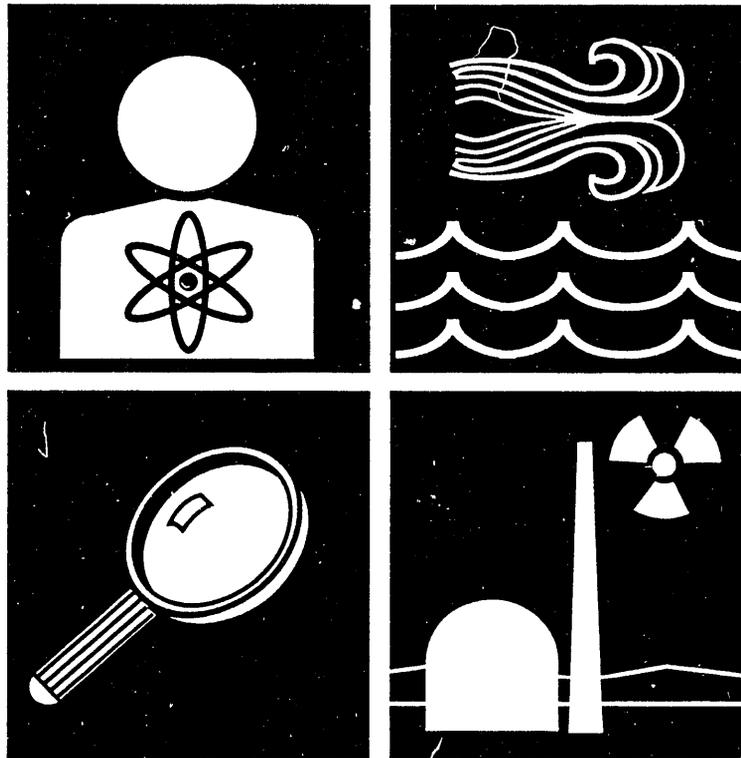


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Letter Report: References For Radioactive Releases to the Atmosphere From Hanford Operations, 1944-1957

R. B. Hall

November 1991



Prepared for the Technical Steering Panel



PNL-7868 HEDR

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LETTER REPORT:
REFERENCES FOR RADIOACTIVE RELEASES
TO THE ATMOSPHERE FROM HANFORD
OPERATIONS, 1944-1957

Hanford Environmental
Dose Reconstruction Project

R. B. Hall

November 1991

Prepared for the
Technical Steering Panel

Pacific Northwest Laboratory
Richland, Washington 99352

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SUMMARY

A search was made for published documents related to discharges of radioactive material from Hanford Site facilities to the atmosphere from 1944-1957. The purpose was to list documents that contain data that might be useful in developing a source term for airborne releases. The source term for the radionuclide that contributes most to dose, iodine-131, is a separate effort. Other source terms will be developed later.

This tabulation of published summaries of atmospheric release data shows the type of measurements that were being made from 1944-1957 and the magnitude of the discharges to the atmosphere.

In the early years, very little data were collected that related to specific radionuclides. However, most of the key radionuclides were known to be present in effluents from occasional specific radionuclide analyses.

CONTENTS

SUMMARY	iii
1.0 INTRODUCTION	1
2.0 TECHNICAL APPROACH	2
3.0 SUMMARY OF RELEASES TO THE ATMOSPHERE	2
3.1 DATA SOURCES	3
3.2 DOSIMETRY-RELATED RADIONUCLIDES	3
3.3 RADIONUCLIDES OF ADDITIONAL CONCERN	4
3.4 OTHER RADIONUCLIDES FOUND IN LITERATURE	5
3.5 CORROBORATING SOURCES	6
3.6 OTHER DOCUMENTS	6
4.0 REFERENCES	6
APPENDIX - DOCUMENTS CONTAINING DATA ON RADIOACTIVE RELEASES TO THE ATMOSPHERE, 1944-1957	A.1

1.0 INTRODUCTION

The goal of the work to which this report contributes is to develop daily release data for key radionuclides selected by the Technical Steering Panel. The documents listed in Appendix A will be used to provide information for source term reconstruction in FY 1992 and FY 1993. The purpose of this letter report was to review published data with foreknowledge that these data consisted of summaries and averages that would not fulfill the daily release requirements needed for complete source term development. However, documents referenced in the appendix tables are used to help develop source terms and provide confirmation of developed source terms.

Documents related to the release of radioactive materials to the atmosphere were reviewed, including data or information on all accidental and unintentional releases and deliberate discharges. Results of a similar search for documents on release of materials to the Columbia River are provided in a companion document, "Letter Report: Reference for Radionuclide Releases to the Columbia River from Hanford Operations, 1944-1957." The development of a source term for releases of iodine-131 for the 1944-1947 period is a separate effort.

Though information related to releases from all Hanford facilities was included in the review, most atmospheric releases were from the separations facilities. Hanford facilities were described in Ballinger and Hall (1991).

Information on two categories of key radionuclides was searched for: dosimetry-related and those of additional public concern. Dosimetry-related radionuclides included iodine-131, ruthenium-103, ruthenium-106, cobalt-60, cerium-144, and tellurium-132. Radionuclides of additional public concern included plutonium-239, iodine-129, strontium-90, cesium-137, argon-41, and krypton-85. Tritium was added to the list because the literature included large releases from one facility during two campaigns in the early 1950s.

The time period of interest extended from 1944 through 1957. Because most of the releases came from separations facilities, the period began with the startup of T Plant in December 1944.

This document fulfills HEDR Milestone 0303A, "Documented Significant Airborne Radionuclides, 1944-1957." All documents listed in the appendix are publicly available and may be found in the DOE Field Office, Richland (RL), Public Reading Room.

2.0 TECHNICAL APPROACH

More than 800 documents were reviewed that contained information relevant to releases to the atmosphere. Also included were those that showed the state of understanding and knowledge at the time and those that described emissions control or lack thereof and problems with equipment and facilities. Publications dated later than 1957 are addressed because that information may well assist in the development of source terms. Document searches were conducted in accordance with Shipler (1991).

The search and recording of applicable documents did not involve data generation or judgement or evaluation of the quality of data included. The ability to measure specific radionuclides did improve greatly over the years. Consequently, it will be necessary at a later date to judge the quality of the data in the documents when daily source terms are developed. Some of the quality control measures used at the time of data generation are included in documents listed in Table A.4, "Routine Sampling and Counting Methods."

3.0 SUMMARY OF RELEASES TO THE ATMOSPHERE

Hanford facilities and processes are discussed in Ballinger and Hall (1991). No filtrations or effluent cleaning devices were utilized in any facility in 1944. Stack sampling equipment was installed in major facilities and some monitoring took place. Iodine-131 emissions from 200 Area stacks were recognized as a problem at once, but knowledge of the milk exposure pathway did not occur for several years after Hanford operations began in 1944. Control measures evolved from extension of cooling time before dissolution to installation of dissolver off-gas scrubbers, and later, silver reactors. Control measures for particle releases included sand filters and

fiberglass filters. These were not effective in restraining the ruthenium that passed through the REDOX filters as a gas and became entrained in ammonium nitrate deposits in the stack. Those problems are discussed in reference documents listed in Table A.1.

3.1 DATA SOURCES

Stack monitoring was conducted by two different groups in the Health Instrument Division in the early years. The Operational groups in the 100 (Reactor) and 200 (Separations) Areas conducted instrumented monitoring (if any); Site Survey or Regional Monitoring staff collected samples. Data collected by the Operational groups were not found. Documents containing iodine data and related documents have been included in the time line in Table A.1. Some of these documents included measurements for radionuclides other than the key ones listed in Section 1.0.

3.2 DOSIMETRY-RELATED RADIONUCLIDES

3.2.1 Iodine-131

Regular measurements of iodine emissions began to appear in published documents in 1950 (e.g., Soldat and Paas, HW21891, in Table A.1). However, there is reason to believe that measurements were being made in 1949 during the "green run" (Jenne and Healy HW17381De1, in Table A.1). Regular monitoring data for T Plant appeared in Regional Monitoring reports listed in Table A.3, beginning in January 1951 (e.g., HW21214). Monitoring data for B Plant appear in September 1951 (HW23133) but monitoring was discontinued in April 1952 when B Plant was shut down.

The data were collected by Regional Monitoring personnel to obtain a source term to relate to environmental sample data. Sampling and counting procedures are listed in Table A.4.

3.2.2 Ruthenium-103 and -106

Reporting of ruthenium discharges from the REDOX Plant began soon after the facility started operating in 1952. The routine report on radioactive contamination in the environs began reporting releases in March 1952

(HW26493). Releases from REDOX were reported regularly thereafter and were begun at the PUREX Facility when it began operating in January 1956. These reports are listed chronologically in Table A.3, "Regional Monitoring Reports." Ruthenium and other fission products were measured by Healy (HW10758); the fact that he was able to analyze those filters is evidence that stack monitoring samples were collected. These data were collected by Health Instruments personnel to obtain a source term for comparison with data collected in the environs. Counting room procedures and some sampling procedures are contained in documents listed in Table A.4.

3.2.3 Cobalt-60

No data were found in the 1944-1957 reports concerning cobalt releases to the atmosphere.

3.2.4 Cerium-144

This radionuclide would be expected to appear with other fission products. No data on the release of this radionuclide were found.

3.2.5 Tellurium-132

It is doubtful that any data on this radionuclide would appear in reports prior to 1957. Its short half life (76 hours) would have meant that it would have largely disappeared from fuel before it was dissolved.

3.3 RADIONUCLIDES OF ADDITIONAL CONCERN

3.3.1 Plutonium-239

The discharge of plutonium to the atmosphere was always a concern to Health Instruments personnel. Seymour (HW7-5463) estimated that about 0.5 grams had been discharged during the first 2 years of operation. Estimates are also included in Anderson's summary report (ARH-3026). Documents related to the P-11 fire (HW22936 and HW25709) discuss the only release unrelated to separations.

3.3.2 Iodine-129

No search was made for data related to this radionuclide because of its very low specific activity. It would be unlikely that any data were collected in the early years of Hanford operations.

3.3.3 Strontium-90 and Cesium-137

These fission products were noted by Healy (HW10758). Although they are known to appear occasionally in data from time, time did not permit identification of documents containing significant data.

3.3.4 Argon-41

This radionuclide was known to exist in reactor gaseous releases. It was first reported by German (DUH13013) in 1945. It resulted from activation of air that occurred in vertical and horizontal control rod thimbles, and in air dissolved in cooling water. Its release from reactor building stacks was monitored by passing a sample through an ionization chamber. No monitoring of gases released from the water was conducted in this time period. Carter (HW50138) discusses the generation of argon-41 and other inert gases.

3.3.5 Krypton-85

No monitoring data for this noble gas are known to exist. The release of krypton-85 from separations facilities could be calculated from data collected by Bennett (HAN-56128Del) and by procedures described by Gumprecht (HW-32753Del).

3.4 OTHER RADIONUCLIDES FOUND IN LITERATURE

3.4.1 Tritium

Table A.2 lists a series of one-page documents that reported releases of tritium from the P-10 Facility at 108-B in the 100 B Area. Other releases of tritium were measured in reactor building exhaust stack gas samples beginning in 1951. These samples were collected by Regional Monitoring staff and analyzed according to procedures described in documents listed in Table A.4. Sulfur-35 and carbon-14 were collected in the sample with tritium and discharge data were reported with the tritium data.

3.5 CORROBORATING SOURCES

Although it is believed that Operational Health Instruments staff collected stack emissions data, none were found. The only other corroborating information are the environmental data.

3.6 OTHER DOCUMENTS

Documents describing stack release problems and reactions to problems were included to show activities and approaches common at the time.

4.0 REFERENCES

Ballinger, M. Y. and Hall, R. B. 1991. A History of Major Hanford Facilities and Processes Involving Radioactive Material. PNL-6964 HEDR, Pacific Northwest Laboratory, Richland, Washington.

Shipler, D. B. 1991. Document Search and Declassification for Phase I of the HEDR Project. PNL-7713 HEDR, Pacific Northwest Laboratory, Richland, Washington.

APPENDIX

DOCUMENTS CONTAINING DATA ON RADIOACTIVE
RELEASES TO THE ATMOSPHERE, 1944-1957

Note: The documents listed in this appendix are all publicly available and can be found in the RL Public Reading Room in Richland, Washington and are available for purchase from the National Technical Information Service.

List of Tables

<u>Table</u>	<u>Title</u>	<u>Page</u>
A.1	Source Term Timeline for Releases to Air	A.2
A.2	Tritium Releases From Hanford	A.1
A.3	Regional Monitoring Reports	A.8
A.4	Routine Sampling and Counting Methods	A.10

TABLE A.1. Source Term Timeline for Releases to Air

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	<u>Reference Author</u>	<u>Document</u>
1944		Limits for fission products in air and water	WE Cohn	DUH4517
1943	Ru, I	Questions the fate of volatile ruthenium and iodine in dissolver off-gas	L Squires	DUH12040
1944 to 1986		Timelines - Standards, plant wide operations, and 200 Area waste disposal, including unplanned releases		RHO-HS-ST-10 Vol. 1
1944 to 1968		Chronology of Separations Operations	RW McCullugh JR Cartmell	ARH-780
1944 to 1990		History of major Hanford facilities	MY Ballinger RB Hall	PNL-6964HEDR
1944 to 1972		Cooling time data for fuel charged to dissolvers	RE Roberts	HWN-1991Del
1944 to 1972	U, Pu, I-131	Radionuclides in gaseous wastes	JD Anderson	ARH-3026
1945 to 1954	Kr-85	Production of Kr-85	CA Bennett	HAN-56128Del
1945		105-F start-up - active gas release	LL German	DUH13013
1945		Estimates of radioactive limits on plants	HM Parker	HW7-3217
1945		Calculation of the expected sensitivity of the xenon monitor	CC Gamertsfelder	HW3-2208
1945	Xe, I-131	Generation and emission data during dissolving	JL Dreher	HW3-3003
1945		Stack flow rates	FB Vaughn MF Acken DA Miller	HW3-3241
1946 Jan, Feb, Mar	I-131	Estimated number of curies in fuel dissolved	JW Healy	HW3-3495
1946 Apr, May, June	I-131	Estimated number of curies in fuel dissolved	JW Healy LD Turner	HW3-3603

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	<u>Reference Author</u>	<u>Document</u>
1946 July, Aug	I-131	Estimated number of curies in fuel dissolved	LD Turner	HW3-5055
1946	I-131	Estimates of iodine released	RS Apple	HW7-4074
1946		Tolerance doses, state of knowledge about radiation exposure and measurement	HM Parker	DUH12744
1947 to 1952		Extracts of reports on efforts to control stack releases from separations plants	PM Cleavenger SP Gydesen	HW89072
1945 to 1947	Fission Products	Decay curves and analyses of stack sample filters	JW Healy	HW10758
1946 to 1948		History of particle problem in the 200 Areas and particle removal equipment	JB Work	HW11529
1947 to 1957	I-131	Emission data summary	BV Andersen	HW89066
1947	I-131, Xe	110 curies of I-131 and 15 curies of xenon released per charge of slugs	HM Parker	HW7184 Part 2
1948	Fission Products	Review of the particle problem in the 200 Areas	HM Parker	HW9259
1948	I-131	Estimated number of curies of iodine in fuel dissolved	W Singlevich	HW10242
1948		Water scrubbers and sample lines installed in dissolver off-gas lines	CN Gross	HW10395Del
1948		T Plant fans and ducts replaced	HM Parker	HW10421
1948		Argues against reduction in cooling time	HM Parker	HW10568
1948	I-131 F.P.	Summary data prepared for presentation to the Reactor Safeguards Committee	HM Parker	HW10592
1948	I-131 F. P.	Chronology of 200 Area stack problems and reactive control measures.	FAR Stainken	HW11082
1948		Start-up of sand filters: T Plant 10/15/48, B Plant 10/30/48	CN Gross	HW11410
1949		Emission control status at B and T Plants	CE Lapple	HDC-978

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	<u>Reference Author</u>	<u>Document</u>
1949		Could not use I-131 monitoring instruments; were using scrubbers and counting samples	JB Work	HW12168
1949	I-131	Account of project "green run," including estimated iodine release rate in curies/sec	DE Jenne JW Healy	HW17381Del
1949		Report to Congress - waste disposal practices	HM Parker	HW12895
1949		Discusses sand filters, fiberglass filters and silver reactor	AG Blasewitz	HW15200
1949	I-131	Analysis of scrubber solution indicates 50% of iodine released	AG Blasewitz	HW15367
1950		Hanford Works Stacks: Description of all tall stacks, including powerhouses	JM Smith	HW18143
1950		Two documents discussing reduction of cooling time	HM Parker HM Parker	HW18409 HW18567
1950	Particles	Test of T Plant sand filter efficiency	AG Blasewitz	HW18094
1951		Efficiency evaluation of the dissolver cell silver reactors and fiberglass filters	JB Work	HW19898
1951	I-131 Particles	Measured efficiencies of silver reactors and packed bed fiberglass filters	AG Blasewitz RV Carlisle BF Judson MF Katzer WC Schmidt B Weiddenbaum	HW20332
1950-51	I-131	T-plant emission data before and after silver reactor installations	HJ Paas JK Soldat	HW21891
1951	I-131	Estimates of I-131 held by silver reactors	AG Blasewitz	HW21631
1951	I-131	Re-evaluation of silver reactor performance	AG Blasewitz BF Judson	HW21826
1951	I-131	Plant tests on silver reactors after regeneration	AG Blasewitz	HW21957
1951	Pu-239	P-11 fire investigation, 12/4/51	FE Kruesi	HW22936

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	Reference <u>Author</u>	<u>Document</u>
1951		Critical mass facility; criticality and fire during cleanup	ML Mickelson	HW25709
1952	Ru 103/106 Zr/Nb95	Analysis of REDOX particles	TK Bierlein	HW24885
1952	I-131, Ru Tritium	Iodine and ruthenium from separations stacks; tritium from reactor stacks.	HM Parker	HW-26720E
1952	I-131	Study of iodine suppression by addition of mercury to dissolver	BE Kirkendall	HW25151
1953	I-131	History of silver reactor performance	HF Soule RE Taylor	HW-27580
1953		Identifies ammonium nitrate as carrier for radioactivity from REDOX stack	FE Adley WE Gill RH Scott DE Wisehart HB Perry HF Soule	HW28780
1953	I-131, Ru, Pu-239, Sr-89, Sr-90	A study of particles in T Plant stack	DE Wisehart FE Adley	HW28828
1953	Ru	Letter report on a single REDOX emission	RN Donelson	HW29346
1953	I-131 Tritium	Iodine in separations stacks; tritium in reactor stacks.	HM Parker	HW-30423E
1954	Ru	REDOX stack gas particles	FE Adley	HW32209
1952 to 1954	Ru	History of REDOX Ru releases; incident investigations are referenced.	DP Ebright	HW32473
1954	Kr-85	Production of Kr-85	RO Gumprecht	HW32753Del
1954	Ru	Review of potential Ru problem at PUREX	RE Tomlinson FJ Leitz, Jr.	HW-33479
1953 and 1954	Ru	Appraisal of problems and recommended remedies; REDOX ruthenium	ER Irish	HW-34882
1955	Ru	Progress report on Ru from REDOX	GL Helgeson HF Soule	HW34817

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	<u>Reference Author</u>	<u>Document</u>
1955	I-131	B and T Plant iodine problems of measurement and control	WG Browne	HW36112
1955	I-131	Flow sheet for improved iodine emission control	RE Smith	HW36819
1955		Thermodynamics of iodine removal with silver reactors	LL Burger	HW38311Rev
1955	I-131	Symposium on iodine problems; abstracts of papers presented	OF Hill, et al.	HW39073
1955	I-131	Series of 11 dissolutions to test I-131 removal by two silver reactors in a series	WG Browne HP Maffei	HW39485
1955		Views on I-131 emission limits	HM Parker	HW36207
1956 Apr & May	I-131	T-Plant iodine emission measurements	GL Helgeson	HW44879
1956	NO _x	Good report of oxides of nitrogen released from REDOX and PUREX. Includes sample methods.	CP Skillern	HW-51667
1957	Noble gases	Gases in reactor effluent water; no data	LA Carter	HW50138
1957	Ru	Performance study of PUREX deep bed fiberglass filter	CP Skillern	HW-52789
1952 to 1957		Summary of environmental contamination incidents (airborne materials)	JM Selby JK Soldat	HW-54636
1957	I-131	Investigations of PUREX silver reactor incident	OC Schroeder	HW-55223
1952 to 1957	I-131, Ru	History of 200 Area airborne contamination and control efforts	RE Roberts	HW-55569RD
1962		Status of gaseous effluent monitoring	D McConnon	HW69205Rev
1962	I-131	Sampling systems at REDOX and PUREX; description and efficiency	JD McCormack	HW73288

TABLE A.2. Tritium Releases from Hanford

<u>Time Period</u>	<u>Reference</u>	
1953	DG Sturges	HAN 51086
	RW Cook	HAN 51270
	RW Cook	HAN 51808
	DG Sturges	HAN 52165
	DG Sturges	HAN 53113
	CA Priode	HAN 53178
1954	DG Sturges	HAN 53327
	CA Priode	HAN 53349
	DG Sturges	HAN 53450
	RW Cook	HAN 53887
	CA Priode	HAN 53978Del
	DG Sturges	HAN 54002Del
	DG Sturges	HAN 54114Del
	SG English	HAN 54172Del
	SG English	HAN 54282Del
	AT Gifford	HAN 54289Del
	AT Gifford	HAN 54344
	CA Priode	HAN 54514
	DG Sturges	HAN 54549
	DG Sturges	HAN 54644
	CA Priode	HAN 54892Del
	DG Sturges	HAN 54956
	CA Priode	HAN 55656Del
DG Sturges	HAN 55694Del	
CA Priode	HAN 56028	
AT Gifford	HAN 56157	
1955	GM Kavanagh	HAN 60373
1955	AT Gifford	HAN 60517

TABLE A.3. Regional Monitoring Reports

All of these documents included total alpha, total beta measurements on an evaporated sample of reactor effluent water. These documents also included data on release to the atmosphere. Early reports include estimates of iodine-131 in metal charged into dissolvers. Later reports show measured iodine-131 release data, ruthenium release data from 200 Area stacks, and measurements of tritium, carbon-64, and sulfur-35 released from reactor stacks.

<u>Time Period</u>	<u>Comments</u>	<u>Author</u>	<u>Reference</u>	<u>Document</u>
All of 1945	Weekly reports bound together	CC Gamertsfelder		HW7-1115
All of 1946	Annual summary report	LD Turner		HW3-5402
First, 1947		LD Turner		HW-3-5511
Second, 1947		JW Healy		HW-7317
Third, 1947		W Singlevich		HW-8549Del
Fourth, 1947		W Singlevich		HW-9496Del
First, 1948		W Singlevich		HW-10242Del
Second, 1948		W Singlevich		HW-11333Del
Third, 1948		W Singlevich		HW-12677Del
Fourth, 1948		W Singlevich, HJ Paas		HW-13743
First, 1949		W Singlevich, HJ Paas		HW-14243Del
Second, 1949		W Singlevich, HJ Paas		HW-17434Del
Third, 1949		W Singlevich, HJ Paas		HW-18615
Fourth, 1949	Includes I-131 from "green run"	W Singlevich, HJ Paas		HW-17003Del
First, 1950		W Singlevich, HJ Paas		HW-18446
Second, 1950		W Singlevich, HJ Paas		HW-19454
Third, 1950		W Singlevich, HJ Paas		HW-20700
Fourth, 1950		W Singlevich, HJ Paas		HW-21566
First, 1951		W Singlevich, HJ Paas		HW-21214
Second, 1951		W Singlevich, HJ Paas		HW-22313
Third, 1951		W Singlevich, HJ Paas		HW-23133
Fourth, 1951		W Singlevich, HJ Paas		HW-24203
First, 1952		HJ Paas		HW-25866
Second, 1952		HJ Paas		HW-26493
Third, 1952		HJ Paas		HW-27510
Fourth, 1952		HJ Paas		HW-27641
First, 1953		HJ Paas		HW-28009
Second, 1953		HJ Paas		HW-29514
Third, 1953		HJ Paas		HW-30174
Fourth, 1953		HJ Paas		HW-30744

<u>Time Period</u>	<u>Comments</u>	<u>Author</u>	<u>Document</u>
First, 1954		HJ Paas	HW-31818
Second, 1954		HJ Paas, GE Pilcher	HW-33896
Third, 1954		GE Pilcher, JK Soldat, ZE Carey	HW-36504
Fourth, 1954			HW-36505
First, 1955			HW-36506
Second, 1955			HW-38566
Third, 1955			HW-39429
Fourth, 1955			HW-40871
First, 1956		BV Andersen, JK Soldat, MW McConiga	HW-43012
Second, 1956		BV Andersen, JK Soldat, MW McConiga	HW-44215
Third, 1956		BV Andersen, JK Soldat, MW McConiga	HW-46726
Fourth, 1956		BV Andersen, JK Soldat, MW McConiga	HW-48374
First, 1957		MW McConiga, JK Soldat, JM Selby	HW-51009
Second, 1957		MW McConiga, JK Soldat, JM Selby	HW-52803
Third, 1957		MW McConiga, JK Soldat, JM Selby	HW-54841
Fourth, 1957		MW McConiga, JK Soldat, JM Selby	HW-54842

TABLE A.4. Routine Sampling and Counting Methods

<u>Time Period</u>	<u>Data</u>	<u>Comments</u>	Reference <u>Author</u>	<u>Document</u>
1948		Routine methods for radiochemical analysis	SA Reynolds	HW13622
1949		Standard Practices for survey work in 100, 200 and 300 Areas	JM Smith, Jr.	HW46104
1950		Sample counting methods	JW Healy LC Schwendiman RC Thorburn	HW18258
1951		Radiochemical analytical procedures	JW Healy RC Thorburn ZE Carey	HW20136
1951		Calculation of conversion factors used in counting room	JH Wolff	HW22682
1952		Alpha counting procedures	JH Wolff	HW23769
1954		Standard Practices counting manual	LC Schwendiman	HW30492
1956		Standard Practices radiochemical calculation manual	HT Norton PO Jackson DL Reid	HW46436
1957		Regional monitoring program	BV Andersen JK Soldat	HW52038

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