07
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.80		0.0048	2.53E-01
Ce-144	47.66		0.0048	2.29E-01
Cs-137	3.39		0.0048	1.63E-02
La-140	52.80		0.0048	2.53E-01
Nb-95	53.60		0.0048	2.57E-01
Ru-103	24.30		0.0048	1.17E-01
Ru-106	2.74		0.0048	1.31E-02
Sr-89	40.20		0.0048	1.93E-01
Sr-90	3.67		0.0048	1.76E-02
Zr-95	53.60		0.0048	2.57E-01
U-235	1.79E-05		0.0048	8.59E-08
U-238	3.88E-04		0.0048	1.86E-06
U-234	3.88E-04		0.0048	1.86E-06
Pu	6.13E-02		0.0048	2.94E-04
Total Particulates	334.83		0.0048	1.61

NOTES:

(1) "Two ruptured fuel elements were removed during the month. In both cases the detection of the rupture was immediate and insignificant amounts of fission products escaped into the ducts. In one case (A-1-3-S), the location of the offending units were routine and consumed a minimum of time; in the second case (C-1-1-N), some difficulty was experienced.

On November 16, at 2110 hours, probe readings indicated the escape of fission products into the ducts although the helium system gave no indication of a leaking element. The reactor was shut down at 2155 hours and alternate methods of locating the rupture were employed. These methods consisted of probing and making short low power runs with tapes placed in the channel exhaust streams. Eventually, the offending element was located in channel C-1-1-N."

(Reactor Operations Monthly Report, November 1953)

(2) "Locating a ruptured fuel cartridge was a long job with high radiation levels. One run with the north scanner slots open resulted in fluxes of 12,000 slow neutrons/cm2/sec and 110 fast neutrons/cm^2/sec in the control room. Handling rods read up to 1000 r/hr at contact when removed from the Pile."

(Health Physics and Safety Summary, November 1953)

(3)

11/16/53	2110	48 mR/hr. – slow rise
	2155	North Probe 202 mR/hr.
		Reactor shut down

(Reactor Operations Logbook)

Rupture Number 13

Class	Non-generic
Rupture Date	1/17/54
Rupture Time	1030
Channel Number	S B-5-5
Discharge Day	1249
Charge Day	1
Number of Days in Reactor	1248
Average Power Level Over Period in Reactor and Total MWD	16.28 MW; 20,317 MWD
Probe Measurement	2000 mR/hr
Derived Slug Equivalent	2
Wind Speed at the Ground	8.5 m/s
Wind Direction (towards)	SE/SSE
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 5.6E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) For rupture 13, there was a stack release measurement by A. Solari of 6 Ci (see M. Weiss's memo of 1954 in Attachment IIC). If this activity is used in the generic approach, two slug failures would have been expected. However, there is a probe measurement of 2000 mR/hr. which would suggest five slugs were ruptured. It was noted that "...conditions surrounding this event were even more severe than usual and were similar to those experienced on August 24, 1953" and "...at several times during this operation flames were noted in the vicinity of the open container." Under such adverse conditions, it seems reasonable to assume that more than 10% of the particulates escaped the slugs. If an assumption of 2 slugs each released 25% of their particulates, a stack release of 7 Ci would be expected and a probe reading of about 2,200 mR would be expected. Therefore, for rupture 13, this report uses 2 slugs ruptures with 25% of the particulates released, 100% of the noble gases, and 80% of the radioiodines released from each of these slugs.

(2) For the noble gases, 100%, and for the radioiodines 80% of the inventory in 2 slugs is assumed to have been released to the stack. For the particulates 2.5% of the inventory in 2 slugs is assumed to have been released to the stack. For this rupture the particulate inventory in one slug is multiplied by 0.050. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.47	2	1	0.95
Xe-133	54.45	2	1	108.90
Total Noble Gases	54.92			109.85
I-129	1.11E-06	2	0.8	2.22E-06
I-131	24.30	2	0.8	38.9
I-133	54.45	2	0.8	87.1
Total Iodines	78.75			126.00
Ba-140	52.80		0.050	2.64
Ce-144	48.05		0.050	2.40
Cs-137	3.55		0.050	1.78E-01
La-140	52.80		0.050	2.64
Nb-95	53.60		0.050	2.68
Ru-103	24.30		0.050	1.22
Ru-106	2.77		0.050	0.14
Sr-89	40.20		0.050	2.01
Sr-90	3.84		0.050	0.19
Zr-95	53.60		0.050	2.68
U-235	1.79E-05		0.050	8.95E-07
U-238	3.88E-04		0.050	1.94E-05
U-234	3.88E-04		0.050	1.94E-05
Pu	6.13E-02		0.050	3.07E-03
Total Particulates	335.58		0.050	16.78

Notes:

(1) "On January 17, at 1144 hours, the reactor was shut down when the fuel element in the south end of the channel B-5-5 ruptured. This was the 13th ruptured fuel element experienced in 1249 days of operation. In eleven of the previous ruptures, ample warning was given in that the oxygen rate was slow enough to permit confirmation of the condition by observing data on the several systems which indicate such conditions. In this case, as in the rupture of August 24, 1953, no warning sign was given and appreciable amounts of uranium oxide escaped from the container. The reactor was shut down seven minutes after the first indication of the rupture. The helium system had located the leaking element two hours in advance of the rupture. However, since it is not uncommon for an element to develop a leak which may be fed with helium indefinitely without permitting the escape of fission products, the location of a leaking unit is not considered to necessarily indicate a rupture. The offending unit was removed in three portions, using three storage tubes. At several times during this operation, flames were noted in the vicinity of the open container. These were of short duration and required no special measures.

The conditions surrounding this event were more severe than usual and were similar to those experienced on August 24, 1953. The removal of this element consumed 18.67 hours.

Prompt evaluation of the surrounding area by the Health Physics Division again indicated that the loss of fission products from the stack had not been significant compared to the conditions in this area as a result of the recent work in Nevada."

(Reactor Operations Monthly Report, January 1954)

(2) "The new H.P. monitor of stack particulate activity gave an interesting record that showed a release of a few curies of activity as a result of the incident, an unimportant amount in view of the tremendous dilution. Meteorological conditions were favorable for detection by one of the area monitoring stations and a sizeable peak was noted on the dust monitor. Activity was also found in several snow samples taken soon after the incident. The activity was confined to a narrow sector with a maximum near the east site perimeter at station E-7. Because of the rapid decay of the released material and the short time involved, no overexposure of individuals either on or off the Site could have resulted. It was discovered that both the dust monitor used on stack air and the one in the monitoring station showed an increased counting rate at the instant of collection of the main burst of activity rather than after the normally assumed delay. Thus the record is a combination of this gamma ray breakthrough, as it varies with the position of the paper, and the normally assumed behavior. The effect was accentuated by the initial rapid decay of the samples. A study will be made of this effect."

(Health Physics and Safety Summary, January 1954)

(3) 1/17/54 1044 South Probe 2000 mR/hr.
Reactor shut down

(Reactor Operations Logbook)

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Rupture Number 14

Class	Generic
Rupture Date	1/22/54
Rupture Time	0935
Channel Number	S D-1-1
Discharge Day	1255
Charge Day	919
Number of Days in Reactor	336
Average Power Level Over Period in Reactor and Total MWD	20.32 MW, 6828 MWD
Probe Measurement	375 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	6.1 m/s
Wind Direction (towards)	SSW
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 4.0E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 2.9 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 307 Ci the release fraction is 2.9 Ci / 307 Ci = 0.0094. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.14	1	1	0.1
Xe-133	54.45	1	1	54.5
Total Noble Gases	54.59			54.6
I-129	2.98E-07	1	0.8	2.39E-07
I-131	24.30	1	0.8	1.94E+01
I-133	54.45	1	0.8	4.36E+01
Total Iodines	78.75			63.00
Ba-140	52.80		0.0094	4.96E-01
Ce-144	28.00		0.0094	2.63E-01
Cs-137	0.98		0.0094	9.25E-03
La-140	52.80		0.0094	4.96E-01
Nb-95	53.54		0.0094	5.03E-01
Ru-103	24.23		0.0094	2.28E-01
Ru-106	1.44		0.0094	1.36E-02
Sr-89	36.69		0.0094	3.73E-01
Sr-90	1.06		0.0094	1.00E-02
Zr-95	52.27		0.0094	4.91E-01
U-235	1.79E-05		0.0094	1.68E-07
U-238	3.88E-04		0.0094	3.65E-06
U-234	3.88E-04		0.0094	3.65E-06
Pu	6.13E-02		0.0094	5.76E-04
Total Particulates	306.88			2.88

NOTES:

(1) "The reactor returned to normal operation on January 18, and continued to operate until January 22 when it was again shut down for 27.75 hours, for removing a ruptured fuel element. In this case, the conditions were not severe and no appreciable amounts of oxide were lost to the exhaust.

The ruptured element removed on January 22 offered no special problem since its location and removal were accomplished before oxidation had progressed to any significant extent."

(Reactor Operations Monthly Report, January 1954)

(2)

1/22/54	0935	Probe 112 mR/hr.
	1330	Probe 240 mR/hr.
	1400	Probe 375 mR/hr.
		Reactor shut down

(Reactor Operations Logbook)

Rupture Number 15

Class	Non-generic
Rupture Date	2/21/54
Rupture Time	0630
Wet Storage Tube Number	2?
Channel Number	N B-4-5
Discharge Day	1286
Charge Day	1
Number of Days in Reactor	1285
Average Power Level Over Period in Reactor and Total MWD	16.34 MW; 20,997 MWD
Probe Measurement	>2000 mR/hr
Derived Slug Equivalent	3
Wind Speed at the Ground	7.2 m/s
Wind Direction (towards)	NW/WNW
Calculated Distance to Maximum I-131 Deposition / Activity	2.5* km / 8.4E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) For rupture 15, the reference material indicates a similarity with rupture 10. The major difference was the difficulty in removing the ruptured element from the fuel channel in rupture 15. F.P. Cowan notes in the February 1954 HP Summary that the stack monitor indicated about 9 Ci. However, the probe measurements were in excess of 2,000 mR/hr. for this rupture. It seems reasonable to use the 3 slugs listed in Table 1 as lost as the number of ruptured slugs, and, as we did with rupture 13, assume that 25% of the particulates were released from the ruptured slug.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 3 slugs is assumed to have been released to the stack. For the particulates, 2.5% of the inventory of 3 slugs is assumed to have been released to the stack. For this rupture, the particulate inventory in 1 slug is multiplied by 0.075. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.49	3	1	1.46
Xe-133	54.45	3	1	163.35
Total Noble Gases	54.94			164.81
I-129	1.14E-06	3	0.8	2.74E-06
I-131	24.30	3	0.8	5.83E+01
I-133	54.45	3	0.8	1.31E+02
Total Iodines	78.75			189.00
Ba-140	52.80		0.075	3.96
Ce-144	48.26		0.075	3.62
Cs-137	3.65		0.075	2.74E-01
La-140	52.80		0.075	3.96
Nb-95	53.60		0.075	4.02
Ru-103	24.30		0.075	1.82
Ru-106	2.79		0.075	2.09E-01
Sr-89	40.20		0.075	3.02
Sr-90	3.95		0.075	2.96E-01
Zr-95	53.60		0.075	4.02

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
U-235	1.79E-05		0.075	1.34E-06
U-238	3.88E-04		0.075	2.91E-05
U-234	3.88E-04		0.075	2.91E-05
Pu	6.13E-02		0.075	4.60E-03
Total Particulates	336.03		0.075	25.20

NOTES:

(1) "On February 21, 1954, the fuel element in the north end of channel B-5-5 ruptured. The conditions surrounding this rupture were similar in many respects to those encountered with the ruptures of August 24, 1953, and January 17, 1954. In each case, large amounts of activity abruptly appeared in the exhaust stream necessitating an immediate cessation of operations. In the latter case, the reactor was shut down approximately one minute after the first indication of the incident.

The removal of this ruptured element was accomplished by drilling through approximately 4 feet of the channel length with a long thin-walled hollow drill, the O.D. of which was 2.5 inches.

Throughout the entire operation the stack effluent was continuously monitored by Health Physics Division and additional ground checks were made in the direction of the wind from the stack. These results indicated that the exhaust filters were effective in controlling the contamination from the incident."

(Reactor Operations Monthly Report, February 1954)

(2) "The stack dust monitor showed that about 8.9 curies of particulate activity were released as a result of this incident. No effects chargeable to the incident were found in the records of any of the area monitoring stations and a survey with geiger counters revealed no detectable fallout. 0.7 curies were released during the period just before shutdown, 2.6 curies during the shutdown and 5.6 curies during the start-up and the 18-hour period after the start-up. The maximum rate of release of activity was about 25 mCi/min. During normal operation of the pile this monitor indicates release of filterable activity at the rate of about 0.14 mCi/min. Thus for the 3-day period of the shutdown, about 0.6 curies would normally have been released."

(Health Physics and Safety Summary, February 1954)

(3) 2/21/54
0632 North Probe >2000 mR/hr, reactor shut down.

(Reactor Operations Logbook)

Rupture Number 16

Class	Non-generic
Rupture Date	4/11/54
Rupture Time	0810
Channel Number	N B-0-1
Discharge Day	1334
Charge Day	1227
Number of Days in Reactor	107
Average Power Level Over Period in Reactor and Total MWD	18.90 MW; 2022 MWD
Probe Measurement	>2000 mR/hr
Derived Slug Equivalent	8
Wind Speed at the Ground	6.1 m/s
Wind Direction (towards)	NNE
Calculated Distance to Maximum I-131 Deposition / Activity	2.5* km / 2.8E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) Rupture 16 is a confusing rupture. The probe reading was greater than 2,000 mR/hr. and some increase in activity was seen in the dust monitor at two of the environmental monitoring stations. There were no reported difficulties removing the slug and no evidence of burning. The most confusing issue was the notation in Table 1 that 0 slugs were lost. It seems most reasonable to assume that 8 slugs ruptured, and the release fraction given for the generic ruptures is most applicable.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 8 slugs is assumed to have been released to the stack. For the particulates, 1% of the inventory of 8 slugs is assumed to have been released. For this rupture, the particulate inventory in 1 slug is multiplied by 0.08. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	4.50E-02	8	1	3.60E-01
Xe-133	54.45	8	1	4.36E+02
Total Noble Gases	54.49			435.96
I-129	9.50E-08	8	0.8	6.08E-07
I-131	24.30	8	0.8	155.50
I-133	54.45	8	0.8	348.48
Total Iodines	78.75			503.98
Ba-140	52.64		0.08	4.21
Ce-144	11.45		0.08	9.16E-01
Cs-137	3.16E-01		0.08	2.53E-02
La-140	52.80		0.08	4.22
Nb-95	47.29		0.08	3.78
Ru-103	20.61		0.08	1.65
Ru-106	5.63E-01		0.08	4.50E-02
Sr-89	30.20		0.08	2.42
Sr-90	3.41E-01		0.08	2.73E-02
Zr-95	37.08		0.08	2.97
U-235	1.79E-05		0.08	1.43E-06
U-238	3.88E-04		0.08	3.10E-05
U-234	3.88E-04		0.08	3.10E-05
Pu	6.13E-02		0.08	4.90E-03
Total Particulates	253.4		0.08	20.27

NOTES:

(1) "On April 11, 1954, all of the exhaust gas instruments suddenly indicated the presence of fission products in the exhaust ducts and the reactor was immediately shut down. The helium system, however, gave no indication of a leaking unit. As is now the practice in such cases, oil soaked tapes were dropped in front of each row of holes and the pile was restarted and operated for a few minutes at low power. The location of the difficulty was determined to be in Channel B-O-1 by the adherence of fission products to the tapes. The removal of the fuel in this channel corrected the difficulty."

(Reactor Operations Monthly Report, April 1954)

(2) "Failure of a fuel element on April 11 resulted in release of activity detectable on the dust monitors in two of the area monitoring stations. The effect was small however, and no activity could be found on the ground with survey instruments. There was the usual large quota of extra H.P. work connected with subsequent charging and discharging operations."

(Health Physics and Safety Summary, April 1954)

(3) 4/11/54 0812 North Probe greater than 2000 mR/hr.

(Reactor Operations Logbook)

Rupture Number 17

Class	Generic
Rupture Date	6/20/54
Rupture Time	0915
Channel Number	S B-3-5
Discharge Day	1404
Charge Day	1
Number of Days in Reactor	1403
Average Power Level Over Period in Reactor and Total MWD	16.81 MW 23, 23,584 MWD
Probe Measurement	120 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	4.7 m/s
Wind Direction (towards)	NNE
Calculated Distance to Maximum I-131 Deposition / Activity	2.5* km / 4.5E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 0.94 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 337 Ci the release fraction is 0.94 Ci / 337 Ci = 0.0028. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.53	1	1	0.53
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.98			54.98
I-129	1.25E-06	1	0.8	9.97E-07
I-131	24.30	1	0.8	1.49E+01
I-133	54.45	1	0.8	4.36E+01
Total Iodines	78.75			63.00
Ba-140	52.80		0.0028	1.48E-01
Ce-144	48.83		0.0028	1.37E-01
Cs-137	3.97		0.0028	1.11E-02
La-140	52.80		0.0028	1.48E-01
Nb-95	53.60		0.0028	1.50E-01
Ru-103	24.30		0.0028	6.80E-02
Ru-106	2.85		0.0028	7.97E-03
Sr-89	40.20		0.0028	1.13E-01
Sr-90	4.30		0.0028	1.20E-02
Zr-95	53.60		0.0028	1.50E-01
U-235	1.79E-05		0.0028	5.01E-08
U-238	3.88E-04		0.0028	1.09E-06
U-234	3.88E-04		0.0028	1.09E-06
Pu	6.13E-02		0.0028	1.72E-04
Total Particulates	337.32		0.0028	0.94

NOTES:

(1) "The element in the south end of B-3-5-S ruptured on June 21. The helium system located the leak simultaneously with the indication of fission products in the exhaust."

(Reactor Operations Monthly Report, p. 6)

(2) 6/21/54 0925 South Probe 110 to 120 mR/hr.
0930 Reactor shut down

(Reactor Operations Logbook)

Rupture Number 18

Class	Non-generic
Rupture Date	7/6/54
Rupture Time	0020
Channel Number	S A-5-2 or S B-5-2 *
Discharge Day	1419
Charge Day	53
Number of Days in Reactor	1366
Average Power Level Over Period in Reactor and Total MWD	17.62 MW; 24,069 MWD
Probe Measurement	80 mR/hr
Derived Slug Equivalent	5
Wind Speed at the Ground	1.8 m/s
Wind Direction (towards)	S
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 3.5E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) For rupture 18, it is difficult to assess the stack release because although the probe read 80 mR/hr, there was a notation in the Reactor Operations Report that "...white hot sparks and a dull red glow were noted several times while the element was being disturbed. Several slugs were probably oxidized badly during this event." Unlike ruptures 22 and 23, this cartridge was easily removed. It would seem appropriate, therefore, that the 5 slugs listed in Table 1 as lost should be accepted as ruptured for this event and then use the generic approach to estimate release fraction. Therefore, 5 slugs were ruptured and 100% of the noble gases, 80% of iodines, and 1% of the particulates were released. For this rupture the particulate inventory in 1 slug is multiplied by 0.05. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.51	5	1	2.57
Xe-133	54.45	5	1	272.25
Total Noble Gases	54.96			274.82
I-129	1.21E-06	5	0.8	4.85E-06
I-131	24.30	5	0.8	97.20
I-133	54.45	5	0.8	217.80
Total Iodines	78.75			315.00
Ba-140	52.80		0.05	2.64
Ce-144	48.67		0.05	2.43
Cs-137	3.87		0.05	1.94E-01
La-140	52.80		0.05	2.64
Nb-95	53.60		0.05	2.68
Ru-103	24.30		0.05	1.22
Ru-106	2.83		0.05	1.42E-01
Sr-89	40.20		0.05	2.01
Sr-90	4.19		0.05	2.10E-01
Zr-95	53.60		0.05	2.68
U-235	1.79E-05		0.05	8.95E-07
U-238	3.88E-04		0.05	1.94E-05
U-234	3.88E-04		0.05	1.94E-05
Pu	6.13E-02		0.05	3.07E-03
Total Particulates	336.93			16.85

NOTES:

(1) "The fuel element in channel B-5-2-S ruptured on July 6, at 1330 hours. All fission product detectors responded properly and the element was promptly located using the information from the helium system with confirmation by probing.

The removal of the unit was accomplished with a minimum of difficulty and contamination, although white hot sparks and a dull red glow were noted several times while the element was being disturbed. Several slugs were probably oxidized badly during this event."

(Reactor Operations Monthly Report, July 1954)

(2) "A defective fuel cartridge released considerable activity inside the pile on July 6th but was discharged with relatively small amounts of contamination. A tool used to probe the suspected channel read 120 r/hr at 3 feet and readings over the canal rose to 1 r/hr after discharge. A very strict traffic control system was set up and effectively minimized the spread of contamination."

(Health Physics and Safety Summary, July 1954)

(3) "On July 6th another rupture occurred. It was evident that it was a fairly bad rupture as it tripped a number of the building radiation monitors. The suspected channel was probed. When withdrawn to the charging face, the probe was found to read more than 120 r/hr at 3 feet. The probe was hastily shoved back into the plenum and was then discharged into the canal. The cartridge was discharged with remarkably little difficulty and relatively small amounts of contamination. Following the work, the highest smear on the pile top was 8300 c/m."

(Memo Keene to Gemmell, August 1954)

(4)

7/6/54	1328	South Probe 80 mR/hr.
	1333	Reactor Shut down

(Reactor Operations Logbook)

Rupture Number 19

Class	Generic
Rupture Date	6/19/55
Rupture Time	0130
Channel Number	N D-8-3
Discharge Day	1767
Charge Day	11
Number of Days in Reactor	1766
Average Power Level Over Period in Reactor and Total MWD	17.88 MW; 31576 MWD
Probe Measurement	144 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	1.9 m/s
Wind Direction (towards)	NE
Calculated Distance to Maximum I-131 Deposition / Activity	4.4 km / 2.5E+03 Bq/m^2

(1) The probe measurement was used to estimate that 1.1 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 340 Ci the release fraction is 1.1 Ci / 340 Ci = 0.0032. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.64	1	1	0.64
Xe-133	54.45	1	1	54.45
Total Noble Gases	55.09			55.09
I-129	1.57E-06	1	0.8	1.25E-06
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.80		0.0032	1.69E-01
Ce-144	49.85		0.0032	1.60E-01
Cs-137	4.95		0.0032	1.58E-02
La-140	52.80		0.0032	1.69E-01
Nb-95	53.60		0.0032	1.72E-01
Ru-103	24.30		0.0032	7.78E-02
Ru-106	2.95		0.0032	9.44E-03
Sr-89	40.20		0.0032	1.29E-01
Sr-90	5.35		0.0032	1.71E-02
Zr-95	53.60		0.0032	1.72E-01
U-235	1.79E-05		0.0032	5.73E-08
U-238	3.88E-04		0.0032	1.24E-06
U-234	3.88E-04		0.0032	1.24E-06
Pu	6.13E-02		0.0032	1.96E-04
Total Particulates	340.46			1.09

NOTES:

(1) "There is reasonably good evidence that a small amount of fission products escaped from one of these elements. On July 19, a gradual rise in the north duct activity was noted. At this time four known leakers were in the north half of the reactor and had been located by the helium system. The reactor was shut down and these leakers were probed. The element in ND-8-3 proved to be the source of the difficulty. It was removed along with the other three leakers and normal operation was resumed."

(Reactor Operations Monthly Report, June 1955)

(2) 6/19/55 0130 Probe 144 mR/hr.
1520 Slow shutdown of the Reactor

(Reactor Operations Logbook)

Rupture Number 20

Class	Generic
Rupture Date	10/11/55
Rupture Time	0600
Channel Number	N D-9-1
Discharge Day	1882
Charge Day	1
Number of Days in Reactor	1881
Average Power Level Over Period in Reactor and Total MWD	18.07 MW, 33,990 MWD
Probe Measurement	500 mR/hr
Derived Slug Equivalent	2
Wind Speed at the Ground	20A (90 minutes) 2.0 m/s 20B (60 minutes) 1.4 m/s 20C (60 minutes) 1.4 m/s
Wind Direction (towards)	20A (90 minutes) S 20B (60 minutes) WSW 20C (60 minutes) W
Calculated Distance to Maximum I-131 Deposition / Activity	20A (90 minutes) 2.25* km / 1.3E+04 Bq/m^2 20B (60 minutes) 2.25* km / 6.5E+03 Bq/m^2 20C (60 minutes) 1.75* km / 8.8E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 3.9 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 341 Ci the release fraction is 3.9 Ci / 341 Ci = 0.0114. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was slightly more than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there were 2 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 2 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	0.68	2	1	1.35
Xe-133	54.45	2	1	108.90
Total Noble Gases	55.13			110.25
I-129	1.67E-06	2	0.8	2.67E-06
I-131	24.30	2	0.8	38.88
I-133	54.45	2	0.8	87.12
Total Iodines	78.75			126.00
Ba-140	52.80		0.0114	0.60
Ce-144	50.02		0.0114	5.70E-01
Cs-137	5.25		0.0114	5.99E-02
La-140	52.80		0.0114	6.02E-01
Nb-95	53.60		0.0114	6.11E-01
Ru-103	24.30		0.0114	2.77E-01
Ru-106	2.97		0.0114	3.39E-02
Sr-89	40.20		0.0114	4.58E-01
Sr-90	5.68		0.0114	6.47E-02
Zr-95	53.60		0.0114	6.11E-01
U-235	1.79E-05		0.0114	2.04E-07
U-238	3.88E-04		0.0114	4.42E-06
U-234	3.88E-04		0.0114	4.42E-06
Pu	6.13E-02		0.0114	6.99E-04
Total Particulates	341.29			3.89

NOTES:

(1) "One element ruptured on October 12 and caused the loss of an insignificant amount of fission products to the exhaust stream. The helium system probes, stack air activity and exit filter activity instrument systems responded promptly, facilitating the detection and location of the ruptured element."

(Reactor Operations Monthly Report, October 1955)

(2) 10/12/55 0925 North Probe 500 mR/hr.
1056 Reactor shut down

(Reactor Operations Logbook)

Rupture Number 21

Class	Generic
Rupture Date	5/1/56
Rupture Time	0000
Wet Storage Tube Number	897
Channel Number	S A-1-3
Discharge Day	2084
Charge Day	1205
Number of Days in Reactor	879
Average Power Level Over Period in Reactor and Total MWD	21.11 MW , 18,556 MWD
Probe Measurement	900 mR/hr
Derived Slug Equivalent	3
Wind Speed at the Ground	1.8 m/s
Wind Direction (towards)	N/NNE - S/SSW
Calculated Distance to Maximum I-131 Deposition / Activity	4.4 km / 7.9E+03 Bq/m^2

(1) The probe measurement was used to estimate that 7 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 330 Ci the release fraction is 7 Ci / 330 Ci = 0.021. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was slightly more than would be expected from two slugs, it was assumed in estimating the noble gas and radioiodine releases that there were 3 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 3 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	3.45E-01	3	1	1.04
Xe-133	54.45	3	1	163.35
Total Noble Gases	54.80			164.39
I-129	7.81E-07	3	0.8	1.87E-06
I-131	24.30	3	0.8	58.32
I-133	54.45	3	0.8	130.68
Total Iodines	78.75			189.00
Ba-140	52.80		0.021	1.11
Ce-144	44.43		0.021	9.33E-01
Cs-137	2.53		0.021	5.32E-02
La-140	52.80		0.021	1.11
Nb-95	53.60		0.021	1.13
Ru-103	24.30		0.021	5.10E-01
Ru-106	2.48		0.021	5.21E-02
Sr-89	40.20		0.021	8.44E-01
Sr-90	2.74		0.021	5.75E-02
Zr-95	53.60		0.021	1.13
U-235	1.79E-05		0.021	3.76E-07
U-238	3.88E-04		0.021	8.15E-06
U-234	3.88E-04		0.021	8.15E-06
Pu	6.13E-02		0.021	1.29E-03
Total Particulates	329.55			7.0

NOTES:

(1) "On May 1, 1956, evidence of fission product contamination of the exhaust air was detected and the reactor was shut down. A fuel element in Channel A-1-3-South was indicated by the helium system as likely to be the ruptured element and was discharged. From the amount of contamination of the handling rods it was proved that this element had ruptured."

(Reactor Operations Monthly Report, May 1956)

(2)

5/1/56	0143	Probe 400 mR/hr.
		Slow shut down started
	2235	Probe read 900 mR/hr. after shut down

(Reactor Operations Logbook)

Rupture Number 22

Class	Non-generic
Rupture Date	5/3/56
Rupture Time	2005
Channel Number	S B-2-5
Discharge Day	2094
Charge Day	18
Number of Days in Reactor	2076
Average Power Level Over Period in Reactor and Total MWD	18.33 MW ; 38,053 MWD
Probe Measurement	>2000 mR/hr
Derived Slug Equivalent	5
Wind Speed at the Ground	6.8 m/s; 0.0004 l/s rain
Wind Direction (towards)	NW
Calculated Distance to Maximum I-131 Deposition / Activity	1.5* km / 2.6E+05 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) For rupture 22, the situation is complicated. In Phillips' analyses, shown in section 4.2, a value of 60 Ci at 2.5 hours is given, which must be increased to 120 Ci for the immediate post-release. Based on the generic approach, that would suggest about 33 slugs were involved. This is difficult to understand since only 5 slugs were missing in the canal. The more reasonable expectation was that the release fraction from the fuel slugs was greater than the assumed 10% based on the Oak Ridge studies. If 75% of the 5 slugs had been oxidized and released from the fuel element, about 130 Ci would have been released from the stack. For this analysis, an assumption is used of 5 slugs lost, and the particulate release fraction from a slug is 75%.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 5 slugs is assumed to have been released to the stack. For the particulates, 75% of the particulates are assumed to have been released from the fuel cartridge and 7.5% released to the stack. For this rupture, the particulate inventory in 1 slug is multiplied by 0.38. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	7.35E-01	5	1	3.68
Xe-133	54.45	5	1	272.25
Total Noble Gases	55.19			275.93
I-129	1.84E-06	5	0.8	7.38E-06
I-131	24.30	5	0.8	97.20
I-133	54.45	5	0.8	217.80
Total Iodines	78.75			315.00
Ba-140	52.80		0.38	20.06
Ce-144	50.23		0.38	19.09
Cs-137	5.76		0.38	2.19
La-140	52.80		0.38	20.06
Nb-95	53.60		0.38	20.37
Ru-103	24.30		0.38	9.23
Ru-106	3.00		0.38	1.14
Sr-89	40.20		0.38	15.28
Sr-90	6.23		0.38	2.37
Zr-95	53.60		0.38	20.37

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
U-235	1.79E-05		0.38	6.8E-06
U-238	3.88E-04		0.38	1.47E-04
U-234	3.88E-04		0.38	1.47E-04
Pu	6.13E-02		0.38	2.33E-03
Total Particulates	342.59			130.16

NOTES:

(1) "On May 2, 1956, Fission product contamination of the reactor exhaust cooling gas stream was again detected and the reactor was shut down. Unfortunately the helium system failed to indicate a definite suspect and it became necessary to make a "tape" run at 1 Mw for 15 minutes. Subsequent probing of channels in the area indicated by the "tape" run showed the fuel element in Channel B-2-5-South as the ruptured element. The rupture had progressed far enough to require more than usual force to be applied in order to discharge the element. Even though the shutdown following the detection of the rupture was made quickly, atmospheric conditions resulted in a small amount of fission product particles being deposited north of the reactor site."

(Reactor Operations Monthly Report, May 1956)

(2) "The meteorology Group records for the above time period that indicate that effluent from the Reactor should have passed directly over the north gate, across the northwest site boundary, and just south of the intersection of Smith's Road and Route 25."

"I think it would be most interesting to determine whether material from the Reactor slug rupture did reach the ground anywhere along this path because we have an unusual set of conditions to deal with. The wind direction was 130 degrees with a speed of 8 m.p.s. The temperature records show a lapse condition (-0.7 degrees Celsius) between 410 and 130 feet, and a very slight inversion (+0.2 degrees Celsius) between 150 an 37 feet above ground. I would normally expect that this very weak inversion layer would prevent small particulates from reaching the ground within any reasonable distance, but in this particular case, we know that light rain was occurring, and it is quite probable that some of the material was subsequently washed away by approximately 0.2 inches of rain.

As shown on the attached map, I would suggest a careful survey of the north gate, the indicated section of the northwest site boundary, the road running northeast from Smith's Road to Route 25, Smith's Road and Route 25 itself.

If I am not mistaken, this is the first rupture we have ever had that was accompanied by the light rainfall. As you know, it is my belief that the rain acts as a very affective scrubbing mechanism even at low levels, and a careful survey of this region should reveal the validity of the contention."

(Memo Smith to Gemmell, May 3, 1956)

(3) "The rupture of a fuel element early in the month led to the release of considerable activity from the stack. As a result of precipitation at the time, measurable activity was found in a narrow sector to the northwest. Local spots of surface contamination were detected with geiger counters well to the north of route 25. An attempt is being made to estimate the amount of activity release by assaying the activity deposited on the stack dust sampler."

(Health Physics and Safety Summary, May 1956)

(4) "An attempt is being made to determine the particle size of radioactive material found on dead vegetation as a result of fall-out from the 22nd. Rupture occurring in the BNL Reactor on May 2nd. It was discovered that the active particles (if entirely particulate and not molecular) are greatly exceeded in number by the inactive particles, and it is almost impossible to distinguish between them in the microscope. The problem evolved into one of removing the particulate material from the dead vegetation using ultrasonic methods then separation of the particles by settling in an alcohol column. Since the particles are not spherical, Stokes Law is only crudely applicable. The problem appears to be enlarged by the wide spectrum of densities observed. Results of this problem are expected to be only approximate and await analysis of samples withdrawn from 12 positions on the alcohol column.

An attempt is also being made to determine the quantity of particulate activity released from the reactor stack as a result of the same rupture. Approximately 5×10^{-6} of the exhaust air is sampled and it is assumed that the same fraction of the particulate activity was collected on the filter paper. Several methods are being tested utilizing Soxhlet extraction techniques to remove the fission products from the filter paper and putting them into solution. An aliquot may then be taken from this known volume and the total activity collected by the filter paper determines, then the total activity released by the reactor stack may be approximated."

(Memo Phillips to Gemmell, June 6, 1956)

(5) "A process has been developed to estimate the quantity of particulate activity released from the reactor stack as a result of fuel element ruptures. The process consists of a series of HNO_3 extractions from Hollingsworth Voss filter media.

Rupture no 22, which occurred 5/2/56, has been partially evaluated using the above mentioned extraction process. Approximately 5×10^{-6} of the exhaust air was sampled and it is assumed that the same fraction of the particulate activity was collected on the filter paper. The assay indicates that the remaining activity which exists today, after 115 days decay, amounts to 2.5 curies. Decay studies indicates that the activity levels remaining 4 ½ days after release from the stack amounted to approximately 10 curies. Insufficient decay data was collected to be able to estimate the number of curies released at any earlier date."

"An alcohol settling column was used to determine the particle size of dusts removed from contaminated leaves. The radioactivity found on the dead vegetation was a result of fallout from the rupture which occurred in the BNL reactor on May 2. The data from the settling column indicated the following results:

1) The particles (both active and inactive) removed from the leaves ranged from 0.5 to 4.5 microns in diameter
2) The greatest percentage of the particles were about one micron in diameter, and less than three percent were greater than two microns in diameter
3) The greatest percentage of activity removed from the leaves was associated with particles of 1 to 2 microns in diameter."

(Memo Phillips to Gemmell, August 31, 1956))

(6) 5/2/56 2035 Probe reading 2000 mR/hr 2 feet down on the handle
2040 Reactor shutdown

(Reactor Operations Logbook)

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Rupture Number 23

Class	Generic
Rupture Date	10/19/56
Rupture Time	0100
Channel Number	S A-3-4
Discharge Day	2255
Charge Day	1783
Number of Days in Reactor	472
Average Power Level Over Period in Reactor and Total MWD	20.58 MW; 9714 MWD
Probe Measurement	625 mR/hr
Derived Slug Equivalent	2
Wind Speed at the Ground	6.8 m/s
Wind Direction (towards)	SSW/SW
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 4.8E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 4.9 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 316 Ci the release fraction is 4.9 Ci / 316 Ci = 0.016. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was slightly more than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there were 2 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 2 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	1.92E-01	2	1	3.84E-01
Xe-133	54.45	2	1	108.90
Total Noble Gases	54.64			109.28
I-129	4.19E-07	2	0.8	6.71E-07
I-131	24.30	2	0.8	3.89E+01
I-133	54.45	2	0.8	8.71E+01
Total Iodines	78.75			126.00
Ba-140	52.80		0.0155	8.18E-01
Ce-144	34.28		0.0155	5.31E-01
Cs-137	1.38		0.0155	2.13E-02
La-140	52.80		0.0155	8.18E-01
Nb-95	53.60		0.0155	8.31E-01
Ru-103	24.29		0.0155	3.77E-01
Ru-106	1.81		0.0155	2.81E-02
Sr-89	40.11		0.0155	6.22E-01
Sr-90	1.49		0.0155	2.31E-02
Zr-95	53.31		0.0155	8.26E-01
U-235	1.79E-05		0.0155	2.77E-07
U-238	3.88E-04		0.0155	6.01E-06
U-234	3.88E-04		0.0155	6.01E-06
Pu	6.13E-02		0.0155	9.50E-04
Total Particulates	315.93			4.90

NOTES:

(1) "On Oct 19, the pile was shut down at 0635 due to a fuel element rupture."

"The first indication of the rupture on October 19 was evidenced by radioactive contamination of the air in both exhaust cooling air ducts.

The fuel element in SA-3-4 was determined to be the rupture and contamination in both ducts was caused apparently by the fuel element in the south half of the channel. This was due to the fact that the fuel element protruded about 1 ½ inches into the north half of the channel."

(Reactor Operations Monthly Report, October 1956)

(2)

10/19/56	0130	North Probe 100 mR/hr.
	0455	Slow Reactor shut down
	0535	Probe 625 mR/hr.
	0630	Reactor shut down

(Reactor Operations Logbook)

Rupture Number 24

Class	Generic
Rupture Date	11/14/56
Rupture Time	2000
Channel Number	S A-1-1
Discharge Day	2281
Charge Day	1910
Number of Days in Reactor	371
Average Power Level Over Period in Reactor on Total MW	20.56 MW; 7,628 MWD
Probe Measurement	450 mR/hr
Derived Slug Equivalent	2
Wind Speed at the Ground	4.7 m/s
Wind Direction (towards)	ENE
Calculated Distance to Maximum I-131 Deposition / Activity	3.5* km / 7.5E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 3.5 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 310 Ci the release fraction is 3.5 Ci / 310 Ci = 0.0113. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was slightly more than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there were 2 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 2 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	1.52E-01	2	1	3.05E-01
Xe-133	54.45	2	1	1.09E+02
Total Noble Gases	54.60			109.20
I-129	3.30E-07	2	0.8	5.27E-07
I-131	24.30	2	0.8	38.88
I-133	54.45	2	0.8	87.12
Total Iodines	78.75			126.00
Ba-140	52.80		0.0113	5.97E-01
Ce-144	29.82		0.0113	3.37E-01
Cs-137	1.09		0.0113	1.23E-02
La-140	52.80		0.0113	5.97E-01
Nb-95	53.57		0.0113	6.05E-01
Ru-103	24.26		0.0113	2.74E-01
Ru-106	1.55		0.0113	1.75E-02
Sr-89	39.88		0.0113	4.51E-01
Sr-90	1.17		0.0113	1.33E-02
Zr-95	52.70		0.0113	5.96E-01
U-235	1.79E-05		0.0113	2.02E-07
U-238	3.88E-04		0.0113	4.38E-06
U-234	3.88E-04		0.0113	4.38E-06
Pu	6.13E-02		0.0113	6.93E-04
Total Particulates	309.69			3.50

NOTES:

(1) "The first indication that a ruptured fuel element existed in the reactor was evidenced by radioactive contamination of the south exhaust cooling air duct on November 14, 1956. The fuel element in SA-1-1 was determined to be the rupture and was discharged to the canal."

(Reactor Operations Monthly Report, November 1956)

(2) "Fuel Element Rupture

The reactor was shut down when a sharp increase in activity was noted in the cooling air. The only cartridge showing a leak was discharged although a probe of that channel showed very little loose activity. The reactor was stared with no further difficulty. This was the "cleanest" rupture shutdown in memory. It is believed that the rupture was a small hole letting out gas and that the reactor was shutdown before a serious break developed."

(Memo, Boutelle to Gemmell, December 10, 1956)

(3) 11/14/56 2055 South Probe 450 mR/hr.
2105 Reactor shut down

(Reactor Operations Logbook)

Rupture Number 25

Class	Non-generic
Rupture Date	1/15/57
Rupture Time	1250
Channel Number	S A-3-3
Discharge Day	2343
Charge Day	1710
Number of Days in Reactor	633
Average Power Level Over Period in Reactor and Total MWD	20.44 MW; 12,939 MWD
Probe Measurement	Not available
Derived Slug Equivalent	4
Wind Speed at the Ground	2.3 m/s
Wind Direction (towards)	SW/SSW
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 2.2E+04 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) As indicated in section 4.2 L. Phillips estimated 7.2 curies of particulates were released from the stack as of 2.5 hours after release. In section 4.4, it was shown that this value should be increased to 14.4 curies to estimate the release at the time of the rupture. A 14.4 curie release of particulates would be consistent with a rupture of 4 slugs. This approach is used in this report.

(2) For the noble gases, 100%, and, for the radioiodines, 80% of the inventory in 4 slugs is assumed to have been released to the stack. For the particulates 1% of the inventory of 4 slugs is assumed to have been released to the stack. For this rupture the particulate inventory in 1 slug is multiplied by 0.04. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	2.54E-01	4	1	1.02
Xe-133	54.45	4	1	217.80
Total Noble Gases	54.70			218.82
I-129	5.62E-07	4	0.8	1.80E-06
I-131	24.30	4	0.8	7.78E+01
I-133	54.45	4	0.8	1.74E+02
Total Iodines	78.75			252.00
Ba-140	52.80		0.04	2.11
Ce-144	39.51		0.04	1.58
Cs-137	1.84		0.04	7.35E-02
La-140	52.80		0.04	2.11
Nb-95	53.60		0.04	2.14
Ru-103	24.30		0.04	0.97
Ru-106	2.14		0.04	8.56E-02
Sr-89	40.19		0.04	1.61
Sr-90	1.99		0.04	7.95E-02
Zr-95	53.55		0.04	2.14
U-235	1.79E-05		0.04	7.16E-07
U-238	3.88E-04		0.04	1.55E-05
U-234	3.88E-04		0.04	1.55E-05
Pu	6.13E-02		0.04	2.45E-03
Total Particulates	322.77			12.91

NOTES:

(1) "Fuel Element Rupture (S-A-3-3)

On January 15, 1957, operations were normal until a rise of 35% in the stack air activity was noted at 1250 indicating gaseous radioactive contamination of the exhaust cooling air. Probe readings in both ducts showed no increase. Inspection of a particulate activity recorder upstream of the filters showed no increase. At 1258 the pile was shut down by pressing the manual Trip No. 1 button. A few seconds before the pile was shut down the exit air filter monitor and the south probe reading increased approximately 60%, indicating radioactive particulate contamination of the exhaust cooling air from the south half of the pile. While the necessary rigging was being done, a recheck was made of several fuel elements which had previously been detected as leakers by means of the Helium Leak Detection System. One showed greatly increased leak rate and when the shield was opened a red glow was observed in the channel. The element was discharged to the canal and the pile started up with normal operation being reached by 0715 on January."

(Reactor Operations Monthly Report, January 1957)

(2) "Studies of the activity detected by the Health Physics stack monitor at the pile during the recent fuel element rupture indicate that the amount of particulate activity released, as of one-half hour after the incident, was about 25 curies. A memorandum is being prepared giving detailed results and techniques in connection with the evaluation of the data obtained from the stack monitor."

(Health Physics and Safety Summary, January 1957)

(3) "At 1300 on January 15, 1957 cartridge number 32 x 72 ruptured in channel SA-3-3. This fuel element was of the old type located in the inner ring of the normal fuel zone, a position at approximately 350 ° fin temperature. A rapid temperature cycle during an unscheduled shutdown on January 14 is considered the primary cause of this, the 25th rupture.

As a result of this rupture, a significant quantity of radioactive particulate was introduced into the pile cooling effluent. This effluent is filtered by the primary air outlet filters. These "glastex" filters are composed of a woven glass fiber cloth which is claimed to be more than 95% efficient for removal of all particles down to 5μ and 25 to 30% efficient for smaller particles. These efficiencies have been partly substantiated by particles size studies made under normal operating conditions and of "wash out" after rupture number 22.

(Progress Report, Phillips to Gemmell, June and August 1956)

After filtration, a small fraction of the effluent is sampled approximately iso-kinetically at the base of the stack by a continuous dust monitor. The dust is collected on a moving Hollingsworth and Vose type H-70 filter paper strip with a collection efficiency greater than 98% for 0.24μ diameter particles. About ten minutes after collection this filter strip moves past an anthracene scintillation counter. This monitor cannot be easily calibrated in terms of activity per unit volume of effluent without a large expenditure of equipment and time because of the problems of very high initial activity levels and "breakthrough", namely spurious counts due to radiation from the freshly collected fission products penetrating the shield surrounding the crystal. A series of nitric acid extractions of filter paper samples gives a simple solution to the problem without deviating from the present sampling methods.

Nitric acid extractions of filter paper samples collected during the rupture were made on January 18th. The analysis of these extractions and the application of the ratio between sampling and effluent flow rates made possible the determination of the number of curies existing in the environment at the time if the extraction. This information, plus decay data obtained from the rupture sample, determines the number of curies existing in the environment at any time after its release from the stack."
(Memo Phillips to Cowan, March 25, 1957)

(4) 1/15/57 1258 South Probe increased by 18 mR/hr.

(Reactor Operations Logbook)

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Rupture Number 26

Class	Generic
Rupture Date	3/1/57
Rupture Time	0840
Channel Number	S B-0-5
Discharge Day	2388
Charge Day	2305
Number of Days in Reactor	83
Average Power Level Over Period in Reactor and Total MWD	19.10 MW; 1585 MWD
Probe Measurement	39 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	4.1 m/s
Wind Direction (towards)	S
Calculated Distance to Maximum I-131 Deposition / Activity	2.25* km / 4.7E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 0.3 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 236 Ci the release fraction is 0.3 Ci / 236 Ci = 0.0013. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	3.50E-02	1	1	3.50E-02
Xe-133	54.45	1	1	54.45
Total Noble Gases	54.48			54.48
I-129	7.37E-08	1	0.8	5.90E-08
I-131	24.28	1	0.8	1.94E+01
I-133	54.45	1	0.8	4.36E+01
Total Iodines	78.73			62.98
Ba-140	52.20		0.0013	6.79E-02
Ce-144	9.13		0.0013	1.19E-02
Cs-137	2.45E-01		0.0013	3.19E-04
La-140	52.80		0.0013	6.86E-02
Nb-95	43.41		0.0013	5.64E-02
Ru-103	18.67		0.0013	2.43E-02
Ru-106	0.45		0.0013	5.80E-04
Sr-89	26.54		0.0013	3.45E-02
Sr-90	2.65E-01		0.0013	3.45E-04
Zr-95	32.09		0.0013	4.17E-02
U-235	1.79E-05		0.0013	2.33E-08

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
U-238	3.88E-04		0.0013	5.04E-07
U-234	3.88E-04		0.0013	5.04E-07
Pu	6.13E-02		0.0013	7.97E-05
Total Particulates	235.86			0.31

NOTES:

(1) "At 0815 on March 1, 1957, the alarm sounded which indicates a fuel element was leaking helium gas at an abnormal rate. A check of the helium system was made showing the fuel element in the south end of channel B-O-5 was leaking helium gas at a high rate.

At 0840 a stack air alarm was noted indicating the presence of gaseous radioactive contamination in the exit cooling air. A rise in the probe readings in the south duct of approximately 25% was noted as well as a gradual rise in the reading on the south monitor which indicates the presence of particulate radioactive contamination of the cooling air in the south exit duct.

A slow shutdown of the reactor was started at 0938 and was complete by 1120. All fuel elements which leaked at a rate too high to tolerate on the regular leak detection system were discharged. Readings of more than 2 R/hr proved the fuel element in the south end of the channel B-O-5 had ruptured.

Area surveys showed that no significant amounts of radioactive material passed through the filters."

(Reactor Operations Monthly Report, March 1957)

(2) "At about 0840 on March 1 evidence of a rupture was noted on the activity monitors. The activity was not increasing rapidly, so the pile was shutdown slowly. The shield was not opened until 1700 and the ruptured element was discharged soon after. Apparently the rupture had not progressed enough to release large quantities of particulate fission products. The job of removing the ruptured cartridge did not result in a particularly large amount of contamination."

(Memo, Boutelle to Gemmell, March 1957)

(3) 3/1/57 0907 South Probe 31-39 mR/hr.

(Reactor Operations Logbook)

Rupture Number 27

Class	Generic
Rupture Date	7/23/57
Rupture Time	0200
Wet Storage Tube Number	1058
Channel Number	N D-10-3
Discharge Day	2532
Charge Day	3
Number of Days in Reactor	2529
Average Power Level Over Period in Reactor and Total MWD	18.58 MW, 46,989 MWD
Probe Measurement	100 mR/hr
Derived Slug Equivalent	1
Wind Speed at the Ground	1.8 m/s
Wind Direction (towards)	S/SSW/SSE
Calculated Distance to Maximum I-131 Deposition / Activity	27A: 3.9 km / 1.9E+03 Bq/m^2 27B: 3.9 km / 9.5E+03 Bq/m^2 27C: 3.4 km / 7.2E+03 Bq/m^2

(1) The probe measurement was used to estimate that 0.78 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 345 Ci the release fraction is 0.78 Ci / 345 Ci = 0.0023. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was less than would be expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there was 1 slug equivalent.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 1 slug equivalent is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	8.62E-01	1	1	8.62E-01
Xe-133	54.45	1	1	54.45
Total Noble Gases	55.31			55.31
I-129	2.25E-06	1	0.8	1.80E-06
I-131	24.30	1	0.8	19.44
I-133	54.45	1	0.8	43.56
Total Iodines	78.75			63.00
Ba-140	52.80		0.0023	1.19E-01
Ce-144	50.46		0.0023	1.14E-01
Cs-137	6.92		0.0023	1.56E-02
La-140	52.80		0.0023	1.19E-01
Nb-95	53.60		0.0023	1.21E-01
Ru-103	24.30		0.0023	5.49E-02
Ru-106	3.03		0.0023	6.86E-03
Sr-89	40.20		0.0023	9.09E-02
Sr-90	7.49		0.0023	1.69E-02
Zr-95	53.60		0.0023	1.21E-01
U-235	1.79E-05		0.0023	4.05E-08
U-238	3.88E-04		0.0023	8.77E-07
U-234	3.88E-04		0.0023	8.77E-07
Pu	6.13E-02		0.0023	1.39E-04
Total Particulates	345.27			7.80E-01

NOTES:

(1) "During the morning of July 23, 1957 (0200 hours) the presence of slightly higher than normal amounts of radioactivity in the exhaust air system was indicated by the filter monitors...Despite the rather meager information that a fuel element had ruptured, the fact that several elements indicated high He leak rates led to the decision to shut the reactor down...The handling tool contamination increased by a factor of twenty when the element in channel ND-10-3 was removed, indicating that it had ruptured."

(Reactor Operations Monthly Report, July 1957)

(2) 7/23/57 0200 North Probe from 38 to 58 mR/hr.
0230 North Probe 58 mR/hr.
0540 North Probe 100 mR/hr.
0550 Slow shut down of the Reactor

(Reactor Operations Logbook)

Rupture Number 28

Class	Generic
Rupture Date	10/3/57
Rupture Time	0500
Channel Number	S B-6-5
Discharge Day	2604
Charge Day	1518
Number of Days in Reactor	1086
Average Power Level Over Period in Reactor and Total MWD	20.50 MW; 22,263 MWD
Probe Measurement	800 mR/hr
Derived Slug Equivalent	2
Wind Speed at the Ground	5.7 m/s
Wind Direction (towards)	SE
Calculated Distance to Maximum I-131 Deposition / Activity	2.5* km / 6.2E+03 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) The probe measurement was used to estimate that 6.2 Ci of particulates were emitted from the stack (section 4.3, Part II).

(2) Since the particulate inventory of this slug was 333 Ci the release fraction is 6.2 Ci / 333 Ci = 0.0186. The specific particulate radionuclide releases are partioned by this release fraction.

(3) The assumed particulate release fraction from one slug is .01 and since the particulate release fraction was nearly twice that expected from one slug, it was assumed in estimating the noble gas and radioiodine releases that there were 2 slug equivalents.

(4) For the noble gases, 100% and for the radioiodines, 80% of the inventory of 2 slug equivalents is assumed to have been released from the stack. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	4.19E-01	2	1	8.37E-01
Xe-133	54.45	2	1	1.09E+02
Total Noble Gases	54.87			109.74
I-129	9.65E-07	2	0.8	1.54E-06
I-131	24.30	2	0.8	3.89E+01
I-133	54.45	2	0.8	8.71E+01
Total Iodines	78.75			126.00
Ba-140	52.80		0.0186	9.82E-01
Ce-144	46.84		0.0186	8.71E-01
Cs-137	3.11		0.0186	5.78E-02
La-140	52.80		0.0186	9.82E-01
Nb-95	53.60		0.0186	9.97E-01
Ru-103	24.30		0.0186	4.52E-01
Ru-106	2.67		0.0186	4.97E-02
Sr-89	40.20		0.0186	7.48E-01
Sr-90	3.36		0.0186	6.25E-02
Zr-95	53.60		0.0186	9.97E-01
U-235	1.79E-05		0.0186	3.33E-07
U-238	3.88E-04		0.0186	7.22E-06
U-234	3.88E-04		0.0186	7.22E-06
Pu	6.13E-02		0.0186	1.14E-03
Total Particulates	333.35			6.20

NOTES:

(1) "Following the reloading operation, the reactor ran routinely for approximately four days when it was shut down due to evidence of fission products in the exhaust gas stream. Since the background activity was increasing slowly, a routine, 2 ° Celsius per minute, shutdown was carried out. As indicated by the Helium System, the offending element was found to be SB-6-5. Although this element was in the inner region of normal fuel elements, thermocouples in this region indicated temperatures well below operating limit."

(Reactor Operations Monthly Report, 9/20-10/30 1957)

(2) 10/3/57 0044 Probe 20 mR/hr.
1115 Probe 800 mR/hr.
1152 Reactor shut down

(Reactor Operations Logbook)

Rupture Number 29

Class	Not Applicable
Rupture Date	10/28/57
Rupture Time	1400
Channel Number	D-0-8
Discharge Day	10/28
Charge Day	9/29
Number of Days in Reactor	30
Average Power Level Over Period in Reactor	13.8 MW, 414 MWD
Probe Measurement	>2000 mR/hr
Derived Slug Equivalent	0.17
Wind Speed at the Ground	5.4 m/s
Wind Direction (towards)	ESE
Calculated Distance to Maximum I-131 Deposition / Activity	3.0* km / 5.8E+02 Bq/m^2

* Predicted maximum deposition was on the Laboratory site, value given is at the site boundary.

(1) This was the rupture of a U_3O_8 sample.
Rupture 29 was not a fuel-slug rupture, but that of a 225 gram sample of uranium oxide, U_3O_8. To enable a comparison with a slug rupture, the uranium content of the sample is required, i.e., there are 209 grams of uranium in 225 grams of U_3O_8. Two hundred and nine grams is about 17% of the amount of uranium in a slug which contains 1,175 grams of uranium. The Reactor Operations Log for October 1957 noted "...that all of its approximately 225 grams of U_3O_8 had been lost." If a release fraction of 1 is used for this rupture, and a 10% release through the filter about 2.7 Ci would have been released. Using the generic approach based on rupture 21, a probe reading of 400 mR/hr would have been expected. Since the Operations Logbook indicated a probe reading of greater than 2 R (2,000 mR/hr), this would have suggested a release of about 15 Ci of particulates. Since this rupture consisted of a complete release of the sample, the iodine release would have been greater than that used in the generic approach. Even with this relatively small addition to the probe reading, the overall result is that the generic approach is apparently overestimating the release by about a factor of 5, which seems quite appropriate for an approach, based on such little information and with such large uncertainties.

(2) For the noble gases, 100%, and, for the radioiodines, 100% of the inventory in this sample is assumed to have been released to the stack. For the particulates, 10% of the sample inventory is assumed to be released to the stack. For this rupture, the particulate inventory is multiplied by 0.1. The Table below presents the release information.

Radionuclide	Inventory (Ci, 1 Slug)	Slug Equivalents	Release Fraction	Curies Released
Kr-85	1.27E-02	0.17	1	2.16E-03
Xe-133	53.38	0.17	1	9.07
Total Noble Gases	53.39			9.08
I-129	2.66E-08	0.17	1	4.53E-09
I-131	22.46	0.17	1	3.82
I-133	54.45	0.17	1	9.26
Total Iodines	76.91			1.31E+01
Ba-140	42.35	0.17	0.1	7.20E-1
Ce-144	3.51	0.17	0.1	5.97E-2
Cs-137	8.87E-02	0.17	0.1	1.51E-3
La-140	52.80	0.17	0.1	8.98E-1
Nb-95	24.18	0.17	0.1	4.11E-1
Ru-103	9.97	0.17	0.1	1.70E-01
Ru-106	0.17	0.17	0.1	2.88E-2
Sr-89	12.98	0.17	0.1	2.21E-1
Sr-90	9.60E-02	0.17	0.1	1.63E-3
Zr-95	15.07	0.17	0.1	2.56E-1
U-235	1.79E-05	0.17	0.1	3.04E-7
U-238	3.88E-04	0.17	0.1	6.60E-6
U-234	3.88E-04	0.17	0.1	6.60E-6
Pu	6.13E-02	0.17	0.1	1.04E-3
Total Particulates	161.29			2.74

NOTES:

(1) "The Presence of fission products in the exhaust gas system was noted during the Day Shift on October 28. Although the indications were similar to those of a ruptured fuel element, a quickly conducted investigation of the U_3O_8 slug being irradiated for the Hot Lab's iodine production program, proved that it had ruptured. This slug was promptly pulled into the shield where it remained for the remainder of the operating period. When later removed from the shield, it was noted that all of its approximately 225 grams of U_3O_8 had been lost."

(Reactor Operations Monthly Report, 9/20-10/31 1957)

(2) 10/28/57 1446 North probe reading >2000 mR/hr.

(Reactor Operations Logbook)

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APPENDIX II-B

OAK RIDGE X-10 SOURCE-TERM ESTIMATES CHEMRISK (1993) APPENDIX E

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VOLUME II – PART A
DOSE RECONSTRUCTION FEASIBILITY STUDY

TASKS 1 & 2
A summary of Historical Activities
On the Oak Ridge Reservation with Emphasis on Information
Concerning Off-Site Emission of Hazardous Material

Prepared by
ChemRisk
A Division of Mclaren/Hart
for the
Tennessee Department of Health and the
Oak Ridge Health Agreement Steering Panel

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APPENDIX E

SOURCE TERM ESTIMATES FOR X-10

Estimates of quantities of radionuclides released to the air or available for release as a result of historical X-10 operations have been prepared for the following areas:

- Radioactive Lanthanum (RaLa) Processing
- Thorex Processing of Short-Decay Irradiated Thorium
- Chemical Separation of Plutonium from Clinton Pile Fuel
- Graphite Reactor Fuel Slug Ruptures
- Argon-41 from Graphite Reactor Cooling Air
- Tritium from Isotope Processing Programs

Each of these areas is discussed in this section, and estimated peak annual release quantities, emission rates, and predicted air concentrations for 18 radionuclides that have been assembled to support the screening process are presented in Table 5-1.

Emissions from Radioactive Lanthanum Separation Operations

The quantities of radionuclides that were available for release from ORNL processing of reactor fuel for separation of radioactive lanthanum (RaLa processing) were estimated based on the RaLa production information summarized in the Task 1 & 2 report and some assumptions and simple calculations. Table 2-7 in the Task 1 & 2 report presents data concerning the ORNL RaLa runs, including run dates, numbers of fuel slugs processed, curies of barium dissolved, curies (Ci) of barium shipped, and yield of the separation process. Complete information in all of these areas is not currently available for each RaLa run. In order to support the screening process, values for missing data were estimated based on the following relationships, which have been characterized based on the considerable data that are available:

- curies dissolved per slug
- curies shipped per slug
- recovery efficiency (Ci shipped ÷ Ci dissolved)

Values of these relationships were used to estimate the numbers of slugs processed and/or curies dissolved for RaLa runs for which such data have not yet been located. An average value of one of the above relationships, calculated over a period near in time and similar in nature of operations to each run with missing data, was used to fill in missing values. This similarity of operations is important because the curie content of the slugs used in RaLa processing increased significantly as supply shifted from ORNL graphite reactor slugs to four-inch Hanford slugs and later included eight-inch Hanford slugs.

With the estimates in place, the magnitude of ORNL RaLa processing over the period from 1944 to 1956 can be summarized as follows:

Number of Slugs Processed:	34,000
Curies of Barium Dissolved:	1,300,000
Curies of Barium Shipped:	560,000

The quantities of barium shipped were measured near the time of final separation of lanthanum, and therefore do not include a significant contribution from lanthanum-140.

The amounts of the selected fission products that were available in each graphite reactor slug used for RaLa processing in 1947 were estimated based on a neutron flux of 1 x 10^{12} neutrons/cm^2-sec, an irradiation period of 40 days, and a cooling period of 1 day after removal from the reactor. The fission product content of each slug was estimated using the following equation:

$$A_i = (1\times10^{12}\ n/cm^2\text{-sec})(577\times10^{-24}cm^2)(N)(yield_i)(1-e^{-\lambda_i t_{irr}})(e^{-\lambda_i t_{cool}})(2.703\times10^{-11}\ Ci/atom\text{-sec})$$

where:

A_i	=	activity of radionuclide i in each fuel slug (Ci)
1 x 10^{12} n/cm^2-sec	=	maximum graphite reactor flux
577 x 10^{-24}	=	fission cross section for uranium-235
N	=	number of U-235 atoms per slug
$yield_i$	=	fission yield of radionuclide i for uranium-235
λ_i	=	decay constant of radionuclide i (sec^{-1})
t_{irr}	=	irradiation time in reactor (sec)
t_{cool}	=	cooling time after removal from reactor (sec)
2.703 x 10^{-11} Ci/atom-sec	=	conversion from atoms/sec to curies

A cross section is a probability that a certain reaction will occur between a nucleus and an incident particle or photon; in this case, the probability that an incident neutron will cause a U-235 atom to fission. The radioactivity content of each slug was multiplied times an estimated 9300 slugs processed in 1947 to estimate the total radionuclide inventory in processed fuel for that year.

Release fractions were applied to radionuclide inventories to estimate quantities released. The following release fractions were used:

- Noble Gases 100%
- Iodine 80%
- Particulates (i.e., others) 0.1%

The noble gas release fraction of 100% is based on the nonreactive nature of xenon and krypton. The release fraction for iodine is based on analyses of iodine release fractions at the Hanford plant performed as part of the Hanford dose reconstruction project. The release fraction for

particulate radionuclides is based on measured particulate emissions from RaLa processing at the Idaho Chemical Processing Plant during 1957 compared to the estimated radionuclide inventories in the materials testing reactor (MTR) fuel used as the barium source at that plant.

The plutonium content of the graphite reactor slugs in 1947 was estimated based on a plutonium formation rate of 36.5 micrograms per kilowatt-hour of reactor exposure obtained from graphite reactor operations reports. The fission rate corresponding to the neutron flux stated earlier was converted to a reactor exposure over 40 days (in kilowatt-hours) and multiplied times the 36.5 microgram Pu/kW-hr value to yield the micrograms of plutonium formed per slug over 40 days of exposure. A specific activity value of 0.0613 Ci/g was used to convert that mass to its curie equivalent. A release fraction of 0.1% was applied to estimate plutonium emissions.

Uranium emissions were estimated based on 2.6 pounds of natural uranium per slug, an isotopic composition of 99.276% uranium-238 and 0.71% uranium-235, and specific activity values of 3.3×10^{-7} Ci/g for uranium-238 and 2.14×10^{-6} Ci/g for uranium-235. A release fraction of 0.1% was applied to the quantities of the uranium isotopes to estimate releases to the atmosphere. Release estimates for 1947 are shown in Table E-1.

Radionuclide emissions for Oak Ridge RaLa processing of Hanford slugs during 1952 were estimated using the same method as above, with the following differences:

- a fission rate of 1.26×10^{14} fissions/sec-slug was calculated based on a power level of 2.25 watts/gram
- reactor irradiation time was 80 days
- cooling time was 5 days
- slug mass was 1800 grams
- an estimated total of 1300 slugs were dissolved

Release estimates for RaLa processing in 1952 are show in Table E-2.

Emissions from Thorex Short-Decay Runs

Quantities of radionuclides available in the processing of short-decayed (20-60 days of decay) irradiated thorium that occurred in 1956 and 1957 were estimated based on documented characteristics of the material that was dissolved. Quantities of thorium dissolved in the four short-decay runs are documented by McDuffee (1957) and McDuffee and Yarbro (1958). A 1957 memorandum by W.L. Albrecht documented the activities of protactinium-233 (Pa-233) and fission products in thorium receiving irradiation of the extent documented for the short-decay feed material. Data derived from the Albrecht memo are shown in Table E-3. Pa-233, an activation product of thorium-232 and the parent of uranium-233, was by far the most prominent radionuclide present. After 30 days of decay, each kilogram of irradiated thorium metal that was processed contained over 14,000 curies of Pa-233.

Quantities of Pa-233 and fission products available for each of the 14 dissolving batches of Thorex Runs HD-19, SD-1, SD-2, and SD-3 were estimated by multiplying the quantity of thorium metal dissolved in each batch by the curie content of each kilogram of metal based on the Albrecht data. Reductions were made in the quantities estimated to have been available for

TABLE E-1

ESTIMATED EMISSIONS FROM X-10 RaLa PROCESSING OF X-10 SLUGS IN 1947

Nuclide	Half-Life (seconds)	Decay Constant (sec^{-1})	Fission Yield	Ci/slug at time t	Total Ci Available	Release Fraction	Release Total (Ci)
I-131	$6.96 \times 10^{+5}$	9.96×10^{-7}	2.90×10^{-2}	$8.63 \times 10^{+6}$	$8.03 \times 10^{+6}$	80%	$6.42 \times 10^{+6}$
I-132	$8.14 \times 10^{+3}$	8.52×10^{-5}	4.40×10^{-2}	$9.39 \times 10^{+3}$	$8.73 \times 10^{+1}$	80%	$6.98 \times 10^{+1}$
I-133	$7.31 \times 10^{+4}$	9.48×10^{-6}	6.50×10^{-2}	$9.60 \times 10^{+6}$	$8.93 \times 10^{+6}$	80%	$7.14 \times 10^{+6}$
I-129	$5.36 \times 10^{+14}$	1.29×10^{-15}	1.00×10^{-2}	1.50×10^{-6}	1.39×10^{-6}	80%	1.11×10^{-6}
Ce-144	$2.45 \times 10^{+7}$	2.82×10^{-8}	6.10×10^{-2}	$1.90 \times 10^{+6}$	$1.76 \times 10^{+6}$	0.1%	$1.76 \times 10^{+1}$
Cs-137	$9.46 \times 10^{+8}$	7.32×10^{-10}	5.90×10^{-2}	5.00×10^{-2}	$4.65 \times 10^{+2}$	0.1%	4.65×10^{-1}
Kr-85	$3.39 \times 10^{+8}$	2.04×10^{-9}	3.00×10^{-3}	7.07×10^{-3}	$6.58 \times 10^{+1}$	100%	$6.58 \times 10^{+1}$
Xe-133	$4.55 \times 10^{+5}$	1.52×10^{-6}	6.50×10^{-2}	$1.90 \times 10^{+1}$	$1.77 \times 10^{+5}$	100%	$1.77 \times 10^{+5}$
Zr-95	$5.67 \times 10^{+6}$	1.22×10^{-7}	6.40×10^{-2}	$7.31 \times 10^{+6}$	$6.80 \times 10^{+4}$	0.1%	$6.80 \times 10^{+1}$
Nb-95	$3.02 \times 10^{+6}$	2.29×10^{-7}	6.40×10^{-2}	$1.15 \times 10^{+1}$	$1.07 \times 10^{+5}$	0.1%	$1.07 \times 10^{+2}$
Ru-103	$3.41 \times 10^{+6}$	2.03×10^{-7}	2.90×10^{-2}	$4.82 \times 10^{+6}$	$4.48 \times 10^{+4}$	0.1%	$4.48 \times 10^{+1}$
Ru-106	$3.18 \times 10^{+7}$	2.18×10^{-8}	3.80×10^{-3}	9.22×10^{-2}	$8.58 \times 10^{+2}$	0.1%	8.58×10^{-1}
Sr-89	$4.55 \times 10^{+6}$	1.52×10^{-7}	4.80×10^{-2}	$6.49 \times 10^{+6}$	$6.04 \times 10^{+4}$	0.1%	$6.04 \times 10^{+1}$
Sr-90	$8.74 \times 10^{+8}$	7.93×10^{-10}	5.80×10^{-2}	5.32×10^{-2}	$4.95 \times 10^{+2}$	0.1%	4.95×10^{-1}
Ba-140	$1.11 \times 10^{+6}$	6.27×10^{-7}	6.30×10^{-2}	$1.77 \times 10^{+1}$	$1.65 \times 10^{+5}$	0.1%	$1.65 \times 10^{+2}$
La-140	$1.45 \times 10^{+5}$	4.79×10^{-6}	6.30×10^{-2}	$1.40 \times 10^{+1}$	$1.30 \times 10^{+5}$	0.1%	$1.30 \times 10^{+2}$
Pu	$7.69 \times 10^{+11}$*	9.01×10^{-13}*	NA	8.54×10^{-4}	$7.94 \times 10^{+0}$	0.1%	7.94×10^{-3}
U-235	$2.24 \times 10^{+16}$	3.10×10^{-17}	NA	1.79×10^{-5}	1.66×10^{-1}	0.1%	1.66×10^{-4}
U-238	$1.42 \times 10^{+17}$	4.87×10^{-18}	NA	3.90×10^{-4}	$3.63 \times 10^{+0}$	0.1%	3.63×10^{-3}

NA = Not Applicable

* Value is for plutonium-239

TABLE E-2

ESTIMATED EMISSIONS FROM X-16 RaLa PROCESSING OF HANFORD SLUGS IN 1952

Nuclide	Half-Life (seconds)	Decay Constant (sec^{-1})	Fission Yield	Ci/slug at time t	Total Ci Available	Release Fraction	Release Total (Ci)
I-131	$6.96 \times 10^{+5}$	9.96×10^{-7}	2.90×10^{-2}	$6.41 \times 10^{+1}$	$8.34 \times 10^{+4}$	80%	$6.67 \times 10^{+4}$
I-132	$8.14 \times 10^{+3}$	8.52×10^{-5}	4.40×10^{-2}	$1.57 \times 10^{+14}$	$2.04 \times 10^{+11}$	80%	$1.63 \times 10^{+11}$
I-133	$7.31 \times 10^{+4}$	9.48×10^{-6}	6.50×10^{-2}	$3.58 \times 10^{+0}$	$4.79 \times 10^{+3}$	80%	$3.83 \times 10^{+3}$
I-129	$5.36 \times 10^{+14}$	1.29×10^{-15}	1.00×10^{-2}	3.04×10^{-7}	3.95×10^{-4}	80%	3.16×10^{-4}
Ce-144	$2.45 \times 10^{+7}$	2.82×10^{-8}	6.10×10^{-2}	$3.64 \times 10^{+1}$	$4.73 \times 10^{+4}$	0.1%	$4.73 \times 10^{+1}$
Cs-137	$9.46 \times 10^{+8}$	7.32×10^{-10}	5.90×10^{-2}	$1.01 \times 10^{+0}$	$1.32 \times 10^{+3}$	0.1%	$1.32 \times 10^{+0}$
Kr-85	$3.39 \times 10^{+8}$	2.04×10^{-9}	3.00×10^{-3}	1.43×10^{-1}	$1.86 \times 10^{+2}$	100%	$1.86 \times 10^{+2}$
Xe-133	$4.55 \times 10^{+5}$	1.52×10^{-6}	6.50×10^{-2}	$1.15 \times 10^{+2}$	$1.49 \times 10^{+5}$	100%	$1.49 \times 10^{+5}$
Zr-95	$5.67 \times 10^{+6}$	1.22×10^{-7}	6.40×10^{-2}	$1.18 \times 10^{+2}$	$1.53 \times 10^{+5}$	0.1%	$1.53 \times 10^{+2}$
Nb-95	$3.02 \times 10^{+6}$	2.29×10^{-7}	6.40×10^{-2}	$1.57 \times 10^{+2}$	$2.04 \times 10^{+5}$	0.1%	$2.04 \times 10^{+2}$
Ru-103	$3.41 \times 10^{+6}$	2.03×10^{-7}	2.90×10^{-2}	$6.82 \times 10^{+1}$	$8.87 \times 10^{+4}$	0.1%	$8.87 \times 10^{+1}$
Ru-106	$3.18 \times 10^{+7}$	2.18×10^{-8}	3.80×10^{-3}	$1.79 \times 10^{+0}$	$2.33 \times 10^{+3}$	0.1%	$2.33 \times 10^{+0}$
Sr-89	$4.55 \times 10^{+6}$	1.52×10^{-7}	4.80×10^{-2}	$9.96 \times 10^{+1}$	$1.29 \times 10^{+5}$	0.1%	$1.29 \times 10^{+2}$
Sr-90	$8.74 \times 10^{+8}$	7.93×10^{-10}	5.80×10^{-2}	$1.08 \times 10^{+0}$	$1.40 \times 10^{+3}$	0.1%	$1.40 \times 10^{+0}$
Ba-140	$1.11 \times 10^{+6}$	6.27×10^{-7}	6.30×10^{-2}	$1.62 \times 10^{+2}$	$2.10 \times 10^{+5}$	0.1%	$2.10 \times 10^{+2}$
La-140	$1.45 \times 10^{+5}$	4.79×10^{-6}	6.30×10^{-2}	$2.71 \times 10^{+1}$	$3.53 \times 10^{+4}$	0.1%	$3.53 \times 10^{+1}$
Pu	$7.69 \times 10^{+11}$ a	9.01×10^{-13} a	NA	1.74×10^{-2}	$2.26 \times 10^{+1}$	0.1%	2.26×10^{-2}
U-235	$2.24 \times 10^{+16}$	3.10×10^{-17}	NA	2.73×10^{-5}	3.55×10^{-2}	0.1%	3.55×10^{-5}
U-238	$1.42 \times 10^{+17}$	4.87×10^{-18}	NA	5.95×10^{-4}	7.74×10^{-1}	0.1%	7.74×10^{-4}

A = Not Applicable

a Value is for plutonium-239

TABLE E-3

FISSION PRODUCT AND PROTACTINIUM-233 CONTENT OF SHORT-DECAY IRRADIATED THORIUM

Radionuclide	Ci per kg of Thorium after 30 d of Decay
Total Fission Products	340
Kr-85	0.12
Zr-95	72
Nb-95	68
Ru-103	9.0
Ru-106	0.90
I-131	5.0
I-132 (Te-132)	0.17
Xe-133	3.1
Ba-140/La-140	54
Ce-141	54
Ce-144	14
Pa-233	14,000

Reference: Albrecht, 1957.

batch HD-19-A to account for an irradiation level of 3300 grams Mass-233 per metric ton of thorium instead of the 4000 g/t value that was the basis of the Albrecht data and for a decay period of 109 days instead of 30. Reductions were made in the quantities estimated to have been available for batches HD-19-B and -C to account for irradiation levels of 1910 grams Mass-233 per metric ton of thorium instead of the 4000 g/t value that was the basis of the Albrecht data.

Quantities of uranium-233 that were contained in the dissolved metal were estimated by multiplying the kilograms of uranium reported to have been dissolved in each batch by 9.48, the number of curies of U-233 per kilogram of U-233.

Release fractions of 100%, 80% and 0.1% were applied to noble gases, iodine and particulates, respectively. Estimated quantities of radionuclides that were released in the course of the Thorex short-decay processing of thorium metal are shown in Table E-4. Available data appear to indicate that calendar year 1957, due to processing of short-decay thorium in the Thorex pilot plant, was the period of peak airborne emissions of Pa-233 from the Oak Ridge Reservation.

Emissions from Chemical Separation of Plutonium from Clinton Pile Fuel

Estimates of quantities of plutonium, uranium, and fission products available in the course of early processing of graphite reactor fuel slugs for recovery of fissionable plutonium were prepared based on material processing rates, estimated process efficiencies, and rates of production of plutonium and fission products in the natural uranium fuel slugs.

The chemical processing pilot plant operated full scale from January 1944 until production ended in January 1945 (Jones, 1985). The bismuth phosphate process was used to recover 326.4 grams of plutonium (Johnson and Schaffer, 1992). The efficiency of separation of plutonium from fission products was improved from 40% to 90% (Jones, 1985). Taking the average plutonium recovery efficiency to be 65% (the midpoint of 40% and 90%), the total amount of plutonium processed was estimated to have been $326.4 \div 0.65 = 502$ grams. Based on a specific activity of 0.0613 Ci/g, this corresponds to 30.8 curies of plutonium.

Given that the pile first went critical on November 4th, 1943 and that chemical processing involved one-third ton of uranium per day by late January 1944 (Thompson, 1963), it appears that decay periods for the slugs processed early in the campaign could not have been very long. A semi-monthly progress report issued in August 1944 indicated that slugs involved in recent dissolvings had been approximately 60 days old (Leverett, 1944). A decay period of 30 days was selected for the purposes of screening calculations.

The fission rate per ton of uranium processed was estimated based on a neutron flux of 5×10^{11} neutrons/cm^2-sec. The radionuclide content of each ton of uranium processed was estimated using the equation given in the beginning of this appendix, with that fission rate substituted for the first three terms on the right hand side, an irradiation time of 40 days, and a cooling period of 30 days. These quantities were multiplied times 0.3 ton per day processed times 365 days to yield the totals of each radionuclide processed.

TABLE E-4

ESTIMATED RADIONUCLIDE EMISSIONS
ORNL THOREX SHORT-DECAY RUNS
(July 1956 through November 1957)

Batch	Metal Dissolved (kilograms)		Activation Products Available (Ci)	
	Th	U	U-233	Pa-233
HD-19-A	239.3	0.79	$7.49 \times 10^{+0}$	$3.75 \times 10^{+5}$
HD-19-B	351.8	0.673	$6.38 \times 10^{+0}$	$2.42 \times 10^{+6}$
HD-19-C	30.8	0.059	5.59×10^{-1}	$2.12 \times 10^{+5}$
SD-1-A	382.7	0.926	$8.81 \times 10^{+0}$	$5.52 \times 10^{+6}$
SD-1-B	335.7	0.422	$4.00 \times 10^{+0}$	$4.84 \times 10^{+6}$
SD-1-C	16.3	0.025	2.37×10^{-1}	$2.35 \times 10^{+5}$
SD-2-A	436.2	1.481	$1.40 \times 10^{+1}$	$6.32 \times 10^{+6}$
SD-2-B	261.7	0.783	$7.42 \times 10^{+0}$	$3.77 \times 10^{+6}$
SD-2-C	264.3	8.38	$7.94 \times 10^{+0}$	$3.81 \times 10^{+6}$
SD-2-D	331.4	9.15	$8.67 \times 10^{+0}$	$4.78 \times 10^{+6}$
SD-2-E	161.6	0.502	$4.76 \times 10^{+0}$	$2.33 \times 10^{+6}$
SD-3-A	324	0.834	$7.91 \times 10^{+0}$	$4.67 \times 10^{+6}$
SD-3-B	301.4	0.768	$7.28 \times 10^{+0}$	$4.34 \times 10^{+6}$
SD-3-C	129.1	0.331	$3.14 \times 10^{+0}$	$1.86 \times 10^{+6}$
1956 Total	622	1.52	$1.44 \times 10^{+1}$	$3.01 \times 10^{+6}$
1957 Total	2,946	7.83	$7.42 \times 10^{+1}$	$4.25 \times 10^{+7}$
TOTAL	3,568	9.35	$8.86 \times 10^{+1}$	$4.55 \times 10^{+7}$
Release Fraction			0.1%	0.1%
1957 Emissions (Ci):			7.42×10^{-2}	$4.25 \times 10^{+4}$

TABLE E-4
(CONTINUED)

ESTIMATED RADIONUCLIDE EMISSIONS
ORNL THOREX SHORT-DECAY RUNS
(July 1956 through November 1957)

Batch	Fission Products Available (Ci)									
	Zr-95	Nb-95	Ba/La-140	Ce-141	Ce-144	Ru-103	I-131	Xe-133	Ru-106	Kr-85
HD-19-A	6.18×10^{-3}	2.79×10^{-3}	1.48×10^{-2}	1.98×10^{-3}	2.20×10^{-4}	4.45×10^{-2}	1.09×10^{-0}	1.89×10^{0}	1.53×10^{-2}	2.28×10^{-1}
HD-19-B	1.21×10^{-2}	1.14×10^{-2}	9.08×10^{-3}	9.08×10^{-3}	2.27×10^{-3}	1.51×10^{-2}	8.32×10^{-2}	5.22×10^{-2}	1.51×10^{-2}	1.97×10^{-1}
HD-19-C	1.06×10^{-2}	9.94×10^{-3}	7.95×10^{-3}	7.95×10^{-3}	1.99×10^{-3}	1.32×10^{-2}	7.29×10^{-1}	4.57×10^{-1}	1.32×10^{-1}	1.72×10^{-0}
SD-1-A	2.76×10^{-2}	2.59×10^{-2}	2.07×10^{-2}	2.07×10^{-2}	5.17×10^{-3}	3.45×10^{-2}	1.90×10^{-2}	1.19×10^{-2}	3.45×10^{-3}	4.48×10^{-1}
SD-1-B	2.42×10^{-2}	2.27×10^{-2}	1.81×10^{-2}	1.81×10^{-2}	4.54×10^{-3}	3.02×10^{-2}	1.66×10^{-2}	1.04×10^{-2}	3.02×10^{-3}	3.93×10^{-1}
SD-1-C	1.17×10^{-2}	1.10×10^{-2}	8.81×10^{-3}	8.81×10^{-3}	2.20×10^{-3}	1.47×10^{-2}	8.08×10^{-1}	5.07×10^{-1}	1.47×10^{-1}	1.91×10^{-0}
SD-2-A	3.16×10^{-2}	2.96×10^{-2}	2.37×10^{-2}	2.37×10^{-2}	5.92×10^{-3}	3.95×10^{-2}	2.17×10^{-2}	1.36×10^{-2}	3.95×10^{-3}	5.13×10^{-1}
SD-2-B	1.89×10^{-2}	1.77×10^{-2}	1.41×10^{-2}	1.41×10^{-2}	3.54×10^{-3}	2.36×10^{-2}	1.30×10^{-2}	8.13×10^{-3}	2.36×10^{-3}	3.06×10^{-1}
SD-2-C	1.90×10^{-2}	1.79×10^{-2}	1.43×10^{-2}	1.43×10^{-2}	3.57×10^{-3}	2.38×10^{-2}	1.31×10^{-2}	8.21×10^{-3}	2.38×10^{-3}	3.10×10^{-1}
SD-2-D	2.39×10^{-2}	2.24×10^{-2}	1.79×10^{-2}	1.79×10^{-2}	4.48×10^{-3}	2.99×10^{-2}	1.64×10^{-2}	1.03×10^{-2}	2.99×10^{-3}	3.88×10^{-1}
SD-2-E	1.16×10^{-2}	1.09×10^{-2}	8.74×10^{-3}	8.74×10^{-3}	2.18×10^{-3}	1.46×10^{-2}	8.01×10^{-2}	5.02×10^{-2}	1.46×10^{-3}	1.89×10^{-1}
SD-3-A	2.34×10^{-2}	2.19×10^{-2}	1.75×10^{-2}	1.75×10^{-2}	4.38×10^{-3}	2.92×10^{-2}	1.61×10^{-2}	1.01×10^{-2}	2.92×10^{-3}	3.79×10^{-1}
SD-3-B	2.17×10^{-2}	2.04×10^{-2}	1.63×10^{-2}	1.63×10^{-2}	4.07×10^{-3}	2.72×10^{-2}	1.49×10^{-2}	9.37×10^{-3}	2.72×10^{-3}	3.53×10^{-1}
SD-3-C	9.30×10^{-3}	8.72×10^{-3}	6.98×10^{-3}	6.98×10^{-3}	1.74×10^{-3}	1.16×10^{-2}	6.40×10^{-2}	4.01×10^{-2}	1.16×10^{-3}	1.51×10^{-1}
1956 Total	1.93×10^{-2}	1.51×10^{-2}	1.03×10^{-2}	1.19×10^{-2}	4.67×10^{-3}	2.09×10^{-2}	9.06×10^{-2}	5.68×10^{-2}	3.18×10^{-3}	4.42×10^{-1}
1957 Total	2.12×10^{-3}	1.99×10^{-3}	1.59×10^{-3}	1.59×10^{-3}	3.98×10^{-4}	2.65×10^{-4}	1.46×10^{-0}	9.16×10^{-3}	2.65×10^{-3}	3.45×10^{-3}
TOTAL	2.32×10^{-2}	2.14×10^{-2}	1.69×10^{-2}	1.71×10^{-2}	4.45×10^{-3}	2.86×10^{-2}	1.55×10^{-0}	9.73×10^{-2}	2.97×10^{-3}	3.89×10^{-1}
Release Fraction	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%	80%	100%	0.1%	100%
1957 Emissions (Ci):	2.12×10^{-3}	1.99×10^{-3}	1.59×10^{-3}	1.59×10^{-3}	3.98×10^{-4}	2.65×10^{-4}	1.17×10^{-0}	9.16×10^{-3}	2.65×10^{-3}	3.45×10^{-3}

The amount of uranium available was estimated to be 0.3 tons per day times 365 days, or 219,000 pounds. This amount of natural uranium was estimated to be 0.71% U-235 and 99.28% U-238 by weight, yielding totals of 1.5 and 210 curies of uranium-235 and uranium-238 available, respectively.

Release fractions of 100%, 80%, and 0.1% were applied to inventories of noble gases, iodine, and particulates available, respectively, to estimate quantities released to the atmosphere. Estimated quantities of radionuclides that were released in the course of pilot plant chemical separation of plutonium are shown in Table E-5. Available data appear to indicate that calendar year 1944, due to processing of graphite reactor fuel for chemical separation of plutonium, was the period of peak airborne emissions of iodine-129, cerium-144, cesium-137, zirconium-95, niobium-95, ruthenium-103, ruthenium-106, strontium-89, strontium-90, plutonium, uranium-235, and uranium-238 from the Oak Ridge Reservation.

Emissions from Graphite Reactor Fuel Slug Ruptures

The quantities of uranium, plutonium, and fission products released as a result of ruptures of the aluminum cans which encased graphite reactor fuel slugs were estimated. The natural uranium metal that comprised these slugs oxidized upon contact with air, and uranium oxide particles and liberated fission products in pile exhaust air went unfiltered from 1944 to 1948. Fifty slug rupture events from 1944 through 1948 were documented by Cagle and Emlet in 1948. Data available concerning the slugs that ruptured include position in the reactor (row, position in row, radial coordinate), date charged to the reactor, date ruptured, total age in days, accumulated kilowatt-hours of exposure, and temperature zone.

The average neutron flux in the graphite reactor was reportedly 5.0×10^{11} neutrons per cubic centimeter per second, and each fuel slug contained approximately 1175 grams of natural uranium metal (Rupp and Cox, 1955). With natural uranium being 0.71% U-235 by weight, each slug contained 2.15×10^{22} U-235 atoms. Based on a U-235 fission cross-section of 577 barns (577×10^{-24} cm^2), the average graphite reactor neutron flux resulted in 6.2×10^{12} fissions per second in each slug.

The fission product content of each slug that ruptured was estimated based on the fission rate derived above and the length of time the slug had spent in the reactor. The age of each slug, in hours, was estimated by dividing the reported accumulated kilowatt-hours of reactor exposure by 3500 kilowatts, the average reactor power level. The fission product content of the slug at the time of rupture was then calculated based on the fission rate, the fission yield of each fission product nuclide, and the rates of decay of each fission product after it was formed using the equation shown earlier in this appendix. All slug rupture events were assumed to have involved single slugs, except for the events of November 30, 1947 and August 25, 1948, which involved 13 and 5 slugs, respectively (Cagle and Emlet, 1948). Reports indicate that "much" of the released uranium oxide fell to the water-filled canal below the reactor air outlet (Emlet, 1947; Cagle and Emlet, 1948). No data or information was located to support a release fraction for particulates from slug ruptures. For the purposes of screening calculations, 10% of the particulate fission product activities present in each slug at the time of rupture were assumed to be released when the uranium oxidized based on professional judgement. Release fractions of 100% and 80% were applied to noble gas and iodine inventories, respectively.

TABLE E-5

ESTIMATED RADIONUCLIDE EMISSIONS
CLINTON LABORATORIES CHEMICAL SEPARATION OF PLUTONIUM

Nuclide	Half-Life (seconds)	Decay Constant (sec^{-1})	Fission Yield	Ci/Ton at End of Cooling	Total Ci Processed	Release Fraction	Release Total (Ci)
I-131	$6.96 \times 10^{+5}$	9.96×10^{-7}	0.029	$2.70 \times 10^{+2}$	$2.95 \times 10^{+4}$	80%	$2.36 \times 10^{+4}$
I-132	$8.14 \times 10^{+3}$	8.38×10^{-5}	0.044	2.73×10^{-91}	2.99×10^{-89}	80%	2.39×10^{-89}
I-133	$7.31 \times 10^{+4}$	9.48×10^{-6}	0.065	1.75×10^{-7}	1.91×10^{-5}	80%	1.53×10^{-5}
I-129	$5.36 \times 10^{+14}$	1.27×10^{-15}	0.010	5.58×10^{-4}	6.11×10^{-4}	80%	4.89×10^{-4}
Ce-144	$2.45 \times 10^{+7}$	2.78×10^{-8}	0.061	$6.60 \times 10^{+2}$	$7.23 \times 10^{+4}$	0.1%	$7.23 \times 10^{+1}$
Cs-137	$9.46 \times 10^{+8}$	7.20×10^{-10}	0.059	$1.86 \times 10^{+1}$	$2.04 \times 10^{+3}$	0.1%	$2.04 \times 10^{+0}$
Kr-85	$3.39 \times 10^{+8}$	2.01×10^{-9}	0.003	$2.62 \times 10^{+0}$	$2.87 \times 10^{+2}$	100%	$2.87 \times 10^{+2}$
Xe-133	$4.55 \times 10^{+5}$	1.50×10^{-6}	0.065	$1.70 \times 10^{+2}$	$1.86 \times 10^{+4}$	100%	$1.86 \times 10^{+4}$
Zr-95	$5.67 \times 10^{+6}$	1.20×10^{-7}	0.064	$2.02 \times 10^{+3}$	$2.22 \times 10^{+5}$	0.1%	$2.22 \times 10^{+2}$
Nb-95	$3.02 \times 10^{+6}$	2.25×10^{-7}	0.064	$2.45 \times 10^{+3}$	$2.69 \times 10^{+5}$	0.1%	$2.69 \times 10^{+2}$
Ru-103	$3.41 \times 10^{+6}$	2.03×10^{-7}	0.029	$1.10 \times 10^{+3}$	$1.20 \times 10^{+5}$	0.1%	$1.20 \times 10^{+2}$
Ru-106	$3.18 \times 10^{+7}$	2.14×10^{-8}	0.004	$3.26 \times 10^{+1}$	$3.57 \times 10^{+3}$	0.1%	$3.57 \times 10^{+0}$
Sr-89	$4.55 \times 10^{+6}$	1.50×10^{-7}	0.048	$1.67 \times 10^{+3}$	$1.83 \times 10^{+5}$	0.1%	$1.83 \times 10^{+2}$
Sr-90	$8.74 \times 10^{+8}$	7.80×10^{-10}	0.058	$1.98 \times 10^{+1}$	$2.17 \times 10^{+3}$	0.1%	$2.17 \times 10^{+0}$
Ba-140	$1.11 \times 10^{+6}$	6.16×10^{-7}	0.063	$1.43 \times 10^{+3}$	$1.56 \times 10^{+5}$	0.1%	$1.56 \times 10^{+2}$
La-140	$1.45 \times 10^{+5}$	4.71×10^{-6}	0.063	4.02×10^{-2}	$4.40 \times 10^{+0}$	0.1%	4.40×10^{-3}
Pu	$7.69 \times 10^{+11}$ •	9.01×10^{-12} •	NA	NA	$3.08 \times 10^{+1}$	0.1%	3.08×10^{-2}
U-235	$2.24 \times 10^{+16}$	3.10×10^{-17}	NA	NA	$1.51 \times 10^{+0}$	0.1%	1.51×10^{-3}
U-238	$1.42 \times 10^{+17}$	4.87×10^{-18}	NA	NA	$2.11 \times 10^{+2}$	0.1%	2.11×10^{-1}

NA = Not Applicable

• Value is for plutonium-239

Quantities of plutonium available from the ruptured slugs were estimated based on there being an average of 60.5 grams of plutonium present per ton in uranium irradiated for 1000 days or more (Emlet, 1947). This concentration applied to the mass of uranium liberated from ruptured slugs yielded an estimate of plutonium available from each event. Quantities of uranium available were estimated based on the number of slugs that ruptured and the mass (2.6 pounds) and composition of the natural uranium (0.71% U-235 and 99.276% U-238) that each slug contained.

With the multiple-slug ruptures in November, 1947 appears to be the year in which emissions from ruptured slugs would have been the greatest. In November 1948, the graphite reactor filter house went into operation. While slug ruptures continued past 1948 (there were 41 in 1956 (Seagren and Cox, 1957)), emissions of particulate radionuclides were substantially decreased by the filters, and non-filterable emissions do not appear to have approached the magnitude of other operations which are being evaluated in the screening process.

Estimated quantities of radionuclides that were released from slug ruptures in the graphite reactor in 1947 are shown in Table E-6. Available data appear to indicate that slug ruptures were not the most significant airborne emission source for any of the identified radionuclides. Ten of the radionuclides included in the assessment of slug rupture emissions could be elevated to roughly the magnitude of the current most significant airborne emission source of the nuclide in question if the particulate release fraction were to increase significantly from the 10% used in the screening calculations. The following values of particulate release fraction would be required for emissions of the identified radionuclides from graphite reactor slug ruptures in 1947 to rival the most significant emissions of that nuclide:

cesium-137	15%
strontium-90	15%
plutonium	26%
ruthenium-106	30%
cerium-144	34%
lanthanum-140	50%
barium-140	81%
zirconium-95	89%
strontium-89	96%
niobium-95	100%

Emissions of Argon-41 in Graphite Reactor Cooling Air

Ar-41 was created by neutron activation of stable argon-40 in graphite reactor cooling air. The release rate of Ar-41 from the graphite reactor stack was estimated to be 470 curies per day when the pile was operated at a power level of 3.6 megawatts (Morgan, 1949). The graphite reactor operated from November 1943 to November 1963, and annual emissions are [illegible] to have varied significantly from the corresponding annual emission of 172,000 curie.

TABLE E-6

ESTIMATED RELEASES FROM OAK RIDGE GRAPHITE REACTOR SLUG RUPTURES

Date	KWh in Rx	Slugs Rel'd	I-131	I-133	I-129	Ce-144	Cs-137	Kr-85	Xe-133	Zr-95	Nb-95
Feb-47	$6.8 \times 10^{+7}$	1	4.86	10.89	1.51×10^{-7}	8.80	0.49	6.69×10^{-2}	10.89	10.72	10.72
Feb-47	$2.6 \times 10^{+7}$	1	4.86	10.89	5.79×10^{-8}	5.42	0.19	2.67×10^{-2}	10.89	10.32	10.70
Apr-47	$9.4 \times 10^{+5}$	1	3.01	10.89	2.09×10^{-9}	0.28	0.01	9.92×10^{-4}	8.39	1.19	2.13
Aug-47	$5.6 \times 10^{+6}$	1	4.84	10.89	1.26×10^{-8}	1.54	0.04	5.92×10^{-3}	10.89	5.44	7.89
Oct-47	$9.0 \times 10^{+7}$	1	4.86	10.89	1.99×10^{-7}	9.46	0.64	8.62×10^{-2}	10.89	10.72	10.72
Oct-47	$1.2 \times 10^{+7}$	1	4.86	10.89	2.57×10^{-8}	2.91	0.09	1.20×10^{-2}	10.89	8.20	10.02
Nov-47	$9.5 \times 10^{+7}$	1	4.86	10.89	2.11×10^{-7}	9.57	0.68	9.06×10^{-2}	10.89	10.72	10.72
Nov-47	$9.3 \times 10^{+7}$	1	4.86	10.89	2.08×10^{-7}	9.54	0.67	8.94×10^{-2}	10.89	10.72	10.72
Nov-47	$9.4 \times 10^{+7}$	13	63.17	141.59	2.71×10^{-6}	124.12	8.76	$1.17 \times 10^{+0}$	141.59	139.42	139.42
Dec-47	$9.2 \times 10^{+7}$	1	4.86	10.89	2.05×10^{-7}	9.52	0.66	8.86×10^{-2}	10.89	10.72	10.72
Dec-47	$9.2 \times 10^{+7}$	1	4.86	10.89	2.04×10^{-7}	9.51	0.66	8.82×10^{-2}	10.89	10.72	10.72
Dec-47	$9.2 \times 10^{+7}$	1	4.86	10.89	2.04×10^{-7}	9.51	0.66	8.82×10^{-2}	10.89	10.72	10.72
Dec-47	$9.6 \times 10^{+7}$	1	4.86	10.89	2.13×10^{-7}	9.59	0.69	9.17×10^{-2}	10.89	10.72	10.72
Total Liberated in 1947 (Ci):			119.62	272.30	4.40×10^{-6}	209.77	14.25	1.90	269.79	250.37	255.95
Release Fraction:			80%	80%	80%	10%	10%	100%	100%	10%	10%
1947 Release Total (Ci):			$9.6 \times 10^{+1}$	$2.2 \times 10^{+2}$	3.5×10^{-6}	$2.1 \times 10^{+1}$	$1.4 \times 10^{+0}$	$1.9 \times 10^{+0}$	$2.7 \times 10^{+2}$	$2.5 \times 10^{+1}$	$2.6 \times 10^{+1}$

TABLE E-6
(CONTINUED)

ESTIMATED RELEASES FROM OAK RIDGE GRAPHITE REACTOR SLUG RUPTURES

Date	KWh in Rx	Slugs Rel'd	Ru-103	Ru-106	Sr-89	Sr-90	Ba-140	La-140	U-235	U-238	Pu
Feb-47	$6.8 \times 10^{+7}$	1	4.86	0.49	8.04	0.53	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Feb-47	$2.6 \times 10^{+7}$	1	4.84	0.28	7.91	0.20	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Apr-47	$9.4 \times 10^{+5}$	1	0.97	0.01	1.10	0.01	4.80	10.45	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Aug-47	$5.6 \times 10^{+6}$	1	3.35	0.07	4.72	0.04	10.28	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Oct-47	$9.0 \times 10^{+7}$	1	4.86	0.55	8.04	0.68	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Oct-47	$1.2 \times 10^{+7}$	1	4.46	0.14	6.72	0.09	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Nc	$9.5 \times 10^{+7}$	1	4.86	0.56	8.04	0.72	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Nc		1	4.86	0.56	8.04	0.71	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Nov-4.		13	63.2	7.23	104.56	9.30	137.24	137.24	2.32×10^{-4}	5.05×10^{-3}	6.27×10^{-2}
Dec-47	$9.2 \times 10^{+7}$	1	4.86	0.55	8.04	0.71	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Dec-47	$9.2 \times 10^{+7}$	1	4.86	0.55	8.04	0.70	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Dec-47	$9.2 \times 10^{+7}$	1	4.86	0.55	8.04	0.70	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Dec-47	$9.6 \times 10^{+7}$	1	4.86	0.56	8.04	0.73	10.56	10.56	1.79×10^{-5}	3.88×10^{-4}	4.82×10^{-3}
Total Liberated in 1947 (Ci):			115.58	12.11	189.35	15.13	257.88	263.82	4.46×10^{-4}	9.71×10^{-3}	1.21×10^{-1}
Release Fraction:			10%	10%	10%	10%	10%	10%	10%	10%	10%
1947 Release Total (Ci):			11.56	$1.2 \times 10^{+0}$	$1.9 \times 10^{+1}$	$1.5 \times 10^{+0}$	$2.6 \times 10^{+1}$	$2.6 \times 10^{+1}$	4.5×10^{-5}	9.7×10^{-4}	1.2×10^{-2}

Emissions of Tritium from Isotope Processing Programs

While airborne tritium was likely emitted to some extent from ORNL reactor and fuel processing operations, available data indicate that the most significant source of airborne tritium emissions was the handling of tritium that was received from Savannah River, purified, and repackaged for commercial distribution. Documented quantities of tritium shipped from ORNL provide indication of trends of quantities of the nuclide that were processed. According to Isotope Division reports, under 50,000 Ci were shipped each year 1952 through 1958; 1971 shipments totaled 220,000 Ci; shipments in 1986 topped a million curies; and shipments peaked at 2.4 million curies in 1987.

Reporting of airborne tritium emissions from ORNL began in 1972. Like quantities shipped, the reported airborne effluents peaked in 1987. Reported quantities of tritium shipped annually from ORNL and quantities reported to have been released in ORNL airborne effluents are depicted in Figure E-1. Because the information that has been reviewed does not identify any sources of airborne tritium emissions in the 1950s through 1960s that likely approached the magnitude of reported emissions from isotope processing during the 1980s, the peak annual tritium emission of 44,000 curies reported for 1987 was used for screening calculations.

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APPENDIX II-C

SUPPORTING MEMORANDA

S. Block
and
R. Lawler

BROOKHAVEN NATIONAL LABORATORY

M E M O R A N D U M

DATE: January 4, 1952

TO: All Health Physics Surveyors

FROM: Lee Gemmell

SUBJECT: Summary of Meeting Held January 3, 1952

I. Sy Block outlined the ruptured slug incident which happened on the previous evening. The rod was finally pulled out of the Pile around 1 A.M. and a new one was put in its place and the Pile was back on the air before 5 A.M.

The 30-minute air samples taken on the front porch of T-88 and at the Health Physics office window in Chemistry on the same afternoon of the accident, showed no activity. The air sample taken on the following morning at the Igloo Area, downwind from the Pile stack, showed no activity. Two water collectors were placed at the apartment area and the ball diamond during the night. No appreciable activity was found. The results from the Area Monitoring Stations is not yet available.

II. The use of Sy Block's Po-Be source was discussed. Loaning it is a constant source of irritation. However, it was felt that we should cooperate where ever possible with scientists who want it on a short term basis. Sy will continue to use his good judgment as to how best it may be scheduled and to prevent abuse of the privilege.

III. Drs. Kuper and Cowan brought the group up-to-date on the discussions at the last Radiation Safety Committee meeting. The following pertinent points were made:

A. New scientists or visiting scientists will be assigned to an already active group which will act as a "godfather" to the new member of the team.

B. Somewhere along the line (the exact time has not as yet been established) the new scientist will get some orientation from the Safety Division.

C. The Pile Department hopes to employ another man who will be a specialist on apparatus--its safety and ability to do the job for which it is intended.

D. Operations will institute policy of notifying Safety and Health Physics of new space assigned at the Pile and will endeavor to bring Health Physics in on the initial planning of new experiments.

It was also brought out at the meeting that Health Physics was the "eyes and ears" of Safety. We should be on the lookout for infractions of the safety rules and, in general, these infractions should be reported to Safety for proper action. However,

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: February 3, 1954

TO: File

FROM: M. M. Weiss MMW

SUBJECT: Data on the Slug Rupture Which Occurred on the Morning of 1/17/54 (Rupture No. 13) LP.

As is well known by now, the meteorological conditions on this day were such that any activity from the reactor stack was carried in the direction of Station E-7 which is located at 321° from the stack at a distance of about 2500 meters. Before the slug rupture occurred, roof counters at this location indicated the usual activity detected at this distance, downwind from the reactor stack.

The two graphs included below are plots of the activity picked up on the filter paper as a function of time, the time plotted being the time of collection. Using this data and assuming an overall counting efficiency of 10%, it is estimated that 0.17 μcuries of activity was detected with the dust monitor during the 29-hour period bracketing the slug rupture. This total activity was filtered from 197 cubic meters of air (assumed rate of flow = 4 c.f.m.). The average rate of arrival of activity is roughly 0.86×10^{-15} curies/cc; while the peak rate is 1.3×10^{-14} curies/cc. These results are not corrected for decay during the 4-1/2 hours between collection and counting time.

The most active segment of the filter paper was removed from the roll for further analysis. A half-life check reveals a short-lived component or components of the order of 18 hours with a fairly clear-cut long half-lived component with $T_{1/2}$ = 8+ days. This, of course, suggests I^{131} in appreciable quantities. For further substantiation of this hypothesis, a gamma spectrum was obtained on the gray-wedge machine and a very prominent line at about 360 Kev was noted superimposed on the more or less familiar mixed fission products spectrum. This is to be expected if I^{131} is the element responsible. Intensities on the spectrum picture indicate that this line is by far the major contributor to the activity present on the paper.

Several snow samples were also picked up at various recommended locations with the following results. It seems as if the activity was confined to a rather narrow sector with the maximum near the perimeter of the laboratory where E-7 is located.

Location Where Sample Was Taken (from Stack)		Surface Area Sampled	Activity in
Bearing	Distance	in meters^2	d/m/m^2
321°	2500 meters	.56	13,852
316	5750	.84	155
318	6200	.84	1,538
321	5900	.84	1,670
321	2070	.84	4,509
326	1040	.84	3,423
	below stack	.84	83

cc: J.B.H. Kuper

AMOUNT OF ACTIVITY EXHAUSTED THROUGH THE PILE STACK

1/17/54
from

0145 to 1019	.072	cubies
1019 to 1047	1.335	
1047 to 1305	.200	
1305 to 1542	.288	
1542 to 1751	.142	
1751 to 2101	.096	
2101 to 0230 (1/18/54)	.129	
0230 to 0858	.141	

Total activity for the period was 2.4 curies

The above amounts of activity in the stack effluent were computed on the basis of a sampling rate of 6.2 cfm. The smpling rate as measured on 2/2/54 was 2.5 cfm.

A correction factor of 6.2/2.5 = 2.48 should therefore be applied to the above figures as well as to the coordinates of the graph.

Solari

2/3/54

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 2, 1954

PRIVATE

TO: L. Gemmell

FROM: B. Keene BLK

SUBJECT: Progress Report for July 1954

Some difficulty has been encountered in getting the new laundry room into proper operation. A number of the people using the east locker room seem to feel that they are being imposed upon in being requested to place their lab clothing in the hampers in the locker room. As a result, several of them have been dumping their clothing on the floor of the locker room at the place where the hampers used to be. This situation has been called to Powell's attention and he has issued a memo on the subject. So far this has done no good. What steps can be taken to cure this problem are not as yet clear. The situation is admittedly unwieldy at the end of the day shift. There is too much traffic at this time for the size of the laundry room. It is thought that the problem might be eased by allowing clothing under 5 mr/hr to be worn to the lockers for changing, after which the dirty clothing is to be placed in the hampers.

On July 6th another rupture occurred. It was evident that it was a fairly bad rupture as it tripped a number of the building radiation monitors. The suspected channel was probed. When withdrawn to the charging face, the probe was found to read more than 120 r/hr at 3 feet. The probe was hastily shoved back into the plenum and was then discharged into the canal. The cartridge was discharged with remarkably little difficulty and relatively small amounts of contamination. Following the work, the highest smear on the pile top was 8300 c/m.

Upon discharge of the cartridge, readings over the canal water rose to a peak of 1 r/hr. Shortly after the discharge, the building air became contaminated. The exact origin of this airborne contamination is unknown, but there is some evidence to indicate it came from the canal as a result of discharging the hot uranium into cold water.

Since the rupture was discharged in mid-afternoon of a week day, relatively strict traffic control was thought to be necessary. To accomplish this, the personnel elevator was tagged off and the main switch opened. Two trainees were posted at the entrance to the charging area to prevent entry of unauthorized personnel, and to check the shift personnel for contamination upon leaving the area. As a result, only the south 110' became contaminated. It is planned to use a similar setup for future ruptures.

One instance of rather serious contamination occurred. Binns and Lones sorted through a number of ionization chambers which were stored in a shield in the instrument room. These chambers had been in the pile for various lengths of time and had had various amounts of cooling. The fact that the chambers had been unwrapped and moved about was not known to H.P. until 4 or 5 days later when smears in the room

May 1, 1956

Lee Gemmell

Maynard Smith

Meteorological conditions,
0050 to 0117 EST, 5-1-56.

We have checked our records for the above time period in order to determine the possible fate of effluent from the Reactor. We had a mild temperature inversion amounting to +1.0°C between 410 and 37 feet accompanied by a strong northerly wind (360°, 8 meters per second). Such conditions would have produced a very narrow plume of effluent from the Reactor that almost certainly stayed aloft for at least 10 to 15 kilometers provided there were no particulates greater than 10 microns involved.

With such conditions one would not expect to find any trace of material at ground-level at the established Health Physics monitoring stations. I suggest that you inspect the records from station 0-1 (352°, 6.4 km.), since this is the only possibility, but I confidently expect that you will find nothing there.

MES:mcf

cc: R. W. Powell ← THIS COPY FOR
M. Fox
R. M. Brown

May 3, 1956

Ken Gammill

May 2, 1956

Maynard Smith

Meteorological conditions
1925 to 2000 EST, 5/2/56.

The Meteorology Group records for the above time period indicate that effluent from the Reactor should have passed directly over the north gate, across the northeast site boundary, and just south of the intersection of Smith's Road and Route 25.

I think it would be most interesting to determine whether material from the Reactor slug rupture did reach the ground anywhere along this path because we have an unusual set of conditions to deal with. The wind direction was 130° with a speed of 8 m.p.h. The temperature records showed a lapse condition (-0.7°C) between 410 and 150 feet, and a very slight inversion (+0.2°C) between 150 and 37 feet above ground. I would normally expect that this very weak inversion layer would prevent small particulates from reaching the ground within any reasonable distance, but in this particular case, we know that light rain was occurring, and it is quite probable that material was brought to the ground by it. It is, however, also probable that some of the material was subsequently washed away by approximately 0.2 inches of rain.

As shown on the attached map, I would suggest a careful survey of the north gate, the indicated section of the northeast site boundary, the road running northeast from Smith's Road to Route 25, Smith's Road and Route 25 itself.

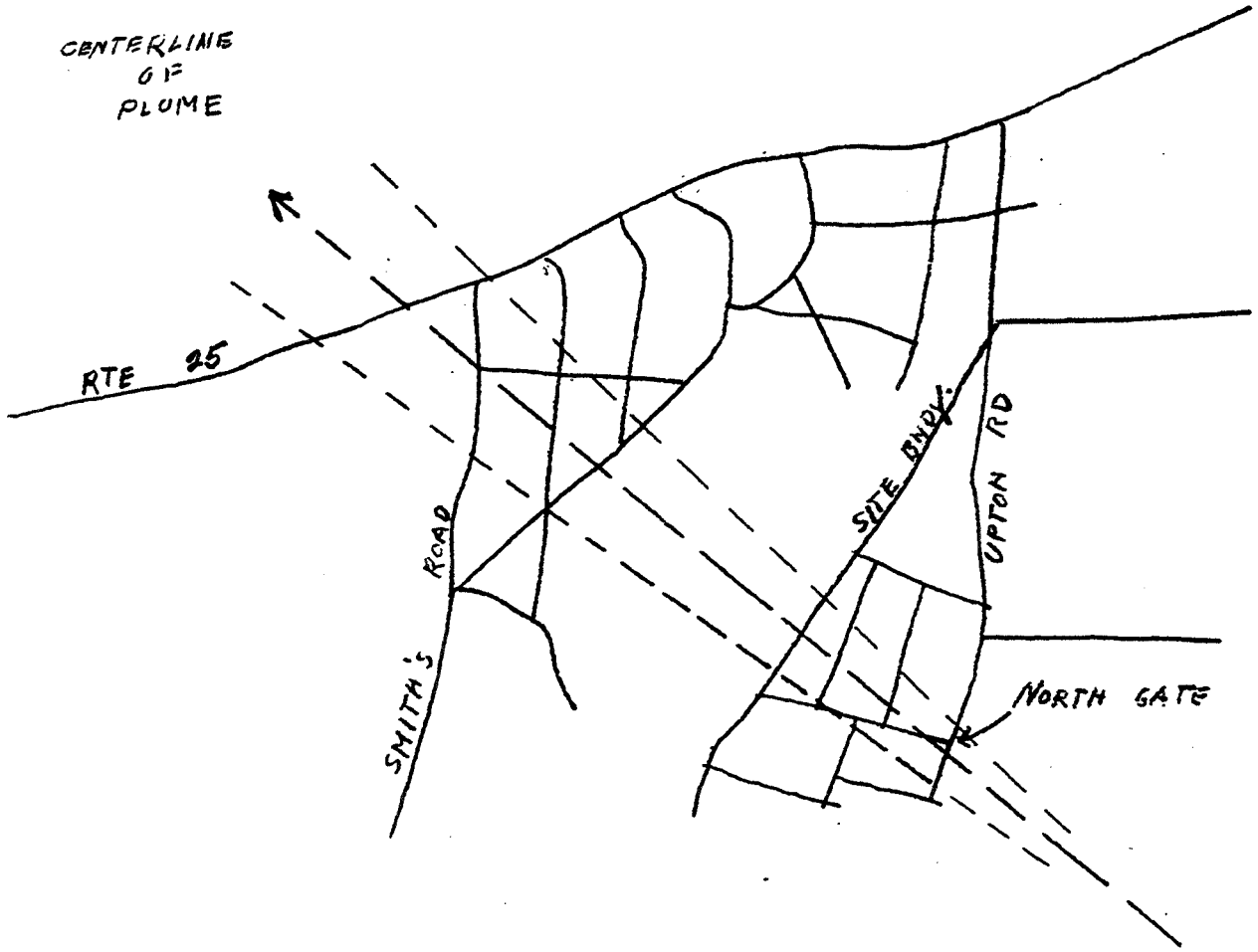
If I am not mistaken, this is the first rupture we have ever had that was accompanied by light rainfall. As you know, it is my belief that the rain acts as a very effective scrubbing mechanism even at low levels, and a careful survey of this region should reveal the validity of the contention.

MES:mcf
cc: M. Fox
R. W. Powell ← THIS COPY FOR
M. Weiss
I. Singer

Path of Reactor Effluent Plume

1925 E - 2000 E

May 2, 1956



June 6, 1956

PRIVATE

Lee Gemmell

L. F. Phillips

Hot Lab Progress Report, May

Five hollow cylinder kilocurie Co^{60} sources have been surveyed to determine if any of the radioactive material was leaking out of their jackets. One Al jacketed source was discovered to be leaking. Preparations are being made to can the source in stainless steel.

A small Co^{60} source was lifted, unshielded, out of the source transfer tank to check radiation levels in air.

A new alarm system has been installed on the liquid "F" waste system. This enables personnel, on weekends, to determine the liquid level in the "F" tanks by signals transmitted over the telephone. This is to prevent the possibility of an "F" waste overflow and also makes unnecessary the trips into the lab. by personnel to check the liquid levels. The "F" waste overflow line has now been connected directly to the sewer.

An attempt is being made to determine the particle size of radioactive material found on dead vegetation as a result of fall-out from the 22nd, rupture occurring in the HML Reactor on May 2nd. It was discovered that the active particles (if entirely particulate and not molecular) are greatly exceeded in number by the inactive particles, and it is almost impossible to distinguish between them in the microscope. The problem evolved into one of removing the particulate material from the dead vegetation using ultrasonic methods then separation of the particles by settling in an alcohol column. Since the particles are not spherical, Stokes Law is only crudly applicable. The problem appears to be enlarged by the wide spectrum of densities observed. Results of this problem are expected to be only approximate and await analysis of samples withdrawn from 12 positions on the alcohol column.

An attempt is also being made to determine the quantity of particulate activity released from the reactor stack as a result of the same rupture. Approximately 5×10^{-6} of the exhaust air is sampled and it is assumed that the same fraction of the particulate activity was collected on the filter paper. Several methods are being tested utilizing Soxhlet extraction techniques to remove the fission products from the filter paper and putting them into solution. An aliquot may then be taken from this known volume and the total activity collected by the filter paper determined. Then the total activity released by the reactor stack may be approximated.

Tests have been made on a sample of "Clean Tread Mat", a product of the Merit Paper and Chemical Corp., for the removal of contamination from shoes. Results of these tests have been given to L. Gemmell.

LFP:rmh

cc: J.B.H. Kuper

August 31, 1956

L. Gemmell

L. Phillips

Progress Report, August

The 14 AEC Radiological Physics Fellows have completed their annual one week summer course of practical training in H.P. operations at the Hot Lab.

T. Brooms of the Glenn L. Martin Co. spent one week at the Hot Lab as a part of his 5 week H.P. indoctrination program.

Leaking Co^{60} sources in the source transfer tank of the Gamma Irradiation Facility have resulted in a continued increase in water contamination. Flushing and dilution methods have been unsatisfactory in decreasing the contamination. Activity levels reached an all time high of 1.5×10^{-3} c/ml which amounts to about 3.5 curies of Co^{60} in the water without considering the activity levels in the sludge at the bottom of the tank. This resulted in the spreading of nuisance levels of contamination about the facility and on equipment, which at present has been cleaned up. At present all sources have been taken out of the tank in Pb pigs. The tank has been drained and scrubbed. Plans have been made to paint the sides and place a stainless steel tray in the bottom of the tank. Most of the Co^{60} kilocurie sources have been checked for leakage using HNO_3. Four of the older Al clad sources were found to be leaking. They will be sent to Oak Ridge to be reclad in stainless steel.

A process has been developed to estimate the quantity of particulate activity released from the reactor stack as a result of fuel element ruptures. The process consists of a series of HNO_3 extractions from Hollingsworth Vose filter media.

Rupture no 22, which occurred 5/2/56, has been partially evaluated using the above mentioned extraction process. Approximately 5×10^{-6} of the exhaust air was sampled and it is assumed that the same fraction of the particulate activity was collected on the filter paper. The assay indicates that the remaining activity which exists today, after 115 days decay, amounts to 2.5 curies. Decay studies indicates that the activity levels remaining 4 1/2 days after release from the stack amounted to approximately 10 curies. Insufficient decay data was collected to be able to estimate the number of curies released at any earlier date.

It is hoped that, on the advent of another rupture, a combination of 1.) early decay data, and 2.) a series of simple extractions will give a good estimate of curies emitted from the stack as a function of time after emission.

A question exists as to "how close" a representative sample is being collected on the filter paper at the time of the rupture. This question deserves further study.

Two particle size studies were made of the reactor off-gas under normal operatin conditions. Dust samples were collected at the base of the stack using type HA millipore filters. The particles were studied using a 1455 power (oil emersion) light field microscope. In both studies the greatest percentage of particles observed were 0.4 microns in diameter and less than two percent were greater than one micron in diameter. This is in good agreement with data observed in July of 1954. Simple agglomerations of small particles were observed, however, no huge (60 μ) odd shaped agglomerations were observed as reported in the 1954 report.

Several decay curves have been plotted of airborne dust samples taken at the base of the stack during normal operating conditions. Air samples using millipore filters and counted on 2π flow counters indicate a large abundance of short-lived particulates. A ten fold decrease in activity is observed in the 1.2 to 45 minute decay interval after completion of 10 minute air samples.

LFP:rmh
cc: J.B.H. Kuper

March 25, 1957

F. P. Cowan

L. F. Phillips

Estimation of Particulate Activity Released into the Atmosphere as a Result of BNL Reactor Fuel Element Ruptures

At 1300 on January 15, 1957 cartridge number 32x72 ruptured in channel 3A-3-3. This fuel element was of the old type located in the inner ring of the normal fuel zone, a position at approximately 350° C fin temperature. A rapid temperature cycle during an unscheduled shutdown on January 14 is considered the primary cause of this, the 25th rupture.

As a result of this rupture, a significant quantity of radioactive particulate was introduced into the pile cooling effluent. This effluent is filtered by the primary air outlet filters. These "glastex" filters are composed of a woven glass fiber cloth which is claimed to be more than 95% efficient for removal of all particles down to 5 μ and 25 to 30% efficient for smaller particles. These efficiencies have been partly substantiated by particle size studies made under normal operating conditions and of "wash out" after rupture number 22. (Progress Report, Phillips to Gemmell June and August 1956)

After filtration, a small fraction of the effluent is sampled approximately isokinetically at the base of the stack by a continuous dust monitor. The dust is collected on a moving Hollingsworth and Vose type H-70 filter paper strip with a collection efficiency greater than 98% for 0.24 μ diameter particles. About ten minutes after collection this filter strip moves past an anthracene scintillation counter. This monitor cannot be easily calibrated in terms of activity per unit volume of effluent without a large expenditure of equipment and time because of the problems of very high initial activity levels and "breakthrough", namely spurious counts due to radiation from the freshly collected fission products penetrating the shield surrounding the crystal. A series of nitric acid extractions of filter paper samples gives a simple solution to the problem without deviating from the present sampling methods.

Nitric acid extractions of filter paper samples collected during the rupture were made on January 18th. The analysis of these extractions and the application of the ratio between sampling and effluent flow rates made possible the determination of the number of curies existing in the environment at the time of the extraction. This information, plus decay data obtained from the rupture sample, determines the number of curies existing in the environment at any time after its release from the stack.

The dust monitor indicated an off-scale deflection at the time of the rupture. A few minutes later the filter paper containing the rupture sample was taken to the Hot Lab, and the decay was observed for the period covering 21 minutes to 3 hours after the rupture with a CS-40 (serial #166) survey meter. (This decay data was

obtained by C. J. Kennerth and A. R. Lukas) A small fraction of the rupture sample was removed from the filter paper and another decay curve is being obtained, using a 2π proportional flow counter, starting 2-1/4 hours after the rupture and continuing to date.

Flow Data

Density of stack air at 59° C = 0.0665 pounds/ft^3

Stack air flow rate (temp. corrected):

$$\frac{1.468 \times 10^6 \text{ pounds/hour}}{60 \text{ min/hr} \times 0.0665 \text{ pounds/ft}^3} = 3.67 \times 10^5 \text{ ft}^3/\text{min}$$

Sampled air flow rate = 1.6 ft^3/min

Fraction of the stack air sampled:

$$\frac{1.6 \text{ ft}^3/\text{min}}{3.67 \times 10^5 \text{ ft}^3/\text{min}} = 4.36 \times 10^{-6}$$

Extraction Data

Practically the entire rupture sample (>99%) was contained on 3-1/4 in^2 of the filter paper. After three HNO_3 extractions and one distilled water extraction a yield of better than 99% was obtained. The sample was taken up to 250 ml volume and eight one ml aliquots were taken. These samples were evaporated and counted in a 2π proportional flow counter calibrated with a Tl^{204} standard. An average disintegration rate of 59,360 d/m ml was obtained. The total number of microcuries on the filter paper at the time of the extraction (0400 on January 18, 63 hours after the rupture) amounted to:

$$\frac{250 \text{ ml} \times 59{,}360 \text{ d/m ml}}{2.22 \times 10^6 \text{ d/m } \mu c} = 6.68 \ \mu c$$

The total number of curies existing in the environment at the time of the extraction amounted to:

$$\frac{6.68 \ \mu c \times 10^{-6} \text{ c}/\mu c}{4.36 \times 10^{-6}} = 1.53 \text{ curies}$$

Graph II, showing the number of curies existing in the environment vs decay time after the rupture, was obtained using this information and the above-mentioned decay data.

Discussion of Graphs

Graph I was drawn after making non-linear zero drift corrections on the decay data obtained with the CS-40.

Since the CS-40 measures dose rate and the 2π proportional counter measures a

count rate, the CS-40 decay data cannot be used to extend the information given by the 2π counter back to the earlier time.

Due to the method of sample collection the data represented in both Graph I and II represent an unknown mixture of volatile and non-volatile constituents. This makes comparison with other fission product decay curves difficult.

Since about 51 mg/cm^2 of absorber (chamber wall $\simeq$ 32 mg/cm^2, 16 cm of air $\simeq$ 19 mg/cm^2) separate the sensitive volume of the chamber from the source the CS-40 detects less of the lower energy β's in comparison with the 2π flow counter. Since the shorter-lived isotopes, in general, decay with greater average β energies the CS-40 decay data exaggerates their relative importance, as is indicated by the graphs.

Comparison of Ruptures No. 21 and 22 with Rupture No. 25

Rupture No.	Date Ruptured	Number of Megawatt Days	Decay Time After Rupture: 2-1/4 Hours	63 Hours	4-1/2 Days	13-1/2 Days	115 Days
21	5/1/56	18,573	3.6 c*	0.8 c*	0.6 c*	0.32 c**	-
22	5/2/56	37,888	60*	13*	10+	-	2.5++
25	1/15/57	12,382	7.2+	1.53++	1.2+	0.63+	-

* Approximate values obtained by using decay data from rupture #25
** From earlier soxhlet extractions
\+ From observed decay studies
++ From direct HNO_3 extractions

If one assumes that the fission products in all three fuel elements had the same approximate decay characteristics, then the values obtained in the table from the known decay data of rupture #25 would be valid.

Rupture #22 had been partly evaluated when the first applications of nitric acid extractions on rupture samples were made in August 1956. However, there were insufficient decay data for the estimation of activity levels before 4-1/2 days decay. (Progress Report, Phillips to Gemmell, August 1956)

The rupture #21 data were obtained from earlier attempts at extraction using constant boiling hydrochloric acid and a soxhlet extractor which gave poor yields. Decay data were also lacking for this sample.

Discussion

Some of the problems which deserve further study are:

1) A re-evaluation of the efficiency of the sampling method. (A question exists as to "how close" a representative sample is being collected on the filter paper at the time of the rupture.)

2) A dust monitor calibration for low-level particulate activities. (μc/cc of stack effluent)

3) Particle size studies at several locations through the pile cooling system. (One may wish to confirm periodically the efficiency of the exit air filters in removing larger particles.)

4) Special decay studies.

With the gradual replacement of the old type of fuel element with the new design, the probability of another rupture (as we know it) should decrease. When all the old fuel is replaced there should be no need for a high activity level dust monitor system. However, there is a possibility that a defective sample irradiated in the reactor may disintegrate or in some way release particulate activity into the reactor cooling stream from the experimental holes or the fuel channels.

The Nitric Acid Extraction Process

Since nothing has ever been started concerning the exact steps in the extraction process, a detailed procedure will follow for the benefit of anyone continuing the work in the event of another rupture.

A decay study must be made with a small fraction of the rupture sample ($\sim 2 \times 10^5$ c/m) using a 2π proportional flow counter. This decay study should start as soon as possible after the rupture occurs and continue beyond the time that the extractions are analyzed.

The following extraction steps are not necessarily the best or most efficient but they worked and were obtained by trial and error extractions from smears (~ 1 rad/hr at 5 cm) of a plenum shovel and air samples ($\sim 1/2$ rad/hr at 5 cm) obtained in the plenum of the reactor during shutdown.

Procedure

I. Sample Preparation

1) Cut filter paper samples down to a minimum area and center them on a glass petri dish.

2) Using a CS-40 survey meter, measure the dose rates at several distances above the sample. These values will be used later to approximate the yield.

II. The Extraction

1) Place the samples in a 250 ml pyrex centrifuge bottle with 3 glass boiling beads.

2) Add 70 ml conc. HNO_3 and boil for 10 minutes.
Caution - these bottles are thick walled and even though made of pyrex they will crack very easily. Heating and cooling must be done gradually.

3) After cooling, record the count rate with an end-window GM tube held at the bottom of the bottle. All count rates will be made at this same position to indicate relative progress of each extraction.

4) Centrifuge at a maximum of 2,000 rpm for about 5 minutes. (International centrifuge, size 1, type SB, 1/3 H.P.)

5) Carefully remove the bottle from the centrifuge and record the count rate. If the count rate increases significantly above that observed in step 3, then the yield may be poor as a considerable amount of the activity may still exist with the filter paper fibers.

6) Carefully decant the supernate into a 250 ml beaker using a Fisher filtrator. Record the count rates of the supernate and residue for later comparisons.

7) Repeat steps 2 through 6 until the count rate from the residue is reduced to several times background.

8) Repeat steps 2 through 6 using 70 ml distilled water instead of acid.

9) Wash residue into a glass petri dish and dry. The surface area of the residue should be kept approximately equal to that of the original sample. Compare dose rates with that of the unprocessed samples for estimates of % yield.

10) Evaporate the supernate to less than 250 ml, place in 250 ml volumetric flask and let cool to room temperature. The dilution factor is determined by the total activity present.

11) Bring the supernate up to volume and pipette several samples into Al planchets. The volume of the samples will depend upon their specific activity.

12) Dry slowly under a heat lamp and count in a 2π proportional flow counter previously calibrated using a Tl^{204} standard. Record the time at which the counts are made.

III. Calculations

1) Calculate the number of $\mu c/ml$ of solution.

2) Calculate (A_1) the total number of μc on the filter paper (effective when the sample was counted in the flow counter).

$$A_1 = 250 \text{ ml} \times \mu c/\text{ml}$$

3) Estimate the number of curies existing in the environment after a decay period from the time of the rupture to the time the samples were counted in the flow counter.

$$\text{Curies} = \frac{A_1 \times 10^{-6} \text{ c}/\mu c}{(\text{fraction of the effluent sampled})}$$

4) Determine decay corrections and plot the number of curies existing in the environment as a function of decay time after the rupture.

DECAY DATA RUPTURE No. 25 t_o = APPROX. TIME OF RUPTURE = 1300, 1/15/195.