

**FINAL**

**ATMOSPHERIC SOURCE TERMS  
FOR THE IDAHO CHEMICAL  
PROCESSING PLANT, 1957–1959**

Contract No. 200-2002-00367  
Task Order No. 1, Subtask 1

A final report to the  
Centers for Disease Control and Prevention  
Atlanta, Georgia 30335

SC&A, Inc.  
6858 Old Dominion Drive, Suite 301  
McLean, Virginia 22101

SENES Oak Ridge, Inc.  
102 Donner Drive  
Oak Ridge, Tennessee 37830

Authors:

Robert P. Wichner, *SENES* Oak Ridge, Inc.  
John-Paul Renier, *SENES* Oak Ridge, Inc.  
A. Iulian Apostoaei, *SENES* Oak Ridge, Inc.

July 2005

---

## TABLE OF CONTENTS

EXECUTIVE SUMMARY .....	ES-1
1.0 SCOPE AND APPROACH .....	1-1
1.1 Scope .....	1-1
1.2 Approach.....	1-1
1.2.1 Operational RaLa Releases .....	1-1
1.2.2 Idaho Chemical Processing Plant Criticality Approach.....	1-2
2.0 THE RADIOACTIVE LANTHANUM (RaLa) PROCESS .....	2-1
2.1 Background .....	2-1
2.1.1 Requirement.....	2-1
2.1.2 Work at Oak Ridge National Laboratory.....	2-1
2.1.3 Early Process Decisions .....	2-2
2.1.4 The RaLa Development Program at the Idaho Chemical Processing Plant .....	2-2
2.2 Idaho RaLa Process Reporting.....	2-4
2.2.1 Primary Sources of Information.....	2-4
2.2.2 Secondary Sources .....	2-4
2.3 Chemical Flow Sheet .....	2-4
2.4 Process Equipment.....	2-9
2.5 Iodine and Other Fission Product Distribution Tests.....	2-17
2.6 Operational History and Process Modifications .....	2-20
2.7 Off-Gas Flow System .....	2-23
2.7.1 The Idaho Chemical Processing Plant Ventilation and Off-Gas System.....	2-23
2.7.2 Description of the Dissolver .....	2-25
2.7.3 Description of the Scrubber .....	2-28
2.7.4 Operation of the Charcoal Beds.....	2-28
2.8 Summary and Conclusions .....	2-28
3.0 IODINE BEHAVIOR IN THE RALA SYSTEM .....	3-1
3.1 Radioactive Properties of Iodine Radionuclides.....	3-1
3.1.1 Birth and Decay Diagram .....	3-1
3.1.2 Iodine and Tellurium Inventory in the Feed .....	3-3
3.1.3 Radioiodine Gamma Ray Emissions .....	3-3
3.2 Iodine Behavior in the Caustic Dissolver .....	3-6
3.3 Iodine Behavior in the Centrifuge and Acidified Waste Tank .....	3-7
3.4 Iodine Behavior in the Scrubber—Effect of Sodium Thiosulfate .....	3-7
3.5 Mercury Complexing in Acid Solutions .....	3-8

---

---

**TABLE OF CONTENTS (continued)**

3.6	Behavior of Iodine in the Stack Monitor Sampler .....	3-9
3.7	Tellurium Chemistry in the Alkaline and Acid Dissolvers.....	3-9
3.8	Production of Organic Iodide.....	3-10
3.9	Summary and Conclusions .....	3-11
4.0	<b>RADIOIODINE PRODUCTION IN THE MATERIALS TESTING REACTOR, 1957–1959.....</b>	<b>4-1</b>
4.1	Introduction.....	4-1
4.2	Materials Testing Reactor Description .....	4-2
4.3	Estimation of Barium-140 and Radioiodine Production in the Materials Testing Reactor .....	4-5
	4.3.1 1957 Method .....	4-5
	4.3.2 Isotope Buildup Using ORIGEN2 .....	4-8
4.4	RaLa Fuel Element Irradiation Histories .....	4-9
	4.4.1 Documented RaLa Fuel Elements and Irradiation Histories .....	4-9
	4.4.2 Surmised RaLa Fuel Elements and Irradiation Histories.....	4-9
	4.4.3 Recreated Rala Fuel Elements and Irradiation Histories .....	4-12
4.5	ORIGEN2 Results.....	4-19
	4.5.1 Radioiodine Production .....	4-19
4.6	Reported Iodine-131 Production in the Materials Testing Reactor .....	4-20
5.0	<b>THE STACK MONITOR .....</b>	<b>5-1</b>
5.1	Introduction.....	5-1
5.2	General Features of the Stack Monitor System .....	5-1
	5.2.1 Flow Diagram .....	5-1
	5.2.2 System Operation and the Iodine Sampler.....	5-3
	5.2.3 Early Stack Monitor Experience .....	5-5
	5.2.4 Relation of Stack Sampler Activity to Release.....	5-5
5.3	The Stack Monitor Datasheets .....	5-7
	5.3.1 General Features .....	5-7
	5.3.2 Sample Datasheets .....	5-8
5.4	Stack Monitor System Evaluation .....	5-13
	5.4.1 Evaluation of Sampler Efficiency.....	5-13
	5.4.2 Iodine Deposition Error .....	5-18
	5.4.3 Effect of Variable Iodine Release .....	5-20
	5.4.4 Possible Radiation Detection Errors .....	5-21
5.5	Summary and Conclusions Regarding the Stack Monitor Datasheets.....	5-23
	5.5.1 Summary of Stack Sampler Error Evaluation.....	5-23
	5.5.2 Conclusions.....	5-24

## TABLE OF CONTENTS (continued)

6.0	IODINE RELEASES FROM THE RALA PROCESS, FEBRUARY 1957 TO DECEMBER 1959.....	6-1
6.1	Early Iodine Releases.....	6-1
6.2	Existing Reports of Iodine Releases from the RaLa Process.....	6-1
	6.2.1 The Hayden Notegrams .....	6-1
	6.2.2 The Health and Safety Division Annuals.....	6-2
	6.2.3 The Historical Dose Evaluation Study.....	6-2
6.3	Iodine, Tellurium, and Xenon Nuclides Fed to the Dissolver .....	6-4
6.4	Recommended Daily Releases of Iodine-131 .....	6-5
6.5	Evaluation of Iodine-132 Release Data .....	6-5
	6.5.1 Cases of High Iodine-132 Release Readings .....	6-5
	6.5.2 Possible Contributing Factors to the Observed High Iodine-132 Releases .....	6-6
	6.5.3 Extrapolating Iodine-132 Readings Back to February 1957.....	6-7
6.6	Estimation of Iodine-133 Releases .....	6-9
6.7	The Complete Daily Release Record.....	6-11
6.8	Comparison of the Recommended Releases with the Historic Dose Evaluation Study (DOE 1991).....	6-12
6.9	Fractional Releases per Run.....	6-12
6.10	Error Range of Releases.....	6-15
	6.10.1 Error Range of Iodine-131 Releases .....	6-15
	6.10.2 Error Range of Iodine-132 Releases .....	6-16
	6.10.3 Error Range of Iodine-133 Releases .....	6-16
6.11	Summary of Chapter 6 .....	6-16
7.0	CRITICALITY ACCIDENT, OCTOBER 16, 1959.....	7-1
7.1	Description of the Criticality Accident .....	7-1
	7.1.1 Timing and Location.....	7-1
	7.1.2 Material Transfers During and Post Event.....	7-1
7.2	Estimated Number of Fissions .....	7-5
	7.2.1 Radiochemical Analyses Results .....	7-5
	7.2.2 Possible Errors in Estimating the Severity of the Event.....	7-7
7.3	Leakage Pathways from the Idaho Chemical Processing Plant Building .....	7-7
7.4	The Stack Monitor Record Following the Accident .....	7-8
	7.4.1 The Iodine-131 and Iodine-132 Record.....	7-8
	7.4.2 Search for $\gamma$ -Interferences with the Iodine-132 Reading .....	7-9
7.5	Radionuclide Production and Decay.....	7-10
	7.5.1 Radioiodine Production and Decay .....	7-10
	7.5.2 Radiobromine Production and Decay .....	7-11
7.6	The Post-Criticality Iodine Source Term .....	7-12

## TABLE OF CONTENTS (continued)

	7.6.1 Iodine Release Model .....	7-12
	7.6.2 Measured Iodine-131 Releases Due to the Criticality .....	7-14
	7.6.3 Releases of the Other Radioiodines .....	7-14
7.7	Radiobromine Release Following the Criticality.....	7-15
7.8	Xenon Release Following the Criticality.....	7-16
7.9	Krypton Release Following the Criticality .....	7-19
7.10	Comparison of Criticality Accident Releases with the Idaho National Engineering Laboratory Historic Dose Evaluation .....	7-20
	7.10.1 Similarities/Differences in Approach .....	7-20
	7.10.2 Comparison of Releases.....	7-21
8.0	UNCERTAINTY ANALYSIS FOR THE ESTIMATED RELEASES OF IODINE FROM THE IDAHO CHEMICAL PROCESSING PLANT. ....	8-1
	REFERENCES .....	R-1

## APPENDICES

APPENDIX A	REFERENCES: RALA PROCESS DESIGN AND OPERATION
APPENDIX B	SAMPLE ORIGEN2 OUTPUT, MATERIALS TESTING REACTOR IRRADIATION FOR RALA RUN 2, 2-DAY COOLING TIME
APPENDIX C	BEHAVIOR OF TELLURIUM IN THE ALKALINE AND ACID DISSOLVERS
APPENDIX D	NOBLE GAS, IODINE, AND TELLURIUM PRODUCTION IN THE IDAHO CHEMICAL PROCESSING PLANT CRITICALITY ACCIDENT OF OCTOBER 16, 1959
APPENDIX E	THE HAYDEN DAILY RELEASE NOTEGRAMS
APPENDIX F	DAILY RELEASES OF IODINE-131, IODINE-132, AND IODINE-133 FROM FEBRUARY 1, 1957, TO DECEMBER 30, 1959— DETERMINISTIC VALUES

## LIST OF TABLES

Table ES-1	Summary of monthly radioiodine releases (Ci) based on estimated daily releases .....	ES-11
Table ES-2	Percent of iodine-131 released for selected RaLa runs.....	ES-13
Table ES-3	Activities of iodine isotopes produced during October 16, 1959, accident, as estimated using ORIGEN2 computer code .....	ES-14
Table ES-4	Estimated radioiodine releases (Ci) from 1959 criticality accident at the Idaho Chemical Processing Plant.....	ES-15
Table ES-5	Estimated releases of all isotopes of bromine, krypton, and xenon from 1959 criticality accident at the Idaho Chemical Processing Plant .....	ES-15
Table ES-6	Estimated atmospheric releases of radioactive iodine from Idaho Chemical Processing Plant during 1957–1959 (Ci).....	ES-18
Table 2-1	Nominal RaLa feed material .....	2-5
Table 2-2	Percent of selected fission products dissolved into the caustic supernate .....	2-18
Table 2-3	Iodine-131 material balance* for several early RaLa runs.....	2-18
Table 2-4	Distribution of iodine-131 in RaLa process streams with estimated release to the stack .....	2-19
Table 2-5	Record of RaLa runs and operational changes .....	2-21
Table 2-6	Data for initial runs involving irradiated fuel .....	2-23
Table 3-1	Typical iodine and tellurium inventories in 2-day cooled Materials Testing Reactor fuel (curies).....	3-3
Table 3-2	Principal radioiodine gamma ray emissions .....	3-5
Table 3-3	Effect of mercury concentration on iodine removal by air sparging .....	3-8
Table 4-1	Fuel element data for the first five RaLa runs .....	4-9
Table 4-2	Brief review of fuel cycle data of the candidate Materials Testing Reactor fuel elements used in the analyses of the RaLa runs .....	4-15
Table 4-3	Radioiodines fed to the dissolver determined from ORIGEN2.....	4-20
Table 4-4	Activity of iodine-131 (Ci) fed to the dissolver.....	4-21
Table 5-1	Emendations to the iodine-131 release record of February 1957 relative to the datasheets .....	5-10
Table 5-2	Emendations to the iodine-131 release record of March 1957 relative to the datasheets .....	5-12
Table 5-3	Efficiency determinations of the “Production” Sampler, Location D .....	5-17
Table 5-4	Sampler efficiency determinations for the temporary sampler.....	5-17
Table 5-5	Effect of iodine deposition on estimated releases, April, March, and June 1957 tests .....	5-19
Table 6-1	Curies released per RaLa run, reported in DOE (1991).....	6-3
Table 6-2	Iodine, tellurium, and xenon nuclides fed to the dissolver per typical 2-day cooled run* (including precursor tellurium) in curies.....	6-4

## LIST OF TABLES (continued)

Table 6-3	Examples of high iodine-132 daily releases, in curies.....	6-6
Table 6-4	Comparison of iodine releases with historic dose evaluation.....	6-13
Table 6-5	Percentage of iodine-131 per run.....	6-14
Table 7-1	Total number of fissions determined from the molybdenum-99, barium-140, and lanthanum-140 analyses.....	7-6
Table 7-2	Daily release record of iodine-131 and iodine-132 from October 13–23, 1959.....	7-8
Table 7-3	Summary of radioiodines considered in this study.....	7-10
Table 7-4	Iodine-131 releases due to the criticality accident.....	7-14
Table 7-5	Releases per day (Ci) of iodine-132, iodine-133, iodine-134, and iodine-135 following the Idaho Chemical Processing Plant criticality of October 16, 1959.....	7-15
Table 7-6	Summary of radiobromine considered in this study.....	7-15
Table 7-7	Radiobromine releases (Ci) following the criticality.....	7-15
Table 7-8	Summary of radioxenons considered in this study.....	7-16
Table 7-9	Releases per day (Ci) of xenon-133, xenon-135m, xenon-135, xenon-137, and xenon-138 following the Idaho Chemical Processing Plant criticality of October 16, 1959.....	7-18
Table 7-10	Short-lived xenons contributing to an occupational immersion dose.....	7-19
Table 7-11	Releases per day (Ci) of krypton-83m, krypton-85m, krypton-87, krypton-88, and krypton-89 following the Idaho Chemical Processing Plant criticality of October 16, 1959.....	7-19
Table 7-12	Short-lived radiokryptons, greater than 100 Ci at 1 minute.....	7-19
Table 7-13	Total (all days) halogen and noble gas release comparison: this study versus DOE (1991).....	7-22
Table 8-1	Estimated atmospheric releases of radioactive iodine from the Idaho Chemical Processing Plant during 1957–1959 (Ci).....	8-2

## LIST OF FIGURES

Figure ES-1	Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), February 1957 .....	ES-8
Figure ES-2	Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), June 1958 .....	ES-9
Figure ES-3	Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), August 1958 .....	ES-9
Figure ES-4	Daily releases of iodine-131 during January 1958 and after the 1959 criticality accident adjusted by the uncertain bias correction factor. ....	ES-17
Figure 2-1	Simplified chemical flowsheet.....	2-6
Figure 2-2	Illustration of air-lifting RaLa solutions .....	2-8
Figure 2-3	RaLa equipment layout in L-Cell .....	2-11
Figure 2-4	Dissolution flowsheet.....	2-12
Figure 2-5	Detailed flowsheet — centrifugation .....	2-14
Figure 2-6	Centrifuge details .....	2-15
Figure 2-7	South face of cell L — access corridor level .....	2-16
Figure 2-8	RaLa main control panel — access corridor level .....	2-17
Figure 2-9	Photograph of gas flows into the stack as of 1973.....	2-24
Figure 2-10	RaLa off-gas flows downstream from the scrubber.....	2-26
Figure 2-11	RaLa and Idaho Chemical Processing Plant off-gas flow schematic, 1957–1959.....	2-27
Figure 3-1	Iodine birth and decay diagrams .....	3-2
Figure 4-1	Mass-140 production from thermal fissions in uranium-235.....	4-1
Figure 4-2	Cross section of fuel assembly.....	4-2
Figure 4-3	Fuel-assembly units .....	4-3
Figure 4-4	Horizontal section through Materials Testing Reactor and core arrangement ....	4-4
Figure 4-5	View of the south and west faces of the Materials Testing Reactor .....	4-5
Figure 4-6	Illustration of RaLa fuel element irradiation history .....	4-6
Figure 4-7	One page of a data book illustrating calculations of barium-140 and iodine-131 content of Materials Testing Reactor elements.....	4-7
Figure 4-8	Core map for Charge 82C .....	4-11
Figure 4-9	Irradiation record for Charges 82A, 82B, and 82C.....	4-13
Figure 4-10	Reduced information regarding core loading following June 1958.....	4-14
Figure 5-1	Stack sampling system configuration during sampler tests, April–June 1957.....	5-2
Figure 5-2	Early stack monitor systems — February 1957 (top) and April 1957 revisions.....	5-4
Figure 5-3	Stack monitor datasheet for February 1957 from document MC 60312 .....	5-9
Figure 5-4	Iodine-131 daily releases during February 1957 .....	5-11
Figure 5-5	Stack monitor datasheet for May 1958 from document MC 60312 .....	5-14

---

**LIST OF FIGURES (continued)**

Figure 5-6	Iodine-131 and iodine-132 releases following Run 14 started on May 28, 1958 .....	5-15
Figure 5-7	A portion of a datasheet for April 1957 showing the determination of sampler efficiency .....	5-16
Figure 5-8	Detection error due to sample strength .....	5-22
Figure 6-1	Observed iodine-132/iodine-131 release ratios for four runs .....	6-8
Figure 6-2	Observed iodine-132/iodine-131 release ratios for three consistent runs .....	6-9
Figure 6-3	Iodine-133/iodine-131 ratio estimated for a typical Materials Testing Reactor irradiation .....	6-11
Figure 7-1	Cross section of the Process Building CPP-601 looking north from the waste tank rooms .....	7-2
Figure 7-2	Continuous air monitor chart in the storage vessel room .....	7-3
Figure 7-3	Waste tank interconnect .....	7-4
Figure 7-4	Daily releases of iodine-131 following RaLa Run 34, October 13, 1959 .....	7-9
Figure 7-5	Inventory of radioiodines produced during the criticality and its evolution with time for 8 days after the criticality .....	7-11
Figure 7-6	Inventory of radiobromines as a function of time after criticality .....	7-12
Figure 7-7	Inventory of long-lived xenons as a function of time after the criticality .....	7-17
Figure 7-8	Inventory of intermediate-lived xenons as a function of time after the criticality .....	7-17
Figure 7-9	Inventory of radiokryptons as a function of time after the criticality .....	7-18
Figure 8-1	Daily releases of iodine-131 during January 1958 and after the 1959 criticality accident, adjusted by the uncertain bias correction factor .....	8-3

## EXECUTIVE SUMMARY

### Introduction

This report provides a study of atmospheric release of gaseous radionuclides and radioactive aerosols (in the Addendum) from the Idaho Chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory (INEL) during the 3-year period, February 1957 through December 1959. Two earlier studies, the INEL Historical Dose Evaluation (HDE) sponsored by the Department of Energy (DOE, 1991) and a scoping study sponsored by the Centers for Disease Control and Prevention (Till, 2000), indicated that the principal hazards were radioiodine emissions, particularly I-131, from the operation of the Radioactive Lanthanum (RaLa) process within the ICPP. The 3-year period of study included the initial operating years of the RaLa process during which the highest emissions occurred.

RaLa processed fully irradiated, short-cooled fuel from the Materials Testing Reactor (MTR) to recover the radioactive fission product, Ba-140. The product was shipped to Los Alamos for diagnostic purposes. Since only RaLa processed short-cooled fuel, all radioiodine emissions detected in the ICPP off-gas were due to RaLa.

The reported routine releases of I-131 are based on measurements by a sampling system installed in the ICPP off-gas stack. Less important and less certain are the measured releases of I-132. Iodine-133 releases were not measured, but were determined in this study by a correlation.

Routine releases of radioactive aerosols are reported in the Addendum to the main report. Aerosol measurements were of two types, alpha emitters captured on filters and so-called b-I (beta minus iodine) readings resulting from solid fission products captured by the iodine sampler. Aerosol releases were long-lived actinides and fission products that may have emanated from any process within the ICPP, not necessarily RaLa.

Emissions due to the criticality accident on October 16, 1959, at the ICPP are given in the latter portion of the main report. Shorter-lived iodine and bromine isotopes released during this event are reported.

### Approach to Estimating Routine Releases from RaLa Process

To the extent that is possible, the intention in this study is to report releases based on verifiable, direct measurements. By verifiable, it is meant that the data sources are primary reports or documents (i.e., written contemporaneously by project personnel) describing methodology in sufficient detail for evaluation. Fortunately, such information is available for I-131, the key hazard nuclide, from the Stack Monitor Datasheets (SMDs).

Estimation of releases becomes progressively less certain when such verifiable, primary data are not available and other means of estimation must be resorted to.

## **Approach to Estimating Releases from the 1959 Criticality Accident**

The approach to estimating releases from the 1959 criticality accident at the ICPP involved three steps.

The first step was to obtain a complete description of the accident, including identification of the vessels involved in the criticality event, movements of the solution from vessel to vessel, and the release pathways from the vessels to the atmosphere.

The second step was to verify a previous estimate of the number of fissions during the criticality event by evaluating possible sources of error in the methodology.

Finally, based on estimated releases of I-131 from stack monitoring data and a calculation of the inventory of I-131 produced in the accident, a model is developed to estimate releases of I-132, I-133, I-134, I-135, and isotopes of bromine, krypton, and xenon. Measured releases of I-132 during the accident appear to be erroneous.

## **Description of the RaLa Process**

Knowledge of RaLa process details is helpful in rationalizing the measured releases, especially for interpreting daily variations. A complete description of the chemical and mechanical features of the process at start-up is available from pre-operational design reports. However, only unreferenced, secondary reports were found describing the design and operational changes instituted after operation commenced. Therefore, we have only vague and unverified information on the process changes made during early operation that ultimately succeeded in reducing emissions.

Features of the RaLa process described in this report include the chemistry of the process, the equipment used in the process, and the off-gas system. Also included are all test data that we were able to locate on the behavior of iodine in the system. Based on the available information, the RaLa process is chemically and mechanically complex, and estimates of radioiodine releases based on principles of chemistry and mass transfer would be highly uncertain even with full knowledge of the process variables.

It is apparent from the schedule of early development activities that the final design of the RaLa process, which was initiated in July 1954, could not have accounted for all the process features that were tested in cold pilot plant or hot laboratory tests. As a result, deficiencies in the process were revealed immediately upon startup of operations in February 1957, and process modifications continued through 1959.

In particular, examination of the early development work indicates that the behavior of iodine in the system was only minimally studied and was not clearly understood. Design assumptions regarding the purported high retention of iodine in the caustic supernate in the dissolver evidently were not realized.

An important early design deficiency was the weak seals on each centrifuge shaft. The centrifuge bowls, which were used as process vessels, were effectively unsealed during early

RaLa runs, with the result that much of the iodine evolved downstream from the caustic dissolver and entered the cell air system, thus bypassing the scrubber.

Various sources of information indicate that several modifications directed at increasing the retention of iodine were instituted after the start of RaLa operations in February 1957:

- Centrifuge seals were improved to reduce leakage from this process vessel into the cell air that bypassed the scrubber.
- Process temperatures were lowered to reduce volatility.
- The frequency of sampling was reduced (each sample required agitation by a nitrogen jet, which not only added gas volume to the off-gas removal system but also enhanced volatilization of dissolved gases).
- Charcoal absorber beds were added to the RaLa dissolver off-gas system to reduce iodine emissions.
- Mercury nitrates were added to the acid solution to promote sequestering of iodine in the waste tanks.

All gaseous radionuclides released from processes in the main ICPP process building entered into either the ICPP dissolver off-gas system, the RaLa dissolver or vessel off-gas systems, or the ICPP cell air system. All such systems were routed to the stack by various paths. Thus, all iodine released from the ICPP was counted by the stack monitor system. However, possible leakage into the off-gas cell that housed the charcoal beds would have bypassed the stack monitor.

An additional feature of the off-gas system was a 10,000-ft<sup>3</sup> gas storage vessel, which was installed in early 1957 and connected to the RaLa dissolver off-gas line. The intention was to feed RaLa off-gas into this vessel during periods of calm winds, and then to release stored gases to the atmosphere during periods of high dispersion. Thus, it is not possible to correlate releases with RaLa operations without knowledge of operation of this storage vessel. However, we have found no record of its operation or the time period during which it was an active part of the system.

### **Behavior of Iodine in the RaLa Process**

Although the behavior of iodine in the RaLa process vessels is generally understood, more detailed information would be required to enable quantitative estimates of flows between vessels and volatilization to the off-gas systems.

The general principles of iodine behavior in sodium hydroxide solutions, the effect of acidification, the role of sodium thiosulfate in enhancing iodine solubility in basic solutions, and the effect of using mercury salts on sequestering of iodine in acid solutions are briefly reviewed in this report. The selection of sodium hydroxide plus sodium thiosulfate solution as a stack

sampler fluid was appropriate, and difficulties with using sodium hydroxide alone, which became apparent later, were avoided. However, in a few cases where data on the behavior of iodine in the system are available, it is apparent that the actual behavior did not conform completely to predictions.

Sodium acetate used in one of the separations steps of the RaLa process may have contributed to formation of organic iodide compounds, although most of the iodine was removed from the process prior to this step. Use of a 5% nitric acid solution in the scrubber beginning in March 1959 should have been effective in removing iodine in organic form.

Unusually high releases of I-132 were observed on the first day of RaLa runs. No clear chemical basis for these releases could be established. A difference in solubility of the longer-lived precursor Te-132 and iodine, which could explain the high releases of I-132, is not supported by an evaluation of tellurium chemistry presented in this report.

### **Radioiodine Production in the Materials Testing Reactor**

The feed for the RaLa process were fuel elements irradiated in the MTR. One fuel element was used per RaLa run. When sufficient information on the fuel element irradiation history was available, radionuclide inventories in the fuel elements were estimated. However, the MTR progress reports do not identify the RaLa elements. Irradiation histories were discerned with some uncertainty (by comparing discharge dates with the run schedule and assuming 2-day cooling) for a few of the feed elements when the partial information permitted.

The intention was to support estimates of iodine releases based on stack monitoring data by calculating an iodine mass balance for each RaLa run. In addition, essential information on cooling times from reactor shutdown to insertion into the RaLa process were found for only 6 of the 37 RaLa runs during the 3-year period of interest.

Nonetheless, sufficient core loading data are given in early MTR progress reports to allow a re-creation, with difficulty and considerable uncertainty, of the irradiation history of the first 14 RaLa runs up to May 1958. Estimation of radioiodine inventories was not possible for the remaining 23 runs. Re-creation of irradiation histories is a complex undertaking, owing to the operational characteristics of the MTR and the need to service other programs. Typically, a fuel element destined for the RaLa process may have been irradiated up to 12 different times in 4 or 5 different core locations.

Contemporary methods of estimating inventories of Ba-140, I-131, and other key radionuclides in selected fuel elements were based on assuming a uniform flux of neutrons in the reactor and fission cross sections for thermal neutrons. These methods were somewhat approximate. When information on irradiation history was available, the method of estimating radioiodine inventories in discharged MTR fuel elements used in this report is based on a standard isotopes production code, ORIGEN2. It was first necessary to model the MTR core to determine the variation in neutron flux and the proper spectrum-averaged fission cross sections. Inventories of I-131, I-132, I-133, and I-135 in feed materials are reasonably well established for the first 5 RaLa runs, but the inventories in the remaining 8 of the first 13 RaLa runs are somewhat uncertain, due to the need to make assumptions about the irradiation and decay times.

## Stack Monitor System

Daily radioiodine releases estimated in this study are based on information given in SMDs. SMDs give estimates of I-131 releases that were based on measurements of activity in the stack sampler beginning with the first RaLa run on February 1, 1957, and continuing through December 1959 (and beyond). I-132 releases were reported beginning in May 1958. Other radioiodines were not monitored.

The SMDs were the primary source of data reproduced in later reports. Letter summaries of daily releases began in July 1957, in internal letters called Hayden Notegrams. These undoubtedly were the unreferenced sources of data used in the Health and Safety Division Annual Reports, and were subsequently adopted in DOE's HDE published in 1991.

Effluents routed to the stack consisted of cell ventilation exhaust, ICPP vessel off-gas, ICPP dissolver off-gas, and a separate line for RaLa dissolver off-gas. Numerous interconnections between off-gas lines were possible, leading to some contradictions in available documentation, but it is most likely that the valving was set so that all RaLa vessels vented into the RaLa dissolver off-gas line. The significance of this is that the RaLa dissolver off-gas discharged through the iodine scrubber and through the charcoal beds after June 1958, whereas the ICPP vessel off-gas and dissolver off-gas did not. Since no other ICPP dissolver processed fuel with short cooling times, all I-131 and I-132 monitored in the stack may be attributed to RaLa runs.

The stack monitor system used in the 1957-1959 time frame is not described in any available project reports. Characteristics of an early version of the system used up to November 1957 can be inferred from project letters and information in the SMDs. No description of the stack monitor system after mid-1957 has been found.

Initially, stack monitoring was based on a flow system in which the activity of the collected iodine solution was continually measured by a scintillation detector. However, due to difficulties of operation, this method was soon replaced by a static system in which samples were collected over a 24-hour period and counted on a daily basis. Activities of I-131 and I-132 in stack releases were estimated based on the activities in samples collected in the 24-hour static sampler by assuming a constant source over the 24-hour sampling period, a collection efficiency of 50%, and measured flows in the stack gas relative to the sampler line. The assumed sampler efficiency of 50% appears to have been an early administrative decision.

## Evaluation of Sources of Error in the Stack Monitor System

Despite the lack of published reports and the absence of information after mid-1957, a reasonable evaluation of the reliability of the stack monitor system can be based on internal letters and notes included among the SMDs. The factors that may contribute to errors in estimating activities of I-131 and I-132 released include:

- Variations in iodine capture efficiency
- An unknown degree of iodine loss in the sampler lines

- Variations in iodine release rates over a 24-hour sampling period
- Impurities in the sample that emit gamma rays that interfere with the primary gamma rays emitted in decay of I-131 and I-132

Our evaluation of these possible sources of error in the stack monitor system is summarized as follows:

- The assumed iodine capture efficiency of 50% appears to be reasonable based on measurements conducted in 1957 and reported in project letters and SMDs.
- Evaluation of losses of iodine due to deposition in the sampler lines is inconclusive. Reported test results, although they may not be definitive, do not show an effect due to deposition in the sampler line. Shortening of the sampler line to about 50 feet, which may have occurred in November 1957, presumably had a beneficial effect in reducing iodine losses.
- Measurements of I-131 are relatively insensitive to variations in release rates over a 24-hour sampling period, due to the relatively long half-life of this isotope. In contrast, measurements of I-132 (half-life of 2.3 hours) are highly sensitive to variations in release rates during the sampling period. A peaking of I-132 releases early in the 24-hour sampling period, for example, would have resulted in significant underestimates of the total activity released during that time. Since radioactive decay is exponential in time and I-132 has a short half-life compared with the sampling period, variations in release rates of I-132 would, in most cases, result in an underestimate of the total release over 24 hours.
- No significant interferences from gamma rays emitted by other radionuclides have been found that could have affected measurements of I-131 and I-132 by gamma spectroscopy to a significant extent. The high I-132 readings on the days dissolving of fuel in RaLa runs occurred are not explained.

This evaluation leads to a conclusion that the stack sampler appears to provide a satisfactory basis for estimating daily I-131 releases to the atmosphere.

### **Estimated Releases of Radioiodines from the RaLa Process Operations— Deterministic Results**

Corrected daily releases of I-131 from February 1, 1957, to December 30, 1959, obtained from SMDs, are tabulated in this report. When there was no reported release for a given day, the release is estimated by interpolation of readings for adjacent days.

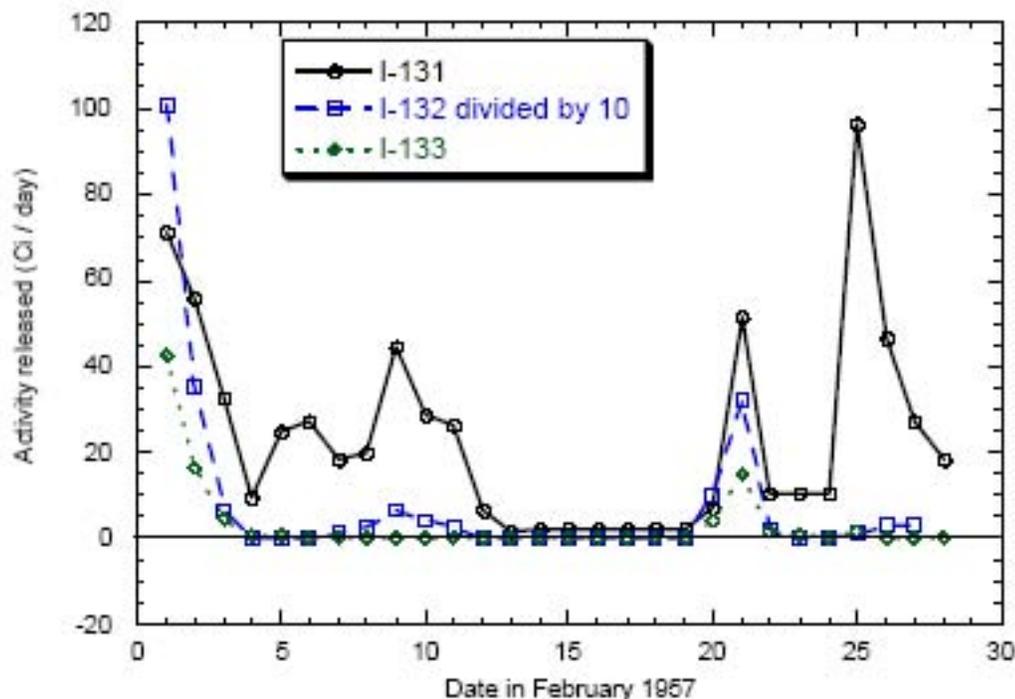
Daily measurements of I-132 releases began in May 1958. Releases of I-132 back to February 1957 are estimated in this report using an empirical relationship based on measured daily releases during later runs and observed I-132/I-131 activity ratios in these releases. Daily I-132 releases also are tabulated in this report. As noted previously, the high I-132 readings that

invariably occurred on the first day of RaLa runs, when dissolving occurred, are not yet explained. There are several factors that should be considered in interpreting the estimated I-132 releases. These include the following:

- Occasional releases of I-132 appear to have been quite high, e.g., about 50-80 times higher than releases of I-131 on the days of dissolving in May and June 1958, and much higher following the ICPP criticality accident on October 16, 1959.
- It appears unlikely that a possible cause of the high readings for I-132 is an impurity in the samples that emits gamma rays that interfere with important I-132 emissions.
- In principle, the I-132 release peaks could be explained by differences in the transport path between I-131 and I-132 and the chemistry of the long-lived precursor, Te-132. For example, if tellurium were insoluble in the alkaline dissolver (unlike iodine), it would pass to the acid dissolver where it would decay to iodine and evolve rapidly from the acid solution. This would be an especially effective release mode in the early runs with weak centrifuge shaft seals. In contrast, I-131 would be largely soluble in the alkaline dissolver, pass into the waste tanks, and be released later when the waste tanks were acidified. However, a tentative evaluation of tellurium chemistry in the alkaline dissolver, presented in [Appendix C](#), casts doubt on this explanation.

Daily releases of I-133, which are also tabulated in this report, are estimated based on the measured daily releases of I-131 and I-133/I-131 activity ratios calculated using the isotopes generation code ORIGEN2. Since the decay characteristics of the tellurium precursors relative to those of the two iodine isotopes are similar, this method should be reasonably reliable.

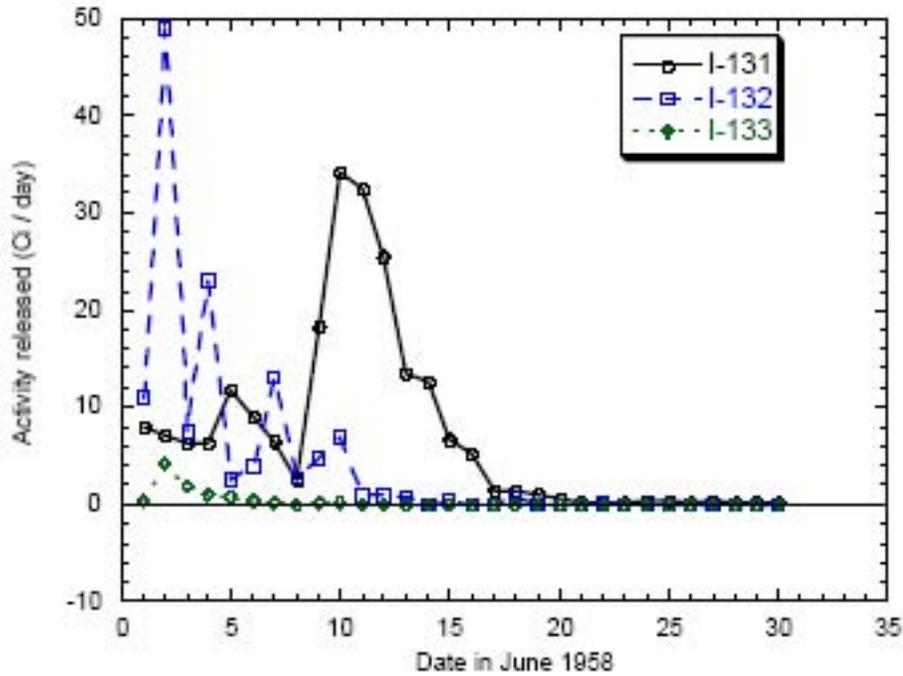
Estimated daily releases of I-131, I-132, and I-133 during February 1957, the first month of RaLa operations, are shown in [Figure ES-1](#). There were two runs during this month, marked by the peaks on February 1 and 21. As described previously, the I-132 and I-133 releases were estimated by a numerical procedure, rather than measured. These data illustrate the high daily releases that occurred in early RaLa runs. The peaks in the I-131 releases observed on February 9 and 25 could not be predicted without detailed knowledge of process operations, including decontamination, solution acidification, and operation of the 10,000-ft<sup>3</sup> off-gas storage vessel.



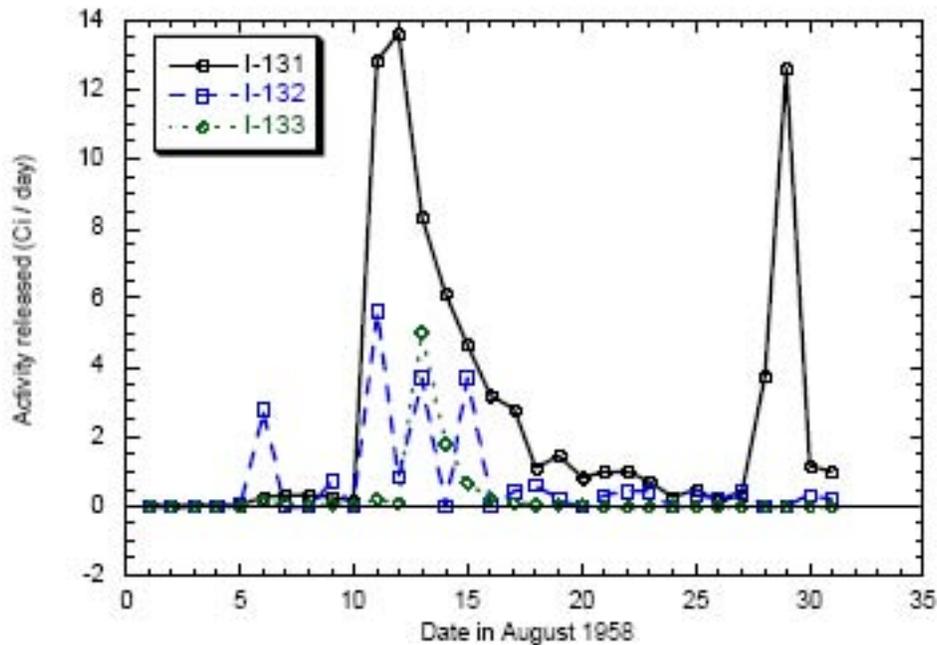
**Figure ES-1 Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), February 1957**  
(Run 1, Feb 1-3; Run 2, Feb. 20-21)

Figures ES-2 and ES-3 show estimated releases for the months of June and August 1958, the latter being the first month of operation with the charcoal filters in place. In both figures, the estimated I-132 releases are obtained from measurements reported in SMDs. The June run took place on the June 2, as indicated by the high I-132 reading on that date. The August run began on the August 12, as indicated by the peak in the I-131 readings. The peak in I-131 readings on August 29 cannot be explained without detailed knowledge of process operations. A notable feature of the data in these two figures is the generally lower releases following installation of the charcoal filters.

The data in Figures ES-1 through ES-3 illustrate the formidable difficulty of predicting iodine releases from a complex process, such as RaLa operations, based on principles of chemistry and mass transfer. Such details as solution transfers, decontamination, stabilization of solutions by acidification, and leakages through seals are of paramount importance, compared with more traditional efforts to account for scrubber efficiency.



**Figure ES-2 Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), June 1958**  
(Run 15, June 2)



**Figure ES-3 Iodine-131, iodine-132, and iodine-133 releases from the RaLa process (Ci/day), August 1958**  
(Run 17, August 13; first run with charcoal beds)

Table ES-1 lists monthly summaries of the daily releases of I-131, I-132, and I-133 estimated in this report. One or, occasionally, two RaLa runs usually occurred in each month, excluding the months designated by an asterisk. A general but irregular downward trend in releases over time is seen, and the effect of installing the charcoal absorbers in reducing releases from August 1958 onward is evident.

Some of the variations in releases shown in Table ES-1 are inexplicable without detailed knowledge of process operations. For example, there were fairly high releases in November 1957, but no RaLa run occurred during this month. Also, there is no available explanation for the high I-132 releases that occurred during several months.

A comparison of the estimated releases of radioiodines obtained in this study with the releases estimated in the 1991 HDE study by DOE can be summarized as follows:

- This study presents estimated daily releases of I-131, I-132, and I-133, as required for dose reconstruction (see Appendix F). The HDE presents I-131 and I-132 releases per RaLa run, which occurred roughly once per month.
- The HDE provides no estimates of I-132 releases per run prior to May 1958. This study provides estimates of daily I-132 releases back to the start of RaLa operations in February 1957. In addition, the HDE presents only annual estimates of I-133 releases, whereas this study provides estimates of daily I-133 releases.
- The estimated I-131 releases and the estimated I-132 releases from May 1958 onward in the two studies are similar. Such agreement is expected when the two sets of estimates are based ultimately on SMDs, either directly in our study or via Health and Safety Division Annual Reports in the HDE.
- The annual release of I-132 for 1957 reported in the HDE differs from the estimated annual release obtained in this study by less than 20%.
- The annual releases of I-133 for 1957 and 1958 estimated in this study are three to four times lower than the releases reported in the HDE. The release during 1959 estimated in this study is about 50% higher. We have no explanation for these differences, but they do not appear to be significant since both are low compared with the releases of I-131. Moreover, the dose per Ci of I-133 is considerably lower than for I-131.

**Table ES-1 Summary of monthly radioiodine releases (Ci) based on estimated daily releases**

Year	Month	Radionuclide		
		I-131	I-132	I-133
<b>1957</b>				
	FEBRUARY	661	2162	91
	MARCH*	97	16	0.03
	APRIL	76	56	1.7
	MAY	44	70	3.7
	JUNE	143	65	5.9
	JULY*	23	18	0.03
	AUGUST*	0.14	0 0	
	SEPTEMBER	34	96	1.89
	OCTOBER	278	913	40
	NOVEMBER*	226	57	0
	DECEMBER*	2.5	0 0	
	<b>TOTAL</b>	<b>1585</b>	<b>3453</b>	<b>144</b>
<b>1958</b>				
	JANUARY	31	89	3.9
	FEBRUARY	75	129	5.5
	MARCH	130	137	3.7
	APRIL	49	173	7.9
	MAY	276	1544	44
	JUNE	212	128	8.7
	JULY*	1.5	2.3	0
	AUGUST†	79	21	8.5
	SEPTEMBER*	25	7.6	0
	OCTOBER	110	804	21
	NOVEMBER	35	117	5
	DECEMBER*	3.8	7.3	0
	<b>TOTAL</b>	<b>1027</b>	<b>3159</b>	<b>108</b>
<b>1959</b>				
	JANUARY*	0.25	1.3	0
	FEBRUARY	112	188	14
	MARCH	47	62	6.4
	APRIL	7.8	124	1.1
	MAY	3.1	44	0.41
	JUNE	2.4	65	0.49
	JULY	10	309	1.1
	AUGUST	3.8	64	0.4
	SEPTEMBER	8.1	55	0.56
	OCTOBER‡	15	125 127	
	NOVEMBER	13	114	1.7
	DECEMBER	1.8	28	0.28
	<b>TOTAL</b>	<b>224</b>	<b>1179</b>	<b>153</b>

\* No runs during this month

† First run with charcoal absorbers

‡ Includes October 16 criticality accident

Uncertainties in reported releases of I-131 are judged to result mainly from uncertainties in the assumed efficiency of the sampler and in the extent of iodine deposition in the sampler line. Reported releases of I-132 are in doubt due to the inability to explain the consistently high readings on days dissolving occurred. If the high readings prove to be valid, an important additional source of uncertainty in estimating releases of short-lived I-132 would be the variation in releases over time during a 24-hour sampling period. Since I-133 releases are calculated based on reported releases of I-131 and calculations of relative inventories of the two isotopes in processed fuel, uncertainties in I-133 releases should be similar to those for I-131. Some judgments on uncertainties in the radioiodine source terms, expressed in terms of probability distributions of releases, are presented in this report.

Table ES-2 presents a summary of the estimated activities of I-131 in fuel fed to the dissolver per RaLa run, in cases where it could be calculated based on available information, compared with the estimated releases from the stack. The decay time is assumed to be two days when specific information is not available. Inventories of I-131 entering the RaLa process cannot be estimated beyond Run 14, due to a complete absence of information on fuel irradiations. Total I-131 released per run was estimated by summing all daily releases prior to the next run. These data indicate that about 1.2% of all I-131 entering the RaLa process was released in the earliest runs, and that the release fraction decreased to about 0.5% after the first year of operation.

### **Releases from 1959 Criticality Accident at the Idaho Chemical Processing Plant— Deterministic Results**

In the early morning of October 16, 1959, an air sparging operation in a critically safe vessel caused an inadvertent transfer of highly enriched uranium solution to a waste tank that was not designed for that purpose. Critical conditions were achieved, and it was later estimated that  $4 \times 10^{19}$  fissions occurred over a period of several minutes. Sufficient heat and pressure were generated to boil off most of the liquid and drive the remainder into an adjacent waste tank.

The number of fissions during the criticality accident was estimated by analyzing the remaining solutions for the activities of a few carefully selected fission products (Mo-99, Ba-140, and La-140). Samples were taken from the final locations after dilution, poisoning, and washing to recover solids that were deposited during evaporation. The method required that none of these particular fission products were lost in the evaporation process, and that the quantity of liquid after dilution was known. A key step was acquiring representative samples from all final solutions.

Based on our evaluation of the available information, we believe that the estimated number of fissions given above is reasonably accurate. This conclusion is based on the following considerations:

**Table ES-2 Percent of iodine-131 released for selected RaLa runs**

<b>RaLa Run</b>	<b>Date</b>	<b>Decay Time (days)</b>	<b>I-131 Fed to the Dissolver (Ci)</b>	<b>I-131 Release (Ci)</b>	<b>Iodine Release (Percent)</b>
1957					
1	Feb. 1-3	5*		381.0	
2	Feb. 20-21	2*	23100	281.0	1.22
3	April 5-6	5*		85.5	
4	May 19-20	7*		42.1	
5	June 24-25	6*	17700	158.8	0.90
6	Sept. 11	2†	21700	103.1	0.48
7	Oct. 8	2†	24400	126.2	0.52
8	Oct. 21	2†	11700	81.9	0.70
1958					
9	Jan. 6	8‡	7000	35.3	0.50
10	Feb. 12	2†	25800	154.6	0.60
11	March 13-14	2†	22800	55.6	0.24
12	Apr. 16	2†	27300	34.9	0.13
13	Apr. 30-May 1	2†	26900	176.7	0.66
14	May. 28	2†	24200	111.2	0.46
15	June. 2	2†	¶	205.8	

\* Obtained from [Legler et al. \(1957\)](#), IDO-14414

† Assumed to be the nominal 2-day cooling period

‡ Cited in [Stevenson \(1958\)](#), IDO-14444

¶ Inventory estimation beyond Run 14 not possible due to lack of irradiation data

- An accurate mass balance of uranium was obtained, which indicates that only a small fraction of the non-volatile fission products used as markers in the analysis was lost due to evaporation.
- Estimates of the number of fissions based on analyses of Mo-99 and Ba-140 in wash solutions after the event agreed within 3%. Analysis of La-140 gave an estimate that was about 25% less.
- Since no duplicate samples were withdrawn, it is not possible to assess the accuracy of sampling. However, the usual sampling protocol of 30 minutes of nitrogen sparging should have resulted in a reasonably representative sample.

A calculation using the ORIGEN2 computer code gave the following estimates of the activities of iodine isotopes produced in the accident ([Table ES-3](#)).

**Table ES-3 Activities of iodine isotopes produced during October 16, 1959, accident, as estimated using ORIGEN2 computer code**

Nuclide	Maximum Activity (Ci)	Time Span of Maximum Activity
I-131	26.7	4 hours–15 days
I-132	103.0	10 hours
I-133	582.0	4 hours
I-135	1,930.0	1–20 minutes

Twenty-one hours after the accident, at midnight, a routine stack monitoring sample was taken and counted for I-131 and I-132 using gamma spectroscopy. The results were converted to estimated releases for that day based on the usual assumptions (a constant release rate and a sampler efficiency of 50%). The stack monitoring data gave the following estimates of releases for October 16, 1959: 3.73 Ci of I-131 and 9,780 Ci of I-132.

Compared with the estimated maximum activities of these isotopes produced in the accident, the high reading of I-132 in the stack monitor certainly appears to be erroneous, but for unknown reasons; this conclusion agrees with the 1991 DOE study.<sup>1</sup> In contrast, the estimated I-131 release based on the stack monitoring data appears to be reasonable.

Since all ICPP vessels were closed, the principal release path to the atmosphere was the vent line from each vessel, which fed into the vessel off-gas system and then directly to the stack. Therefore, releases should have occurred quickly after entry into the vessel off-gas line. The releases probably were sporadic, due initially to pressure surges from the heat produced by the fissions and later to nitrogen sparged into the vessels to facilitate solution transfers and sampling. The stack monitoring data indicate that I-131 releases continued for at least seven days after the criticality occurred.

Releases of volatile fission products from the solution were estimated based on a model that assumed release at a constant fractional rate of the inventory that remained in the vessel, taking into account decay and prior volatilization. The total inventory of a radionuclide as a function of time was estimated using the ORIGEN2 code. The fractional release rate of I-132, I-133, I-134, I-135, and isotopes of bromine was estimated based on the measured release of I-131 and its calculated inventory. The fractional release rate of isotopes of krypton and xenon was assumed to be 10 times higher than the fractional release rate of the halogens. It was assumed that release of radionuclides attached to aerosols did not occur.

Estimated radioiodine releases from the criticality accident, as obtained from the stack monitoring data for I-131 and modeled for the other isotopes assuming a constant fractional release rate, are given in [Table ES-4](#). The total release of radioiodines is estimated to be 429 Ci.

<sup>1</sup> We have not been able to identify the source of error, which possibly also affected measurements of I-132 in routine RaLa releases. The most obvious source of error is the presence of an impurity in the sample. However, an impurity would have to emit gamma rays at energies that interfere with the prominent I-132 emissions, be caught in the iodine trap and be present in a high concentration, and have a half-life of at least several hours in order to be present 21 hours after the accident. We have not found any such impurity.

**Table ES-4 Estimated radioiodine releases (Ci) from 1959 criticality accident at the Idaho Chemical Processing Plant**

Date	I-131	I-132	I-133	I-134	I-135
Oct. 16	3.73	16.4	74.4	87.0	140.0
Oct. 17	2.44	12.5	33.1	0	13.2
Oct. 18	0.97	8.13	12.0	0	0.86
Oct. 19	1.01	5.28	4.32	0	0.10
Oct. 20	1.15	3.43	1.56	0	0
Oct. 21	0.94	2.23	0.56	0	0
Oct. 22	0.87	1.45	0.20	0	0
Oct. 23	0.18	0.94	0.10	0	0
<b>Totals</b>	11.3	50.4	126.0	87.0	154.0

**Total, all radioiodines = 429 Ci**

Our estimates of the total releases of all isotopes of bromine, krypton, and xenon are given in [Table ES-5](#).

**Table ES-5 Estimated releases of all isotopes of bromine, krypton, and xenon from 1959 criticality accident at the Idaho Chemical Processing Plant**

Element	Release (Ci)
<b>Bromine</b>	32.2
<b>Krypton</b>	2,500.0
<b>Xenon</b>	3,290.0

A breakdown of the releases of bromine and the noble gases by isotope and date is given in [Chapter 7](#) of this report.

### Effect of the Uncertainty Evaluation on Releases

The principle sources of uncertainty for routine releases of I-131 due to the uncertain values of the sampler collection efficiency and deposition in the sampling line. Evaluation of some poorly documented sampling efficiency and deposition test data leads to the reasonable judgment of the distribution of these sources of error are described in [Section 6.10.1](#). The net effect of these uncertainties and assumed distributions leads to a central estimate of 1.4 for a bias correction factor with the 95% confidence range of 0.8 to 1.8. The upper portion of Figure ES-4 illustrates the net effect of applying the central estimate of the bias correction factor to routine releases reported for January 1958.

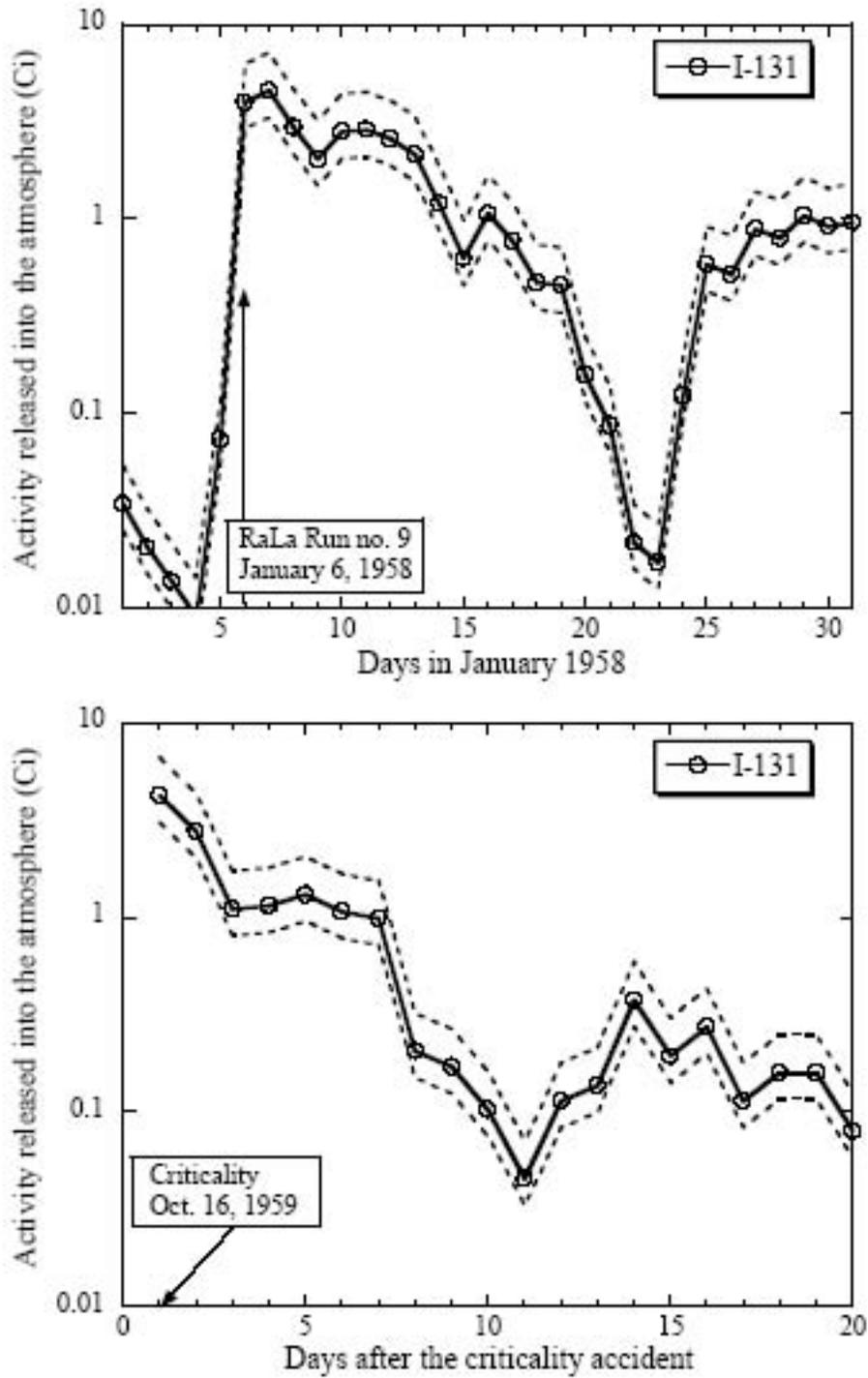
Iodine-132 releases are more uncertain for several reasons. First of all there is some question as to the validity of any of the I-132 data in view of the improbably high releases reported on several occasions. If the reported I-132 releases are assumed to valid, the uncertainty range would still be higher than for I-131 due to the shorter half-life of I-132. The short half-life (2.3

hr) leads, on the average, to an underestimate of the reported reading based on the 24-hour sampling period. Applying the three sources of error to I-132 (sampler error, deposition, and sampler decay correction) leads to a median bias correction factor of 4.4 for I-132, with a 95% confidence range of 1.1 to 12.2.

The method adopted for determining I-133 releases (i.e., using the estimated I-131 releases and accounting for the different inventories levels) leads to the identical uncertainties as defined for I-131.

The upper part of [Table ES-6](#) summarizes the central estimates of I-131, I-132 and I-133 routine releases for the 3-year period, February 1957 to December 1959, together with the upper and lower 95% confidence values.

Since the same collection efficiency, line deposition, and sampler decay corrections apply to the October 16, 1959, criticality accident as well, the same uncertainty distributions were applied for this event for I-131, I-132, and I-133. The lower portion of [Figure ES-4](#) illustrates the application of the uncertain bias correction factor for the release of I-131 during the criticality accident. The lower portion of [Table ES-6](#) lists the central estimate and 95% confidence bounds for the October 16, 1959, criticality accident.



**Figure ES-4 Daily releases of iodine-131 during January 1958 and after the 1959 criticality accident adjusted by the uncertain bias correction factor**  
The solid line is the 50<sup>th</sup> percentile and dotted lines represent the 95% confidence interval.

**Table ES-6 Estimated atmospheric releases of radioactive iodine from Idaho Chemical Processing Plant during 1957–1959 (Ci)**

<b>Routine Releases</b>			
<b>(including October 16, 1959, criticality accident)</b>			
95 % Confidence Interval			
<b>Isotope</b>	Lower bound	Central Estimate *	Upper Bound
$^{131}\text{I}$	2,400	3,200	5,100
$^{132}\text{I}$	25,000	37,000	58,000
$^{133}\text{I}$	340	470	730
<b>October 16, 1959, criticality accident</b>			
$^{131}\text{I}$	9.4	13	20
$^{132}\text{I}$	130	230	450
$^{133}\text{I}$	110	140	230

\* The 50<sup>th</sup> percentile of the probability distribution describing the uncertainty in the estimated releases.

## 1.0 SCOPE AND APPROACH

### 1.1 Scope

The principal objective of the source term effort is evaluation of radioiodine releases from the Idaho Chemical Processing Plant (ICPP) in the years 1957–1959. Essentially all of the routine radioiodine releases came from the Radioactive Lanthanum Project (RaLa), which chemically processed nominally 2-day cooled fuel irradiated in the Materials Testing Reactor (MTR). The first 3 years of operation historically had the highest releases over the 11 years of process operation.

The second objective is estimation of the radioiodine, bromine, and noble gas releases from the accidental criticality in an ICPP waste tank, which occurred during this period on October 16, 1959.

In addition, the scope includes an estimate of radioactive aerosol releases to the atmosphere. These may be composed of, or contain, longer-lived actinides and fission products. As such, their point of origin could have been any of the chemical processes in the ICPP and not solely RaLa.

### 1.2 Approach

#### 1.2.1 *Operational RaLa Releases*

The preferred option for developing a source term for a complex chemical process such as RaLa is to base it on verifiable measurements of release cited in primary documents. When this possible — as it happens to be for RaLa releases of I-131, though not with resounding certainty — the most unequivocal results are obtained with the smallest possible error band. A primary document is one that is written by project personnel near the time of performance of the work, e.g., topical reports, progress reports, archived datasheets, and contemporary letters. Secondary documents such as conference articles written many years after the fact or progress reports written by non-project personnel may be useful for background, but usually present information that cannot be substantiated.

Verifiability is often difficult, especially for a 50-year-old project. Technical reports containing evaluations and design descriptions of the stack monitoring system would have been extremely valuable in this regard. Unfortunately, none have been found, evidently lost along the way. However, early notes, letters, and datasheets offer fragmentary evidence that a serious effort was made to develop a reliable stack monitoring system.

The principal components of the stack monitor system that could have contributed in various degrees to error in the release measurements are the following:

- Uncertainty in the stack sampler efficiency
- Unknown degree of deposition in the sampling lines

- Radioactivity determination of the sampler solution

Fragmentary notes indicate that these issues were at least addressed by project personnel and supervisors.

If the first option, direct measurement, is not available, alternate methods must be used which entail progressively more uncertainty and error. For example, a second approach might be use of release measurements taken with test equipment. If this option is available (it was not for RaLa), then it must be realized that test equipment can never duplicate the conditions of an actual operational release under fully radioactive conditions. Unless there is a full understanding of all the factors affecting release, which there rarely is, it would always be debatable as to how closely the test measurements correspond to reality.

A third option occasionally mentioned is a modeling approach in which releases are calculated based on chemical engineering principles. As will become apparent to those who have participated in such an endeavor, calculation of atmospheric releases from a complex process like RaLa cannot be accomplished without resorting to the most gross and broad approximations. Even for less complex systems, complete design and operation details are essential — temperatures, sampling times, sampling methods, flowrates, run schedules, and timing of activities — for all the key process vessels. Virtually none of this sort of information is available. Therefore, even broad approximations would be difficult to formulate due to the lack of survival of virtually all contemporary operational information.

The approach for I-132 was to utilize the release measurements taken beginning April 1958 and extrapolate back to the start of operations in February 1957 by means of a correlation. It should be said that the I-132 readings were always suspect due to some unusually high reported releases on the day of dissolving for some of the runs. However, we had no solid basis for rejecting such high readings throughout most of the project effort. The behavior of I-132 in the system differed from other iodine radionuclides by virtue of its relatively long-lived precursor, Te-132. Thus I-132 releases depend to some extent on Te-132 chemistry in the RaLa system, which is not well known.

There were no I-133 release readings taken by the stack monitor system. Our approach for I-133 releases was based on a correlation using the I-131 release measurements and the relative abundance of I-131 and I-133 in the dissolver calculated from an isotope generation code, ORIGEN2. The logical basis for this approach is that I-131 and I-133 have similarly short-lived tellurium precursors and so should exhibit similar behavior in the system, once the relative abundances are accounted for.

### ***1.2.2 Idaho Chemical Processing Plant Criticality Approach***

The approach for assessing releases from the ICPP criticality accident of October 16, 1959, consists of the following steps:

- (1) The first step is description of the event, including circumstances of the initiating event, the vessels involved in the criticality, location of the vessels in the main

process building, and identification of the release pathways from the vessels to the atmosphere.

- (2) Second, the means for estimating the severity of the event, i.e., the number of fissions, was evaluated. Possible sources of error in estimating the number of fissions were identified.
- (3) A release rate model was developed based on the I-131 release measured 21 hours after the event by the ICPP stack monitor. The release model was then applied to determine I-132, I-133, I-135, radiobromine, xenon, and krypton releases.

## 2.0 THE RADIOACTIVE LANTHANUM (RaLa) PROCESS

### 2.1 Background

#### 2.1.1 Requirement

The motivation for the RaLa process was the need by weapons developers at Los Alamos for large quantities of a low mass, intense, highly energetic source of  $\gamma$ -rays for use in a diagnostic device for evaluating the implosion process. It was decided that the  $\gamma$ -emissions from the fission product lanthanum-140 (La-140) would be most appropriate for this purpose. Lanthanum-140, with a half-life of 40.2 hours, would be acquired by separation of its longer-lived parent, barium-140 (Ba-140, with a half-life 12.9 days), from a mixture of fission products.

The strategy was to intensely irradiate a highly enriched fuel element in the MTR, permit only short cooling, nominally 2 days, and separate barium from fuel in the RaLa process. The product would consist of stable Ba-138 as the major portion, plus radioactive Ba-140. Rapid processing of the discharged fuel was required due to the relatively short half-life of Ba-140. The product was shipped to Los Alamos, where after several days, the La-140 built back to an equilibrium level<sup>1</sup> and was separated from barium in a simple physical process.

#### 2.1.2 Work at Oak Ridge National Laboratory

The initial RaLa process was set up at the Oak Ridge National Laboratory (ORNL) in 1945. Separation of Ba-140 from short-cooled fuel from the Oak Ridge Graphite Reactor produced batches having from 500 to 2,000 curies of Ba-140 (Legler et al. 1955). This was boosted to 10,000 Ci batches in 1951 by using fuel from the Hanford reactors. Subsequent requests for batches of 30,000 Ci were beyond the Oak Ridge facility's capability.

As a consequence, beginning in the early 1950s at ORNL, plans were formulated for a follow-on process using fuel from the then-new MTR at the National Reactor Testing Station (NRTS), as INEL was then called. The MTR had much higher specific power than either the Oak Ridge Graphite Reactor or the Hanford reactors, thereby producing significantly higher amounts of radioactive fission products per mass of fuel charged.

Development of an early MTR-based RaLa process at ORNL was described by Blanco (1952). The major departure in the newer process was the proposed use of an alkaline dissolver as the initial process step instead of the acid dissolver used in the original Oak Ridge process. Since the alkaline solution would dissolve only the aluminum cladding and a few soluble fission products (not barium or uranium), the large mass of aluminum cladding would be effectively removed from the process at an early stage. This would be followed by a second dissolution step using concentrated nitric acid that would dissolve the uranium and most of the fission products.

---

<sup>1</sup> Pure Ba-140 will decay to an equilibrium mixture of Ba-140 and La-140 within a few half-lives of the shorter-lived daughter. At that point, decay of La-140 is effectively controlled by its production rate (the decay of Ba-140), and the curie levels of parent and daughter are nearly equal.

### **2.1.3 Early Process Decisions**

In October 1952, a decision was made to develop a larger capacity RaLa process at the ICPP based on MTR fuel (Anderson et al. 1959). Initially, the American Cyanamid contractor, and subsequently (September 1953) the Phillips Petroleum Company, assumed responsibility for RaLa development.

The general concept of alkaline dissolution of the aluminum cladding was accepted, but it was judged that there were several difficulties with the Oak Ridge approach:

- Separation of the aluminate dissolver solution from the uranium and fission product solids by filtration through a sintered, stainless steel filter was not an attractive prospect. The alternative, conventional centrifugation, was not an option, as tests at Los Alamos showed (Anderson et al. 1959). The problem with conventional centrifugation was that the separated solids resuspended after the rotation stopped due to convection currents generated by the decay heat. Therefore, a more complex form of centrifugation was selected, whereby the supernate was decanted from the centrifuge during rotation by means of skimmer walls.
- The Oak Ridge concept of performing the final separation of barium from strontium by means of ion exchange columns was rejected in favor of either chromate precipitation or an alternate “chloride method.”

There were several other process alterations, but the basic concept of alkaline dissolving of an MTR element and on-line centrifugation of supernate from solids (at several points) determined the general character of the process, and that general character must be described as highly complex. Physical complexity was caused by the need to move process liquids, in some cases including solids, repeatedly back and forth between vessels, and the presence of two centrifuges in the flow sheet being used as both separators and reaction vessels. In addition, the process had to deal with solid, liquid, and gaseous materials, the latter including emissions of radioactive iodines and noble gases and hydrogen. Gaseous radioactive emissions had to be controlled sufficiently to permit adequate dispersion in the atmosphere. In addition the concentration of hydrogen evolved from the dissolver had to be limited to below explosive levels. Further chemical complexity was due to the need to separate a small amount of highly radioactive barium from a large amount of cladding, uranium, and other fission product material. Not all of the attendant difficulties were completely resolved by the time startup operations on short-cooled MTR fuel commenced on February 1, 1957.

### **2.1.4 The RaLa Development Program at the Idaho Chemical Processing Plant**

Engineers were faced with the problem of developing, fabricating, and testing a complex, essentially new, process involving the highest radiation levels yet experienced at the ICPP on a tight schedule. From available reports, it appears that several development activities were put into motion:

- The basic chemistry of the process was reviewed.

- The centerpiece of the development program was a full-scale pilot facility operated on surrogate, non-radioactive materials, except for small amounts of barium and strontium activity added for analytical purposes. No iodine or noble gas evolution or control tests were included in this phase of the program. However, all the unit operations were tested: dissolving, centrifugation, product packaging, precipitation, and fluid pumping by vacuum jet. Equipment and process variables were tested. Results are reported in [Legler et al. \(1955\)](#).
- Small-scale laboratory tests on radioactive materials were initiated, culminating in mid-1955 in the newly constructed Multicurie Cell in the ICPP. These results were reported by [Anderson et al. \(1959\)](#).<sup>2</sup> The tests were evidently conducted on wafer-sized samples of spent MTR fuel in laboratory glassware. No irradiation levels or cooling times are given. This phase of the program appears to have emphasized uranium recovery, barium-strontium separation, and corrosion of process materials, and not radioiodine emissions.
- A few tests were conducted on fission product separation in the alkaline and acid dissolving steps. Apparently, there were only two tests on iodine evolution, both with inconclusive results. In one, it was reported that only 17.1% of the iodine remained in the supernate during caustic dissolving, quite different from later reports of 90% in secondary references. A second test dealt with iodine evolution from the caustic supernate on dilution and acidification. These tests were marred by high surface deposition, but evidently most of the dissolved iodine evolved on acidification.
- Shakedown of the final RaLa production facility began on April 25, 1956, using non-radioactive materials. Eleven cold tests were run in 1956, followed by three tests using progressively shorter cooled MTR elements as feed ([Legler et al. 1957](#)). These preliminary hot tests were labeled 001-RH, 002-RH, etc., followed by production runs labeled 001-RP, etc., beginning February 1, 1957. The program of process evaluation continued into the production run phase through Run 005-RP conducted in June 1957.
- Final iodine inventories in some of the process solutions are reported for the three preliminary “RH” runs and the first five production runs. These and other iodine distribution data are summarized in [Section 2.5](#) regarding iodine distribution tests.
- Development tests were initiated in the Technical Branch of the ICPP and reported in a series of Quarterly Progress Reports (QPR). (Some are listed in the references, QPR-ICPP, various dates.) Some (or all) of the QPR tests were actually parts of the experimental programs cited above and reported therein. Nevertheless, the QPRs were scanned for additional iodine behavior data in the RaLa process. None were found, except possibly some data taken in RaLa Runs 3

---

<sup>2</sup>This report was evidently delayed. The work was completed some time earlier, probably early 1957.

and 5, reported in the [September 1957 QPR \(IDO-14419\)](#), which is similar to that reported in the Startup Operations Report ([Legler et al. 1957](#)).

Under a tightly compressed schedule, the Blaw Knox Company initiated the final design of the facility in July 1954. Construction of the final process equipment in L-Cell of the ICPP by the J.F. Pritchard Company began in November 1955.

## 2.2 Idaho RaLa Process Reporting

### 2.2.1 Primary Sources of Information

A primary source is written while active on the project, contains test data, and is published by the project, becoming a part of the project archive. Logbooks, letters, and original datasheets may also be considered primary sources. It is essential that source term evaluations be based on primary sources of information.

All of the available primary sources on RaLa operations relevant to the 1957-1959 time frame or process design leading to operation are listed in [Appendix A](#). As can be seen, early work leading to initial hot operation beginning February 1, 1957, is adequately documented. However, no RaLa operations reports have been located following [Legler et al. \(1957\)](#), which covers events only through Run 5 in June 1957.

Fortunately, the Stack Monitor Datasheets are available and provide a near-direct measure of the iodine releases. Evaluations in [Chapter 5](#) seek to establish the degree of reliability of the stack monitor data.

### 2.2.2 Secondary Sources

Secondary sources may be useful for general description and background but usually do not provide sufficient information to verify conclusions. In some cases, information is contradicted by primary sources.<sup>3</sup> Examples of secondary sources are conference papers (several appeared in Air Cleaning Conferences), digests in Nuclear Safety, international conference papers, and summaries written by non-project personnel.

## 2.3 Chemical Flow Sheet

The nominal starting material of the process is a 20-day irradiated MTR fuel element achieving 21.6% burn-up, cooled two days prior to insertion in the dissolver. The composition of normal RaLa feed material is presented in [Table 2-1](#).

---

<sup>3</sup>A notable example is the statement that 90% of the iodine remains with the caustic dissolver solution ([Cederberg and McQueen, 1961](#)). Primary sources show this to be highly variable and never as high as 90%.

**Table 2-1 Nominal RaLa feed material**  
(Legler et al. 1955)

Feed Materials	Weight (activity)
Aluminum	3818 g
Uranium	168.5 g
Barium-138	1.37 g
Barium-140	0.726 g (53,000 Ci)
Strontium-88	0.519 g
Strontium-89	0.541 g (15,000 Ci)
Strontium-90	0.697 g (139 Ci)
Strontium-91	small g (2,400 Ci)
Other fission products	30-50 g
Iron	16 g
Silica	30 g
Copper	4 g

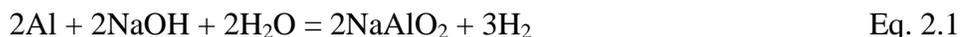
The basic process is to dissolve first aluminum in a caustic dissolver solution and physically separate the uranium and most of the fission products (including the product Ba-140), which are insoluble in caustic, from the aluminate solution. After decanting the caustic aluminate solution, the solids would be dissolved in concentrated nitric acid. There follows a train of chemical steps, first separating uranium and plutonium from the fission products, culminating in separating the chemically similar fission products barium and strontium. The final steps are several purifications of the barium product.

The chemical steps in the process are described below. Iodine evolved from several stages in the RaLa process.

A simplified chemical flow sheet is shown in [Figure 2-1 \(Legler et al. 1957\)](#). (A more comprehensive flow sheet is provided in [Legler et al., Figure 29](#).)

### *Step 1: Fuel Dissolution*

The process begins with batch dissolution of one MTR fuel element in 5 M sodium hydroxide producing dissolved sodium aluminate and evolving large quantities of hydrogen.



The resulting uranium and other solids become small particulates in the dissolver solution. The noble gases in the MTR fuel element evolve completely, joining the hydrogen in the dissolver off-gas (DOG). In addition, [Legler et al. \(1957\)](#) state that cesium and tellurium dissolve.<sup>4</sup> Some portion of the iodine also dissolves, which is retained in the alkaline dissolver solution. The test data on iodine dissolution are reviewed in [Section 2.5](#).

<sup>4</sup>Test data show that 90%+ of the cesium dissolves, but no test data have been found on tellurium. As discussed in [Chapter 3](#), the degree of tellurium dissolution affects the release path for I-132.



There was some concern over the stability of the dissolver solution. Later reactions could form the troublesome gel, hydrated alumina,  $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ . Consequently, once separated, it was desirable to stabilize the dissolver solution by acidification with nitric acid, a process that evolves the iodine unless a sequestering agent is simultaneously added. There is no record of when the acidification occurred.

### *Step 2: Dissolver Solution Centrifugation*

The dissolver solution plus solids are steam-jetted to the centrifuge. The aluminate solution is decanted while spinning to a waste tank; the solids remain in the bowl.

The gas jetting method (air, nitrogen, or steam) is a factor in the evolution of iodine. An air-jetting test, [Figure 2-2](#), illustrates the method used throughout the RaLa process for transferring liquids between reaction vessels. The liquid, in this case the slurry, is carried along in the direction of a jet of high-pressure gas, in this case air. The test shows that the method is capable of elevating liquid 20 feet. The agitation provides opportunity for gas evolution. In addition, the volume of gas or vapor added to the process is relieved by venting either to the DOG or to the vessel off-gas (VOG) system.

### *Step 3: Cake Dissolution*

The uranium and remaining fission products are dissolved in the centrifuge bowl by adding 6 M of nitric acid. The silica and perhaps copper remain as solids. The solution is steam-jetted to the boil down tank. The solids are slurried up and jetted to a waste tank.

### *Step 4: Barium and Strontium Separation*

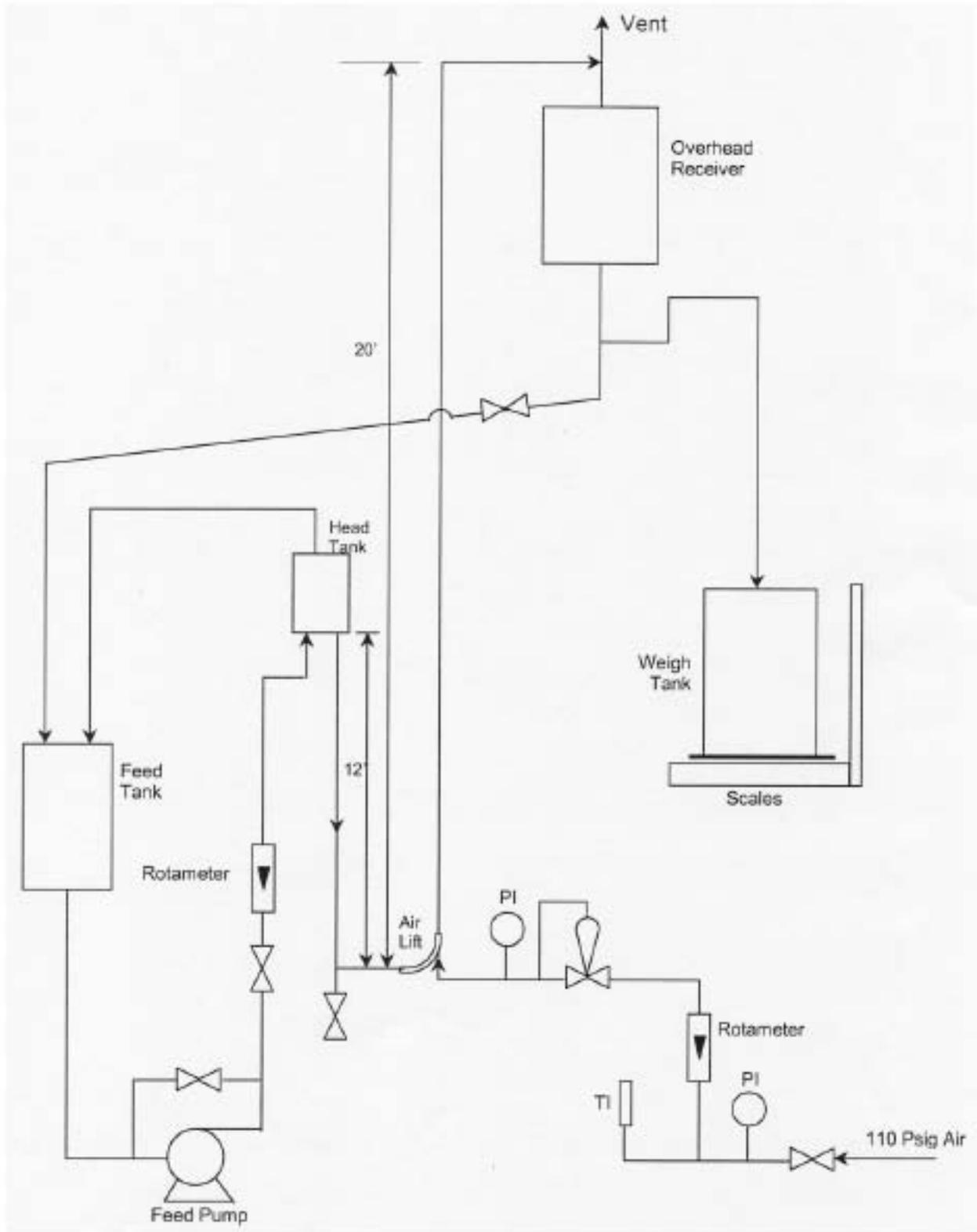
Stable barium carrier is added to the solution, and evaporation initiates the precipitation of barium and strontium nitrate. The concentrate is jetted back to the centrifuge where the precipitation of barium and strontium is completed by the addition of nitric acid. Centrifugation then separates the uranium solution, which is decanted off to the uranium storage vessel for subsequent charging into the ICPP uranium recovery process.

### *Step 5: First Chromate Precipitation*

The nitrate cake in the centrifuge bowl is dissolved in water. Sodium dichromate is added to produce barium and strontium chromate. Sodium acetate<sup>5</sup> is added as a pH buffer. At the resulting pH, the barium chromate precipitates, but strontium chromate remains in solution. Centrifugation decants the strontium chromate solution to a strontium process tank, where it is ultimately recovered.

---

<sup>5</sup>The organic acetate anion may contribute to organic iodide formation.



**Figure 2-2 Illustration of air-lifting RaLa solutions**  
(Legler et al. 1957)

### *Step 6: Second Chromate Precipitation*

The barium chromate cake in the dissolver bowl is dissolved in 0.2 M nitric acid and the chromate precipitation step is repeated to improve the efficiency of strontium – barium separation. Centrifugation separates the strontium containing supernate to a waste tank.

### *Step 7: Conversion to Barium Nitrate*

The cake is converted to barium nitrate solid by the addition of fuming nitric acid. The excess acid is centrifuged off to a waste tank.

### *Step 8: Final Nitrate Precipitation*

The barium nitrate solid in the centrifuge bowl is dissolved in water and reprecipitated by the addition of fuming nitric acid.

### *Step 9: Product Drying*

The barium nitrate is redissolved and transferred to a product cup, where it is reprecipitated by evaporation to dryness.

## **2.4 Process Equipment**

The complexity of the process is illustrated by flow sheet and equipment drawings taken from the startup report (Legler et al. 1957).

Figure 2-3 shows the arrangement of process vessels in Cell L. In addition to these, product make-up vessels are located on the level above Cell L and waste tanks on the level below. Identified in the figure are the following vessels:

L-101	Dissolver
L-105	Dissolver Supernate Tank
L-108	Caustic Waste Tank #1
L-109	Caustic Waste Tank #2
L-102	Off-Gas Scrubber
L-103	Scrubber Waste Holdup Tank #1
L-153	Scrubber Waste Holdup Tank #2
L-400	Centrifuge #1
L-106	Centrifuge #1 Feed Tank
L-450	Centrifuge #2
L-156	Centrifuge #2 Feed Tank
L-107	Uranium Storage Tank
L-113	Strontium Storage Tank
L-112	Strontium Collection Tank
L-104	Centrifuge #1 Feed Pot
L-154	Centrifuge #2 Feed Pot

L-111	Strontium Batch Tank
L-112	Strontium Collection Tank

Design drawings for many of these vessels are described in [Legler et al. \(1957\)](#). Note that there are two centrifuge vessels in L-Cell, each equipped with a feed tank. Presumably one is a backup when the other is undergoing maintenance.

[Figure 2-4](#) is a detailed flow sheet of the front end of the process.

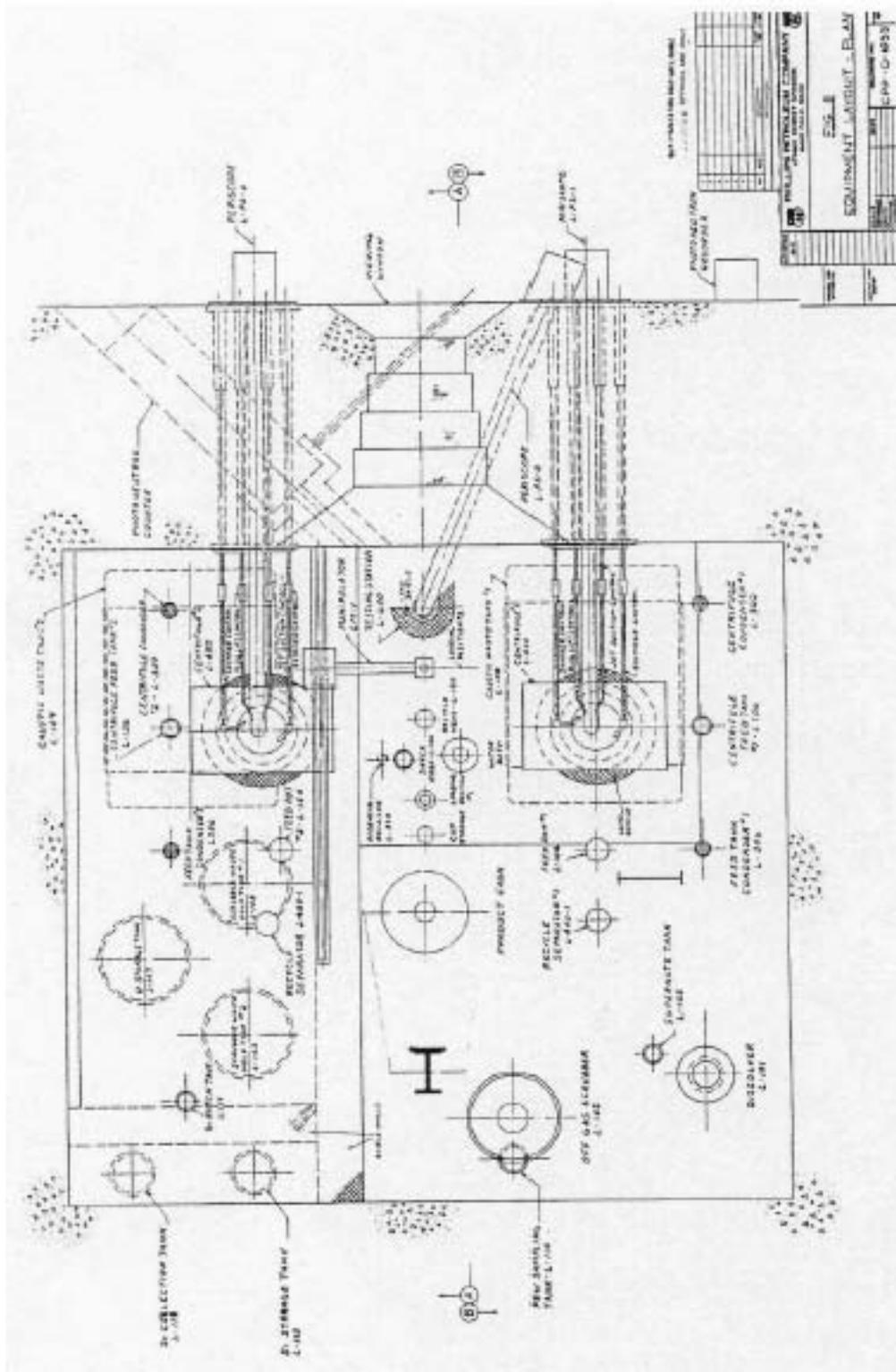


Figure 2-3 RaLa equipment layout in L-Cell  
(Legler et al. 1957)



Adjacent to the scrubber (L-102, shown in the lower left corner) is a list of process vessels whose off-gas flows are fed to the scrubber. Fourteen vessels are listed as venting through the scrubber, including several identified in the list above. Significantly, a large portion of the iodine evolves from two caustic waste tanks (L-108 and L-109) following acidification of the alkaline supernate. In addition, the dissolver (L-101) and the two scrubber waste tanks (L-103 and L-153) show vent lines to the inlet of the scrubber. The off-gas from the centrifuge feed tanks (L-104 and L-154) is also shown to be routed into the scrubber.

Therefore, all the RaLa process vessels can be vented into the DOG, which passes through the scrubber. However, [Ayers and Burn \(1960\)](#) state that by a valving change, the off-gas may also be routed into the VOG, which bypasses the scrubber.<sup>6</sup> It is not clear why this flexibility existed or when the vessels vented to either the DOG or VOG. Presumably, all RaLa vessels vented to the RaLa DOG.

[Figure 2-4](#) also distinguishes between nitrogen lifts and steam jets as means for impelling flows. A typical steam jet is shown by the icon for L-500, adjacent to the dissolver. A nitrogen lift icon is shown adjacent to the scrubber. Both of these methods of impelling liquids add gas volume to the process system that must be relieved by venting of vessels into the off-gas system.

The dissolver (L-101) is fed by product feed tanks, PM-120-0 and PM-121-0 (which could also feed the scrubber, L-102). Cooling coils shown in the upper part of the vessel form a reflux condenser. Agitation of the liquid in the dissolver is provided by a nitrogen sparger, which serves to accelerate the dissolving process. It also promotes evolution of volatiles: noble gases, hydrogen, and possibly some iodine from the liquid. The steam jet (L-500) recirculates liquid in the dissolver. Other steam jets, L-104 or L-105, impel flow into the centrifuge feed pots (L-104 and L-154).

The scrubber (L-102) is seen at lower left. A nitrogen lift provides recirculation of the liquid collected in the base of the vessel. When spent or at the end of the run, scrubber liquid collected in the base of the vessel is steam-jetted to the two scrubber waste holdup tanks (L-103 and L-153).

[Figure 2-5](#) is the detailed flow sheet around the centrifuge, showing the two centrifuges in the process, each with its related service vessels.

[Figure 2-6](#) shows the complexity of the centrifuge. Examination shows that the rotating bowl was open to permit entry of various probes (i.e., feed lines, skimmers, etc.). Evidently the outer casing provided the barrier preventing free access to cell air. Note the “carbon seal,” where the shaft exits through the upper plate. [Legler et al. \(1957\)](#) wrote that these seals easily wore down, contaminating the centrifuge bowl with carbon particles. Tests both using other materials (tygon) and removing the seals altogether were done. The latter, however, permitted excessive

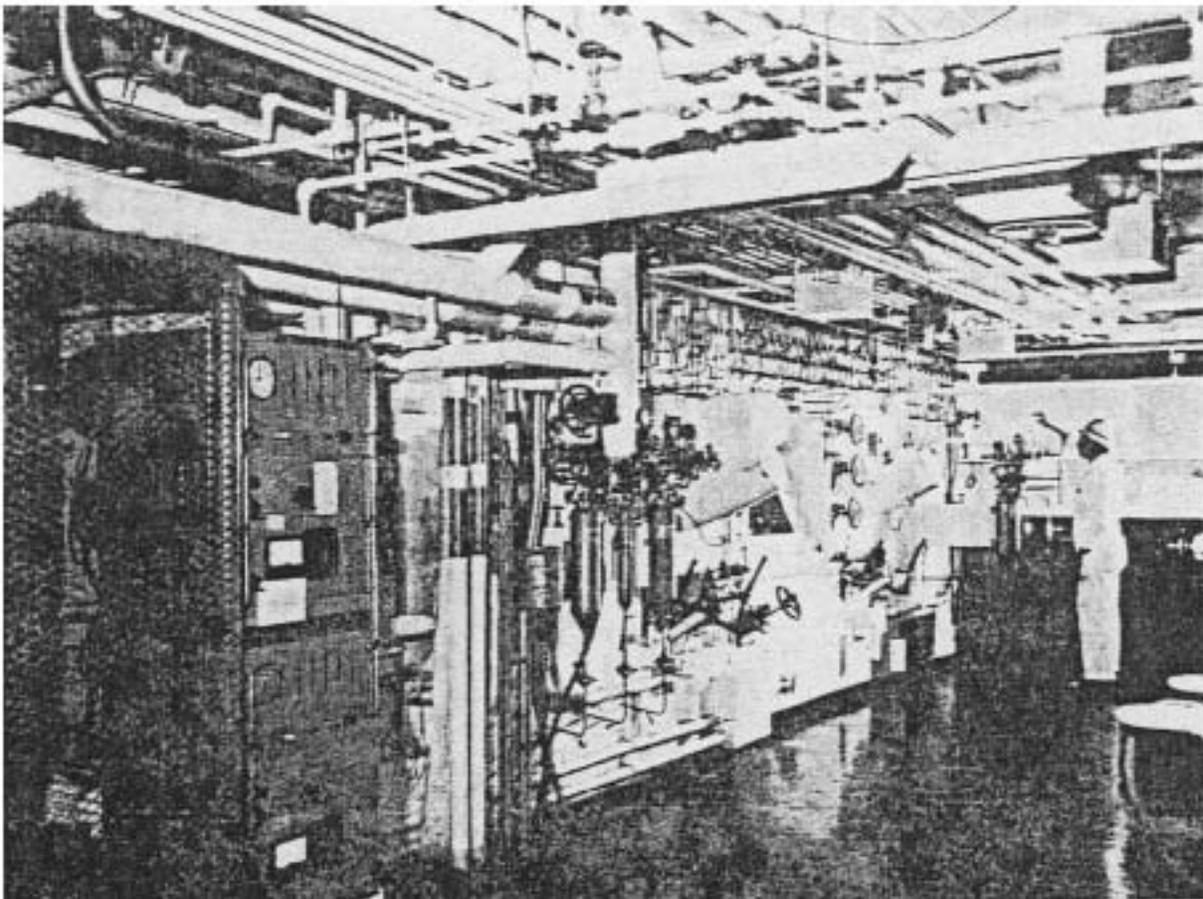
---

<sup>6</sup>[Legler et al. \(1957\)](#) provides a bit of confusion. In his Appendix H, [Legler et al.](#) state, “... a vessel off-gas system provides common vacuum venting for all the process vessels except the dissolver L-101, and off-gas scrubber L-102; and a dissolver off-gas system providing vacuum venting for the dissolver and off-gas scrubber.” It may be set in this mode with appropriate valving.

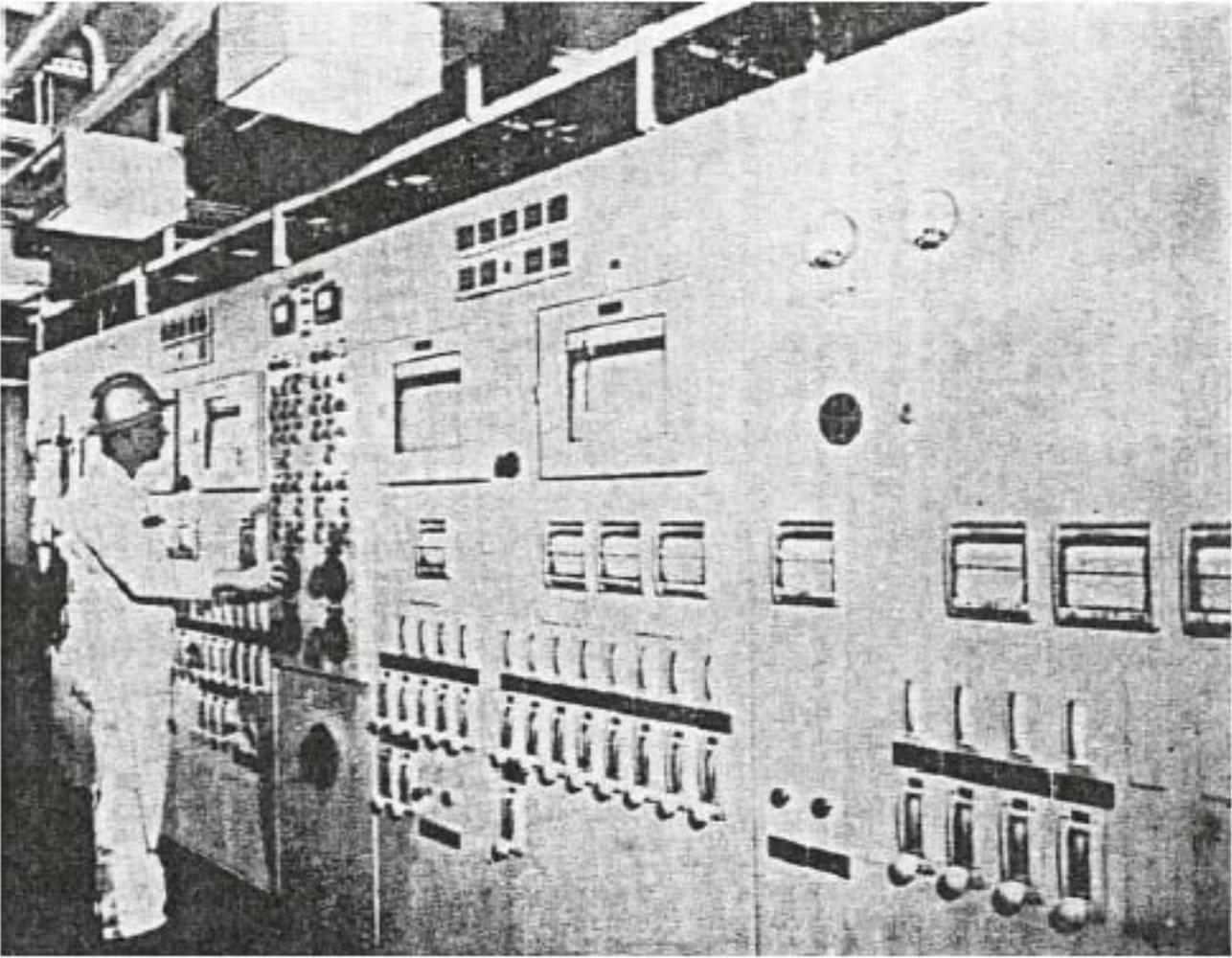




leakage to cell air. Later reports (e.g., [Cederberg and MacQueen 1961](#)) state that centrifuge seals were improved at a later date. [Figures 2-7 and 2-8](#) show views of the Cell L face and the main RaLa control panel, respectively ([Legler et al. 1957](#)).



**Figure 2-7** South face of cell L — access corridor level



**Figure 2-8 RaLa main control panel — access corridor level**

## **2.5 Iodine and Other Fission Product Distribution Tests**

No consistent picture of iodine behavior in the RaLa system was developed in the testing program despite the recognition that iodine releases could limit operations. The disparate and contradictory measurements that were made are summarized below.

The Laboratory Development Program ([Anderson et al. 1959](#)) reported two tests of the fraction of selected fission products that dissolved into the caustic supernate on dissolution of an irradiated MTR wafer. One of these tests included an iodine measurement. [Table 2-2](#) gives the percent of fission products dissolved in caustic supernate in two different tests.

**Table 2-2 Percent of selected fission products dissolved into the caustic supernate**  
(Anderson et al. 1959)

Selected Fission Products	Test 1	Test 2 (filtered)
Ruthenium	10.7	0.10
Zirconium	0.045	--
Niobium	0.89	--
Iodine	17.1	--
Barium	0.30	--
Strontium	0.65	0.10
Cesium	96	99.5
Technetium	--	0.49

As can be seen, a surprisingly small quantity of iodine dissolved into the caustic supernate in this laboratory test.

Legler et al. (1957) report iodine concentrations in final process solutions at the end of preliminary Runs 001-RH, 002-RH, and 003-RH, and the first 5 RaLa production runs, Runs 001-RP to 005-RP (Table 2-3).

**Table 2-3 Iodine-131 material balance\* for several early RaLa runs**  
(Legler et al. 1957)

RaLa Run Number	Solution <sup>†</sup>					
	1	2	3	4	5	6
001-RH	No data					
002-RH					0.8	
003-RH	69.1	11.2	0.4	6.6	7.0	8.3
001-RP	--	10.5	0.02	112.8	9.6	2.6
002-RP	--	7.4	2.3	79.7	14.2	5.8
003-RP	--	0.2	5.2	27.7	5.5	--
004-RP	--	0.5	0.3	57.5	48.8	--
005-RP	50.8	0.2	0.3	10.0	18.6	--

\* The percent of I-131 measured in the final repository relative to the theoretical total amount of I-131 at that time

† Solutions:

1. Dissolver supernate (evidently before acidification?)
2. Uranium supernate
3. First chromate supernate
4. Other waste supernates
5. Scrubber solution
6. Decontamination solutions

As seen in [Table 2-3](#), the I-131 material accountability was quite erratic. According to the discussion in [Legler et al. \(1957\)](#), the erratic readings were caused by the sampling procedure that introduced air bubbles into the liquid, tending to strip out the iodine from acid solutions, leading to the possibility of iodine deposition in the sampling tubes.

Clearly, the caustic supernate is a significant final repository for iodine based on the two measurements, but more complete retention was expected during the design phase of the project than the 69.1% and 50.8% observed for Runs 003-RH and 005-RP.

The highly variable iodine inventory in the scrubber solution at the end of the run is also surprising. In two production runs, the percentage of I-131 captured by the scrubber was measured as less than 10% of the total amount at the end of the run. The significance of such low capture levels of I-131 on release to the atmosphere depends on both the iodine pathway through the process and on the quantities held in other process solutions. But clearly, such low retention in the scrubber liquid implies low importance of the scrubber relative to other final repositories of iodine.

Note that significant amounts of I-131 show up in decontamination solutions. This may explain occasional high releases observed several days following a dissolving run.

A third set of iodine distribution data is given in the September 1957 QPR ([QPR-ICPP, IDO-14419](#)). Since these tests were also performed during Runs 3 and 5, they may, in some way, be related to the tests reported in [Legler et al. \(1957\)](#).

Results of the measurements in the QPR are listed in [Table 2-4](#).

**Table 2-4 Distribution of iodine-131 in RaLa process streams with estimated release to the stack (QPR, IDO-14419)**

Hours After Start	Process Solution	Percent of I-131 Found		I-131 in Off-Gas, curies	
		Run 3	Run 5	Run 3	Run 5
0-7	Caustic supernate*	84.4	62.0	0.1	0.05
7-10	Acid dissolver	0.2	0.2	2.0	2.0
10-14	Chromate precipitation	5.2	0.1	~0	~0
14-16	Nitrate precipitation	--	--	0.5	3.5
16-20	Product drying condensate	5.0	19.0	0.3	2.5
0-20	Cell off-gas	0.007	0.01	1.0	2.0
0-20	Dissolver off-gas <sup>†</sup>	0.02	0.05	2.9	8.1
0-20	Scrubber <sup>‡</sup>	5.2	18.7	--	--

\* Before neutralization.

† Written as “total to dissolver off-gas.” This includes off-gas from the other vessels. It is not stated whether or not this upstream or downstream from the scrubber.

‡ The scrubber liquid is given as 1.0 M NaOH + 0.1 M Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>, indicating that the scrubber liquid was improved from 1 M NaOH prior to Run 3.

These tests must have used scintillation counters set up adjacent to the process equipment and the cell off-gas line, monitoring I-131 activity during a run. Further indication is that the dissolver off-gas and cell off-gas are given separately. This could not be done by the stack monitor system, which samples the combined flows in the stack. There must have been off-gas monitors operated by the Technical Division in addition to the stack monitors operated by the Health Physics Group of the ICPP. Evidently, these tests and any further ones that may have been conducted were never written up as a project report (or the report has been lost).

Table 2-4 is reproduced essentially as presented in the cited [QPR \(QPR-ICPP, IDO-14419\)](#). Note that the percent of total iodine found in the scrubber liquid closely agrees with [Legler et al. \(1957\)](#) for the same RaLa runs: 5.2% and 18.7%. The experiments may have been related and reported separately. As noted above, the reason for the large difference in percentage of iodine captured by the scrubber between Runs 3 and 5 is not known.

Another interesting observation is that the sum of the five operations in the top part of the table equals the entry for "Total Dissolver Off-Gas." This is an indication that the other RaLa process vessels vented to the DOG during the test, and hence onto the scrubber.<sup>7</sup> As noted earlier, information on off-gas flow routing from RaLa process vessels is equivocal as given by the descriptions in [Legler et al. \(1957\)](#).

A relatively large percentage of the total I-131 at the end of the run was found in the caustic dissolving solution before neutralization, 84.4% and 62.2%. A portion of the I-131 decays, depending on when acidification occurs. Most of the balance subsequently entered the off-gas after acidification. (Later, possibly March 1959, for Run 24, mercury salts began to be used to limit volatilization of iodine from acid solutions.)

## 2.6 Operational History and Process Modifications

No primary sources of information have been found on RaLa operational history and process modifications after [Legler et al. \(1957\)](#) or after Run 5. The available information from secondary sources is summarized in Table 2-5. The RaLa run record in Table 2-5 is essentially identical to that adopted by the [HDE \(DOE 1991\)](#).

The operational changes noted in [Table 2-5](#) were intended to reduce evolution of iodine from process solutions. Results reported in the [Health and Safety Annual \(1959\)](#) and [Bower and Buckham \(1961\)](#) indicate that the steps were successful. According to [Bower and Buckham \(1961\)](#), I-131 releases to the atmosphere were reduced from 1.03% of inventory charged averaged over Runs 1-5, to 0.018% averaged over Runs 25 and 26 in April 1959. These reported data are not referenced to permit verification.

According to [Cederberg and MacQueen \(1961\)](#), the change of scrubber solution from 1 M NaOH to 1 M NaOH + 0.1 M Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> (sodium thiosulphate) improved scrubber efficiency from 90% to 97%. No reference or other support is given for this assertion. However, there is a somewhat speculative chemical basis to expect some degree of improved efficiency.

---

<sup>7</sup>All the RaLa vessels had off-gas connections with both the DOG and VOG venting systems.

**Table 2-5 Record of RaLa runs and operational changes**All run dates are from [H&SD Annual \(1959\)](#)

<i>Run Number</i>	<b>Run Dates</b>	<b>Operational Changes, Comments</b>	<b>Reference</b>
<b>1957</b>			
<b>1</b>	February 1–3		
<b>2</b>	February 20–21		
Between Runs 2 and 3		Installed 10,000 ft <sup>3</sup> gas accumulator	<a href="#">Project Letters (1957)</a>
<b>3</b>	April 5–6	Scrubbing solution changed from NaOH to NaOH + Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub>	<a href="#">Legler et al. (1957)</a>
<b>4</b>	May 19–20		
<b>5</b>	June 24–25		
<b>6</b>	September 11	Improved seals on centrifuge	<a href="#">Bower and Buckham (1969);</a> <a href="#">Project Letters (1957)</a>
<b>7</b>	October 8		
<b>8</b>	October 21		
<b>1958</b>			
<b>9</b>	January 6		
<b>10</b>	February 12		
<b>11</b>	March 13–14		
<b>12</b>	April 16		
<b>13</b>	April 30–May 1		
<b>14</b>	May 28		
<b>15</b>	June 2		
<b>16</b>	August 6	Carbon filter beds first utilized	<a href="#">H&amp;SD Annual (1959)</a>
<b>17</b>	August 13		
<b>18</b>	October 1	Reduced sampling	<a href="#">Bower and Buckham (1969)</a>
<b>19</b>	October 22		
<b>20</b>	October 22–23	Probable run numbering error, no run	
<b>21</b>	November 12		
<b>1959</b>			
<b>22</b>	February 4		
<b>23</b>	February 25		
<b>24</b>	March 18	Scrubbing solution changed to acid plus Hg(NO <sub>3</sub> ) <sub>2</sub>	<a href="#">Bower and Buckham (1969)</a>
<b>25</b>	April 8	Discontinued heating centrifuge jacket Added Hg(NO <sub>3</sub> ) <sub>2</sub> to final supernate receiver	<a href="#">Bower and Buckham (1969)</a>
<b>26</b>	April 29		
<b>27</b>	May 19		
<b>28</b>	June 10	Added Hg(NO <sub>3</sub> ) <sub>2</sub> to uranium supernate receiver	<a href="#">Bower and Buckham (1969)</a>
<b>29</b>	July 6–7	Start of "2-day" runs	<a href="#">H&amp;SD Annual (1959)</a>
<b>30</b>	July 21–22		
<b>31</b>	August 11–12		

The improved seals on the centrifuge, reported as occurring prior to Run 6 (September 11, 1957), were intended to reduce the leakage from the centrifuge bowl into the cell air space. As discussed in the next section, the cell air contamination joins the building ventilation flow, bypassing the scrubber, and therefore has the potential for contributing significantly to the stack activity.

The carbon filters were installed prior to Run 16 (August 6, 1958) in an adjacent building (CPP-631), downstream from the scrubber. Since all process vessels vented through the scrubber, an additional absorber in this off-gas line could have a significant reduction of iodine emissions. [Cederberg and MacQueen \(1961\)](#) assert a 97.4% efficiency for the charcoal beds, without reference or other backup. Since the absorption efficiency of charcoal is known to diminish with age, moisture, and acid in the air, this high efficiency may be representative of an early test. Average efficiency over the lifetime of the bed is likely much lower and would depend to some degree on the frequency of regeneration.

[Cederberg and MacQueen \(1961\)](#) do not mention additives to the charcoal to make it effective for absorbing organic iodine. Since it is expected that such additives would be mentioned, it may be assumed that there were none, and that the installed charcoal beds evidently did not affect the emission of organic iodine.

The reduced sampling schedule cited as occurring prior to Run 18 on October 1, 1958, would have the effect of generally reducing the off-gas flow and therefore generally reducing iodine emissions. There are two reasons for this, but the effect is difficult to quantify. First, the sampling procedure calls for agitation of the vessel contents to increase the chance of obtaining a representative sample. Agitation was either by jetting in nitrogen or by recirculating the liquid by jet-lifting. Either method adds gas volume to the tank and adds to the rate of gas evolution. Second, sample acquisition requires a jet-pumping procedure adding further to the volume of gas added to the system, which would then be relieved by venting to the off-gas system.

The scrubber solution change to nitric acid containing mercury salts (March 18, 1959, prior to Run 24) was motivated by operational considerations and likely did not improve scrubber efficiency. No reports have been found on scrubber efficiency with the acid solution (or for that matter, any scrubber efficiency). According to [Cederberg and MacQueen \(1961\)](#), the main objection to the sodium hydroxide – sodium thiosulfate scrubbing solution was that it caused formation of solid carbonates by reaction with CO<sub>2</sub> in air, fouling flow-metering equipment.

[Table 2-6](#), taken from [Legler et al. \(1957\)](#), provides data for the first five RaLa runs. The charged fuel elements are identified, which identifies the irradiation history from the MTR records provided in the MTR monthly progress reports. The important parameter, preprocessing cooling time, is provided for these five runs. Note that the nominal 2 days was achieved only once in these first five runs.

**Table 2-6 Data for initial runs involving irradiated fuel**  
(Legler et al. 1957)

Run Number	Element	Run Dates	Pre-processing Cooling Time (days)	Total Charged Uranium	Calculated Ba-140 (Ci)	Ba-140 in Product (Ci)
001-RP	P-126	February 1-3	5	143	38,000	18,000
002-RP	P-146	February 20-21	2	158	43,700	49,000
003-RP	P-227	April 5-6	5	192	23,800	18,600
004-RP	P-237	May 19-20	7	172	29,150	21,600
005-RP	P-285	June 24-25	6	163	32,000	14,500

## 2.7 Off-Gas Flow System

### 2.7.1 The Idaho Chemical Processing Plant Ventilation and Off-Gas System

The airflow system on the ICPP consisted of three elements (Ayers and Burn, 1960; Girton et al. 1973), described as follows:

#### *The Building Ventilating System*

Work areas in the main process building (CPP-601), the adjacent laboratory building (CPP-602), and hot pilot plant (CPP-640) were kept under negative pressure by fans drawing about 100,000 scfm (standard cubic feet per minute).<sup>8</sup> The direction of flow was from the office areas, through the main corridors, into the process cells through louvers, from the cells through adjustable vents, and into vent corridors that ran along the outside walls.

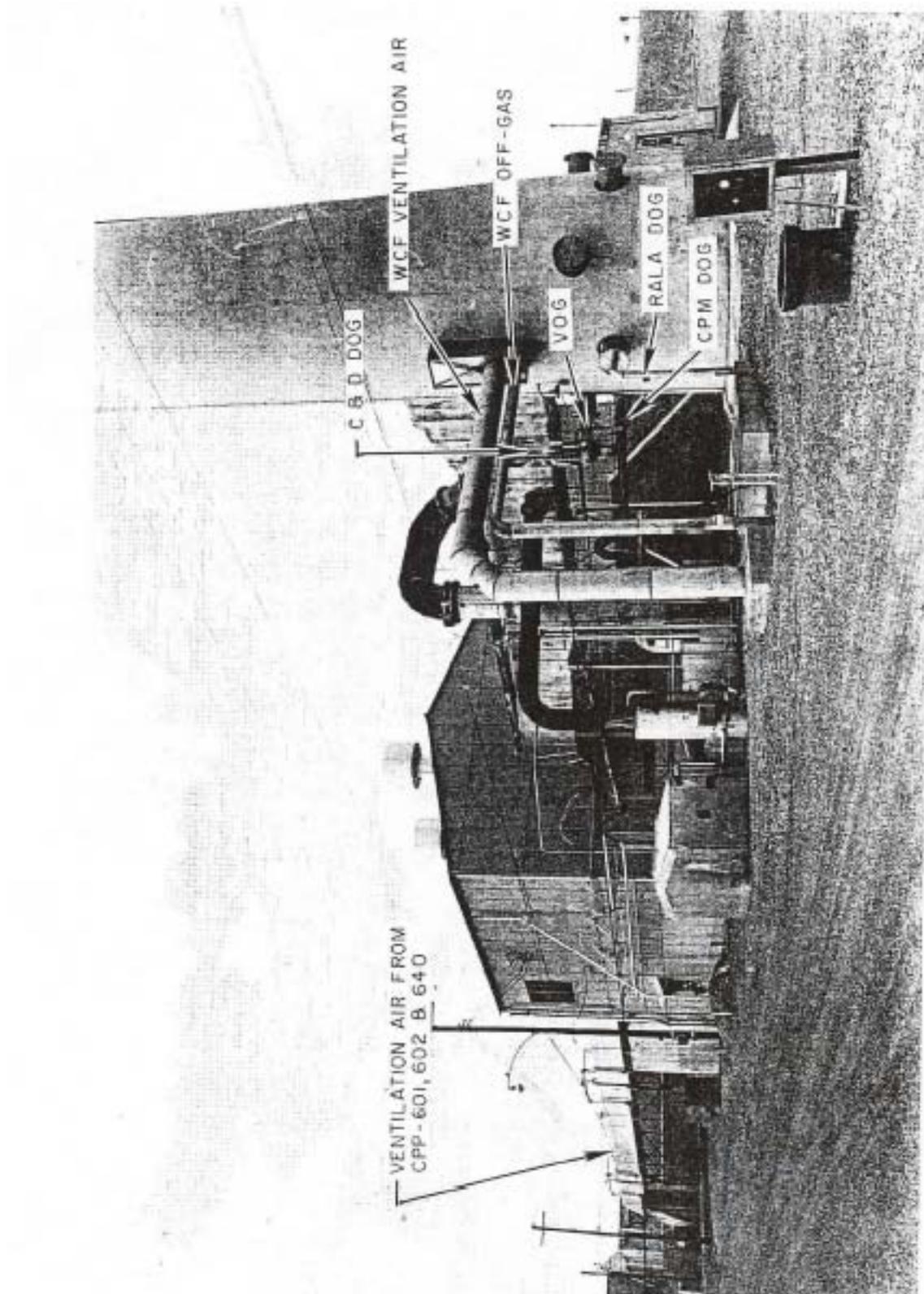
The exhaust was led to the stack through a 10-ft wide by 7-ft high insulated duct that is clearly seen in Figure 2-9 (Girton et al. 1973). The main process building is in the background. The duct passes through the Intermediate Waste Disposal Building (CPP-604) untouched.

#### *The Vessel Off-Gas System*

All ICPP vessels, including storage vessels in the building and adjacent liquid waste vessels but excluding non-RaLa dissolvers, were connected to the VOG system held under negative pressure by two 2,500 scfm capacity blowers. (Evidently, all of the RaLa vessels including the dissolver were also connected to the VOG, but with valving changes, could also be led into a separate RaLa dissolver off-gas system (Ayers and Burn 1960).

The VOG was led into the filter room of the Waste Disposal Building (CPP-604), shown in the foreground of Figure 2-9, and then on to the stack. At the time of Ayers and Burn's paper (1960), the filters were bypassed while being improved from one type of glass wool filter medium to another.

<sup>8</sup>Ayers and Burn (1960) say there were two 90,000-scfm blowers, but recorded stack flows were on the order of 100,000 scfm.



**Figure 2-9 Photograph of gas flows into the stack as of 1973**  
(Girton et al. 1973)

The VOG flow rate is given by [Girton et al. \(1973\)](#) as 1,000 scfm, but this must be highly approximate as the suction fans are deadheaded by sealed vessels. All flow appears to result from gas volume added to the system by means of jet lifts, nitrogen mixing, or sampler operations. The exit duct from the Waste Disposal Building is seen in Figure 2-9 as it enters the stack.

### *The Dissolver Off-Gas System*

Descriptions in [Ayers and Burn \(1960\)](#) and [Girton et al. \(1973\)](#) differ somewhat, possibly reflecting the difference in time between the writings. The present investigation is concerned mainly with the RaLa DOG, which in Figure 2-9 is clearly shown as a separate line entering the stack.

[Ayers and Burn \(1960\)](#) state that all RaLa vessels, including the dissolver and centrifuge, may send off-gas to either the VOG system or to the special RaLa DOG. This is corroborated by a figure in [Cederberg and Bower \(1959\)](#). Note that the detailed flow sheet around the dissolver, [Figure 2-4](#), clearly shows that all the RaLa vessels can vent to the scrubber (i.e., the DOG) with valves in each line that may modify the routing. There is no discussion on why this was the case or when the off-gas flows were routed into either system.

When routed into the DOG, the off-gas passed through the scrubber and then through one of two parallel charcoal beds (after they were installed) in the RaLa Off-Gas Building (CPP-631) located adjacent to the main process building. However, [Ayers and Burn \(1960\)](#) state that the charcoal beds were bypassed during the time of noble gas release, i.e., during the caustic dissolving process.<sup>9</sup>

Flow from the charcoal beds (or bypass flow around the beds) proceeded through a valve box (CPP-726) into the 10,000-ft<sup>3</sup> storage vessel (CPP-727), after its fabrication in March 1957. Valve box controls permitted DOG flow either into this gas storage vessel or out of the storage vessel and into the stack CPP-708. [Figure 2-10 \(Smith and Bradford 1981\)](#) is useful in corroborating a portion of the RaLa DOG.<sup>10</sup>

[Figure 2-11](#) summarizes the RaLa and ICPP off-gas flow as it existed in 1957-1959 from the various discussions.

### **2.7.2 Description of the Dissolver**

The dissolver is described in [Legler et al. \(1957\)](#) (page 54). Figure 11 of that reference presents a detailed drawing.

---

<sup>9</sup>This probably had more to do with hydrogen evolution than noble gas evolution.

<sup>10</sup>The later report, [Girton et al. \(1973\)](#), states that the RaLa DOG was routed to the Waste Treatment Building (CPP-604) where it would be either treated for noble gas removal or passed into the “CPM-DOG cleanup system.” This may reflect a later development. Also, [Chamberlain \(1972\)](#) states that the RaLa DOG routing into the Waste Treatment Building existed in 1972. However, it has not been noted in 1957–1959 reports.

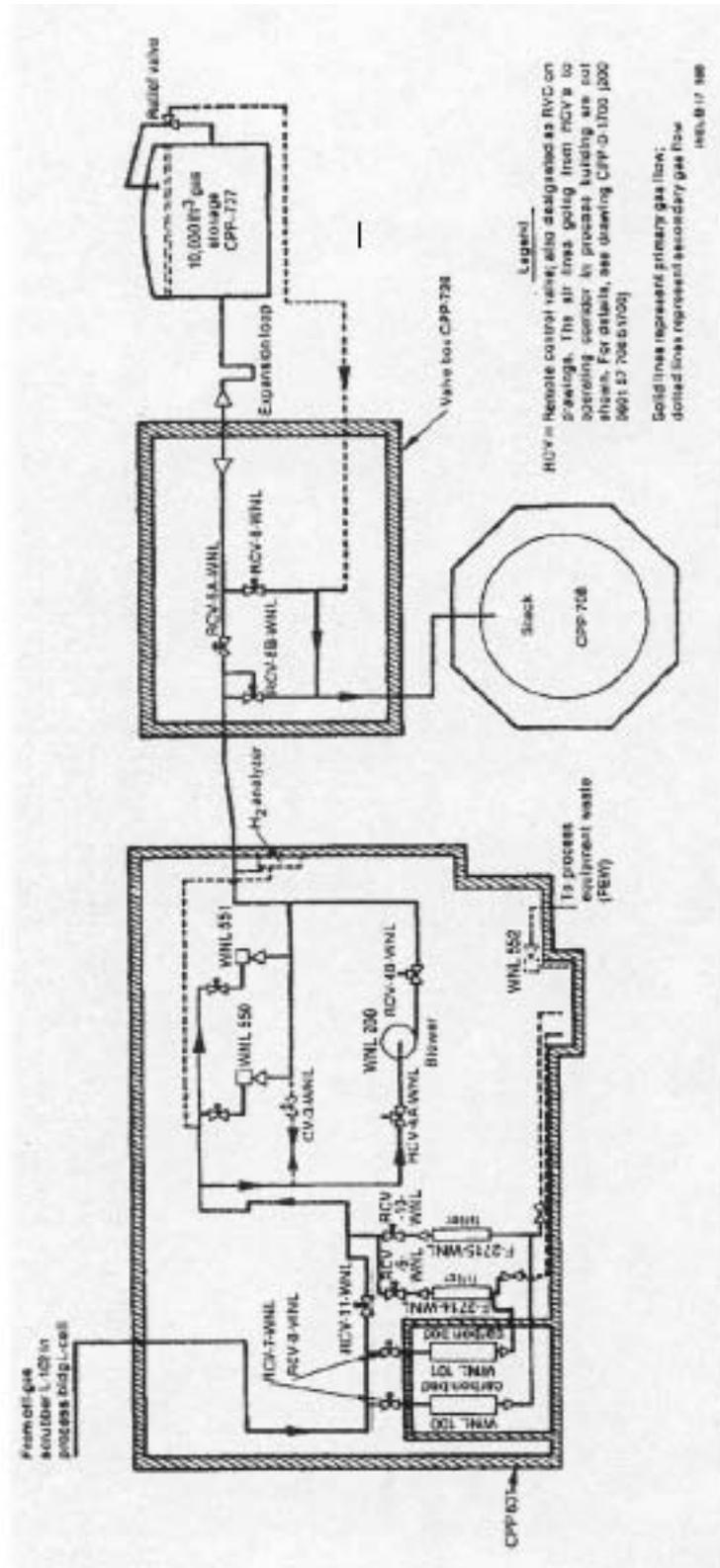


Figure 2-10 RaLa off-gas flows downstream from the scrubber (Smith and Bradford 1981)

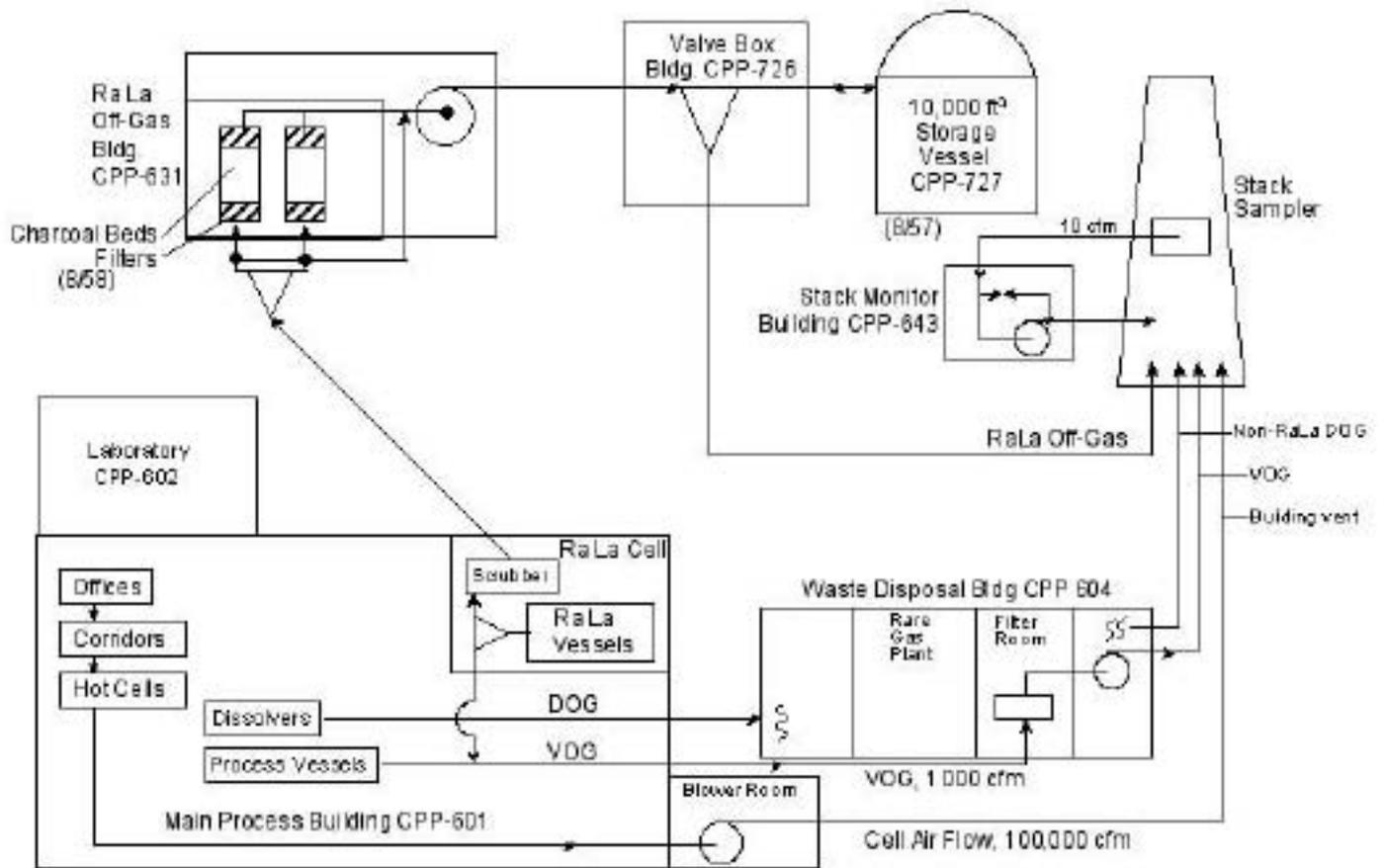


Figure 2-11 RaLa and Idaho Chemical Processing Plant off-gas flow schematic, 1957–1959

### 2.7.3 *Description of the Scrubber*

A brief physical description of the scrubber is given by [Legler et al. \(1957; page 54\)](#). There is no detailed drawing in this reference.

[Figure 2-4](#), reproduced from [Legler et al. \(1957\)](#), illustrates the highly flexible arrangements of process connections to and from the scrubber (L-102). The capability is shown for off-gas input from all the RaLa vessels, including the scrubber waste tanks (L-103 and L-153) as well as from the “VOG,” possibly meaning the ICPP VOG. The contents of the scrubber waste tanks are shown as steam-jetting to the waste tank L-110.

### 2.7.4 *Operation of the Charcoal Beds*

A general description of the charcoal beds is provided by [Cederberg and MacQueen \(1961\)](#). [Ayers and Burns \(1960\)](#) state that the beds have stainless steel filters (presumably on the inlet and outlet) and that the beds are bypassed during the noble gas evolution phase of RaLa operation, i.e., the caustic dissolving.<sup>11</sup> There were provisions for regenerating the bed with steam and replacement of charcoal if necessary. [Ayers and Burns](#) state further that the beds were usually dried out before a run, but that regeneration or replacement was not necessary during the first two years of operation.

## 2.8 Summary and Conclusions

- Review of the RaLa chemical process shows that it must be described as complex. It would be impossible to calculate iodine releases from chemical and mass transfer principles even with full knowledge of process variables. (Releases presented in Chapter 7 are based on modified stack monitor readings.)
- Search of the available database found no primary sources for RaLa operation during the 1957–1959 years (i.e., official project reports, log books, etc.) after September 1957, except for the unpublished Stack Monitor Datasheets.
- It is apparent from the schedule of early activities that the final design, initiated in July 1954, could not have accounted for all the process features being tested in the cold pilot plant or the hot laboratory tests. As a result, process deficiencies were revealed on startup and were corrected during early operation.
- Examination of the early development work indicates that iodine behavior in the system was only minimally studied and never clearly understood. Design assumptions regarding the purported high retention of iodine in caustic supernate were not realized.
- An important early design deficiency was the weak seals on each centrifuge shaft. Effectively, the dissolver bowls were not sealed during the early runs.

---

<sup>11</sup>The reason is not given, but bypassing the beds may have had more to do with hydrogen evolution than noble gas evolution. This could have been a safety precaution. Hydrogen is evolved along with the noble gases during caustic dissolving.

Consequently, much of the iodine that evolved downstream from the caustic dissolver entered the cell air system, bypassing the scrubber.

- It is not clear why all RaLa vessels were valved so as to vent to either the DOG, which led to the scrubber, or to the VOG, which did not. No discussion was found regarding this. Presumably, the vessels vented to the DOG most of the time.
- All iodine leakages from either the DOG or VOG into the ICPP cell air system would ultimately be routed to the stack and thus be detected by the stack monitor system.
- An attempt was made to identify possible iodine release paths that would bypass such detection. The main process building, held under negative pressure, could not be such a source. However, leakage from the Off-Gas Cell (CPP-631) that housed the charcoal beds could be such a theoretical possibility.

### 3.0 IODINE BEHAVIOR IN THE RALA SYSTEM

As seen from the fragmentary data in the last section, iodine was distributed in various process solutions and evolved at various stages. In addition, it will be seen when the stack monitor data are reviewed that releases of I-132, though less significant than I-131, were occasionally quite high. Attempts to rationalize the observed iodine releases should start with an evaluation of the chemical behavior of iodine in the process.

#### 3.1 Radioactive Properties of Iodine Radionuclides

##### 3.1.1 Birth and Decay Diagram

Figure 3-1 illustrates the birth and decay schemes for I-131, I-132, and I-133 (Voigt 1976).

The top row of Figure 3-1 is the cumulative yield up to that atomic number in atoms produced per 100 fissions. Note that these iodine isotopes are born predominantly as antimony (Sb), which  $\beta$ -decays rapidly to tellurium (Te).

For mass-131, 10% of the Sb-131 decays to a fairly persistent Te-131m, which has a 30-hour half-life. A little more than half of the Te-131m would decay in the nominal 2-day cooling period after MTR irradiation. Therefore, about 4% of the mass-131 entered the dissolver as Te-131m, the balance as I-131.

The decay scheme for I-132 shows that it spends a significant time as Te-132, which has a 78.2-hour half-life. Therefore, the mass-132 entering the dissolver consisted of the following:

- A small amount of I-132 that was the remnant of the equilibrium amount produced in the reactor, most of which decayed away in the 2-day cooling period
- The Te-132 parent
- Iodine-132 produced from the Te-132 decay during the 2-day cooling period

In a mixture of fission products, the effective half-life of I-132 would actually be closer to 78.2 hours than its intrinsic value of 2.3 hours, since its concentration is controlled by the decay of the parent, Te-132. Iodine-132 that becomes separated from the parent, as, for example, in the off-gas line, decays with its intrinsic rate. The longer half-life of Te-132 could have caused a separation of I-132 from I-131 or I-133, as, for example, by a different solubility in the dissolver.

Precursor behavior for I-133 is the simplest. It is largely born as Te or below, all of which decays in the 2-day cooling period. Therefore, essentially all of the mass-133 nuclides entered the dissolver as I-133. In this respect, it is similar to I-131. Therefore, it should be expected that I-131 and I-133 will largely track each other along the transport pathway, accounting for the different rates of decay.

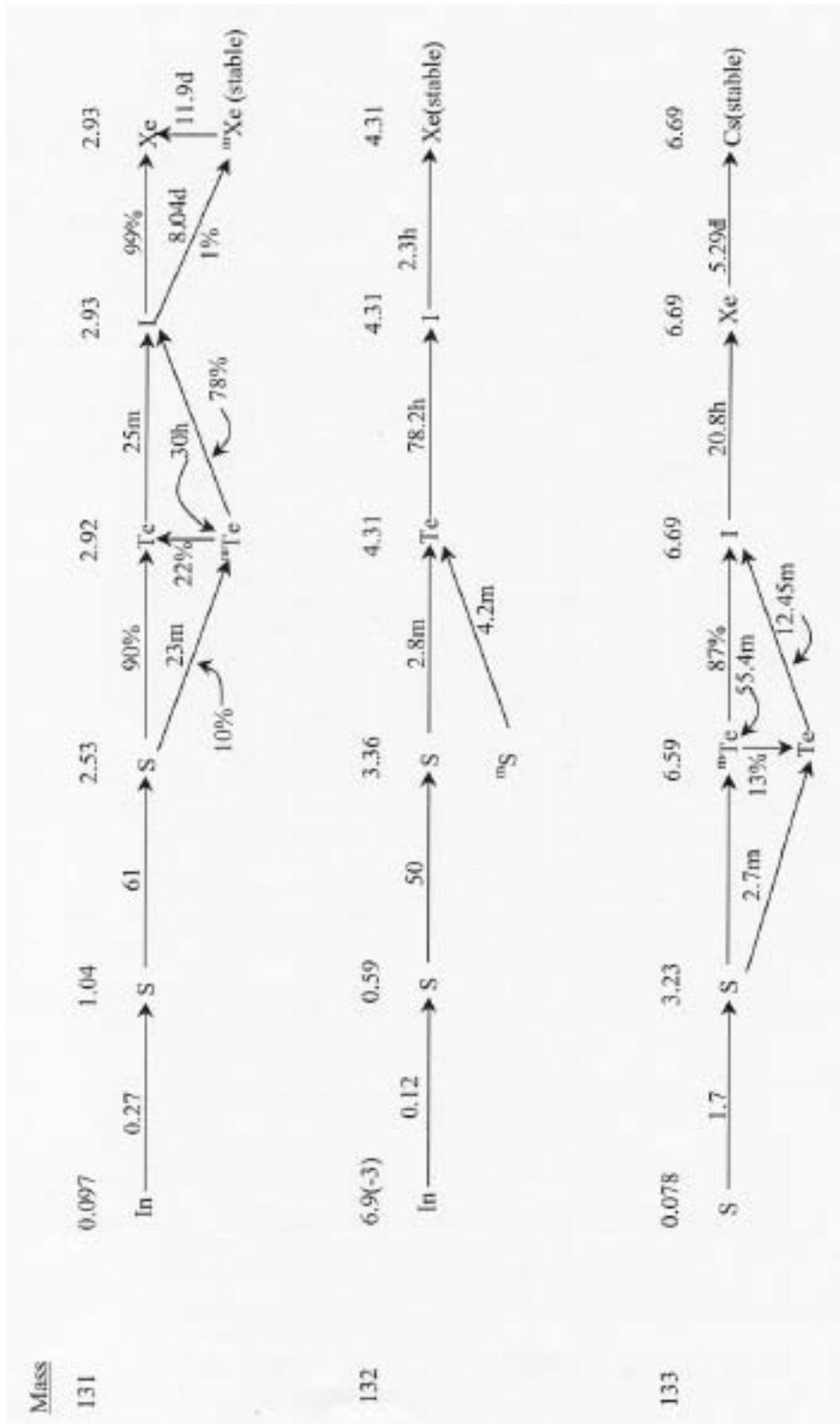


Figure 3-1 Iodine birth and decay diagrams (Voight 1976)

In repositories in which it decays, iodine forms the daughter product, xenon (Xe), notably radioactive Xe-131m with an 11.9-day half-life, and Xe-133 with a 5.29-day half-life.

### 3.1.2 Iodine and Tellurium Inventory in the Feed

Some typical abundances in 2-day cooled MTR fuel fed to the dissolver are given in [Table 3-1](#). The large inventory of I-132 (totaling 65,200 Ci) is more than counter-balanced by its low dose factor—about 150 times lower than I-131 for breathing dose to the thyroid. However, on occasion, I-132 releases were observed to be quite high; on two occasions more than 50 times higher than I-131, and generally about 15 times higher on the day of the dissolving. In addition, a very large I-132 release was monitored following the ICPP criticality accident on October 16, 1959, which was obviously incorrect. For these reasons, some interest in I-132 needs to be maintained.

The inventory of I-133 in the feed is seen to be lower than I-131; in addition I-133 has a lower dose factor for breathing to the lung. Moreover, its formation/decay diagram indicates its release path should track that of I-131. Hence, its release should follow similar rules, accounting for different inventories and decay rates. Therefore, although it was never monitored, it can be assumed that the effect of I-133 is small relative to I-131.

**Table 3-1 Typical iodine and tellurium inventories in 2-day cooled Materials Testing Reactor fuel (curies)**

Isotopes	Inventory (Ci)
Te-131	375
Te-131m	1,660
I-131	24,000
Te-132	32,100
I-132	33,100
Te-133	nil
Te-133m	nil
I-133	17,100

### 3.1.3 Radioiodine Gamma Ray Emissions

A [Project Letter \(1957\)](#) states that an early  $\gamma$ -spectroscopy system was installed in the gas sampler room of the RaLa off-gas system in early 1957. According to an expert ([Dyer 2002](#)), early  $\gamma$ -spectroscopy systems based on sodium iodide crystal scintillation crystals generally counted contributions from  $-10\%$  to  $+10\%$  from the central peak energy.<sup>1</sup> Therefore, the possibility of counting errors from impurities was higher than in later, more discriminating  $\gamma$ -spectroscopy systems.

<sup>1</sup>Dr. F.F. Dyer, now retired from Oak Ridge National Laboratory, is a radiochemist who has been active for many years going back to the early 1960s.

A second hurdle was acquisition of standard samples. The standard is an isotopically pure sample used to calibrate the counts per minute reading into curies. The 16-month delay in the start of I-132 stack monitor reporting may have been due to the greater difficulty in making an I-132 standard. The absence of I-133 readings may have been due to the virtual impossibility of making an I-133 standard.

Table 3-2 lists all  $\gamma$ -energy peaks of I-131, I-132, and I-133 over 4% intensity (Kocher 1981).<sup>2</sup> The dominant peaks, the ones that the measurement would have focused on, are in bold type. Note that the principal I-131  $\gamma$ -emission is at 365.4 keV energy occurring with 81.2% efficiency. The assumption of a +/-10% error in the energy band around the peak yields a detection range of 328-401 keV. Table 3-2 shows that there are no significant emissions from either I-132 or I-133 to interfere with the I-131 determination. Therefore, there should have been no problem reading I-131, assuming no other contamination.

The principal I-132 emission is at 667.7 keV energy, occurring with 98.7% efficiency. Assuming a +/-10% error band yields a detection range of from 601-734 keV. There would be a 7.26% interference in this range from I-131. In other words, if this peak were used, the reading would be enhanced by 7.26% due to I-131 interference. On the other hand, a less intense peak at 772.6 keV could have been used with no other iodine interference.

The principal I-133 peak occurs at 529.9 keV with 86.3% efficiency. A +/-10% error range places two I-132  $\gamma$ -emissions in this range. Thus, if an I-133 determination were made, the reading would be enhanced by about 21% due to two I-132 interferences.

In summary, I-131 determinations could be made with no interference from other radioiodines, even using the old model NaI detectors. Iodine-132 and I-133 readings could be taken with relatively minor interferences from other radioiodines. The principal presumption is that there were no unknown contaminants.

---

<sup>2</sup>Intensity is the number of  $\gamma$ -emissions per 100 disintegrations at that energy level.

**Table 3-2 Principal radioiodine gamma ray emissions (greater than 4% intensity)**

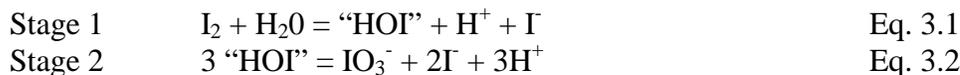
Energy Range keV	I-131				I-132				I-133			
	Energy Peak keV	Intensity %	NaI Range plus 10% minus 10%		Energy Peaks keV	Intensity %	NaI Range plus 10% minus 10%		Energy Peaks keV	Intensity %	NaI Range plus 10% minus 10%	
0-100												
101-200												
201-300	284.3	6.05	312.73	255.87								
301-400	<b>364.5</b>	<b>81.2</b>	<b>400.95</b>	<b>328.05</b>								
401-500												
501-600					505.9	5.03	556.49	455.31	<b>529.9</b>	<b>86.3</b>	<b>582.89</b>	<b>476.91</b>
					522.7	16.1	574.97	470.43				
601-700	637	7.26	700.7	573.3	630.2	13.7	693.22	567.18				
					<b>667.7</b>	<b>98.7</b>	<b>734.47</b>	<b>600.93</b>				
					669.8	4.9	736.78	602.82				
					671.6	5.2	738.76	604.44				
701-800					<b>772.6</b>	<b>76.2</b>	<b>849.86</b>	<b>695.34</b>				
801-900					812.2	5.6	893.42	730.98	875.3	4.47	962.83	787.77
901-1000					954.6	18.1	1050.06	859.14				
1001-1100												
1101-1200												
1201-1300												
1301-1400					1398.6	7.1	1538.46	1258.74				

### 3.2 Iodine Behavior in the Caustic Dissolver

The caustic dissolver solution was highly concentrated sodium hydroxide (NaOH), initially at a concentration of about 5 M (moles per liter). The purpose was to dissolve the aluminum cladding, thereby removing this large mass from the rest of the chemical process. The reaction produced dissolved sodium aluminate and evolved copious amounts of hydrogen gas. Mixing in the dissolver was provided by jets of nitrogen, which also served to dilute the hydrogen to below combustible limits (Legler et al. 1957). Indications are that the dissolver temperature was near the boiling point of 100°C, at least for the early runs, but may have been reduced later for better control of the dissolving reaction.

The alkaline dissolving process pretty well fragmented the uranium in the uranium-aluminum alloy fuel, exposing it to the action of the NaOH. The working hypothesis of the original design appears to have been to take advantage of the well-known solubility of iodine in NaOH to trap most or all of the iodine in the caustic solution.<sup>3</sup> As seen by the fragmentary test data in the previous section, the design hypothesis did not prove to be entirely correct.

The iodine exposed in the fragmented uranium-aluminum alloy fuel dissolves in the NaOH solution by means of the hydrolysis reaction with water. The NaOH environment serves to enhance the reaction. The hydrolysis reaction is given below as Stage 1, followed by a disproportionation reaction of the intermediate product, HOI.



HOI is written in quotation marks because some intermediate product like it must exist, but has not been separated as a pure compound (Toth et al. 1984). Toth states that Stage 1 is reported to be rapid, going to completion in seconds. Stage 2 is much slower and highly dependent on conditions. In a fairly hot (65°C), strongly basic solution of pH 10, Stage 2 proceeds to equilibrium on the order of hours. At lower temperatures and less basic solutions, the second stage may take days to complete. Therefore, Stage 2 would not be a factor in either the dissolver or scrubber.

The intermediate “HOI” may have some volatility by analogy with similar halogen compounds. Therefore, some volatilization of iodine from the dissolver solution could have occurred despite the highly favorable equilibrium constant of the reaction.

Iodine trapped within the uranium-aluminum alloy fuel fragments and undissolved iodine would proceed along with the other undissolved material to the next stage of the process.

---

<sup>3</sup>The plan had a loop-hole in that the dissolver solution needed to be stabilized by acidification soon after removal to the dissolver storage vessel. Acidification released the dissolved iodine until later use of mercury suppression.

### 3.3 Iodine Behavior in the Centrifuge and Acidified Waste Tank

In the centrifuge, the fragmented uranium is dissolved in nitric acid, exposing the iodine trapped in the fuel to the acid environment. The aluminate supernate solution, decanted off to the dissolver waste tank, is acidified with nitric acid soon after the transfer in order to stabilize the solution and render it compatible with other ICPP waste solutions.

Prior to the recognition of the beneficial effect of the mercury complexing agent (possibly in March 1959), much of the dissolved iodine in the waste tanks evolved as iodine bubbles when the solution was acidified. This was due to the reversal of the hydrolysis reaction caused by the high concentration of  $H^+$  ions in the acid solution. The effect drives the reaction back to the left, according to the well-known principle of Le Chatelier, which states that a reaction (Eqs. 3.1 and 3.2) proceeds in the direction to relieve an imposed stress. Here the imposed stress is the high concentration of  $H^+$  in the acid, which is relieved by the leftward direction of the reaction.

A similar effect occurred in the centrifuge bowl during nitric acid dissolution of uranium. The portion of the iodine that escaped the alkaline dissolver by being trapped in the uranium fragments was exposed to the acid solution in the centrifuge. Much of this iodine would evolve as bubbles due to the ineffectiveness of the hydrolysis reaction in acid media.

In sum, iodine retention in the acidic RaLa solutions was low until the technique of mercury stabilization was adopted, possibly in March 1959. (Mercury stabilization is discussed in Section 3.5.)

### 3.4 Iodine Behavior in the Scrubber—Effect of Sodium Thiosulfate

During the first few months of operation, the scrubber solution was 1 M NaOH. During this period, Cederberg and MacQueen (1961) allege that the scrubber efficiency was 90%.<sup>4</sup> Contributing to this relatively low scrubber efficiency may have been some degree of volatility of the hydrolysis product, HOI. An additional factor may have been the slow kinetics of the hydrolysis reaction. Even though Stage 1 is stated as going to completion in a matter of seconds, contact times in the scrubber were also a matter of seconds. Therefore, insufficient contact time may also have been a factor in the relatively low initial scrubber efficiency. In sum, NaOH proved to be a rather poor scrubber liquid despite the highly favorable equilibrium of the hydrolysis reaction.

After a few months of operation with 1 M NaOH, 0.1 M sodium thiosulfate ( $Na_2S_2O_3$ ) was added to the solution. According to Cederberg and MacQueen (1961), doing so increased the scrubber efficiency to 97%.<sup>5</sup> Sodium thiosulfate is a reducing agent whose effect would have been to reduce the volatile form,  $I_2$ , to the dissolved anion,  $I^-$ , according to the following reaction.



---

<sup>4</sup>This statement is unsupported in their text and unreferenced.

<sup>5</sup>This statement is unsupported in their text and unreferenced.

This appears to be the basis of the observed improved iodine dissolving efficiencies using thiosulfate. It evidently was free of kinetics or volatile intermediate limitations.

According to [Bower and Buckham \(1969\)](#), the scrubber solution was changed to nitric acid containing mercury salts as iodine sequestering agents in March 1959. The major objection to the alkaline scrubber solution was the formation of solid carbonates by reaction with CO<sub>2</sub> in air ([Cederberg and MacQueen 1961](#)). It is unlikely that this change in scrubber solution improved efficiency. However, by this time the charcoal absorbers had been installed in the dissolver off-gas line in series with the scrubber. Therefore, the precise efficiency of the scrubber was not as significant as before.

### 3.5 Mercury Complexing in Acid Solutions

For the operational reasons cited above, the scrubber solution was changed to a 5% nitric acid containing 0.001 M mercuric nitrate (Hg(NO<sub>3</sub>)<sub>2</sub>) and 0.001 M mercurous nitrate (HgNO<sub>3</sub>) in March 1959. Presumably at about the same time, the same combination of mercury salts was used to minimize iodine evolution from the acid and acidified RaLa solutions, such as the centrifuge bowl during uranium dissolution and the dissolver waste tank after acidification of the caustic.

Sequestering of iodine in acid solutions by complexing with mercury salts was first observed in isotope production activities at Oak Ridge in the 1940s. The first detailed study was in reprocessing tests at Hanford ([Holm 1951](#)).<sup>6</sup> Holm reported the following effect ([Table 3-3](#)), using a simulated dissolver solution under conditions of 10<sup>-5</sup> M concentration of iodine, 45°C temperature and 1.7 cc/min. liter air sparging rate.

**Table 3-3 Effect of mercury concentration on iodine removal by air sparging**  
([Holm 1951](#))

Mercury concentration	% evolved in 1 hour iodine added as:		
	I <sup>-</sup>	I <sub>2</sub>	IO <sub>3</sub> <sup>-</sup>
0.0	40.3	45.5	5.52
10 <sup>-6</sup>	25.8	30.0	1.34
10 <sup>-5</sup>	1.31	5.93	0.65
10 <sup>-4</sup>	0.006	0.05	0.009
10 <sup>-3</sup>	--	0.001	--

The top row of the table shows that air sparging of an acid solution of iodine without mercury removed only about 50% of the iodine added as I<sup>-</sup> or I<sub>2</sub> in 1 hour of sparging, and a much lower portion of iodate. This indicates that iodine evolution in the centrifuge during acid dissolution of

<sup>6</sup>Later studies at Hanford showed that it could not be used for redox reprocessing due to mercury contamination of the product.

uranium was probably incomplete, explaining the unexpected presence of iodine in later stages of the RaLa process.

Table 3-3 also shows that addition of as little as  $10^{-4}$  M mercuric nitrate ( $\text{Hg}(\text{NO}_3)_2$ ) to an acid solution containing iodine significantly inhibited evolution by air sparging. Increasing the mercury concentration to  $10^{-3}$  M had a further beneficial effect.

It is not recorded who came up with the RaLa composition of 0.001 M mercuric plus 0.001 M mercurous nitrate. No reports have been found on the degree of iodine evolution with this particular mercury composition, but presumably it was at least as good as that given above for mercuric nitrate alone.

It is implied that the mercury-complexed acid solution improved scrubber efficiency. This is doubtful and unsupported. The change was made for operational reasons.

Addition of mercury to process solutions poses a mercury disposal problem. Recycle of mercury appears to be feasible (Holladay 1979), but was evidently not a part of the Idaho RaLa process. Therefore, the mercury must have stayed with the waste solution and was treated at the Waste Calcining Facility.

### 3.6 Behavior of Iodine in the Stack Monitor Sampler

According to a letter by Rich (Rich 1957), the stack sampler solution, at least in June 1957, was an alkaline solution of 0.05 M NaOH plus 0.05 M  $\text{Na}_2\text{S}_2\text{O}_3$ . As discussed above, addition of thiosulfate evidently overcomes the deficiencies of the pure NaOH scrub solution by improving kinetics and reducing possible volatile intermediate iodine compounds.

### 3.7 Tellurium Chemistry in the Alkaline and Acid Dissolvers

The chemistry of tellurium is a factor in assessing the release of I-132 because of the relatively long half-life of the precursor, Te-132. The formation/decay diagram, Figure 3-1, shows that tellurium chemistry can have only a minor effect on I-131 release and would have no effect on I-133 for 2-day cooled fuel.

As seen in Chapter 5, I-132 releases were apparently about 15 times higher than I-131 on a curie basis on the day of the dissolving. On two occasions the ratio was much higher, from 50 to 80 times that of I-131. The cause is not clear<sup>7</sup>. A possibility is that the tellurium and iodine separated in the caustic dissolver due to differences in solubility, setting the I-131 and I-132 on different release pathways ultimately leading to different release fractions. However, this speculation is not borne out by reported chemical opinions (see Appendix C). No project data have been found on tellurium distribution in RaLa solutions to aid this evaluation.

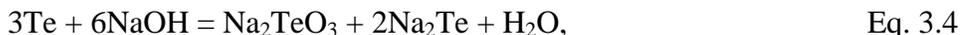
---

<sup>7</sup>Subsequent analysis of the criticality event (Chapter 7) indicates that the I-132 readings were probably incorrect.

Regarding the caustic dissolution process, [Legler et al. \(1957\)](#) state that "... aluminum, cesium and tellurium are actually dissolved..." in this step. However, the statement is not backed by test data or references.

An evaluation of tellurium chemistry in the caustic dissolver and centrifuge bowl by Richardson<sup>8</sup> is given in [Appendix C](#). The major points of his letter report are outlined below.

According to Richardson, at least two sources state that tellurium disproportionates in hot NaOH according to the following, with the tellurium species on the right being soluble.



One reference indicates that the reaction reverses itself as the solution cools. Accordingly, tellurium behavior in the (hot) caustic dissolver should be similar to that of the other soluble species, iodine and cesium.

Richardson further states that on cooling and subsequent acidification, as in both the dissolver waste tank and centrifuge bowl, the tellurium should revert to one of several possible insoluble forms. If so, then the portion of tellurium proceeding to the centrifuge may have precipitated and joined the insoluble material, e.g., silica. In that case, some I-132 may have evolved from the solid waste repository of the RaLa process.

Richardson also states that volatile tellurium hydride ( $\text{TeH}_2$ ) is not likely to form as indicated by the difficulty of producing such species in the laboratory. A cited source states that the hydride is unstable above 0°C and decomposes in air.

Nonetheless, a small quantity of tellurium was observed in at least some of the off-gas line samples ([Rich 1957](#)). Rich states that spot checks during Run 4 (March 1957) showed a curie level of Te-132 in the off-gas sampler equal to about 2% of that of I-131. Either a tellurium volatile species formed by a radiation-induced process unachievable under ordinary laboratory conditions or was carried along in aerosol particles.

### 3.8 Production of Organic Iodide

Organic iodide compounds form when molecular iodine,  $\text{I}_2$ , comes into contact with a variety of organic materials in a radiation field. The radiation may be either an external source or an internal one provided by the radioiodine itself. Organic iodides may also form to a small degree without imposed radiation, especially in a complex mixture of gases ([Beahm et al. 1985](#)). However, the organic and iodine free radicals produced by radiation are the principal source of organic iodide formation.

Without an obvious source of organic material, a minute amount of organic iodide is often found in reprocessing fluids, generally much less than 1% of the total iodine.

---

<sup>8</sup>Dr. William S. Richardson is a retired professor of inorganic chemistry from Auburn University.

There are several sources of organics in the RaLa/ICPP system. Most apparent are the organic solvents used in the extraction step of reprocessing such as tributyl phosphate (TBP). But, as the ICPP air flow diagram shows (Figure 2-11), there would be no contact with organic-laden VOG from the ICPP reprocessing cells with radioiodine from the RaLa system until the well diluted flows combine in the stack.

A more direct source of organic material is the sodium acetate used as a pH buffer in Step 5 of the RaLa process (see Section 2.3). Though most of the iodine was removed from the process before this step into either the dissolver waste tank or the scrubber, the test results show that some radioiodine persisted in the RaLa process to later stages. Therefore, the sodium acetate buffer used in Step 5 must be noted as a possible source of organic iodide.

In addition, organic impurities in the charcoal beds may have provided a second source of organic material in the RaLa process.

Organic iodides are much less reactive than molecular iodine and would not be removed in the alkaline scrubber solution. Nor would organic iodides be picked up in the sodium hydroxide, sodium thiosulfate stack sampler solution. However, the 5% nitric acid scrub solution, adopted in March 1959, should have oxidized the organic iodide and released the iodine, enabling molecular iodine capture in either the scrub solution or the charcoal bed.

The charcoal beds would not sorb organic iodides unless specially treated; apparently they were not.

In summary, an unknown amount of organic iodide was formed in the RaLa process, probably mainly from contact with sodium acetate in a late stage, by which time most of the iodine was removed. The organic iodide would be undetected by the stack sampler.

### 3.9 Summary and Conclusions

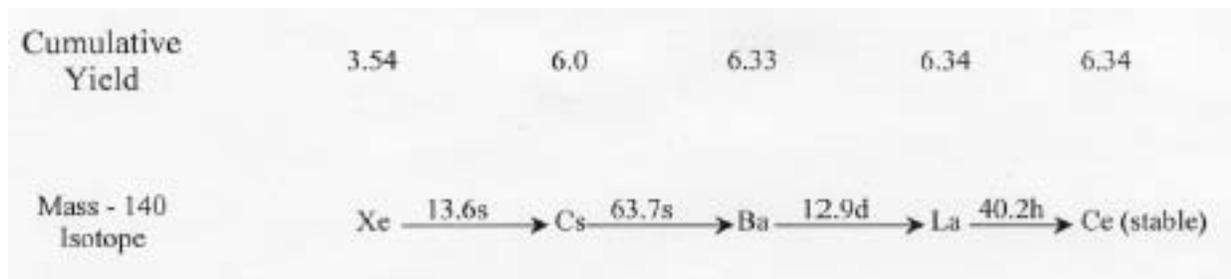
- Iodine behavior in the RaLa process vessels is reviewed. Although generally understood, more information would be required for quantitative estimation of flows between vessels and volatilization to the off-gas.
- The required information would be: (1) volatility data for the various process solutions; (2) process parameters, e.g., temperatures, sparging rates, residence times; and (3) degree of sequestering within UO<sub>2</sub> during caustic dissolving.
- No clear chemical explanation was found for the high I-132 releases observed on the first day of the run (Chapter 5). No  $\gamma$ -energy interferences were found to corrupt the spectrometry reading. A difference in solubility of precursor Te-132 and iodine, a possible explanation, runs counter to chemical evaluation presented in Appendix C.

- An appropriate stack sampler fluid was selected—a sodium hydroxide plus sodium thiosulfate solution. The difficulties with sodium hydroxide alone, which became apparent later, were avoided.
- Sodium acetate, used in a later process step, may have contributed to organic iodide formation, though most of the iodine was removed from the process prior to this step. A second possible source may have been organic residues in the charcoal filters. Adoption of 5% nitric acid solution in the scrubber in March 1959 should have removed organic iodide from the DOG.

## 4.0 RADIOIODINE PRODUCTION IN THE MATERIALS TESTING REACTOR, 1957–1959

### 4.1 Introduction

Intensely irradiated fuel elements from the Idaho MTR were the feed to the RaLa process, one element per run. Figure 4-1 shows the production and decay scheme of the mass-140 fission product in the fuel element.



**Figure 4-1 Mass-140 production from thermal fissions in uranium-235**

The top row showing the cumulative yield indicates that the majority of mass-140 fission product is born as xenon (Xe-140), which quickly decays through cesium (Cs-140) to barium (Ba-140). If one assumes that the precursors decay quickly and the irradiation is time-steady, the level of Ba-140 per unit mass of fuel builds up to the following:

$$N_{\text{Ba-140}} = \frac{\text{fr} Y}{\lambda} (1 - e^{-\lambda t}), \quad \text{Eq. 4.1}$$

where:

$N_{\text{Ba-140}}$	=	atoms Ba-140 at time t
fr	=	the fission rate, fissions/sec
Y	=	the yield of Ba-140, atoms per fission
$\lambda$	=	the decay rate parameter, 1/sec

The great advantage of the MTR was its high fission rate per unit mass of fuel and high flux. As an MTR, dimensions and fuel selection were made to maximize the flux (Weinberg et al. 1956) such that its average value at  $2.5 \times 10^{14}$  (Directory 1959) was about 250 times higher than the Oak Ridge Graphite reactor. In addition, the MTR held a great advantage in the use of highly enriched fuel relative to the natural uranium. These two factors combined to produce a Ba-140 level in one MTR fuel element that exceeded the entire Oak Ridge Graphite Reactor.

## 4.2 Materials Testing Reactor Description

The MTR is an evolution of the common swimming pool reactor, modified for both high volume-specific power and high fuel-specific power (Weinberg et al. 1957). This was achieved by use of fully enriched fuel and light water moderation, which provided small size by virtue of high fissile density coupled with a short thermalization distance. In order to maximize heat removal per gram of fuel, it was necessary to arrange the fuel in thin plates cooled on either side by high velocity water.

Figure 4-2 shows the cross section of the MTR fuel element (Handbook 1955). Note the curved plates designed to withstand differential pressure between coolant channels. The fuel plates consisted of 0.020-inch thick alloy of 18.8 wt-% uranium metal sandwiched in aluminum to a total thickness of 0.060 inches. Each element was brazed to aluminum side-plates. A rack of MTR fuel elements is shown in the photograph (Figure 4-3), illustrating their relatively small size.

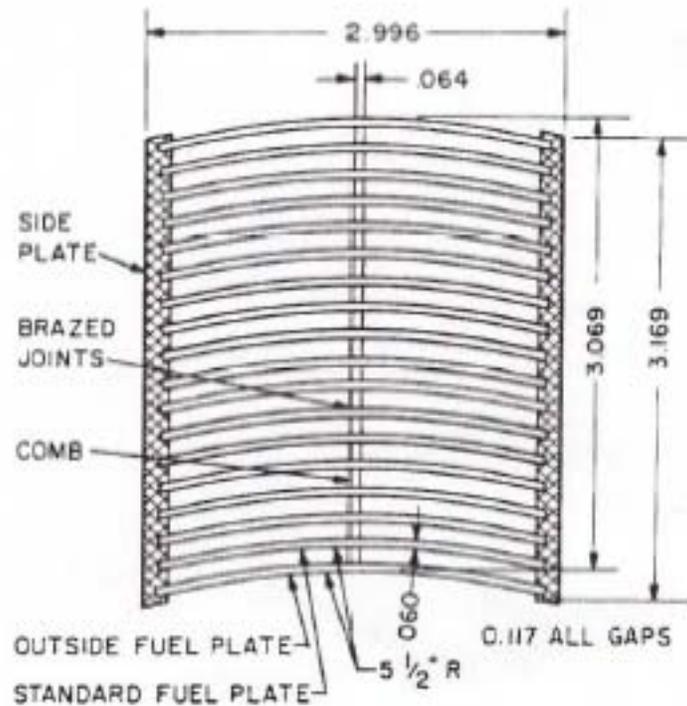
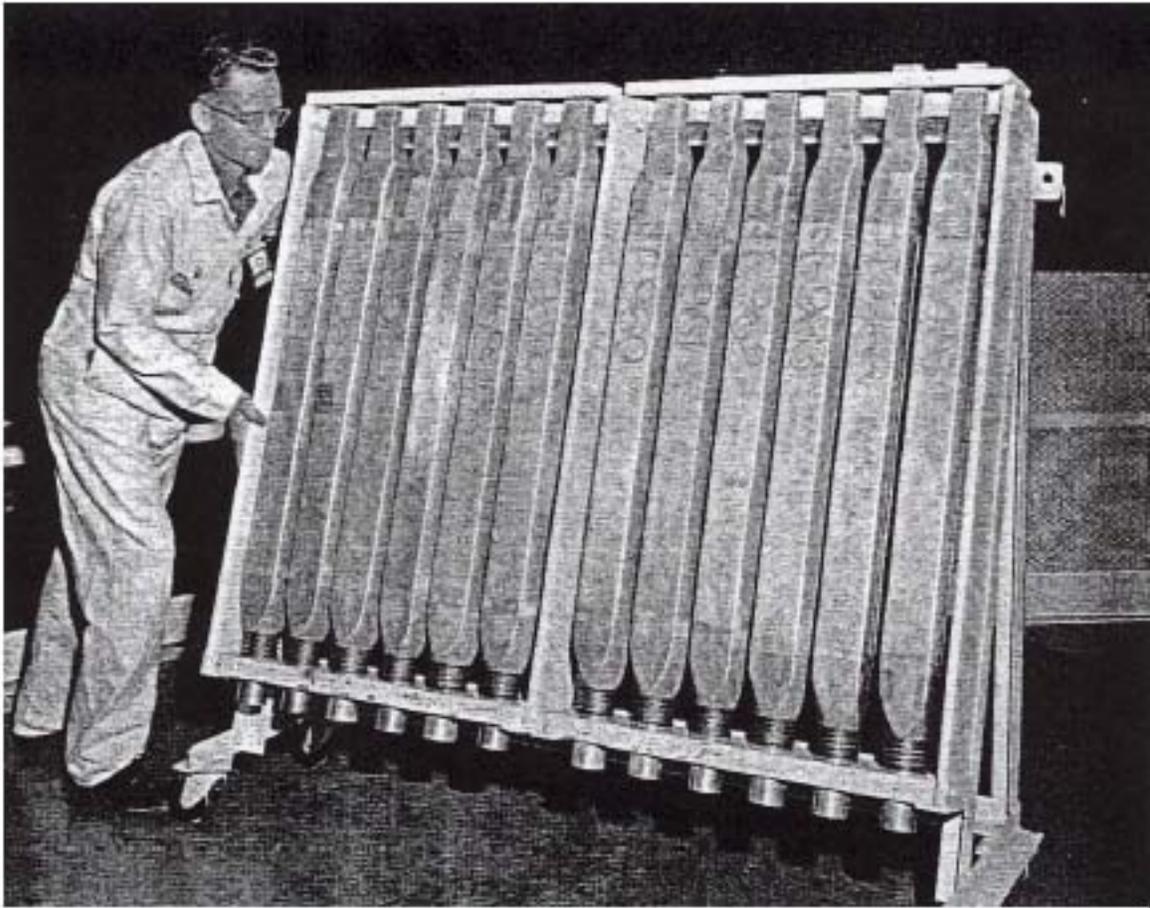
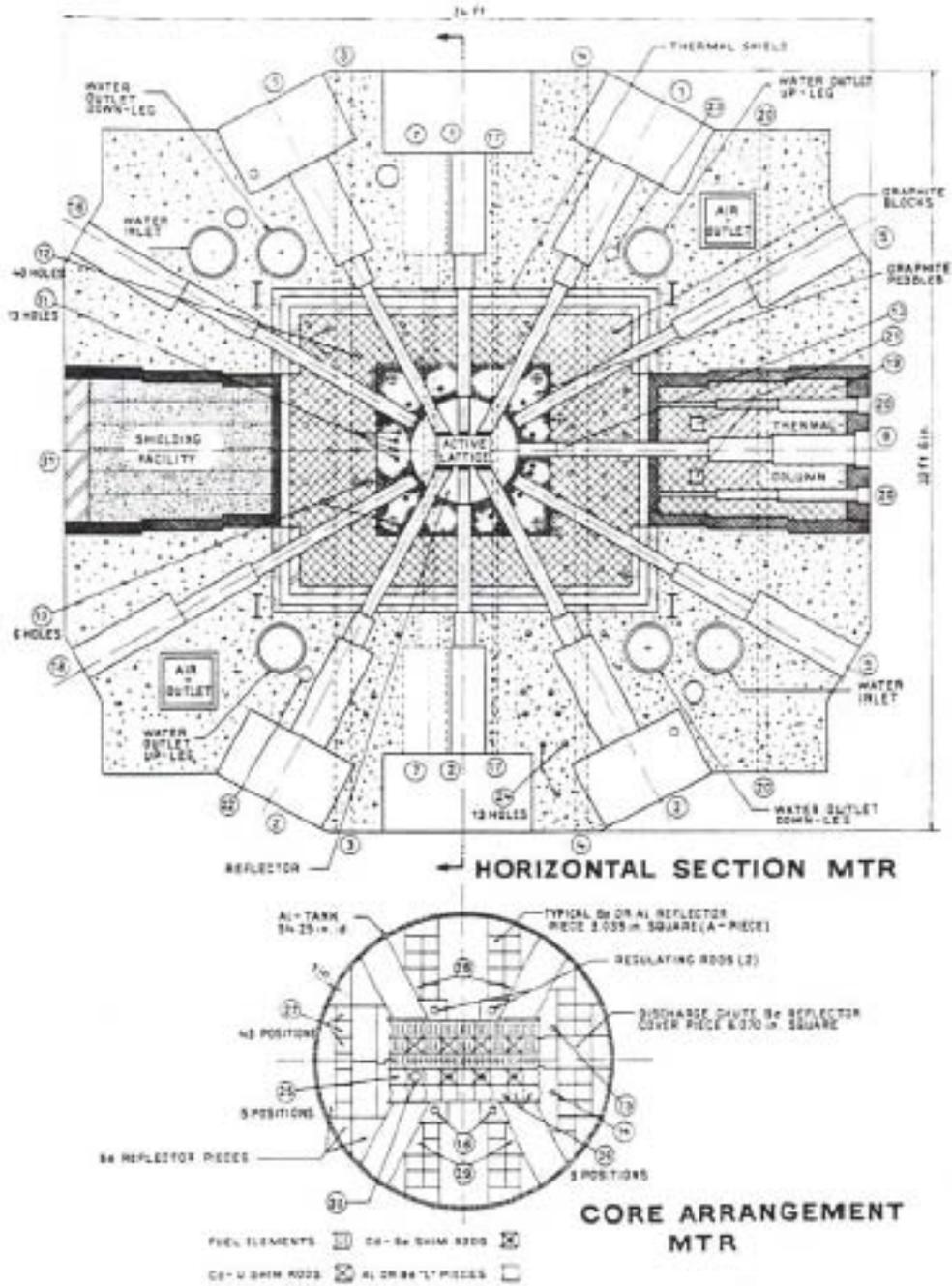


Figure 4-2 Cross section of fuel assembly



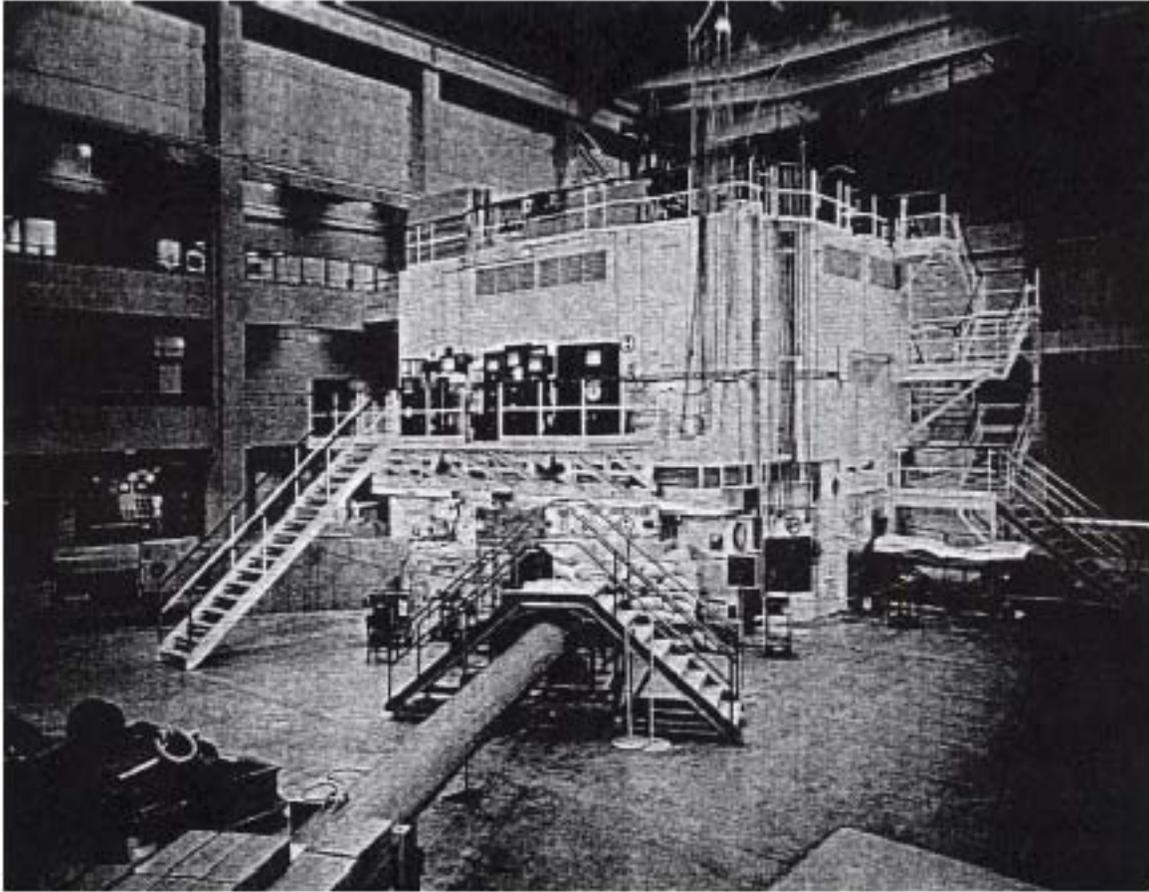
**Figure 4-3 Fuel-assembly units**

A cross-section through the reactor is shown in [Figure 4-4 \(Directory 1959\)](#) with an expanded view of the core shown at the bottom of the figure. The core lattice was a 9 x 5 array providing 45 locations, of which 18 were aluminum or beryllium blanks. An additional four core locations were shim rods for control, leaving only 23 fuel elements. A 54-inch diameter aluminum tank enclosed the core and the surrounding beryllium reflector rods. Outside the tank were graphite balls providing neutron moderation and reflection. Surrounding the graphite balls was a permanent grid of graphite blocks surrounded by about a 12-foot thick concrete shield. [Figure 4-5](#) is a contemporary photograph of the facility ([Handbook 1955](#)).



1 95 LASR-Research 8 - MTR

**Figure 4-4 Horizontal section through Materials Testing Reactor and core arrangement**  
(Directory 1959)



**Figure 4-5** View of the south and west faces of the Materials Testing Reactor

### **4.3 Estimation of Barium-140 and Radioiodine Production in the Materials Testing Reactor**

#### **4.3.1 1957 Method**

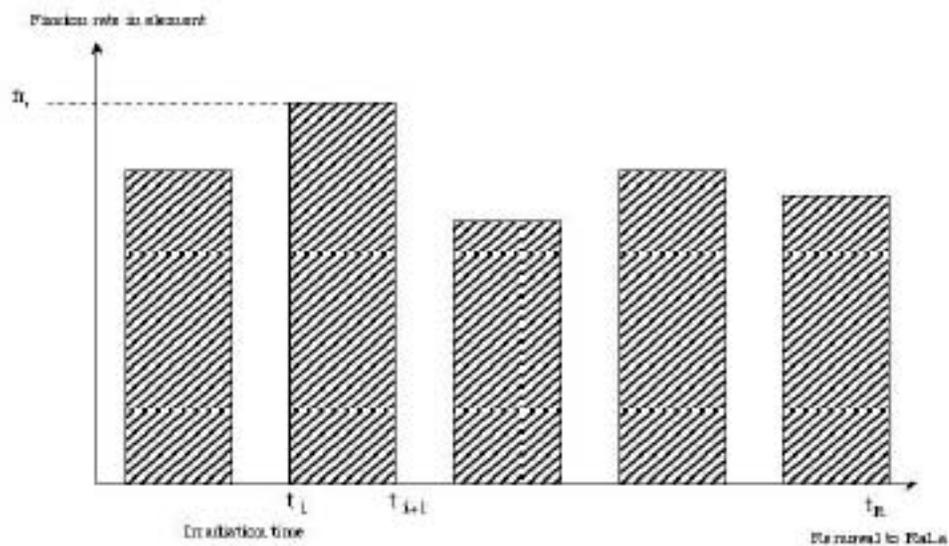
Operational necessities, such as servicing many programs and the need to minimize power peaking in the vicinity of fresh fuel elements, dictated a highly intermittent mode of reactor operation. The general procedure was to run the reactor briefly (about a week), shut down for a day or two to enable re-arrangement of the fuel elements in the core, and restart for another week of operation. Data book notes ([Coykendall 1957](#)) and [MTR Monthly Progress Reports](#) (see complete list in reference section) indicate that RaLa elements were subjected to as many as seven separate reactor irradiations in several different core locations, each separated by several days of non-irradiation before the element was removed to the RaLa process.

[Figure 4-6](#) illustrates such an intermittent irradiation history. The time at power,  $t_{i+1} - t_i$ , may differ for each operational period and the fission rate in the element,  $fr_i$ , may differ somewhat at

each location. At time  $t_R$  the element is removed to the RaLa process. In such a case, Ba-140 and radioiodines accumulate in the element according to a modification of Eq. 4.1.

$$N_{\text{Ba-140}} = \sum_1^N \frac{f r_i \mathbf{Y}}{\lambda} (1 - e^{-\lambda(t_{i+1}-t_i)}) e^{-\lambda(t_R-t_{i+1})} \quad \text{Eq. 4.3}$$

Equation 4.3 states that fission products accumulate additively from each irradiation period, accounting for the decay from the end of each period to  $t_R$ , the time of removal for RaLa.



**Figure 4-6 Illustration of RaLa fuel element irradiation history**

Figure 4-7, a page from a data book assigned to Coykendall (1957), indicates that an expression equivalent to Eq. 4.3 was used to hand-calculate Ba-140 and I-131 accumulation in various fuel elements throughout numerous irradiation campaigns, designated by 81C and 81D through 84D. The numeral in the “charge” designation indicates MTR runs between which the core was reshuffled. The letters, A, B, etc, indicate shutdowns without core reshuffling. Adjacent pages to the one shown in Figure 4-7 indicate that I-133, Te-129, Te-132, Sr-89, and Sr-90 accumulation was also followed by this procedure.

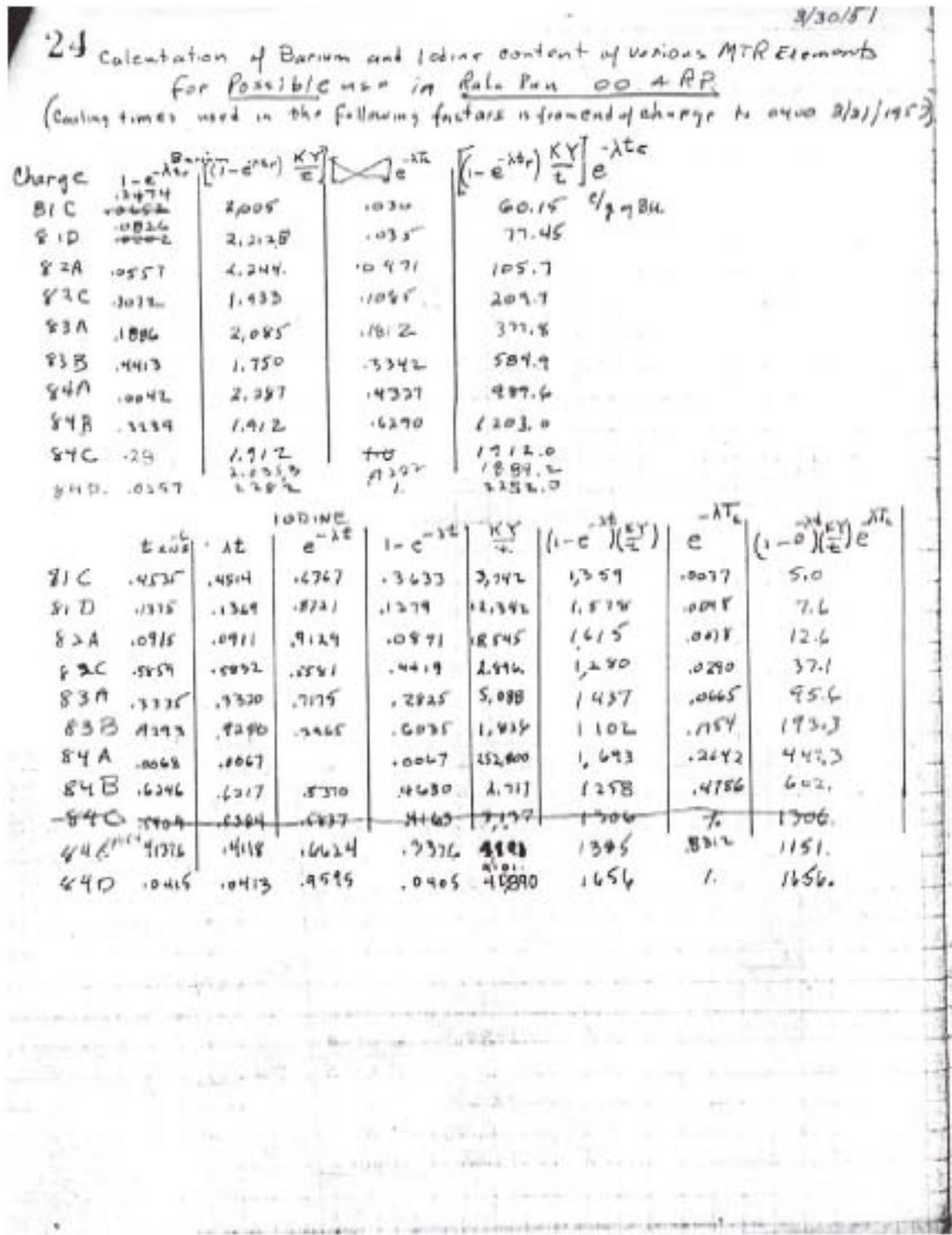


Figure 4-7 One page of a data book illustrating calculations of barium-140 and iodine-131 content of Materials Testing Reactor elements  
 (Coykendall 1957)

As seen from the notebook fragment, hand calculation of isotope buildup was quite laborious and therefore limited to a few of the more important nuclides. In addition, apparently variations of fission density between the different core locations were not taken into account. Therefore, the method must have been fairly inaccurate.

### 4.3.2 *Isotope Buildup Using ORIGEN2*

The isotope generation code ORIGEN2 (Renier and Croff 1980) was used to calculate isotope buildup in designated RaLa fuel elements throughout the irradiation cycle, including reactor downtime and relocation in the core.

As a preparatory step, the flux distribution in the core was determined from its geometrical configuration described in detail in Handbook (1955), the core fissile loading, and the distribution of coolant and construction materials in the core. The flux distribution is required to determine the particular set of reactor neutron spectrum weighted and collapsed cross-sections appropriate for the MTR fuel elements as required by ORIGEN2.<sup>1</sup> In addition, the core model enables evaluation of fission densities for the different locations of the fuel element as it was moved about the core.

The neutron spectrum-weighted cross sections and fission densities were determined with the MCNP4C code (Briesmeister 2000). The MCNP4C code is a general-purpose Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron/photon transport, including the capability to calculate eigenvalues for critical systems. The MCNP4C code can treat any arbitrary three-dimensional geometry configuration bounded by first- and second-degree surfaces. Pointwise cross sections for different reaction types, such as (n,fission), (n,gamma), (n,2n), (n,3n), (n,p), (n,alpha), and others, derived from particular cross sections evaluations such as ENDF/B-VI, are available or can be generated with the NJOY code (MacFarlane and Muir 1994). The thermal scattering S (alpha,beta) data as a function of temperature are available for the different water regions in the MTR reactor.

The neutron spectrum-weighted collapsed cross sections for the MTR fuel elements were used in the ORIGEN2 code (Renier and Croff 1980). The ORIGEN2 code is widely used in calculations of the depletion, buildup, and decay of materials during neutron irradiation. It uses spectrum-weighted one-group cross sections for a variety of neutron reaction types such as (n,fission), (n,gamma), (n,2n), (n,3n), (n,p), (n,alpha), and others. Using different neutron spectra for each of the different MTR fuel elements and different regions, region-dependent spectrum-weighted cross sections were used during depletion. The different isotopes in ORIGEN-2 are subdivided into three distinct groups: activation, actinides, and fission products.

The power histories, downtimes, refueling times, and reinsertion after temporary discharge were modeled in great detail for each of the fuel cycles and each of the “candidate” MTR fuel elements that were or might have been processed in RaLa.

---

<sup>1</sup>The code used in the DOE (1991) study, RSAC-4, used cross sections developed for light water reactors. Use of LWR-weighted cross sections for the generation of isotopes for the MTR could lead to large errors.

Using a given irradiation history, including power levels, downtimes, and different core locations as input, ORIGEN2 calculates fission product production, heavy metal depletion, and production of activation products.

A portion of a typical ORIGEN2 output is provided in [Appendix B](#).

## 4.4 RaLa Fuel Element Irradiation Histories

### 4.4.1 Documented RaLa Fuel Elements and Irradiation Histories

No direct information has been found on the irradiation histories in the MTR of the fuel elements that were fed to the RaLa dissolver. The MTR progress reports do not identify the elements destined to be feed elements to RaLa, although feed elements to other ICPP programs are clearly shown. However, for Runs 6–14, the RaLa elements may be discerned with some uncertainty, and hence their irradiation history (and iodine inventory), by the method outlined below.

Equally important, specific information on the cooling times between reactor shutdown and insertion into the dissolver has been found only for Runs 1–5 and for Run 9. [Legler et al. \(1957\)](#) provides the following information for the first five runs.

**Table 4-1 Fuel element data for the first five RaLa runs**  
(Legler et al. 1957)

Run Number	MTR Element	Cooling Time (days)	Total U-charged (grams)
001	P-126	5	143
002	P-146	2	158
003	P-227	5	192
004	P-237	7	172
005	P-285	6	163

[Stevenson \(1958\)](#) provides an additional snippet that the cooling time for the element fed to RaLa Run 9 was cooled for 8 days.

No other fuel element identification numbers, other than the five listed in [Table 4-1](#), have been found.

### 4.4.2 Surmised RaLa Fuel Elements and Irradiation Histories

Despite the absence of specific information, some irradiation histories may be surmised with a degree of uncertainty by comparing the RaLa run schedule with information in the MTR Progress Reports and applying certain reasonable assumptions. The procedure is as follows:

- (1) Start with the RaLa run schedule, as provided by the Health and Safety Division Annual Report (1959).<sup>2</sup> This provides a run starting date.
- (2) Select a cooling time. This would be either the known cooling times for Runs 1-5 and 9 or an assumed cooling time of 2 days, frequently cited as nominal for RaLa fuel elements.
- (3) Consult the MTR Progress Reports for fuel elements withdrawn from the reactor nearest the date of the RaLa run start, less the assumed cooling time. Several subsequent reports need to be scanned to assure that the elements were not later reinserted.
- (4) If only one element was removed at the designated time (and if it was not reinserted), it is identified as the RaLa element. Usually it was found that several elements, in some cases as many as five, were permanently removed at the designated time. In such case, the selected RaLa element was assumed to be the one with the highest Ba-140 content. This entailed running ORIGEN2 for each of the possibilities.
- (5) Once the element is selected, the irradiation history may be reconstructed from information in prior MTR monthly reports. Note, however, that the monthly reports do not identify the element by serial number, making tracking somewhat difficult. Tracking was accomplished by noting the given insertion and removal dates for specific core locations.

Figure 4-8 is a copy of a page from an [MTR Progress Report \(IDO-16310\)](#) illustrating the type of information available up to June 1958, at which time the format was reduced. After some interpretation, the following information is given in the figure.

- The three rows and nine columns represent the 3 x 9 lattice of the active core. The numbers 11, 37, 39, etc., are row and column designations (i.e., 11 is row-1, column-1).
- The top number of each set represents the mass of uranium in the element in grams (i.e., 189, 200, 163, etc.) at the time of charge into the given location.
- The numbers below the uranium mass indicate core locations in prior MTR “charges” (i.e., irradiation runs). [Figure 4-8](#) represents the charge designated 82C in the title.

---

<sup>2</sup>Note that this is a secondary reference. Moreover, run starting times are given by date only, i.e., there is a +/- 12-hour uncertainty in starting time.

IDO-16328  
Page 15  
MANIS/57

A. REACTOR OPERATIONS (cont'd)

2e(4). Fuel Loading for Charge 82C

Position	1	2	3	4	5	6	7	8	9
Row 1	189 11 82B	200 New	200 New	163 37 79B-80A 39 23 80B 81C	148 39 38 80A 80B 39 37 80C 81A 82A, B 81B	170 29 79B-80B	200 New	200 New	195 29 81D-82A
2	200 New	63-0 24 77D-78 22 79-82B	169 32 79B-80B 23 23 81D 82A	GRHL-29-1 70-5 24 79-82B	153 38 77C-78A 27 79B-80A 81B	64-5 26 79B-82B 14	168 33 31 80B 80C 27 27 81A 81D-82A	GRHL-14-1 46-5 28 79B-82B	200 New
3	196 18 81D-82A	200 New	200 New	168 37 38 80B 80C/79B-80A 80B 34 34 81A 82A	151 33 31 80B 80B 34 16 81B 82B 81C	163 18 79B-80B	200 New	200 New	196 19 81D-82A

Total g content of fuel elements - 4229  
Total g content of fuel shims - 263

Total 4472

1st lines - Gram content at beginning of cycle  
2d and 4th lines - Position(s) previously irradiated  
3d and 5th lines - Charge(s) previously irradiated  
S - Shim rod

Figure 4-8 Core map for Charge 82C  
(MTR Progress Reports)

- As an example, note location 15, believed to be the fuel element fed to RaLa Run 2. Its uranium mass at the time of insertion into charge 82C was 148 grams. Previously it was in location 39 during charge 80A, in location 38 in charge 80B, in location 39 in charge 80C, in location 37 in charge 81A, and in location 14 in charges 82A and 82B.

Each of the charges had an irradiation history, also provided in the MTR Progress Reports. [Figure 4-9](#) illustrates the irradiation history of charges 82A, 82B, and 82C, the last one being the reconstructed removal date for the element fed to RaLa Run 2. Note that the dates of the run and time at power to the hundredth of an hour are given in columns one and two. During charge 82A the reactor was mainly shut down. Most frequently, the reactor operated at 40 MWth. (Its design power is given as 30 MWth).

[Figures 4-8](#) and [4-9](#) show that the purported RaLa Run 2 element was irradiated in 21 separate campaigns in charges 82A, 82B, and 82C alone at core locations 14 and 15. It underwent additional irradiations in its earlier charges at the different core locations. The ORIGEN2 calculation of the isotopic content of this fuel element accounted for all of these irradiation campaigns, including the fission rate dependence on core location.

#### *Runs 1–5 (February–June 1957) and Run 9 (January 1958)*

The cooling times are positively identified for this group of runs, only one of which was the nominal 2 days. Fuel elements are identified for Runs 1-5, but the information is not usable because element numbers are not identified in the available (expurgated) MTR Progress Reports. (See reference list, MTR Progress Reports.) The available MTR Progress Reports provide specific information of fuel element location transfers to recreate irradiation histories by the method outlined above.

#### *Runs 6–14 (September 1957–May 1958), except Run 9*

The nominal cooling time of 2 days was assumed for this group of runs. The available MTR Progress Reports provide specific information of fuel element location transfers to recreate irradiation histories by the method outlined above.

#### *Runs 15–37 (June 1958–December 1959)*

The available (i.e., expurgated) MTR Progress Reports ceased providing information on specific fuel element transfers. An example of the post-May 1958 irradiation information is shown in [Figure 4-10](#). Note that the prior locations of each fuel element cease to be given, making it virtually impossible to recreate irradiation history.

### **4.4.3 Recreated RaLa Fuel Elements and Irradiation Histories**

[Table 4-2](#) lists an abbreviated review of the recreated irradiation and decay time histories for RaLa Runs 1–14.

DATE 02-18-57  
 PAGE 191

A. REACTOR OPERATIONS (Cont'd)

2a. Daily Operating Data

Date	Hours Operated	Avg Operating Power (Mw)	Accumulated Energy - Mwd		
			This Date	Charge No. 82A	To Date
1-28-57	Scrammed at 0158 for Cycle 82				
1-29	0.00	0.0	0	0	39,716
1-30	0.00	0.0	0	0	39,716
1-31	0.00	0.0	0	0	39,716
2-1	13.28	30.9	17	17	39,733
2-2	16.38	40.0	27	44	39,760
				<u>Charge No. 82B</u>	
2-3	16.78	32.8	23	67	39,783
2-4	24.00	40.0	40	107	39,823
2-5	24.00	40.0	40	147	39,863
2-6	24.00	40.0	40	187	39,903
2-7	24.00	39.9	40	227	39,943
2-8	24.00	40.0	40	267	39,983
2-9	24.00	40.0	40	307	40,023
2-10	14.00	40.0	23	330	40,046
				<u>Charge No. 82C</u>	
2-11	14.17	35.7	21	351	40,067
2-12	24.00	39.0	39	390	40,106
2-13	24.00	40.0	40	430	40,146
2-14	24.00	40.0	40	470	40,186
2-15	24.00	40.0	40	510	40,226
2-16	24.00	40.0	40	550	40,266
2-17	24.00	40.0	40	590	40,306
2-18	4.00	40.0	7	597	40,313
Total	366.61	39.1 (Avg)	597		

2b. Power Reductions (Power less than 40 Mw)

Date	Time	Duration (Hr)	Reason
2-1-57	1043	4.25	Approaching full power after shutdown for Cycle 82; 1149 - Manual reverse to $H_L$ for ORNL-2b-52
2-3	0713	3.28	Approaching full power after shutdown for Charge 82B
	1410	0.87	Scram from GEANP-1 (high temperature)
	1535	0.48	Jr. scram from GEANP-1 (high pressure)
	2120	0.42	Jr. scram from GEANP-1 (high discharge probe temperature)
	2227	1.15	Jr. scram from GEANP-1 (high discharge probe temperature)
2-5	1009	0.40	To $H_L$ to discharge WAPD-21
2-7	1952	1.00	To 38 Mw to stop leak on ORNL-2b-53

Figure 4-9 Irradiation record for Charges 82A, 82B, and 82C  
 (MTR Progress Reports)

A. REACTOR OPERATIONS (Cont'd)

2e(3). Fuel Loading for Charge 104B

Position	1	2	3	4	5	6	7	8	9
Row									
1	169 g	168 g	172 g	162 g	154 g	162 g	169 g	168 g	168 g
2	168	117	168	72	152	72	167	118	168
3	172	168	169	165	164	165	167	168	168
4									
5									

(ORNL-14-3)  
99

(ORNL-14-2)  
70

(ORNL-21-15)  
3

Total g content of fuel elements at beginning of cycle - 3,821 g  
 Total g content of fuel shims at beginning of cycle - 379  
 Total 4,200 g

NOTE: Because of the short duration of Charge 104B, no Shim Rod Position vs. Rod graph will be published.

Figure 4-10 Reduced information regarding core loading following June 1958  
(MTR Progress Reports)

**Table 4-2 Brief review of fuel cycle data of the candidate Materials Testing Reactor fuel elements used in the analyses of the RaLa runs**

RaLa Run Number	RaLa Start & Finish Dates	Fuel Element Discharged at end of Cycle	Fuel Final Discharge Date	Cooling Time (days)	MTR Element	Initial Charge Cycle	Total U Charged (grams)	Fuel element
1	2/1/1957	81D	1/28/1957	3d,22hr	81D-35	79A	200	P-126
	2/3/1957		2.00 hr		81D-15	79A	200	
2	2/20/1957	82C	2/18/1957	1d,20hr	82C-15	80A	200	P-146
	2/21/1957		4.00 hr		82C-36	79B	200	
					82C-19	81D	200	
					82C-35	79B	200	
					82C-14	79B	200	
				82C-25	77C	200		
3	4/5/1957	84D	3/31/1957	5d,20hr	84D-38	84A	201	P-227
	2/6/1957		4.00 hr		84D-33	82B	200	
4	5/19/1957	86C	5/12/1957	6d,12hr	86C-35	84A	202	P-237
	5/20/1957		12.0 hr		86C-15	83B	202	
5	6/24/1957	88B	6/18/1957	5d,10hr	88B-23	87B	200	P-285
	6/25/1957		14.3 hr		88B-15	80C	199	
					88B-32	83B	200	
6	9/11/1957	92A	9/9/1957	1d,10hr	92A-34	92A	200	
			14.0 hr		92A-35	89A	200	
					92A-25	86B	200	
					92A-15	90A	200	
					92A-27	91A	200	
7	10/7/1957	93B	10/5/1957	1d,14hr	93B-14	90A	200	
			10.0 hr		93B-35	82C	200	
					93B-13	91A	200	
					93B-15	87B	201	
					93B-25	87A	201	
					93B-34	87A	200	
8	10/21/1957	94A	10/11/1957	9d,14hr	94A-15	88C	200	
			9.6 hr		94A-35	86C	200	
	10/25/1957*	94B	10/23/1957	1d,4hr	94B-15	85B	200	
			21.1 hr		94B-16	85B	199	
					94B-25	91A	200	
					94B-33	94B	200	
					94B-35	89B	200	

**Table 4-2 Brief review of fuel cycle data of the candidate Materials Testing Reactor fuel elements used in the analyses of the RaLa runs (continued)**

<b>RaLa Run Number</b>	<b>RaLa Start &amp; Finish Dates</b>	<b>Fuel Element Discharged at end of Cycle</b>	<b>Fuel Final Discharge Date</b>	<b>Cooling Time (days)</b>	<b>MTR Element</b>	<b>Initial Charge Cycle</b>	<b>Total U Charged (grams)</b>	<b>Fuel element</b>
<b>9</b>	1/6/1957	97B	12/29/1957 1.8 hr	7d,22hr	97B-17	93A	200	
<b>10</b>	2/12/1958	99B	2/10/1958 0.5 hr	2d	99B-15 99B-35 99B-38 99B-12 99B-36 99B-14 99B-17 99B-16 99B-37 99B-32 99B-34 99B-33 99B-25	81D 84C 87A 83A 84D 93A 98C 93A 80C 83A 93A 85B 89A	200 202 199 200 200 200 199 200 200 200 200 199 201	
<b>11</b>	3/13/1958 3/14/1958	101A	3/11/1958 15.8 hr	1d,8hr	101A-15 101A-25 101A-35	99B 99B 99B	200 200 199	
<b>12</b>	4/16/1958	102C	4/14/1958 0.5 hr	2d	102C-11 102C-32 102C-13 102C-39 102C-16 102C-21 102C-37 102C-36 102C-35 102C-27 102C-14 102C-25 102C-34 102C-31	99B 98C 89C 94B 101B 94A 89C 92A 89A 87B 93B 86B 91A 98B	200 199 200 200 200 200 200 200 200 201 200 200 199 200	
<b>13</b>	4/30/1958 5/1/1958	103A	4/28/1958 4.5 hr	1d,20hr	103A-21 103A-27 103A-14	82B 90A 85B	200 200 199	

**Table 4-2 Brief review of fuel cycle data of the candidate Materials Testing Reactor fuel elements used in the analyses of the RaLa runs (continued)**

RaLa Run Number	RaLa Start & Finish Dates	Fuel Element Discharged at end of Cycle	Fuel Final Discharge Date	Cooling Time (days)	MTR Element	Initial Charge Cycle	Total U Charged (grams)	Fuel element
<b>14</b>	5/28/1958	104B	5/26/1958 0.6 hr	2d	103C16A	91A	200	
					103C13A	103A	199	
					103C11A	92A	200	
					103C19A	100A	200	
					103C39A	77D	200	
					103B16A	92B	200	
					103B18A	98B	199	
					103B29A	87B	200	
<b>15</b>	6/2/1958	104B	5/26/1958 0.6 hr	7d	104B-xx	xxx	xxx	
<b>16</b>	8/6/1958	107B	8/4/1958 0.5 hr	2d	107B-xx	xxx	xxx	
<b>17</b>	8/13/1958	107B	8/4/1958 0.5 hr	9d	107B-xx	xxx	xxx	
<b>18</b>	10/1/1958	110B	9/29/1958 0.5 hr	2d	110B-xx	xxx	xxx	
<b>19</b>	10/22/1958	111B	10/20/1958 0.5 hr	2d	111B-xx	xxx	xxx	
<b>20</b>	10/22/1958 10/23/1958	111B	10/20/1958 0.5 hr	1d	111B-xx	xxx	xxx	
<b>21</b>	11/12/1958	112C	11/10/1958 0.5 hr	2d	112C-xx	xxx	xxx	
<b>22</b>	2/4/1959	116B	2/2/1958 0.5 hr	2d	116B-xx	xxx	xxx	
<b>23</b>	2/25/1959	117C	2/24/1959 0.5 hr	1d	117C-xx	xxx	xxx	
<b>24</b>	3/18/1959	118	3/15/1959 14.8 hr	2d,9hr	118-xx	xxx	xxx	
<b>25</b>	4/8/1959	119B	4/6/1959 0.5 hr	2d	119B-xx	xxx	xxx	
<b>26</b>	4/29/1959	120B	4/27/1959 0.5 hr	2d	120B-xx	xxx	xxx	

**Table 4-2 Brief review of fuel cycle data of the candidate Materials Testing Reactor fuel elements used in the analyses of the RaLa runs (continued)**

RaLa Run Number	RaLa Start & Finish Dates	Fuel Element Discharged at end of Cycle	Fuel Final Discharge Date	Cooling Time (days)	MTR Element	Initial Charge Cycle	Total U Charged (grams)	Fuel element
27	5/19/1959	121C	5/18/1959 0.5 hr	1d	121C-xx	xxx	xxx	
28	6/10/1959	122B	6/8/1959 0.5 hr	2d	122B-xx	xxx	xxx	
29	7/6/1959 7/7/1959	123B	6/29/1959 0.5 hr	7d	123B-xx	xxx	xxx	
30	7/21/1959 7/22/1959	124B	7/20/1959 0.5 hr	1d	124B-xx	xxx	xxx	
31	8/11/1959 8/12/1959	125C	8/9/1959 17.9 hr	1d,6hr	125C-xx	xxx	xxx	
32	9/1/1959 9/2/1959	126B	8/31/1959 0.5 hr	1d	126B-xx	xxx	xxx	
33	9/22/1959 9/23/1959	127B	9/20/1959 22.7 hr	1d,1hr	127B-xx	xxx	xxx	
34	10/13/1959 10/14/1959	xxxx	xx/xx/1959 x.x hr	xx	xxxx-xx	xxx	xxx	
35	11/5/1959 11/6/1959	xxxx	xx/xx/1959 x.x hr	xx	xxxx-xx	xxx	xxx	
36	11/30/1959 12/1/1959	xxxx	xx/xx/1959 x.x hr	xx	xxxx-xx	xxx	xxx	
37	12/15/1959 12/16/1959	xxxx	xx/xx/1959 x.x hr	xx	xxxx-xx	xxx	xxx	

\* If RaLa Run 8 starts at 10-25-57 instead of 10-21-57 (Table A-12 of [Legler et al. 1959](#))

## 4.5 ORIGEN2 Results

### 4.5.1 Radioiodine Production

[Appendix B](#) gives a portion of ORIGEN2 output listing curies of fission products and activation products in the fuel element at discharge and at various cooling times. This is calculated for fuel element P-146, discharged from the MTR on February 18, 1957 and charged 2 days later in the dissolver for RaLa Run 2. Thus the RaLa charge corresponds to the 2.0D cooling column.

This element and associated irradiation history was identified by comparing the date of the RaLa run, as given in Health and Safety Division (1959), with the MTR operations schedule given in the MTR Monthly Progress Report series.

[Table 4-3](#) lists the ORIGEN2 predicted inventories of I-131, I-132, I-133, and I-135 delivered to the dissolver, as developed thus far from available and surmised irradiation histories and cooling times.

**Table 4-3 Radioiodines fed to the dissolver determined from ORIGEN2**

Run number	RaLa date	Fuel element	Decay time (days)	Radioiodine inventories to the dissolver, Ci			
				I-131	I-132	I-133	I-135
<b>1957</b>							
1	February 1-3	P-126*	5 *				
2	February 20-21	P-146*	2 *	23,100	30,300	15,200	453
3	April 5-6	P-227*	5 *				
4	May 19-20	P-237*	7 *				
5	June 24-25	P-285*	6 *	17,700	14,700	681	0
6	September 11	‡	2 †	21,700	28,200	14,600	437
7	October 8	‡	2 †	24,400	31,000	15,000	446
8	October 21	‡	2 †	11,700	10,900	72,200	378
<b>1958</b>							
9	January 6	‡	8 §	7,000	3,050	60	nil
10	February 12	‡	2 †	25,800	33,000	16,000	476
11	March 13-14	‡	2 †	22,800	27,500	15,600	467
12	April 16	‡	2 †	27,300	36,400	18,000	537
13	April 30-May 1	‡	2 †	26,900	35,700	17,900	53,100
14	May 28	‡	2 †	24,200	31,100	15,900	489
15	June 2	‡	2 †	¶	¶	¶	¶
16	August 6	‡	2 †	¶	¶	¶	¶
17	August 13	‡	2 †	¶	¶	¶	¶
18	October 1	‡	2 †	¶	¶	¶	¶
19	October 22	‡	2 †	¶	¶	¶	¶
20	October 22-23	**					
21	November 12	‡	2 †	¶	¶	¶	¶
<b>1959</b>							
22-37	February 4 to December 15-16	‡	2 †	¶	¶	¶	¶

\* Obtained from [Legler et al. 1957 \(IDO-14414\)](#)

† Assumed to be the nominal 2-day cooling period

‡ No fuel element serial numbers are recorded in the MTR Progress Reports

§ Cited in [Stevenson 1958 \(IDO-14444\)](#)

¶ Inventory estimation not possible due to lack of irradiation data

\*\* There evidently was no Run 20; numbering error, according to [DOE \(1991\)](#)

## 4.6 Reported Iodine-131 Production in the Materials Testing Reactor

[Bower and Buckham \(1969\)](#), in a later international conference paper, report the average amounts of I-131 fed to the dissolver for groups of RaLa runs ([Table 4-4](#)). The values given were unreferenced and hence unable to be checked.

**Table 4-4 Activity of iodine-131 (Ci) fed to the dissolver**  
(Bower and Buckham 1969)

---

<b>Run number</b>	<b>Average number of curies I-131 fed to the dissolver per run</b>
<b>1-5</b>	16,810
<b>6-15</b>	20,676
<b>16 &amp; 17</b>	14,794
<b>18-23</b>	22,102
<b>24</b>	22,382
<b>25 &amp; 26</b>	22,373
<b>27-40</b>	22,993

---

---

## 5.0 THE STACK MONITOR

### 5.1 Introduction

The stack monitor system provided a nearly direct estimation of the iodine source term by measurement of the concentration in the stack effluent. The complexity of the RaLa system described in [Chapter 2](#) and the lack of operational details preclude estimation of the iodine source term by calculating movements through the system. Fortunately, the stack monitor operated each day for the first three years of operation, from February 1, 1957 to December 1959, and beyond.

The system has evidently never been written up in a project report. However, interpretation of the [Stack Monitor Datasheets \(SMDs\) \(MC 60111\)](#) indicates the mode of operation, and assorted letters written in 1957 ([Project Letters 1957](#)) describe early modifications and provide some crude sketches. Bits of comments from other references add further information.

This section includes the following: (1) a description of the stack monitor system; (2) review of the information available in the SMDs; and (3) an evaluation of the system. The evaluation covers the parts of the system that can contribute to errors: the efficiency of the sampler; iodine losses in the sampling line; the effect of variable stack concentration over the sampling period; and accuracy of the radiation measurement.

### 5.2 General Features of the Stack Monitor System

#### 5.2.1 Flow Diagram

[Figure 5-1](#) shows the overall air and off-gas flows emanating from the main process building, CPP-605, and the relation of the stack sampler to the overall system. The stack monitor draws stack air after it has combined flows from the RaLa dissolver off-gas (DOG), ICPP DOG, ICPP vessel off-gas (VOG), and the air ventilation system sweeping the hot-cells.

Only RaLa within the ICPP processed short-cooled fuel containing radioiodines.<sup>1</sup> Therefore, the iodine readings at the stack monitor could all be attributed to RaLa. However, the  $\beta$ -I and  $\alpha$  levels, attributed to aerosols, were contributed to by all the fuel processing and test operations being conducted in the ICPP.

[Chapter 2](#) cited some uncertainties regarding the gas flow schematic shown in [Figure 2-11](#). The off-gas for all of the RaLa vessels appeared to have had the capability of venting either to the general ICPP VOG or the special RaLa DOG system, the significance being that the ICPP VOG was not scrubbed of iodine. Presumably this flexibility was not often used, and all RaLa vessels apparently vented to the RaLa DOG system essentially all of the time. It is also not clear if the RaLa DOG was led through the Waste Disposal Building, CPP-604. It is shown as bypassing this building, but it might have passed through, opening the option of processing it for noble gas removal.

---

<sup>1</sup>Except for long-lived I-129 emitted from all the dissolvers in the ICPP.

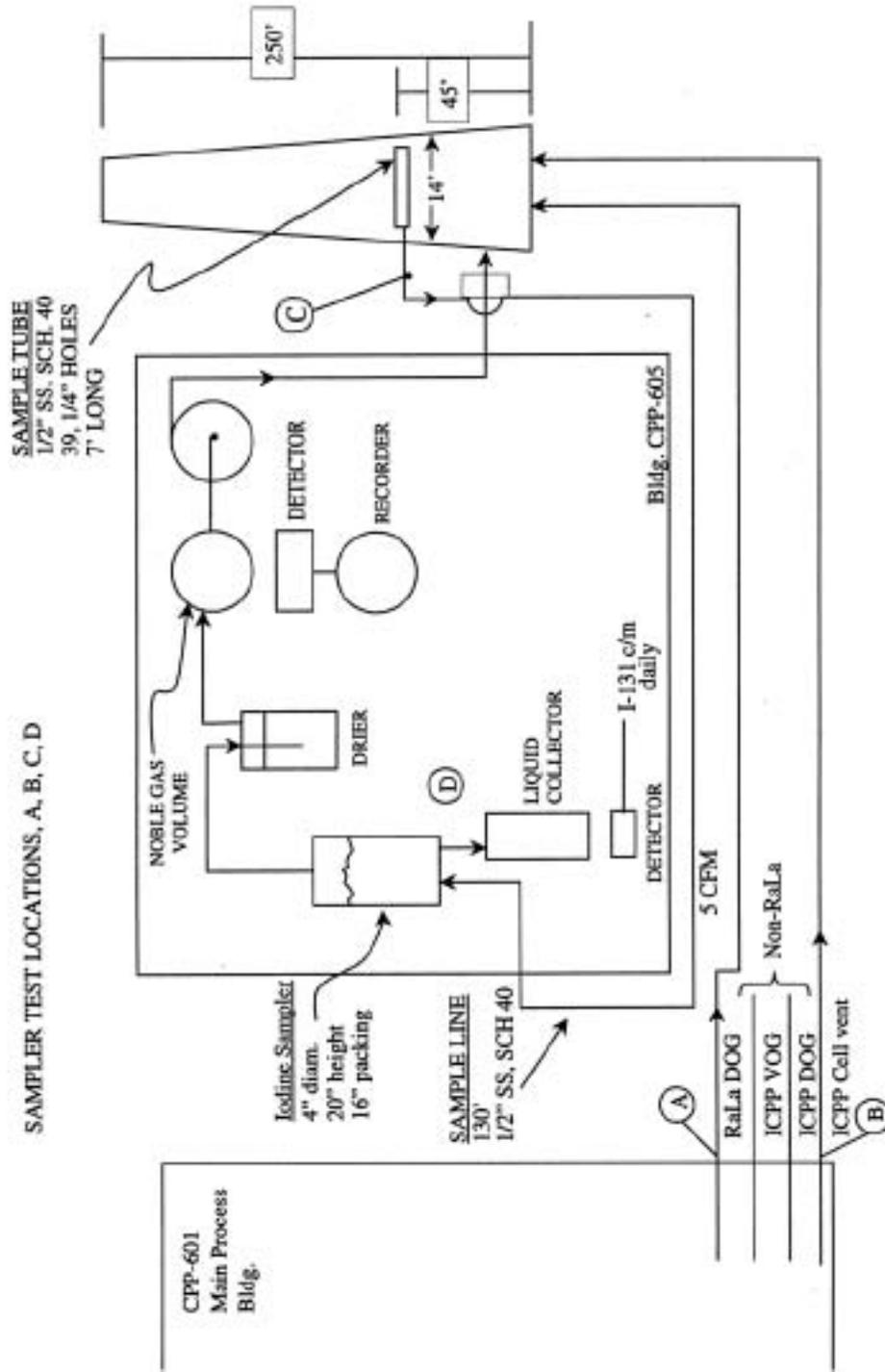


Figure 5-1 Stack sampling system configuration during sampler tests, April–June 1957

Note that the RaLa DOG routing is much simpler than indicated in the flow diagram depicted in [Figure 2-11](#); neither the charcoal beds nor the 10,000 ft<sup>3</sup> gas storage vessel were yet installed. In addition, the stack sampler was installed near CPP-605, farther from the stack than the later installation in CPP-643 at the base of the stack. As a result, the sample line was a rather long 130 feet. There is no record of when the later system in CPP-643 was installed or the differences in the sampler configuration that went along with the newer installation. It may be assumed that the later system was very much like that depicted in [Figure 5-1](#).

### 5.2.2 System Operation and the Iodine Sampler

The operational features of the system can be discerned from the description of the tests conducted in April 1957, provided in [Project Letters \(1957\)](#) and the outline of the SMDs. The central drawing of [Figure 5-2](#), taken from a sketch in an early letter, shows the system set up for tests conducted in April 1957–June 1957 ([Project Letters 1957](#), BLR-8-57A). The later system installed in CPP-643 must be assumed to be similar.

The iodine trap described for the April 1957 tests was a small packed column, 4 inches in diameter, 20 inches high, packed with 16 inches of Raschig ring filler fabricated from ½ inch lengths of ¼ inch tubing. The scrub solution was 0.05 M NaOH plus 0.05 M Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>.

The SMDs indicate that after about a month of initial trials, a steady routine of operation was adopted. A run was started near midnight with fresh scrubber liquid in the sampler and operated for 24 hours, midnight to midnight. The liquid, approximately one liter, was then drained into bottle for determination of the iodine radiation level by means of a scintillation counter. The radiation determinations were the following:

- Iodine-131, by means of a NaI crystal scintillation counter set for the principal I-131 emission
- Iodine-132, beginning April 1958
- β-I, determined by driving off the iodine and β-counting the remaining liquid<sup>2</sup>
- α-counting

[Project Letters \(1957, BLR-43-75A\)](#) state that construction of a new monitoring system was scheduled to begin November 11, 1957. The letter states that the new design will be a “gross improvement,” incorporating all that was learned from earlier operation and from Hanford operations. No description has been found for this later system. Presumably this was when the system was installed in CPP-643 located at the base of the stack. It can only be assumed to be a similar system and iodine trap.

---

<sup>2</sup>The β-I levels were attributed to non-gaseous material caught in the sampler. Early readings (February and March 1957) were erroneous due to an error in the chemical treatment of the sample. The β-I readings and the α-readings are dealt with in the Addendum.

Mr. A. L. Ayers  
File: BLR-8-57A  
June 10, 1957  
Page 13

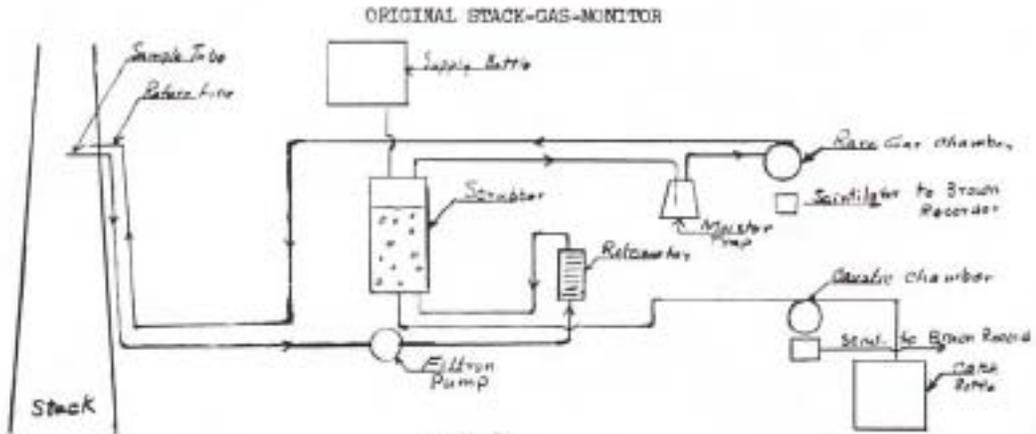


Fig. #1

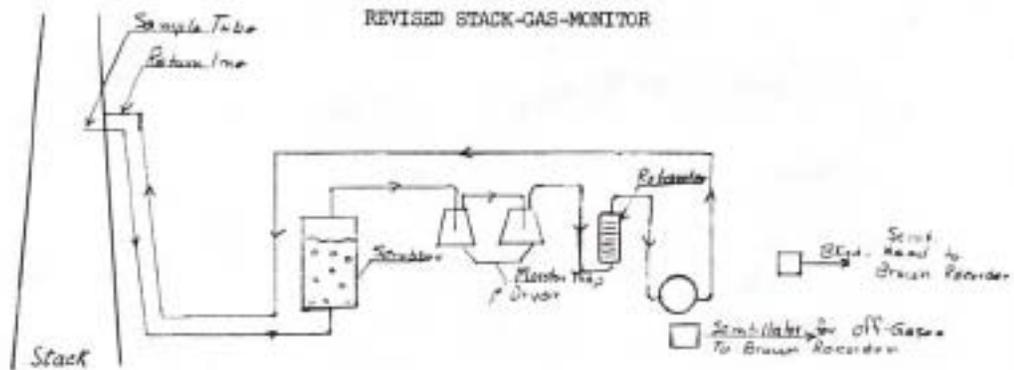


Fig. #2

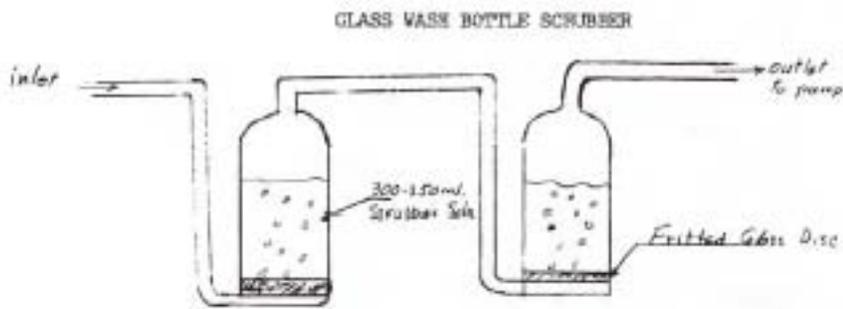


Fig. #3

Figure 5-2 Early stack monitor systems —  
February 1957 (top) and April 1957 revisions  
(Project Letters 1957)

The test samplers, temporarily installed at locations A, B, and C in [Figure 5-1](#), were of a different type from the production sampler at Location D. The temporary test samplers were 300–350 ml laboratory glassware with a glass frit at the base for distributing the inlet gas flow. The configuration shown at the bottom of [Figure 5-2](#) illustrates the temporary sampler and one method (of two) used for determining sampler efficiency. (See Section 5.2.3.)

### 5.2.3 Early Stack Monitor Experience

The top portion of [Figure 5-2](#) illustrates the initial system as operated in February 1957 for Runs 1 and 2, and in March when no runs occurred. Note the iodine trap was, at that time, a small, flow-through packed column with caustic reagent continually supplied by a storage bottle, and effluent fed to a volume that was continually counted for I-131. Dimensions are given in [Figure 5-1](#).

Such a system would provide a continuous record of release from the stack without having to account for decay in the sampler, as was necessary for the static samplers used later. However, the SMDs for February and March seem to indicate that the continuous monitor was never used. Instead, it seems that the liquid was drained periodically and  $\gamma$ -counted.<sup>3</sup> In such a case, the static sampler relations given in the next section apply.

This is not certain, but it will be assumed when examining the February and March record that the (potentially) continuous sampler was actually used in a static mode. This adds a bit of uncertainty in the interpretation of the February and March entries in the datasheets.

### 5.2.4 Relation of Stack Sampler Activity to Release

Iodine atoms collected in sampler as a function of time,  $N(t)$ , are the net result of pickup from the gas source and the decay loss while awaiting counting.

$$dN/dt = f Q_{\text{smp}} C_{\text{stk}}(t) - \lambda N(t) \quad \text{Eq. 5.1}$$

Where:

- $N(t)$  = atoms I-131 (or I-132) collected by time  $t$
- $t$  = time measured from the start of the collection
- $f$  = sampler efficiency
- $Q_{\text{smp}}$  = air flow rate through the sampler
- $C_{\text{stk}}$  = iodine concentration in the stack air, atoms/cm<sup>3</sup>
- $\lambda$  = decay constant

In general, the iodine concentration in the stack gas,  $C_{\text{stk}}$ , is a variable with time, whereas the sample flow,  $Q_{\text{smp}}$ , should have been relatively constant over the sampling period. Solution of

---

<sup>3</sup>This is suggested by the absence of any recording in the datasheets of the liquid flow rate into the continuous holdup tank, a key parameter for the continuous determination. Instead the radiation level and volume of a “catch tank” are given at periodic times. Also, daily releases would have to be obtained by integration of the continuous releases. There is no record of such an integration. All is suggestive that the potentially continuous system was operated in a static mode from the start.

this first-order linear differential equation involves an integration of the variable concentration with the sampling time.

$$N(t_s) = \int_0^{t_s} dt' \exp(-\lambda (t_s - t')) f Q_{\text{smp}} C_{\text{stk}}(t') \quad \text{Eq. 5.2}$$

$N(t_s)$  = the atoms of a particular iodine isotope collected during the sampling time, 0 to  $t_s$ , balancing gain from the variable source and loss due to decay. The corresponding activity level of  $N(t_s)$  atoms is obtained simply by multiplying by the decay constant.<sup>4</sup>

Equation 5.2 is indeterminate; i.e., a single measurement of the sample activity at the end of the sampling period cannot recreate the variation of stack of gas concentration with time or even the total amount of iodine released.

The resolution is to make the sampling period short compared with the half-life, minimizing decay loss during the sampling period. In fact, this is the case for I-131. A 24-hour sampling duration was settled on routinely after Runs 1 and 2, which is short compared with 8.04 days, the I-131 half-life. Therefore, the I-131 measurement should be relatively free of error due to variability over the 24-hour sampling period.

Thus, the I-131 concentration may be assumed to be equal to some average value over the 24-hour sampling period. In such a case, the solution of Eq. 5.2 is the following, after multiplying by the decay constant,  $\lambda$ , to convert atoms,  $N$ , to activity.

$$A_{\text{smp}}(t_s) = f Q_{\text{smp}} C_{\text{stk}} (1 - \exp(-\lambda t_s)) \quad \text{Eq. 5.3}$$

Where  $A_{\text{smp}}(t_s)$  is the activity of the sample at the end of the sampling period,  $t_s$ .

If necessary, the right hand side may be multiplied by a decay factor,  $\exp(-\lambda t_2)$ , to account for decay loss from the time of stopping the sample acquisition to the time of counting,  $t_2$ , being this “time lapse.” Such a term appeared in the early datasheets. It was usually negligible (for I-131) and was later omitted.

The activity release from the stack is obtained by solving Eq. 5.3 for  $C_{\text{stk}}$ , multiplying by the stack flow rate, the sampling duration, and the decay constant.

$$\text{Rel} = \frac{1}{f} \left( \frac{Q_{\text{str}}}{Q_{\text{smp}}} \right) \left( \frac{\lambda t}{1 - \exp(-\lambda t)} \right) A_{\text{smp}} \quad \text{Eq. 5.4}$$

Where:

$A_{\text{smp}}(t_s)$  = activity of a particular iodine isotope in the sampler at end of sampling period

---

<sup>4</sup>This yields the activity in Bequerels.

---

$Rel(t_s)$	=	iodine activity released (Ci or Bq) from the stack during the sampling duration
$Q_{stk}$	=	stack flow rate
$f$	=	sampler efficiency

The release is expressed in whatever unit is selected for the sampler activity. The datasheets used millicuries (mCi) exclusively.

The equivalent of [Eq. 5.4](#) was used throughout the datasheets for calculating activity release to air from the measured sampler activity. In addition, [Eq. 5.4](#) can be used to back-calculate the assumed value of the collector efficiency,  $f$ , from data entries when its value does not explicitly appear (generally the case).

## 5.3 The Stack Monitor Datasheets

### 5.3.1 General Features

The [SMDs \(Doc MC 60312\)](#) are handwritten entries to logbooks in which the radiation levels of the stack samplers were recorded and converted into radioiodine release. The record begins February 1957, when the first two RaLa runs were performed, and continues through December 1959 and beyond, providing a complete set of datasheets for the period of interest.

The SMDs were the primary source of data that ultimately found its way into the [Historical Dose Evaluation \(DOE 1991\)](#). The route was as follows: [SMDs \(Doc MC 60312\)](#) → [Daily Notegrams from R.E. Hayden \(Doc MC 60111\)](#) → [Annual Report of the Health and Safety Division \(AEC 1959\)](#) → [DOE \(1991\)](#).

The record includes several campaigns for the determination of iodine collector efficiency,  $f$ , accompanied by hand sketches illustrating the way the measurements were made, and a crude description of the temporary iodine samplers installed for the April tests. Understanding this portion of the datasheets is aided by several letters, referenced as [Project Letters \(1957\)](#). The “production sampler” used throughout the testing period ending October 1957 is described in [Project Letters](#).

This section illustrates two of the more significant datasheets: (1) February 1957, which includes the first two RaLa runs, and (2) May 1958, when the first I-132 readings were made and by which time the standardized format was adopted. Several other datasheets are discussed.

Occasionally the record contains handwritten notes describing formulae used and results of related supporting experimentation. Some crude sketches show the method used to determine sampler efficiency. The supporting experimentation generally deals with measurements of various sampler collection efficiencies. These are difficult to interpret because of the lack of explanation.

### 5.3.2 *Sample Datasheets*

#### *February 1957 Datasheets*

Figure 5-3 shows a portion of this first datasheet through February 25, which includes Runs 1 and 2, February 1–3 and February 20–21, respectively. Note that there are from two to four determinations each day, and some days are missing. This was later standardized to one 24-hour determination each day.

The rather large volume “catch tank” in column 4, ranging from 3.5 to 18 liters, evidently was used in a static mode, as previously discussed. Later samplers were much smaller. Several different sizes were used, indicating the developmental nature of these early runs.

The primary data in cps (counts/second) are not recorded. Instead, the derivative, d/m/ml, (disintegrations per minute per milliliter) is given as the initial radiation entry. Though not stated, the readings here obviously refer to I-131. The conversion from cps to d/m/ml was not a trivial matter for I-131 and I-132 in 1957. There are two problems discussed more fully elsewhere: (1) a purified standard source must be made and counted by a 100% efficient detector, and (2) the NaI scintillation detectors used for early spectroscopy did not discriminate well between energy peaks. The readings could be subject to large errors due to radioactive impurities in the sample or by interferences from other iodine isotopes. These problems probably delayed regular acquisition of I-132 data until April 1958 and possibly prevented acquisition of I-133 data.

It is highly significant that there is already an entry accounting for sampler efficiency, the factor “f” in Eq. 5.4. Column 11 reads, “Correction for Sampler Eff. (Efficiency).” Comparing it to column 10 shows that the collection efficiency was assumed to be 50% for each determination, evidently administratively selected.

Volumetric stack flows are given in column 5, varying from 82,830 ft<sup>3</sup>/min to (occasionally) 120,000 ft<sup>3</sup>/min for this month.

After some trial and error, the column labeled “Volume Conversion Factor” was determined to be the term  $Q_{stk}/Q_{smp}$  in Eq. 5.4. This identification enables estimation of the assumed sampler efficiency in cases where it does not explicitly appear, which is generally the case. It can be confirmed that the releases to air in the last column were determined from the data in Table 5-1, according to Eq. 5.4, with the collector efficiency assumed to be 50%.

Several emendations are required in the February 1957 datasheet. Procedures probably were not well established for this first run. The most significant alterations required are noted below:

- Note that the dissolving evidently started February 1, as seen by the high reading for the catch tank activity for the second reading on this date, “ $4.8 \times 10^3$  d/m/ml.” Yet the resulting “I<sub>2</sub> to the atmosphere” is not recorded for some reason. This missing value may be estimated as 71.2 Ci based on the catch tank activity.

February 1957

HAR 1140723  
60312

Misc Info				Loadings Analysis									
Time	Time	Level	Volume	Concn	Time	SO <sub>2</sub>	NO <sub>x</sub>	CO	HC	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>
Stack	Sample	Value	SCM	Factor	Day	ppm							
0131	4.5L	92,850	4.1E10		01/31	1.5X10 <sup>3</sup>	3.5E <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	6.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>
0135					01/31	2.2E10 <sup>3</sup>	6.0E10 <sup>3</sup>	1.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>
0140	18L	97,990	4.9E10		01/31	8.0	1.6E10 <sup>3</sup>	6.2E10 <sup>3</sup>	3.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>
0145	18L	92,850	4.2E10		01/31	2.6E10 <sup>3</sup>	7.0E10 <sup>3</sup>	7.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>				
0150	18L				01/31	1.1E10 <sup>3</sup>	1.9E10 <sup>3</sup>	2.9E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>
0155	18L				01/31	2.4E10 <sup>3</sup>	1.2E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>	2.1E10 <sup>3</sup>
0200	4.9E				02/01	0.63	4E						
0205	18L				02/01	2.5E10 <sup>3</sup>	1.7E10 <sup>3</sup>	2.8E10 <sup>3</sup>	4.3E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>
0210	18L				02/01	0.44	2.3E10 <sup>3</sup>	9.0E10 <sup>3</sup>	4.8E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>	5.0E10 <sup>3</sup>
0215	18L				02/01	0.73	1.5E10 <sup>3</sup>	3.2E10 <sup>3</sup>	3.2E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>
0220	18L				02/01	0.50	9.2E10 <sup>3</sup>	1.3E10 <sup>3</sup>	2.1E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>
0225	18L				02/01	0.78	1.5E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.2E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>	2.6E10 <sup>3</sup>
0230	18L				02/01	0.29	7.2E10 <sup>3</sup>	1.9E10 <sup>3</sup>	1.3E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>
0235	18L				02/01	0.25	1.8E10 <sup>3</sup>	2.8E10 <sup>3</sup>	3.9E10 <sup>3</sup>	5.1E10 <sup>3</sup>	5.1E10 <sup>3</sup>	5.1E10 <sup>3</sup>	5.1E10 <sup>3</sup>
0240	18L				02/01	0.37	3.7E10 <sup>3</sup>	7.1E10 <sup>3</sup>	7.3E10 <sup>3</sup>	5.9E10 <sup>3</sup>	5.9E10 <sup>3</sup>	5.9E10 <sup>3</sup>	5.9E10 <sup>3</sup>
0245	18L				02/01	0.46	2.5E10 <sup>3</sup>	5.8E10 <sup>3</sup>	5.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>
0250	18L				02/01	0.33	4.0E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.3E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.0E10 <sup>3</sup>
0255	18L				02/01	0.28	2E	5E	5E	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.2E10 <sup>3</sup>
0300	18L				02/01	0.71	2.6E	4.5E10 <sup>3</sup>	4.8E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>
0305	18L				02/01	0.29	1.5E	2.0E10 <sup>3</sup>	5.1E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.5E10 <sup>3</sup>
0310	18L				02/01	0.47	13.4	2.1E10 <sup>3</sup>	2.9E10 <sup>3</sup>	3.1E10 <sup>3</sup>	3.1E10 <sup>3</sup>	3.1E10 <sup>3</sup>	3.1E10 <sup>3</sup>
0315	18L				02/01	0.40	2.7E10 <sup>3</sup>	5.6E10 <sup>3</sup>	5.2E10 <sup>3</sup>	4.7E10 <sup>3</sup>	4.7E10 <sup>3</sup>	4.7E10 <sup>3</sup>	4.7E10 <sup>3</sup>
0320	18L				02/01	0.2	8.8E10 <sup>3</sup>	6.6E10 <sup>3</sup>	6.7E10 <sup>3</sup>	5.2E10 <sup>3</sup>	5.2E10 <sup>3</sup>	5.2E10 <sup>3</sup>	5.2E10 <sup>3</sup>
0325	18L				02/01	0.77	1.4E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.5E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>
0330	18L				02/01	0.79	2.1E10 <sup>3</sup>	4.2E10 <sup>3</sup>	4.5E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>
0335	18L				02/01	0.33	2.7E10 <sup>3</sup>	3.4E10 <sup>3</sup>	5.0E10 <sup>3</sup>	4.8E10 <sup>3</sup>	4.8E10 <sup>3</sup>	4.8E10 <sup>3</sup>	4.8E10 <sup>3</sup>
0340	18L				02/01	0.23	1.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.1E10 <sup>3</sup>	1.7E10 <sup>3</sup>	1.7E10 <sup>3</sup>	1.7E10 <sup>3</sup>	1.7E10 <sup>3</sup>
0345	18L				02/01	0.19	3.0E10 <sup>3</sup>	3.0E10 <sup>3</sup>	4.1E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>	3.8E10 <sup>3</sup>
0350	18L				02/01	0.21	1.1E10 <sup>3</sup>	3.2E10 <sup>3</sup>	2.4E10 <sup>3</sup>	1.9E10 <sup>3</sup>	1.9E10 <sup>3</sup>	1.9E10 <sup>3</sup>	1.9E10 <sup>3</sup>
0355	18L	120,000	6E10 <sup>10</sup>		02/01	0.22	3.2E10 <sup>3</sup>	4.7E10 <sup>3</sup>	4.2E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>
0400	18L	120,000	6E10 <sup>10</sup>		02/01	0.21	4.0E10 <sup>3</sup>	2.0E10 <sup>3</sup>	2.1E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>	1.6E10 <sup>3</sup>
0405	18L				02/01	0.20	4.1E10 <sup>3</sup>	1.8E10 <sup>3</sup>	3.9E10 <sup>3</sup>	6.0E10 <sup>3</sup>	6.0E10 <sup>3</sup>	6.0E10 <sup>3</sup>	6.0E10 <sup>3</sup>
0410	18L				02/01	0.70	2.2E10 <sup>3</sup>	2.4E10 <sup>3</sup>	2.8E10 <sup>3</sup>	6.5E10 <sup>3</sup>	6.5E10 <sup>3</sup>	6.5E10 <sup>3</sup>	6.5E10 <sup>3</sup>
0415	18L				02/01	0.20	1.8E10 <sup>3</sup>	1.6E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>	3.9E10 <sup>3</sup>
0420	18L	89,000	4.5E10 <sup>10</sup>		02/01	0.21	1.5E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>
0425	18L				02/01	0.29	1.6E10 <sup>3</sup>	3.3E10 <sup>3</sup>	3.3E10 <sup>3</sup>	2.2E10 <sup>3</sup>	2.2E10 <sup>3</sup>	2.2E10 <sup>3</sup>	2.2E10 <sup>3</sup>
0430	18L	88,000	4.5E10 <sup>10</sup>		02/01	0.29	1.5E10 <sup>3</sup>	3.0E10 <sup>3</sup>	3.1E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>	2.3E10 <sup>3</sup>
0435	18L	120,000	6E10 <sup>10</sup>		02/01	0.28	2.1E10 <sup>3</sup>	1.8E10 <sup>3</sup>	1.8E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>
0440	18L	120,000	6E10 <sup>10</sup>		02/01	0.28	2.7E10 <sup>3</sup>	1.7E10 <sup>3</sup>	1.7E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>	1.5E10 <sup>3</sup>

The Data Reported Herein Was Generated  
Using Information in Current  
NO<sub>x</sub> Data Reporting Unit (NDRU) Information  
11-21-97

Figure 5-3 Stack monitor datasheet for February 1957 from document MC 60312 (including RaLa Runs 1 and 2)

- In addition, evidently no reading was taken on February 3, which was probably also a high-release day. The assumption will be made that the release for this date is equal to the average of adjacent dates, or 32.8 Ci.
- There are other corrections that seem warranted. Note also that the entry for February 10 is missing. No sample seems to have been taken. Averaging adjacent samples yields 28.9 Ci release for this date.

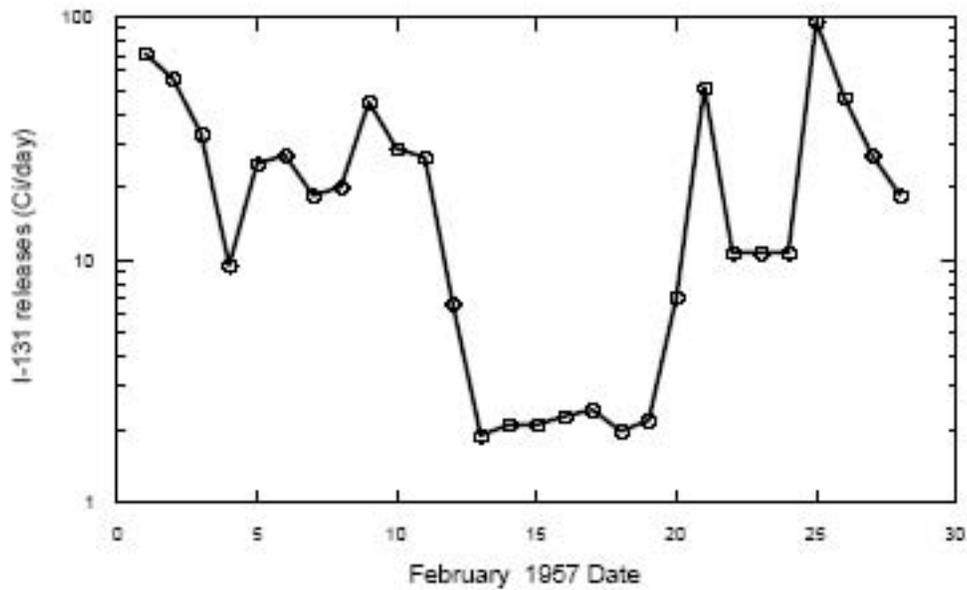
Assumed emendations to the February datasheets are summarized in [Table 5-1](#).

**Table 5-1 Emendations to the iodine-131 release record of February 1957 relative to the datasheets**

Date	Sample	Correction, I-131 release, curies	Reason for correction
February 1	0614	+71.2	Missing calculation of release; catch tank activity given
February 3	Not taken	+32.8	No sample taken; average of adjacent days
February 5	0619	+0.85	Missing calculation
February 8	0624	0	4 samples taken, only 2 reported; collection times are close; therefore, the sample durations probably overlapped
February 9	0778	0	Same situation as above
February 10	0632	+28.9	Entry missing; take average of adjacent days
February 11	0633	+13.2	2 samples taken; one missing; assume it is equal to the other
February 12	0635	+7	Missing sample; average of adjacent days
February 26	0654?	+2	Catch tank activity given, but missing release report
<b>Total release added for February = 156 Ci</b>			

The recommended emendations to the February 1957 datasheet add 156 Ci of I-131 release to the record. [Figure 5-4](#) illustrates the daily releases as given by the emended February datasheets. Note the peak releases on the day of dissolving, but also on other days due to unspecified operations. Release data are discussed more fully in [Chapter 6](#).

Not shown in [Figure 5-3](#) is a second page listing ( $\beta$ -I<sub>2</sub>) and  $\alpha$  catch tank radioactivity levels and calculated releases. Evaluation of these data must be dealt with separately.



**Figure 5-4 Iodine-131 daily releases during February 1957**  
(Run 1, February 1-2; Run 2, February 20-21)

#### *March 1957 Datasheets*

There were no RaLa runs in March. The runs were temporarily suspended due to the questions raised in February for Runs 1 and 2 ([Project Letters 1957](#)). Procedures were still in a formative stage in this second month of operations. The March datasheets omit 9 days of sampling. The omissions were filled in by assuming releases to be equal to the average of adjacent days. As [Table 5-2](#) shows, these additions add 13.2 Ci to the total I-131 released for the month.

It is important to note that about 83 Ci of I-131 were released in March 1957, a month in which no runs were made.

**Table 5-2 Emendations to the iodine-131 release record of March 1957 relative to the datasheets**

<b>Date</b>	<b>Sample</b>	<b>Correction, I-131 release, curies</b>	<b>Reason for correction</b>
<b>March 3</b>	None	+13.2	Missing sample; assumed average of adjacent days
<b>March 15</b>	None	+5.2	Missing samples; assumed average of adjacent days
<b>March 16</b>	None	+5.2	
<b>March 24</b>	None	+1.3	Missing samples; assumed average of adjacent days
<b>March 27</b>	None	+0.2	
<b>March 28</b>	None	+0.2	
<b>March 29</b>	None	+0.2	
<b>March 30</b>	None	+0.2	
<b>March 31</b>	None	+0.2	
<b>Total release added for March = 13.2 Ci</b>			

#### *April 1957 Datasheets*

This was a developmental month for stack monitor operations. Eleven pages in the logbook are devoted to stack monitor activities for this month, including the following:

- The first I-132 readings, soon suspended until May 1, 1958
- Four pages devoted to sampler efficiency evaluations, discussed below
- One page outlining the release calculation, consistent with Eq. 5.4 and confirming that 50% was to be used as the sampler efficiency

The temporary samplers noted in [Figures 5-1](#) and [5-2](#) were installed for system tests. The production sampler size was reduced to 330–1,000 ml compared to the earlier 18-liter size.

#### *May–June 1957 Datasheets*

The May and June datasheets are mixed in the record. They include both RaLa Runs 4 and 5. Some June work seems to have been recorded as late as August. There were more sampler efficiency determinations. Iodine-132 measurements were temporarily halted, and not restarted until May 1, 1958.

### *July 1957 Datasheets*

The sample acquisition routine was stabilized to one sample per day, extending from midnight to midnight. This routine was maintained through December 1959 and beyond.

### *October 1957 Datasheets*

Much extra activity was recorded during this month, undoubtedly related to the sampler system tests described in [Project Letters \(1957\)](#). Runs 7 and 8 occurred during this month. Iodine-131 releases were calculated daily in accordance with the format adopted in July.

### *May 1958 Datasheets*

The I-132 determinations resumed and the datasheet assumed a near final format ([Figure 5-5](#)). Note that high releases are reported for I-132 for May 1 and May 28, the dates associated with RaLa Runs 13 and 14. No explicit collection efficiency is assumed, but one can verify numerically that it was again assumed to be 50%. [Figure 5-6](#) illustrates the daily releases following Run 14 on May 28, 1957. Note the different behavior of I-132 relative to I-131 and its high peak (the I-132 scale multiplier is times 10) release on the day of the dissolving.

### *June 1958–December 1959 Datasheets*

The sampling routine and datasheet format remained similar to that in May 1958.

[Chapter 6](#) shows a comparison of the SMDs with the reported releases.

## **5.4 Stack Monitor System Evaluation**

### **5.4.1 Evaluation of Sampler Efficiency**

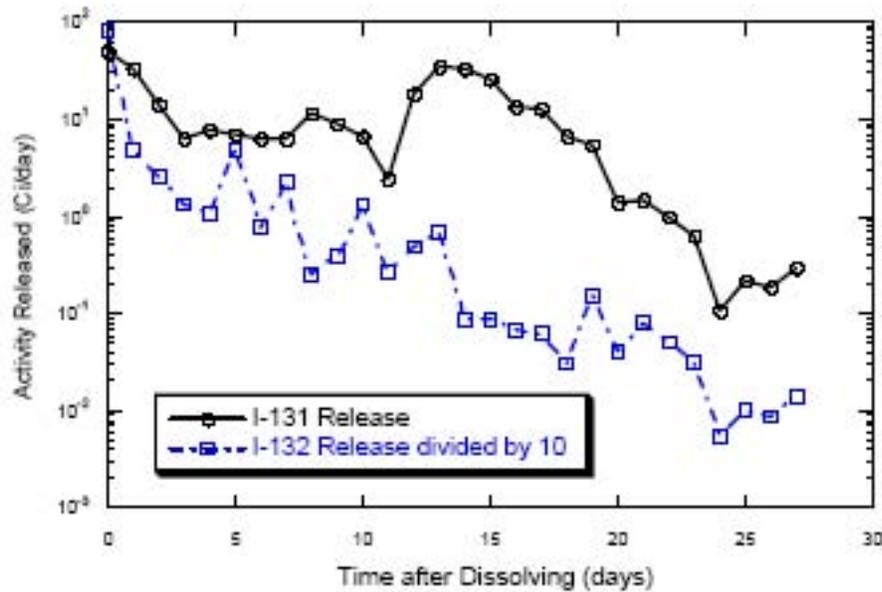
The record shows that a sampler efficiency of 50% was administratively selected for use throughout the operation of the stack sampler system. The earliest datasheets show the sampler efficiency entry of 50%. The entry was later omitted, but the value was maintained throughout, as may be verified by back calculation of the results using [Eq. 5.4](#).

[Project Letters \(1957\)](#) record several measurements of the efficiency of the actual sampler (as it existed up to November 1957) used to acquire the data reported in the SMDs. Extensive modifications of the system were initiated November 11, 1957. No record was found on how these modifications affected the iodine sampler design, if at all. The data summarized below show that the assumed efficiency of 50% was fairly accurate for the early sampler. Some variability from this average is seen.

Numerous sampler efficiency determinations are reported in [Project Letters \(1957\)](#) and in the SMDs for the period April–October 1957. Most of these are for the temporary samplers installed at Locations A, B, and C, shown in [Figure 5-1](#). A few are for the actual production sampler at Location D, at least for the production sampler used from April–November 1957.

General Information										I <sup>132</sup> Analysis			I <sup>131</sup> Analysis						
Day of Month	NPR #	Time Start	Time Stop	Sample Time	Sample Volume	Scrubber Flow CFM	AV Short	Volume Corrected	Time	24h Cond	41/41	41/41	41/41	41/41	41/41	41/41	41/41	41/41	41/41
1	110	24.0	24.0	24.0	970	90,000	4.56	4,560	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
2	115	24.25	24.25	24.25	950														
3	116	24.0	24.0	24.0	960														
4	117	24.0	24.0	24.0	950														
5	118	24.0	24.0	24.0	930														
6	119	24.0	24.0	24.0	950														
7	120	24.0	24.0	24.0	900														
8	121	24.0	24.0	24.0	970														
9	122	24.0	24.0	24.0	970														
10	123	24.0	24.0	24.0	1000														
11	124	24.0	24.0	24.0	990														
12	125	24.0	24.0	24.0	990														
13	126	24.0	24.0	24.0	970														
14	127	24.0	24.0	24.0	990														
15	128	24.0	24.0	24.0	970														
16	129	24.0	24.0	24.0	950														
17	130	24.0	24.0	24.0	920														
18	131	24.0	24.0	24.0	980														
19	132	24.0	24.0	24.0	980														
20	133	24.0	24.0	24.0	980														
21	134	24.0	24.0	24.0	920														
22	135	24.0	24.0	24.0	920														
23	136	24.0	24.0	24.0	920														
24	137	24.0	24.0	24.0	920														
25	138	24.0	24.0	24.0	920														
26	139	24.0	24.0	24.0	920														
27	140	24.0	24.0	24.0	920														
28	141	24.0	24.0	24.0	920														
29	142	24.0	24.0	24.0	920														
30	143	24.0	24.0	24.0	920														
31	144	24.0	24.0	24.0	920														
													1540,290				275,645		
													1540,200						

Figure 5-5 Stack monitor datasheet for May 1958 from document MC 60312 (including RaLa Runs 13 and 14)



**Figure 5-6 Iodine-131 and iodine-132 releases following Run 14 started on May 28, 1958**

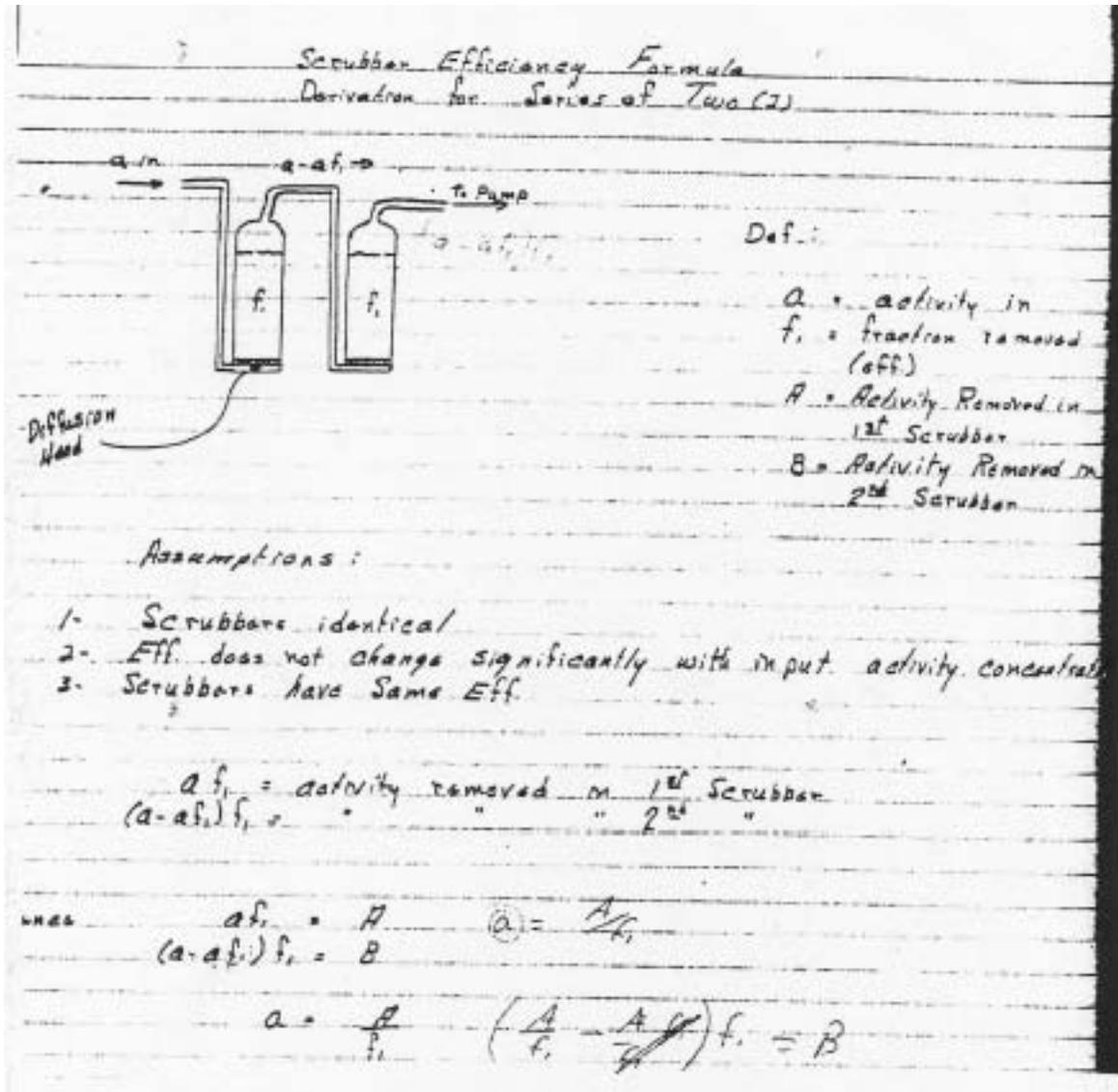
(Note: Scale multiplier for Iodine-132 is 10)

Figure 5-7 is a page from the April datasheet illustrating one of the two methods used to determine the collection efficiency, at least for the temporary samplers. As shown, the temporary samplers were glass bottles with a glass frit at the base for distributing the gas inlet. In comparison, Project Letters (1957) describe the early “production” sampler as a small packed column with dimensions shown in Figure 5-1.

The assumptions of the method are clearly given in Figure 5-7. The source strength for the first sampler is labeled “a” and is in Ci/sec. With “f•a” removed by the first sampler, “a – f•a” is the source for the second sampler. The quantity collected in the sampler may be estimated from Eq. 5.3 for each sampler. For equal sampling duration, the ratio of the activity of the second sampler, B, to the first, A, relates to “f” as follows:

$$f = 1/(1 - B/A) \quad \text{Eq. 5.5}$$

where B/A is the ratio of activities for samplers B and A.



**Figure 5-7 A portion of a datasheet for April 1957 showing the determination of sampler efficiency**  
(Stack Monitor Datasheets)

Another method was used, as implied in some of the datasheets, that involved placing three samplers in series, for example A, B, and C. If the efficiencies are reasonably high, then it can be assumed that all the radioactivity is collected in the three samplers, and the efficiency of sampler A is as follows:

$$f = A/(A + B + C) \qquad \text{Eq. 5.6}$$

The advantage of the second method over the first is that the samplers generally do not have to have equal efficiency, especially useful when using laboratory glass bottle devices.

All of the efficiency determinations for the production sampler at Location D are summarized in [Table 5-3](#).

**Table 5-3 Efficiency determinations of the “Production” Sampler, Location D**

Reference	Efficiency range, %	Number of tests	Average efficiency, %	Standard deviation
BLR-8-57A	46–60		50	
BLR-8-57A	45–60		50	
May Datasheets	30–57	3	0.465	0.147
June Datasheets		4-6	0.50	
June Datasheets		4-6	0.64	

In general, the assumed average efficiency of 50% seems to be borne out.

For comparison, it may be of some interest to look at the sampler efficiencies determined for the temporary samplers at the following locations:

- A – RaLa DOG line near CPP-605
- B – ICPP vent duct near CPP-605
- C – in stack at the 45-foot level
- D – regular stack monitor

Sampler efficiencies at locations A, B, and C are shown in [Table 5-4](#).

**Table 5-4 Sampler efficiency determinations for the temporary sampler  
(Project Letters 1957)**

Sampler*	Sample rate, cfm	Efficiency spread, %	Average
During Run 3, April 1957			
A	0.0112	54-80	60
B	0.27-0.384	92-99.5	95
C	0.27-0.384	60-86	75
During Run 4, May 1957			
A	0.0122	58-60	60
B	0.153	93-99.5	95
C	0.153	62-98	80
During and After Run 5, June 1957			
A	0.0122	61-63	62
B	0.153	88-92	90
C	0.153	75-88	82

\* Samplers A, B, and C were temporary, glassware sample bottles of the type sketched in [Figure 5-2](#). They were installed at the locations indicated in [Figure 5-1](#).

The SMDs list several other efficiency campaigns, the last being in October 1957. However, without a published topical report on these tests, the datasheet entries are difficult to interpret. (For example, samplers E1, E2, and W are cited in October without explanation as to purpose or location.) In general, the efficiency determinations for the temporary samplers improved with time for unspecified reasons.

The fact that the sampler efficiency was an issue from the outset is a positive sign. The 50% efficiency, applied consistently from beginning to end, appears to have been an administrative decision. It was probably considered to be reasonable and conservative (i.e., low).

Several efficiency determinations were reported for Location D, the production location, with the latest being in September 1957. It may be surmised that this was the actual, packed column production sampler described in [Figure 5-1](#) in use up to November 1957. It is not known if this was continued after the November 1957 system revisions.

Results reported for Location D generally support the 50% assumption as a realistic estimate. Various, the ranges of 46-60, 45-60, 45.5 +/- 0.147 sd, and average values of 50% and 64%, are reported. Therefore, a range of 45%-60% with an average around 50% appears to be supported.

#### **5.4.2 Iodine Deposition Error**

The chemically active  $I_2$  molecule is attracted to bare, dry steel surfaces by weak chemical forces resulting in chemisorption. The attraction is much lower for oxidized surfaces. Although the mass transfer and chemisorption rules are known in principle, impurities in the gas and on the surface have a profound effect that renders attempts at prediction of deposition losses generally useless.

One theoretical feature of chemisorption may be worth noting. There are only a limited number of active sites on the surface available for deposition. Sites occupied by the rapidly decaying I-131, I-132, I-133, and I-135 soon become vacant as these isotopes decay to xenon. Some of the vacated sites will be reoccupied by the stable I-127 or long-lived I-129. Therefore, the active sorption sites would be expected to ultimately become occupied with stable or near-stable species. The surface would then be saturated and unreceptive to further  $I_2$  chemisorption.

Wet sample lines undoubtedly retain much larger amounts of  $I_2$  than dry lines. As noted in Chapter 3,  $I_2$  reacts with water to form soluble  $I^-$  (iodide) and ultimately  $IO_3^-$  (iodate). Puddles of water in sampling lines are particularly prone to collect iodine, as evidenced by a notably flawed iodine deposition test conducted at Hanford ([Hanford 1949](#)).

#### *Iodine Deposition Tests*

Two campaigns of iodine deposition tests were conducted in 1957, one in April and one in June. A temporary sampler was installed at the 45-foot level of the stack where the sampler line was

short ([Project Letters 1957](#), BLR-8-57A) (see Location C in [Figure 5-1](#)). Sampler D was the regular stack monitor sampler in operation in April 1957.<sup>5</sup>

As noted in [Figure 5-1](#), the sample line to Location D was 130 feet long and consisted of ½ inch, schedule 40 stainless steel pipe. The length of the sample line to Location C was not given, but was obviously short. The test results comparing estimated releases based on samplers at Locations C and D are summarized in [Table 5-5](#).

**Table 5-5 Effect of iodine deposition on estimated releases, April, March, and June 1957 tests**  
([Project Letters 1957](#), BLR-8-57A and BLR-25-57A)

Location*	Efficiency spread, %	Average efficiency	I-131 release, mCi
C	60-86	75	939
D		50% assumed	1,140
C	60-86	75	498
D		50% assumed	647
C	60-86	75	782
D		50% assumed	920
C	60-86	75	57 <sup>†</sup>
D		50% assumed	529
C	60-86	75	220
D		50% assumed	465
C	62-98	80	1,820
D	46-60	50	3,890
C	62-98	80	286
D	46-60	50	582
C	75-88	82	233
D	45-60	50	810
C	75-88	82	450 <sup>†</sup>
D	45-60	50	3,590

\* Sampler C located at 45 feet elevation of the stack, short sampling line. Sampler D located in CPP-605, sampling line = 130 feet.

† The measurement appears to be low.

As [Table 5-5](#) shows, for some unknown reason I-131 releases based on Sampler D with the 130-ft sampling line always exceeded estimates based on Sampler C with the short line. This unexpected result was undoubtedly due to some bias in the test procedure, e.g., the flow rate through the sampler or the estimated efficiency. Nevertheless, diminished release estimates due to I-131 deposition in the sampling line were not demonstrated.

An earlier attempt to evaluate an I-131 deposition effect on the release estimate was based on a measured surface concentration in a 2-inch length of sampling line after Run 2, February 1957

<sup>5</sup>As noted, stack monitor system modifications were made beginning November 11, 1957. We have found no description of the new system. Possibly, this is when it was moved into CPP-643, at the base of the stack.

(Project Letters 1957, BLR-8-57A).<sup>6</sup> The letter states: “Rough calculations, using these analyses, indicated that 500-1,000 Ci of I-131 activity could have been discharged to the atmosphere during RaLa Run 2, which could have escaped unmonitored, assuming the worst possible conditions, i.e., (1) all the activity was I-131; (2) all the activity deposited in the first day of the run; (3) no activity was removed by continuous air stream; and (4) the entire length of pipe contaminated to the same level as that of the 2” section removed from a horizontal welded run.”

This early test directly contradicts the later, extensive tests reported in Table 5-5. It also demonstrates the perversity of iodine deposition test data taken under non-pristine conditions. Evaluation is difficult, but it seems that assumption (4) is key. One would expect an exponential reduction of deposit along the length of the line, so the location of the 2-inch specimen of sample line is critical. If the sample was taken from near the stack, the impact on the release would have been overestimated.

#### *Moisture in the Sampling Line*

The same reference states that moisture condensing in the sampling line was a problem during the first two runs. Since virtually all of the air volume in the sample line was ventilation air from the main process building, condensation of building humidity on cold days would be the major source. Runs 1 and 2 were conducted in February, indicating a possibility of condensation. The only remedies would be (1) heat the sample line, (2) reduce the humidity of the main process building such that its dew point is below the outside air temperature, and/or (3) minimize the length of line. Also, minimizing horizontal runs and avoiding low spots would be helpful.

None of these possible remedies for condensation was described in Project Letters (1957) for the modifications instituted in April 1957. However, the sampler building was at some point moved from CPP-605 adjacent to the main process building, to CPP-643 located at the base of the stack. Perhaps this was done in the modifications alleged to have begun in November 1957. In its new location, the sample line would have been shortened to about 50 feet, virtually all vertical run. No mention of heating the sample line has been found.

#### *Summary of Sample Line Deposition*

Sample line deposition is a difficult subject to evaluate. However, the extensive tests summarized in Table 5-5, improbable though the results might be, do not show a sample line deposition effect. The high measured I-131 deposit in the 2-inch sample line specimen could have been due to close proximity to the source. Shortening of the sample line to about 50 feet, which may have occurred in November 1957, would have a generally beneficial effect.

#### **5.4.3 Effect of Variable Iodine Release**

As noted above with reference to Eq. 5.2, in general, it is not possible to relate the measured activity in the sampler to the iodine concentration in the stack by simply observing the end point

---

<sup>6</sup>The surface deposit was identified by  $\gamma$ -spectroscopy to consist of 63% I-131, 33% ZrNb, and 4% Ru.

inventory in the bottle. One must assume some concentration variation, the term  $C_{stk}(t)$ , or as a special case, assume it is constant and equal to some average value.

The latter was what was done. Intuitively, this appears to be a valid approach for the case of small decay over the length of the sampling period, as it would be for 8.04 day I-131 in the 24-hour sampling period. However, significant sensitivity to source variability may be expected for the shorter lived I-132 (2.3-hour half-life) or any other short-lived nuclide.

This may be demonstrated by assuming various gas concentration variations,  $C_{stk}(t)$ , and integrating the general equation, Eq. 5.2. Figure 5-8 shows such an estimate. Four different gas concentration variations are assumed to occur over the 24-hour sample period, time-steady labeled  $C_{ave}$ , and burst releases of the same total quantity of I-131 or I-132 at 0 time, 12 hours, and at the end of 24-hour sample run. The ratio of the actual release to the calculated release, based on an assumed steady value for the source, is listed in Figure 5-8.

Note that burst releases of I-131 occurring at either end of the sampling period cause only a 4% error compared with the regular calculation that assumes a steady source. However, I-132 release estimates are seen to be highly sensitive to the actual timing of the release. An I-132 burst occurring near the beginning of the sampling period would be underestimated by a factor of 190 due to underestimation of the decay. An I-132 burst release occurring near the end of the 24-hour sampling period would be overestimated by factor of 7 (i.e., the reciprocal of 0.14) due to overestimation of decay during sampling.<sup>7</sup>

The principle also applies to the  $\beta$ -I and  $\alpha$  determinations, except that in this case the species are not known. However, if these readings were caused by short-lived nuclides, similarly large errors would be incurred due to source strength variability.

#### *Conclusion on Source Variability*

Iodine-131 is relatively immune from source variability error over the 24-hour sampling period. In contrast, I-132 readings are highly sensitive to source strength variations occurring during the sampling period, being greatly underestimated for high early releases and overestimated for high late releases. This applies as well to the  $\beta$ -I determinations.

#### **5.4.4 Possible Radiation Detection Errors**

As noted above, the SMDs report iodine inventory in the sampler bottle in terms of disintegrations per minute per milliliter (d/m/ml), essentially Ci/ml, with an appropriate conversion factor. The actual reading is counts/min (cpm). This is converted to Ci/ml by a calibration curve that accounts for the volume of the sample, the geometry of the counting system, efficiency of the crystal for the particular  $\gamma$ -energy, and the number of emitted gammas per disintegration for that particular energy:

$$Ci/ml = E \times cpm \qquad \text{Eq. 5.7}$$

<sup>7</sup>The October 16, 1959, criticality accident started at about 3 AM.

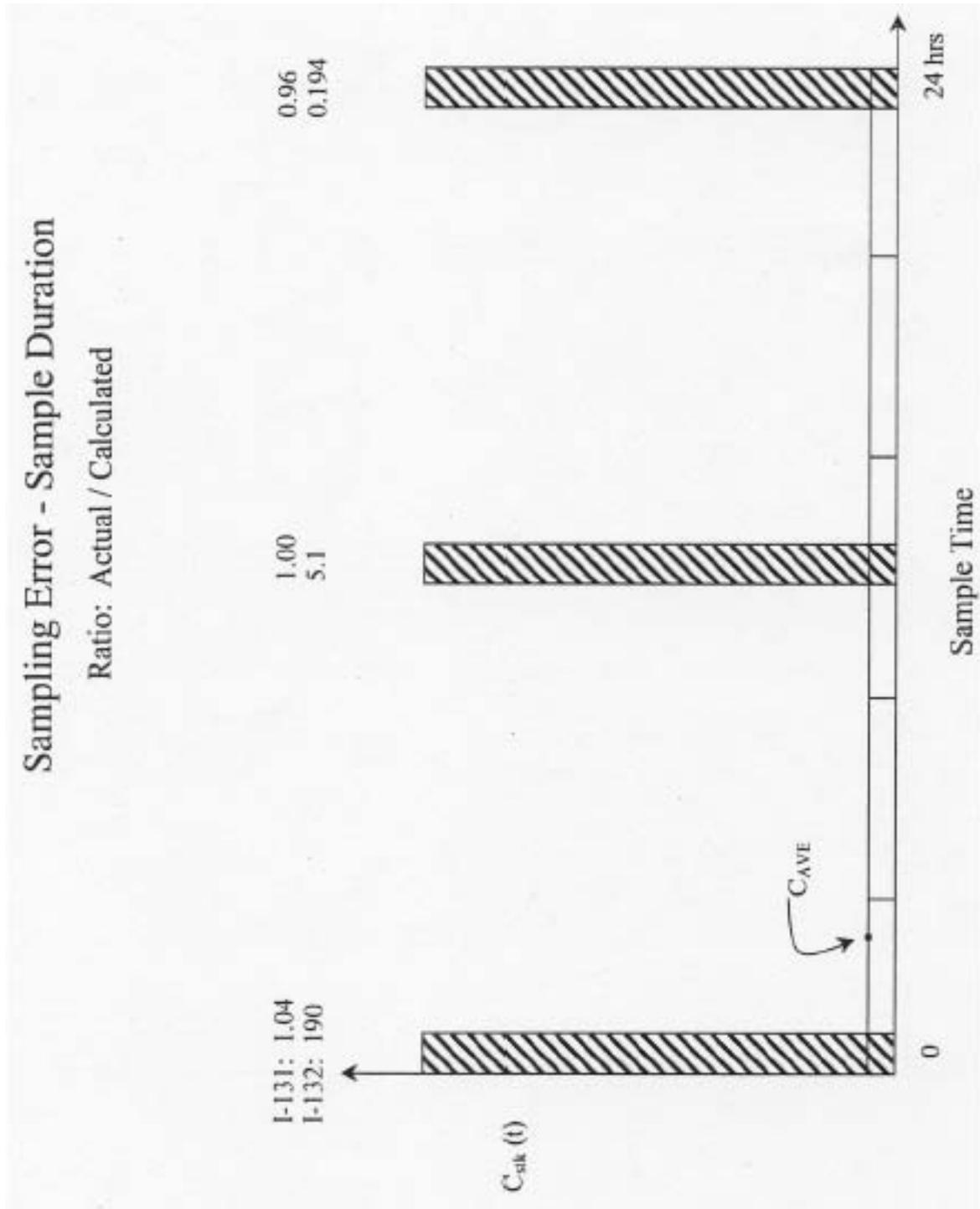


Figure 5-8 Detection error due to sample strength

where E is the efficiency factor for that particular geometry and  $\gamma$ -energy. Determination of E in early  $\gamma$ -spectroscopy was not yet a routine matter. A major hurdle was obtaining a standard sample of the pure isotope. Radiochemical procedures were available in 1957 for I-131 (Klienber [and Cowan 1960](#)) and for the more difficult I-132 (Leddicotte [1961](#)), the latter requiring a tellurium separation.

A more serious question is the effect of impurities in the sample. Early  $\gamma$ -spectrometers, based on NaI scintillation, were not highly discriminating. The cpm reading would include counts from about +10% to -10% of the energy setting. They ought not to have been used on unknown samples and might not select out impurities that happen to emit close to the selected energy (Dyer [2002](#)).

In such a case, the cpm reading is the sum of the iodine emission plus the contaminant:

$$\text{cpm measured} = \text{cpm iodine} + \text{cpm contaminant} \quad \text{Eq. 5.8}$$

A search was conducted using ORIGEN2 for  $\gamma$ -emitters that could have been present in the sample that would affect the reading. Of particular interest was evaluation of the I-132 readings, which appeared to be unusually high on the day of the dissolving and following the October 16, 1959, criticality accident. As noted in [Table 3-2](#), no  $\gamma$ -interferences were found for I-131. Only minor interferences were found for I-132.

In particular, the extremely high I-132 release observed on the first day of the criticality accident cannot be ascribed to interference from impurities in the sample.

## 5.5 Summary and Conclusions Regarding the Stack Monitor Datasheets

### 5.5.1 Summary of Stack Sampler Error Evaluation

#### *Sampler Efficiency*

The sampler efficiency of 50% used to compute releases seems reasonable in the light of measurements conducted in 1957. (The range may be from 40%-60%.)

#### *Iodine Deposition in the Sampler Lines*

Evaluation of this effect is not conclusive. However, the extensive tests summarized in [Table 5-5](#), improbable though the results might be, do not show a sample line deposition effect. Shortening of the sample line to about 50 feet, which may have occurred in November 1957, would have a beneficial effect.

#### *Effect of Variable Iodine Concentrations*

Iodine-131 is relatively immune from source variability over the 24-hour sampling period. In contrast, I-132 readings are highly sensitive to strength variations occurring during the sampling period. The  $\beta$ -I and  $\alpha$  readings would also be subject to source strength variability errors if caused by short half-life nuclides.

### *Radiation Detection Errors*

No significant  $\gamma$ -interferences have been found that may affect I-131 and I-132 collector readings.

#### **5.5.2 Conclusions**

- The stack sampler appears to provide a satisfactory basis for estimating daily I-131 releases to the atmosphere. The major sources of error have been examined. The principal uncertainties are sample line deposition and the lack of description of the newer, post-November 1957 sampler.
- Iodine-132 release estimates are highly sensitive to variations occurring during the 24-hour sampling period. Measurements did not consistently begin until May 1, 1958, and were not recorded in the [Hayden Notegrams](#) until May 25, 1958. Some extremely high I-132 readings are recorded, particularly for May 1958.
- There are some missing daily entries in the [SMDs](#), particularly for I-131 in February-March 1957, during which significant releases must have occurred. Missing days may be filled in by averaging releases from adjacent days. Some missing entries cannot be filled in without uncertainty due to the different procedures used in the early runs.
- The [SMDs](#) may be used to extend the I-131 release record reported in the first semi-public documentation, the [Hayden Notegrams](#), back to February 1, 1957, the initiation date of RaLa Run 1. The Hayden daily reports began July 7, 1957. In addition, several of the Notegrams have evidently been lost. The missing information can be recovered from the datasheets.

---

## 6.0 IODINE RELEASES FROM THE RALA PROCESS, FEBRUARY 1957 TO DECEMBER 1959

### 6.1 Early Iodine Releases

An early memo (Ha-56-57A, in [Project Letters 1957](#)) states that although releases from RaLa Runs 1 and 2 were higher than planned, they still more than adequately met existing release criteria with wind velocities as low as one mile per hour.

Nevertheless, other letters in this collection ([Project Letters 1957](#)) exhibit concern for the higher than expected iodine releases for RaLa Runs 1 and 2 in February 1957, as measured by the early stack monitor. Not helping the situation was the realization that the gas accumulator placed in the off-gas line to store discharged gases was inadequately small. The purpose of the accumulator was to store the off-gas until the wind direction and magnitude proved to be favorable.

The concern over the high releases in Runs 1 and 2 prompted a program of improvements that extended over the first three years of operation. These improvements included (1) a much larger off-gas accumulator, (2) placing (or improving) shaft seals on each centrifuge, (3) lowering process temperatures to reduce iodine volatility, (4) searching for an additive to reduce iodine volatilization from acid solutions, (5) placing an additional iodine absorber in the DOG line, and (6) reducing the number of liquid sampling operations, each of which added gas volume to the system, increasing the off-gas flow.

The cumulative effect of these improvements proved to be highly successful in reducing iodine releases. As a corollary, it must be said that the project was not adequately prepared for dealing with iodine releases at startup. Review of the fission product behavior tests presented in [Chapter 2](#) illustrates that studies of iodine behavior in the process were minimal and contradictory.

### 6.2 Existing Reports of Iodine Release from the RaLa Process

#### 6.2.1 *The Hayden Notegrams*

Internal letters called the [Hayden Notegrams \(Doc MC 60111 or MC 55203\)](#) were the first reports of daily iodine releases from the ICPP stack in a semi-published form. They were written weekly by the head of the Health Physics Department of the ICPP. When spot-checked against the source of the data, they faithfully reproduce the [Stack Monitor Datasheets \(SMDs\) \(Doc MC 60312\)](#). [The Hayden Notegrams](#), however, have the following omissions:

- Only weekly summaries were written prior to July 7. The [SMDs](#) began daily reporting of I-131 from the start of operations on February 1, 1957.
- Daily reports of I-132 releases were not reported until May 28, 1958. The datasheets began reporting them on May 1, 1958.

- Quite a few of the Notegrams are lost from the available collection.
- Therefore, it is better to use the [SMDs](#) for assembling or evaluating release data. Some sample [Hayden Notegrams](#) are given in [Appendix E](#).

### 6.2.2 *The Health and Safety Division Annuals*

The [Hayden Notegrams](#) were the source for releases reported in the nearly contemporaneous [Health and Safety Division Annual Reports](#). However, no references are provided, and the entries are misleading.<sup>1</sup> The Health and Safety Division Annual for 1959 ([AEC 1959](#)) summarized the release data for RaLa Runs 1-37; i.e., from February 1, 1957, through December 16, 1959.

The releases are tabulated per RaLa run, which on the average occurred at a rate of one run per month. As a result, no releases are reported for the several months in which there were no RaLa runs, e.g., March 1957, which showed some high releases according to the datasheets. In addition, it is not clear how many days of release were attributed to each run.

[The Health and Safety Division Annuals](#) provide some useful RaLa operational information available nowhere else. Nominal run dates are given, along with the approximate startup date for the charcoal filters, again with no reference. This is the earliest available source for such operational information.

### 6.2.3 *The Historical Dose Evaluation Study*

The Historical Dose Evaluation Report ([DOE 1991](#)) largely adopted the Health and Safety Division table for iodine releases from RaLa operations. Therefore, most of the comments above apply. [Table 6-1](#) is reproduced from DOE 1991.

The few differences between [Table 6-1](#) and its principal source ([AEC 1959](#)) are summarized below:

- The Health and Safety Division Annual ([AEC 1959](#)) reported the ICPP criticality on October 16, 1959, as an operational release from RaLa Run 34, which started three days earlier. [DOE \(1991\)](#) separated the two, listing the criticality release with “accidental releases.” [DOE \(1991\)](#) also greatly reduced the large reported I-132 release during the accident, stating that it was a physical impossibility. (The discussion of the criticality accident in [Chapter 7](#) concurs with this conclusion.)

---

<sup>1</sup>As read, the release data reported in the Annuals appear to have been developed within the Health and Safety Division of INEL instead of from within the ICPP. Particularly misleading is placement of RaLa releases in the section entitled “Site Survey.” The reader has no indication that the ultimate source of the data was the ICPP stack monitor.

**Table 6-1 Curies released per RaLa run, reported in DOE (1991)**

Run Number	Date of Run	Radionuclide (Ci)				Beta Activity Minus Iodine (Ci)
		I-131	I-132	I-133	I-135	
001	2/1-3/57	230.0	b	b	b	114.6
002	2/20-21/57	351.3	b	b	b	334.7
003	4/5-6/57	80.8	b	b	b	1.3
004	5/19-20/57	42.0	b	b	b	6.6
005	6/24-25/57	158.7	b	b	b	6.8
006	9/11/57	103.4	b	b	b	5.7
007	10/7/57	201.7	b	b	b	14.8
008	10/21/57	234.7	b	b	b	35.7
<b>Total Activity 1957</b>		1402.6	4030	441	6.65	520.2
009	1/6/58	35.1	b	b	b	27.1
010	2/12/58	154.6	b	b	b	26.6
011	3/13-14/58	55.6	b	b	b	12.6
012	4/16/58	34.9	b	b	b	9.2
013	4/30-5/1/58	176.6	626.3	b	b	22.4
014	5/28/58	111.1	935.0	b	b	28.7
015	6/2/58	205.8	119.0	b	b	53.0
016 <sup>c</sup>	8/6/58	27.7	9.9	b	b	14.9
017	8/13/58	75.3	18.7	b	b	77.6
018	10/1/58	70.6	310.6	b	b	62.7
019	10/22/58	45.6	498.8	b	b	30.6
020	10/22-23/58	d	d	d	d	d
021	11/12/58	32.6	120.6	b	b	72.3
<b>Total Activity 1958</b>		1025.5	3380	484	7.67	437.7
022	2/4/59	104.8	171.8	b	b	16.2
023	2/25/59	14.4	18.8	b	b	16.4
024	3/18/59	40.5	60.0	b	b	8.0
025	4/8/59	4.7	24.8	b	b	11.0
026	4/29/59	3.4	129.9	b	b	27.6
027	5/19/59	2.3	14.3	b	b	21.2
028	6/10/59	2.1	64.9	b	b	27.3
029 <sup>e</sup>	7/6-7/59	9.3	79.5	b	b	8.6
030	7/21-22/59	6.4	183.5	b	b	8.5
031	8/11-12/59	2.1	61.4	b	b	18.0
032	9/1-2/59	4.4	29.8	b	b	4.0
033	9/22-23/59	5.0	37.2	b	b	4.6
034	10/13-14/59	10.3 <sup>f</sup>	46.2 <sup>f</sup>	b	b	30.6
035	11/5-6/59	12.0	112.2	b	b	9.2
036	11/30, 12/1/59	.4	.3	b	b	2.0
037	12/15-16/59	1.4	27.1	b	b	5.9
<b>Total Activity 1959</b>		223.5	1061.7	99.8	1.51	219.1

- [DOE \(1991\)](#) included annual estimates of I-132 releases for 1957 and 1958 when data were absent or incomplete. The report states, “The I-132 was adjusted to 3.3 times the I-131 value. This is based on the average of the measured ratio over 1-year period beginning with RaLa run 13 in April 1958. Measured releases over the subsequent year were then used to evaluate the I-132/I-131 ratio of 3.3.”
- The I-132/I-131 release ratio given in [Table 6-1](#) is 2.87 for 1957. It is 3.3 for 1958. However, data for subsequent years (not shown here) show a higher ratio than 3.3.
- [DOE \(1991\)](#) added annual estimates of I-133 and I-135 curie release based on calculated estimates. Evidently these were obtained by multiplying the observed I-131 release by the inventory ratio of I-133/I-131 and I-135/I-131 in a typical 2-day cooled MTR element.<sup>2</sup>

### 6.3 Iodine, Tellurium, and Xenon Nuclides Fed to the Dissolver

[Table 6-2](#) shows the inventory, in curies, of iodine, tellurium, and xenon nuclides in a typical, 2-day cooled fuel element fed to the dissolver. The inventories are based on ORIGEN2 estimates using the reconstructed irradiation history surmised for this element, according the discussions presented in [Chapter 4](#).

**Table 6-2 Iodine, tellurium, and xenon nuclides fed to the dissolver per typical 2-day cooled run\* (including precursor tellurium) in curies**

Nuclides	Mass number			
	131	132	133	135
<b>Te m</b>	1,463	0	0	0
<b>Te</b>	329	30,080	0	0
<b>I</b>	24,400	31,000	14,960	446
<b>Xe m</b>	234	0	1,579	71
<b>Xe</b>	0	0	60,020	3,573

\* Taken from ORIGEN results for Run 7, October 1957

The rows in the table indicate the  $\beta$ -decay direction, from top to bottom. Note that I-132 is in quite close dynamic equilibrium with its precursor, Te-132. In other words, the decay rate of short-lived I-132 (2.3-hour half-life) is controlled by its longer-lived precursor, Te-132 (78.2 hr).

<sup>2</sup>The method is not clear. Relative decay during RaLa processing may have been accounted for. The iodine inventories were calculated from the calculated Sr-91 and Sr-90 levels in a typical 2-day cooled MTR element, given by [Anderson et al. \(1959\)](#). Both calculations assumed one continuous 21.9-day irradiation at constant power in the element, which was not the case. Actually, the irradiation was interrupted several times for fuel element relocation, which would have changed the element power level and modified the results.

## 6.4 Recommended Daily Releases of Iodine-131

The recommended daily releases of I-131 from February 1, 1957 (Run 1) through December 30, 1959 (following Run 37), are given in an Excel spreadsheet in [Appendix F](#). The daily releases are based on the listings on the amended SMDs.

Special attention is needed for the releases for the week following the ICPP criticality accident on October 16, 1959. The October table in Appendix F records the stack monitor readings for I-131. In agreement with [DOE \(1991\)](#), it is concluded that the I-132 recordings were incorrect. The I-132 and I-133 releases for these dates were developed by a model described in Chapter 7.

As noted in [Chapter 5](#), there is some ambiguity in the corrections applied to the February 1957 releases. A different sort of sampler system was used in this first month of operation that attempted to record continuous releases using a large iodine trap feeding a holdup chamber ([Project Letters 1957](#)). However, it appears that the continuous feature was not used, and the iodine trap was counted in the static mode, identical to later sampler systems. The February corrections assume operation in the static mode.

[Tables 5-1](#) and [5-2](#) listed the emendations to the [SMDs](#), which add 156 Ci to I-131 releases for February 1957 and 13.2 Ci for March 1957.

## 6.5 Evaluation of Iodine-132 Release Data

### 6.5.1 Cases of High Iodine-132 Release Readings

Occasionally, some high daily releases of I-132, relative to I-131, are reported in the SMDs. Some illustrations are given in [Table 6-3](#), including the case of the ICPP criticality.

[Table 4-2](#) showed that the curie levels of I-131 and I-132 are roughly comparable in the feed. Therefore, all other things being equal, the observed release rates of these two radioiodines should have been approximately equal.

Note that most of the high I-132 releases occurred on the day of the dissolving and fall rapidly thereafter, as would be expected from its high decay rate relative to I-131. Comparing the May 1 and May 2, 1958, readings and the other consecutive day readings illustrate this. In particular, the October 16, 1959, reading is unusual, reporting an I-132 release 2,640 times higher than I-131 on the day of the criticality event, falling to a ratio of only 35 the day after. Chapter 7 in this report and [DOE \(1991\)](#) concur that the high I-132 release reported on the day of the criticality must be an error. What is not clear is whether or not the high I-132 readings on the day of the RaLa dissolvings are also errors.

**Table 6-3 Examples of reported high iodine-132 daily releases, in curies**

Date	Days after start of run	I-132 release	I-131 release	Ratio I-132/I-131
May 1, 1958	1	488	5.85	83
May 2, 1958	2	11.5	3.2	3.6
May 8, 1958	8	22.2	1.7	13
May 28, 1958	0	835	49.5	17
May 29, 1958	1	50	33.4	1.5
June 2, 1958	0	49	7.1	6.9
June 3, 1958	1	7.5	6.3	1.2
October 1, 1958	0	258	16.3	16
October 2, 1958	1	36.9	10.0	3.7
October 13, 1958	0	44.1	0.06	735
October 14, 1958	1	10.6	0.57	18.6
October 16, 1958	†	9,780	3.7	2,640
October 17, 1958		85	2.4	35

\* I-132 measurements began the next day.

† ICPP criticality accident. The recorded I-132 release for this date is an obvious error.

### 6.5.2 Possible Contributing Factors to the Observed High Iodine-132 Releases

The cause (or causes) of the reported high I-132 readings is not understood. The following may be contributing factors.

#### *Decay of Tellurium-132*

Iodine-132 is in near dynamic equilibrium with its precursor, Te-132. As such, no increase in I-132 curie level is possible due to further Te-132 decay. This could not be the source of the high readings.

#### *Unsteady Source Error*

As discussed in [Chapter 3](#), 2.3-hour half-life I-132 is quite sensitive to counting correction errors due to the long (24-hour) sampling period. A peak release at the start of the sampling cycle results would be **underestimated** by about a factor of 200 due to an underestimate of the decay, based on the release equation that assumes steady release. A peak release at the end of the sampling period would result in an **overestimate** of about a factor of seven due to an overestimate of the quantity decayed. Therefore, unsteady source errors will most frequently cause underestimates of I-132 releases. (Note: the October 16, 1959 criticality accident occurred at 3 AM. Therefore, measured I-132 releases would likely be underestimates.)

### *Impurity Effects*

As noted earlier, the 1957  $\gamma$ -spectroscopy system, based on NaI crystal scintillation, did not have the high discrimination of later systems. Count rates would include gamma contributions from about 10% on either side of the energy setting. Therefore, an impurity emitting near the principal I-132 peak at 668 or 773 keV would be recorded as contributing to I-132. A requirement of this hypothetical impurity is high volatility, because the high readings occur soon after dissolving (or solution boiling in the criticality) and become much lower the second day.

No such impurity has been identified.

### *Different Leakage Pathway Effect*

The long half-life of Te-132 sets I-132 apart from the other radioiodines, permitting, at least in principle, a different leakage pathway. For example, suppose most of the I-131 dissolves in the alkaline dissolver (as was originally thought), while the Te does not dissolve but proceeds to the centrifuge bowl, where it does dissolve in nitric acid. In such a case, I-132 would preferentially evolve in the centrifuge, where any leakage past the seals exited into the cell air space directly into the ventilation air, bypassing the scrubber. If this did occur, I-132 releases would be disproportionately high relative to I-131. The problem with this supposition is that data on iodine retention in the alkaline dissolver is contradictory, and data on tellurium retention in the dissolver is non-existent.<sup>3</sup> In addition, this supposition runs counter to the chemical evaluation of tellurium ([Appendix C](#)), which suggests it is indeed soluble in the hot alkaline solution. Nevertheless, observed I-132 releases seem to behave as if tellurium preferentially escaped dissolution in the alkaline dissolver.

### *Conclusion Regarding Contributing Factors to the High Iodine-132 Releases*

The high I-132 readings observed on the first day of a RaLa run must have been due either to an impurity enhancing the I-132 reading, or to a preferential leakage from the centrifuge to the cell air caused by the non-dissolution of Te in the alkaline dissolver. However, the latter reason could not have affected the high criticality release. An impurity effect may have contributed, but none has been found. Therefore, the cause of the high I-132 readings is not known.

### **6.5.3 *Extrapolating Iodine-132 Readings Back to February 1957***

The daily I-132 release data may be extended back to the start of runs in February 1957 from the initial date of May 1, 1958, if some generally typical I-132/I-131 release ratios can be found. The extrapolation would be based on the following expression:

$$R(\text{I-132}) = R(\text{I-131}) \times \text{ratio (I-132/I-131)} \quad \text{Eq. 6.1}$$

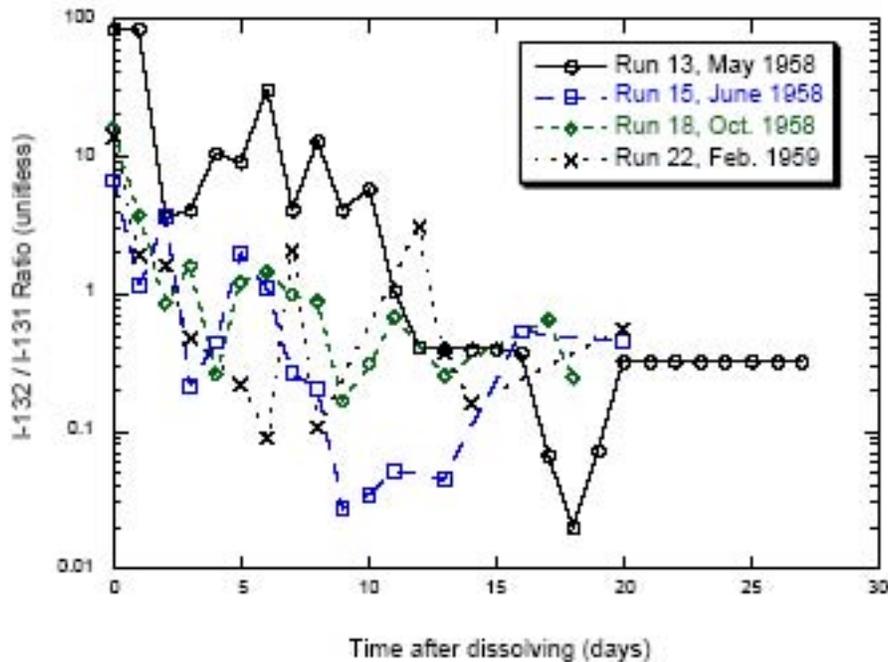
---

<sup>3</sup>However, [Legler et al. \(1957\)](#) states without reference or support that tellurium will dissolve in the alkaline dissolver.

Where:

- $R(I-132)$  = calculated daily I-132 release, Ci  
 $R(I-131)$  = observed I-131 release, Ci  
 Ratio = a correlation of observed I-132/I-131 releases

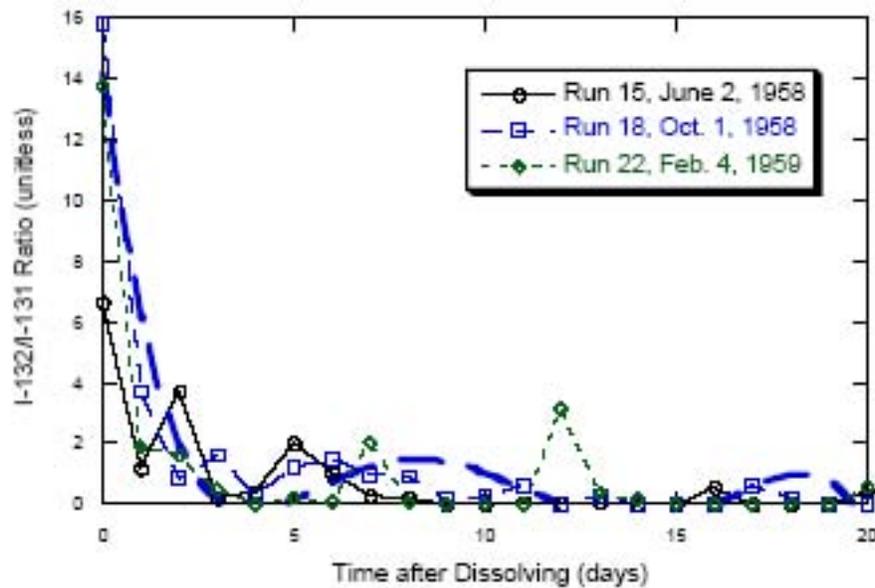
Figure 6-1 illustrates that I-132/I-131 release ratios were not always consistent. The first day of Run 13 showed an I-132 release 82 times higher than I-131, distinctly different from the other three runs shown. However, Runs 15, 18, and 22 appear to show similar I-132/I-131 release ratios.



**Figure 6-1 Observed iodine-132/iodine-131 release ratios for four runs**

The readings of activities for Run 13 produce anomalous I-132/I-131 ratios.

Figure 6-2 shows I-132/I-131 ratios in three RaLa runs (Runs 15, 18, and 22) in which the readings appear to be fairly stable. Note that the initial I-132/I-131 ratio is about 15 for two of the three cases, and about 6 for the third. All are initially higher than the 1 to 1 prediction based on input inventories, but fall roughly to the expected ratio after a few days.



**Figure 6-2 Observed iodine-132/iodine-131 release ratios for three consistent runs**

A fifth order polynomial curve fit is shown as a dashed line with no markers.  
The coefficients of the polynomial are given in the text.

A 6-constant polynomial fit is shown in [Figure 6-2](#).

$$I-132/I-131 = 14.16 - 10.199 t + 2.5735 t^2 - 0.28154 t^3 + 0.013786 t^4 - 0.00024805 t^5$$

Eq. 6.2

The term “t” signifies days following the dissolving, with t=0 for the day of the dissolving. The correlation coefficient for the curve fit is 0.96. After the initial peak ratio of 14.16, there are an additional two peaks, at 8 days and 18 days after dissolving, due to unknown process operations.

[Equation 6.2](#) was used to extrapolate the I-132 releases from their inception, May 1958, back to the initial date of operation, February 1, 1957. The daily releases are listed in the summary table in Appendix F.

## 6.6 Estimation of Iodine-133 Releases

No I-133 release data were taken, possibly because of difficulty with early radiochemistry techniques for making a standard  $\gamma$ -spectroscopy sample. There may not have been sufficient

incentive for the laborious “unfolding” technique<sup>4</sup> because of its lesser importance relative to I-131.

Since there are no precursor effects for either I-131 or I-133 (see [Chapter 3](#)), their release patterns should be similar. A release relation for I-133 may thus be based on the observed I-131 releases, accounting for the different inventory levels as follows.

$$R(I-133) = R(I-131) \times \text{Ratio (I-133/I-131)} \quad \text{Eq. 6.3}$$

Where:

$$\begin{aligned} R(I-133) &= \text{calculated daily release of I-133, Ci} \\ R(I-131) &= \text{observed daily release of I-131} \\ \text{Ratio} &= \text{inventory ratio of I-133/I-131 for each day} \end{aligned}$$

[Figure 6-3](#) shows the I-133/I-131 curie ratio as a function of days into the run for a “typical” MTR irradiation, taken again to be for the recreated Run 7, and for the nominal 2-day cooling period. As seen in [Figure 6-3](#), a curve fit for the isotopic ratio is given in the following equation:

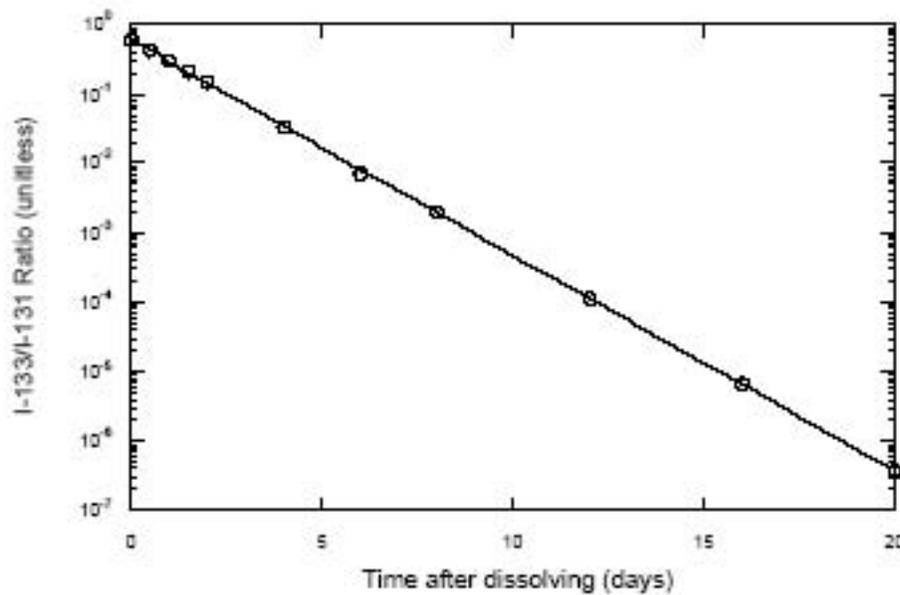
$$I-133/I-131 = 0.6003 * \exp(-0.7139 t) \quad \text{Eq. 6.4}$$

Where “t” is the decay time in days measured from the day of the dissolving, taking t = 0 for the day of the dissolving.

[Equation 6.3](#) was used to generate the I-133 release record given in [Appendix F](#), using curie ratios from [Eq. 6.4](#).

---

<sup>4</sup>In this method, the cpm readings are plotted for about a week, sufficient for complete I-132 and I-133 decay. This enables determination of I-131, which is then subtracted from the total, leaving the curve for the sum of I-132 and I-133. An additional subtraction of I-132 yields I-133.



**Figure 6-3 Iodine-133/iodine-131 ratio estimated for a typical Materials Testing Reactor irradiation**

(i.e., Run 7 using a nominal 2-day cooling period.)

The ratio decreases exponentially with time, starting from the day of dissolving.

## 6.7 The Complete Daily Release Record

[Appendix F](#) lists the I-131, I-132, and I-133 daily releases from February 1, 1957, the first day of RaLa operation, through December 30, 1959, the last day of the period of interest. RaLa run numbers, from Run 1 through Run 37, are given adjacent to the appropriate dates.

The [Appendix F](#) tables were prepared in the following way:

- The [SMDs \(Doc MC 60312\)](#) were used as the primary source.
- Omissions of I-131 releases in the [SMDs](#) were filled in per methods described in [Chapter 5](#). The average of adjacent dates was used for missing entries. The most significant omissions occurred in the first two months of operation, February and March 1957.
- The record of I-132 daily releases was extended back from May 1, 1958, the first entry in the datasheets, to the start of operation using the I-131 daily releases multiplied by a curve fit of I-132/I-131 releases for a stable run, using [Eqs. 6.3](#) and [6.4](#).

- The daily I-133 release record was calculated from the measured I-131 releases, correcting for differences in inventory (Eq. 6.4).

## 6.8 Comparison of the Recommended Releases with the Historic Dose Evaluation Study (DOE 1991)

Monthly releases from [Appendix F](#) are compared with the Historic Dose Estimation Study (DOE 1991) in [Table 6-4](#).

## 6.9 Fractional Releases per Run

A series of process modifications beginning in early 1957 had the effect of reducing iodine emissions. According to Bower and Buckham (1969) initial I-131 releases averaged about 1% of I-131 curies added to the dissolver. According to this reference, I-131 releases were reduced to 0.015% by March 1960 by a series of process improvements including the following.

Improved seals on the centrifuges	March 1957
Charcoal beds installed	August 1958
Reduced sampling	October 1958
Reduced centrifuge temperature	April 1959
Mercury salt added to a process vessel	May 1959

It should be emphasized that source inventory, process modification, and release data reported in [Bower and Buckham \(1969\)](#) are neither supported by any background information, nor are they referenced. As such, they cannot be verified.

The gradual downward trend of daily I-131 releases shown in the [Appendix F](#) tables supports [Bower and Buckham's](#) assertion of reduced fractional releases with time. However, determination of fractional releases per run from verifiable primary sources runs into great difficulty, for Runs 1-13, due to incomplete irradiation data, as reported in [Chapter 4](#). Further reduction in the available irradiation data for Runs 14 and onward makes estimation of the feed inventory impossible.

As much information as we now have (February 2003) on percentage I-131 releases per run are listed in [Table 6-5](#). As seen in the table, percentage releases of I-131 decreased from 1.22% for RaLa Run 2 to 0.52% for RALA Run 7, generally supporting [Bower and Buckham's \(1969\)](#) assertion of gradually reduced percentage release of I-131. Absolute releases are seen to have declined up to about April 1959, when they appear to have leveled out.

**Table 6-4 Comparison of iodine releases with historic dose evaluation  
(DOE 1991)**

	This Study (see <a href="#">Appendix F</a> )			HDE (DOE 1991)		
	I-131	I-132	I-133	I-131	I-132	I-133
<b>1957</b>						
<b>FEBRUARY</b>	661	2162	91	581		
<b>MARCH*</b>	97	16	0.03	0		
<b>APRIL</b>	76	56	1.7	81		
<b>MAY</b>	44	70	3.7	42		
<b>JUNE</b>	143	65	5.9	159		
<b>JULY*</b>	23	18	0.03	0		
<b>AUGUST*</b>	0.14	0	0	0		
<b>SEPTEMBER</b>	34	96	1.89	103		
<b>OCTOBER</b>	278	913	40	436		
<b>NOVEMBER*</b>	226	57	0	0		
<b>DECEMBER*</b>	2.5	0	0	0		
<b>TOTAL</b>	1584.64	3453	144.25	1402	4030	441
<b>1958</b>						
<b>JANUARY</b>	31	89	3.9	35		
<b>FEBRUARY</b>	75	129	5.5	155		
<b>MARCH</b>	130	137	3.7	56		
<b>APRIL</b>	49	173	7.9	35		
<b>MAY</b>	276	1544	44	288	1561	
<b>JUNE</b>	212	128	8.7	206	119	
<b>JULY*</b>	1.5	2.3	0	0	0	
<b>AUGUST†</b>	79	21	8.5	103	28	
<b>SEPTEMBER*</b>	25	7.6	0	0	0	
<b>OCTOBER</b>	110	804	21	116	809	
<b>NOVEMBER</b>	35	117	5	33	121	
<b>DECEMBER*</b>	3.8	7.3	0	0	0	
<b>TOTAL</b>	1027.3	3159.2	108.2	1027	3380	484
<b>1959</b>						
<b>JANUARY*</b>	0.25	1.3	0	0	0	
<b>FEBRUARY</b>	112	188	14	119	191	
<b>MARCH</b>	47	62	6.4	41	60	
<b>APRIL</b>	7.8	124	1.1	8.1	155	
<b>MAY</b>	3.1	44	0.41	2.3	14	
<b>JUNE</b>	2.4	65	0.49	2.1	65	
<b>JULY</b>	10	309	1.1	15.7	263	
<b>AUGUST</b>	3.8	64	0.4	2.1	61	
<b>SEPTEMBER</b>	8.1	55	0.56	9.4	67	
<b>OCTOBER‡</b>	15	125	127	10.3	46	
<b>NOVEMBER</b>	13	114	1.7	12.4	112	
<b>DECEMBER</b>	1.8	28	0.28	1.8	27	
<b>TOTAL</b>	224.25	1179.3	153.44	224.2	1061	99.8

\* No runs in month

† Charcoal absorbers installed

‡ [Appendix F](#) listing includes the October 16 criticality accident; DOE 1991 does not. I-132 and I-133 releases are calculated from release model.

**Table 6-5 Percentage of iodine-131 per run**

RaLa Run Number	Date	Decay Time (days)	I-131 Fed to Dissolver (Ci)	I-131 Release (Ci)	Iodine Release Percent
<b>1957</b>					
1	February 1-3	5 *		381	
2	February 20-21	2 *	2.31E+04	281	1.22
3	April 5-6	5 *		85.5	
4	May 19-20	7 *		42.1	
5	June 24-25	6 *	1.77E+04	158.8	0.90
6	September 11	2 †	2.17E+04	103.1	0.48
7	October 8	2 †	2.44E+04	126.2	0.52
8	October 21	2 †		81.9	
<b>1958</b>					
9	January 6	8 ‡		35.3	
10	February 12	2 †		154.6	
11	March 13-14	2 †		55.6	
12	April 16			34.9	
13	April 30-May 1	2 †		176.7	
14	May 28	2 †	§	111.2	
15	June 2	2 †	§	205.8	
16	August 6	2 †	§	27.7	
17	August 13	2 †	§	75.3	
18	October 1	2 †	§	70.6	
19	October 22	2 †	§	45.6	
20	October 22-23			no run	
21	November 12	2 †	§	32.6	
<b>1959</b>					
22	February 4	2 †	§	104.8	
23	February 25	2 †	§	14.7	
24	March. 18	2 †	§	40.5	
25	April 8	2 †	§	4.7	
26	April 29	2 †	§	3.4	
27	May. 19	2 †	§	2.3	
28	June. 10	2 †	§	2.1	
29	July 6-7	2 †	§	5.3	
30	July 21-22	2 †	§	6.4	
31	August 11-12	2 †	§	2.1	
32	September 1-2	2 †	§	4.5	
33	September 22-23	2 †	§	5.1	
34	October 13-14	2 †	§	14	
35	November 5-6	2 †	§	12.9	
36	November 30-December 1	2 †	§	0.4	
37	December 15-16	2 †	§	1.4	

\* Obtained from [Legler et al. 1957](#)

† Assumed to be the nominal 2-day cooling period

‡ Cited in [Stevenson 1958](#)

§ Inventory estimation not possible due to lack of irradiation data

## 6.10 Error Range of Releases

Dose reconstruction methodology calls for estimation of the error of releases, including the total range of the error and a probability distribution within the range. Rarely is sufficient information available to enable a firm error estimate. The source term analyst is asked to place a best judgment evaluation of such errors.

### 6.10.1 Error Range of Iodine-131 Releases

The factors affecting the correctness of the I-131 releases reported in the SMDs were discussed in Chapter 5. These factors included (1) a sampler efficiency different from the assumed 0.5, (2) iodine deposition in the line, (3) non-steady source in the stack, and (4) possible impurities in the sample emitting at the I-131 energy level. As discussed in Chapter 5, items (3) and (4) appear to have had a small effect on the I-131 readings.

#### *Sampler Efficiency*

A sampler efficiency of 50% appears to have been administratively selected from the start and assumed throughout. In comparison, measurements made on the “production sampler” from May through June 1957 varied from 30–64%, possibly averaging around 55%. (The error estimate is confused somewhat by numerous other measurements on temporary samplers, which showed a much larger range and generally higher efficiencies.)

The impact on the reported release of an efficiency (EFF) other than the assumed 50% is the following:

$$\text{Actual Release} = 0.5/\text{EFF} \qquad \text{Eq. 6.5}$$

The best judgment is that EFF had a constant probability between 0.45 to 0.6, a linearly diminishing probability from 0.6 up to a maximum of 0.7, and a linearly diminishing probability from 0.45 down to minimum of 0.3.

#### *Deposition in the Sampling Line*

Iodine deposition tests described in [Chapter 5](#) showed improbable results. The sampler at the end of a short line (45 feet) gave lower calculated releases than the one at the end of a long line (130 feet) in all nine tests that were conducted. This is opposite from the expected result.

However, in these tests, a measured sampler efficiency was used for the release determination for the short line, whereas a 50% efficiency was assumed for the production sampler. If equal efficiencies are assumed for both samplers, the results of the nine tests divide as follows: in three tests, the long line would show lower calculated releases, as should be expected; in the remaining six tests, the sampler at the end of the longer line would show higher releases. In sum, no sensible trend with sample line length was observed, although there must have been one.

Since no sampler line deposition was assumed in the release determinations, the sample line deposition error can only be one-sided. The recommendation is to assume a one-sided triangular

distribution with a maximum probability at 1.0 (i.e., no effect of sample line deposition), linearly decreasing to a maximum augmentation of 1.5, (i.e., an actual release 1.5 times higher than reported).

### **6.10.2 Error Range of Iodine-132 Releases**

As noted, reported I-132 releases are clouded by uncertainty due to anomalously high readings on the day of dissolving. (This is highlighted by the impossibly high reading recorded on the first day of the ICPP criticality accident discussed in [Chapter 7](#).)

If the I-132 readings are assumed to be valid, they would be subject to the same two errors as described above for I-131.

In addition, they would be subject to errors due to variable source due to the short half-life relative to the sampling duration. As discussed in this section, variable source errors on the average tend to under-predict I-132 releases, but can also result in over-predictions for high releases occurring near the end of the sampling cycle.

A reasonable recommendation for variable source error on the I-132 reading may be the following: a triangular distribution with a peak probability at 2-times enhancement, the minimum probability at 0.5-times reduction, the maximum enhancement at 10 times.

### **6.10.3 Error Range of Iodine-133 Releases**

All I-133 reported releases are based on a comparison with I-131, accounting for differences in inventory. Since I-133 precursor characteristics are similar, correlation errors should be small. Hence, such calculated I-133 readings would be subject to the same errors as assumed for I-131.

## **6.11 Summary of Chapter 6**

- The [SMDs \(Doc MC 60312\)](#) are the primary record of daily iodine releases from the RaLa process. Daily recordings for I-131 began February 1, 1957, and continued to the end of operations.
- Daily recording of I-132 releases began May 1, 1958. There were no I-133 measurements.
- Methods for correcting omissions in the record were described in Chapter 5. There is some uncertainty in the corrections applied to the first month of operation due to the different stack monitor system used at that time.
- The [Hayden Notegrams \(Doc MC 60111\)](#) are the first source of daily I-131 and I-132 releases that are in a reasonably documented form, namely internal letters. For I-131, the daily record begins July 7, 1957, about five months after daily releases were being recorded in the [SMDs](#).

- The releases reported in the Health and Safety Annuals (e.g., [AEC 1959](#)) are summaries expressed as releases per run. Though not referenced, the information was evidently based on the [Hayden Notegrams](#). This format was adopted with modifications by the Historical Dose Evaluation Study ([DOE 1991](#)).
- Evaluation of the high I-132 releases generally observed for the first day of a run did not come to a firm result. Two leading possibilities are either a radioactive impurity emitting at the I-132 energy or an effect due to a difference in solubility of the Te-132 precursor. However, the latter could not have affected the high I-132 release reported for the ICPP criticality on October 16, 1959.
- A correlation for I-132 daily releases was used to extend the I-132 release record from its inception in the [SMDs](#) on May 1, 1958, back to February 1, 1957. The correlation is based on observed I-132/I-131 release ratios as a function of time during the run, using several months of smooth operation.
- A correlation for I-133 releases based on I-131 releases and the inventory ratio I-132/ I-131 was used to estimate daily I-133 releases. The correlation is based on the observation that neither I-131 nor I-133 has significant precursor effects, so they should behave similarly.
- The results of these evaluations yield the daily record of I-131, I-132, and I-133 releases from February 1, 1957 to December 30, 1959, reported in [Appendix F](#).
- Monthly releases from [Appendix F](#) were compared with [DOE \(1991\)](#) in [Table 6-4](#). The I-131 releases compare closely as expected, since they are both based on the stack monitor data. The principal difference is the 182 Ci higher value of this study for 1957, obtained by filling in omissions in the February datasheet. In addition, the I-132 release estimates compare fairly well for 1957 and the initial portion of 1958, when data are absent. The I-133 release estimates were about 3 to 4 times lower than in [DOE \(1991\)](#), but in both cases, I-133 releases are low due to high decay of this short half-life isotope.
- Errors in the reported I-131 readings are judged to be caused by errors in the assumed sampler efficiency and of iodine deposition in the sample line. I-132 readings are in doubt due to the current inability to explain the consistently high readings on the day of the dissolving. An additional source of I-132 error is due to unsteady source. Errors in calculated I-133 releases should be the same as assumed for I-131. Some judgments on the probability distribution of these sources of error are given.
- The limited information currently available on fractional I-131 releases per run supports the view that releases decreased as process improvements were instituted. The percentage release of I-131 for Run 2 (February 1957) is estimated as 1.22% compared with 0.52% for Run 7 (October 1957).

## 7.0 CRITICALITY ACCIDENT, OCTOBER 16, 1959

### 7.1 Description of the Criticality Accident

#### 7.1.1 *Timing and Location*

In the early morning of October 16, 1959, an air sparging operation in a critically safe vessel in Cell B of the main process building caused an inadvertent transfer of highly enriched uranium solution to a waste tank that was not designed for that purpose. As a result, critical conditions were achieved in the waste tank, which later estimates showed to have resulted in about  $4 \times 10^{19}$  fissions over a period of several minutes.

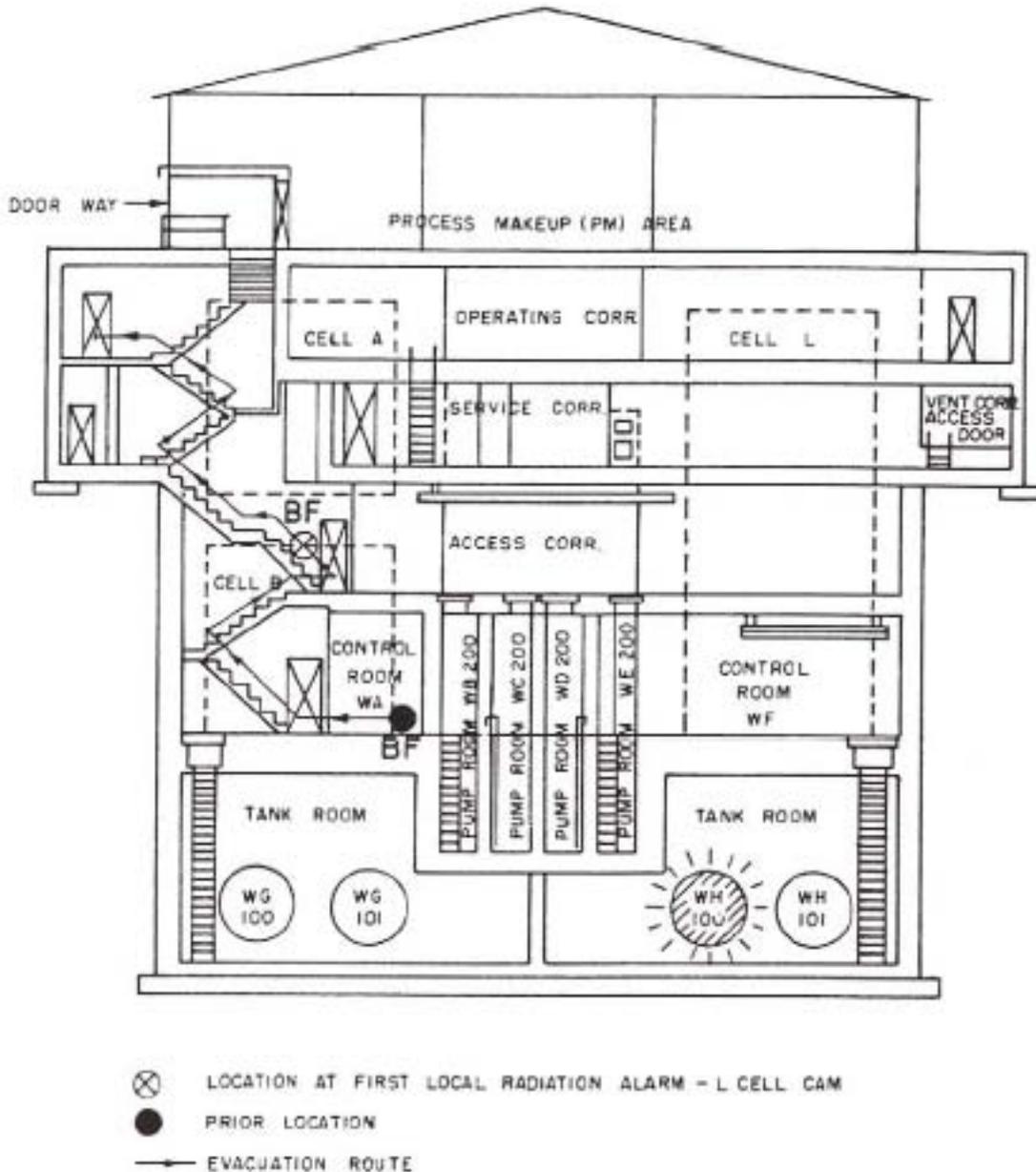
Figure 7-1, taken from the investigating committee's report of the incident (Ginkel et al. 1960), shows the location of the critically safe vessel in Cell B in the southwest corner of building CPP-601. The Process Equipment Waste (PEW) Tank (WH-100) in which the criticality occurred is in the lower level of the southeast corner of the building. Note that WH-100 is located directly below Cell L, the RaLa process cell. Figure 7-2 (Ginkel et al. 1960) shows the trace of the continuous air monitor (CAM) in the cell. Within the accuracy of the strip chart timer, the incident began at 2:50 AM. Three peaks are clearly shown in the next two minutes before the CAM became inoperative due to saturation. (The CAM was probably responding instantaneously to emitted neutrons and gamma rays.)

#### 7.1.2 *Material Transfers During and Post Event*

The severity of the event was estimated by measurement of molybdenum-99 (Mo-99) and barium-140 (Ba-140) fission product concentrations in the final solutions after dilution, poisoning, and vessel transfers. This method requires assurance that all the involved material is sampled for counting.

Figure 7-3 shows the piping interconnections between the waste tanks. WH-100, the criticality vessel, first of all shows a vent line connecting to the vessel off-gas system (VOG), as do all ICPP vessels. Therefore, at least a portion of any liquid or volatile fission products vaporized during the event should have almost immediately shown up in the stack for atmospheric dispersal. (See Figure 2-11 for an outline of the ICPP off-gas system). Note also the piping connections with the companion PEW waste tank, WG-101, as well as the two Cell Floor Drain (CFD) tanks. An additional possible escape route from the PEW tanks appears to be into the floor sump area, which is open to the cell air.

Another important feature of the event is the estimated quantity of heat liberated. At 200 Mev per fission,  $4 \times 10^{19}$  fissions liberates  $3.1 \times 10^5$  kcal heat, sufficient to vaporize about 560 liters of water. Therefore, evidence of vapor transfers should be expected. Fortunately, the connections to WH-100 provided sufficient pressure relief to avoid any physical failures. Ginkel et al. (1960) notes that a pressure rise "in excess of 5 psi" was recorded. The initial boiling of the critical solution would have evolved many short-lived noble gases, iodines, and bromines, a portion of which undoubtedly entered the VOG line directly.



**Figure 7-1 Cross section of the Process Building CPP-601 looking north from the waste tank rooms**  
 (Ginkel et al. 1960)

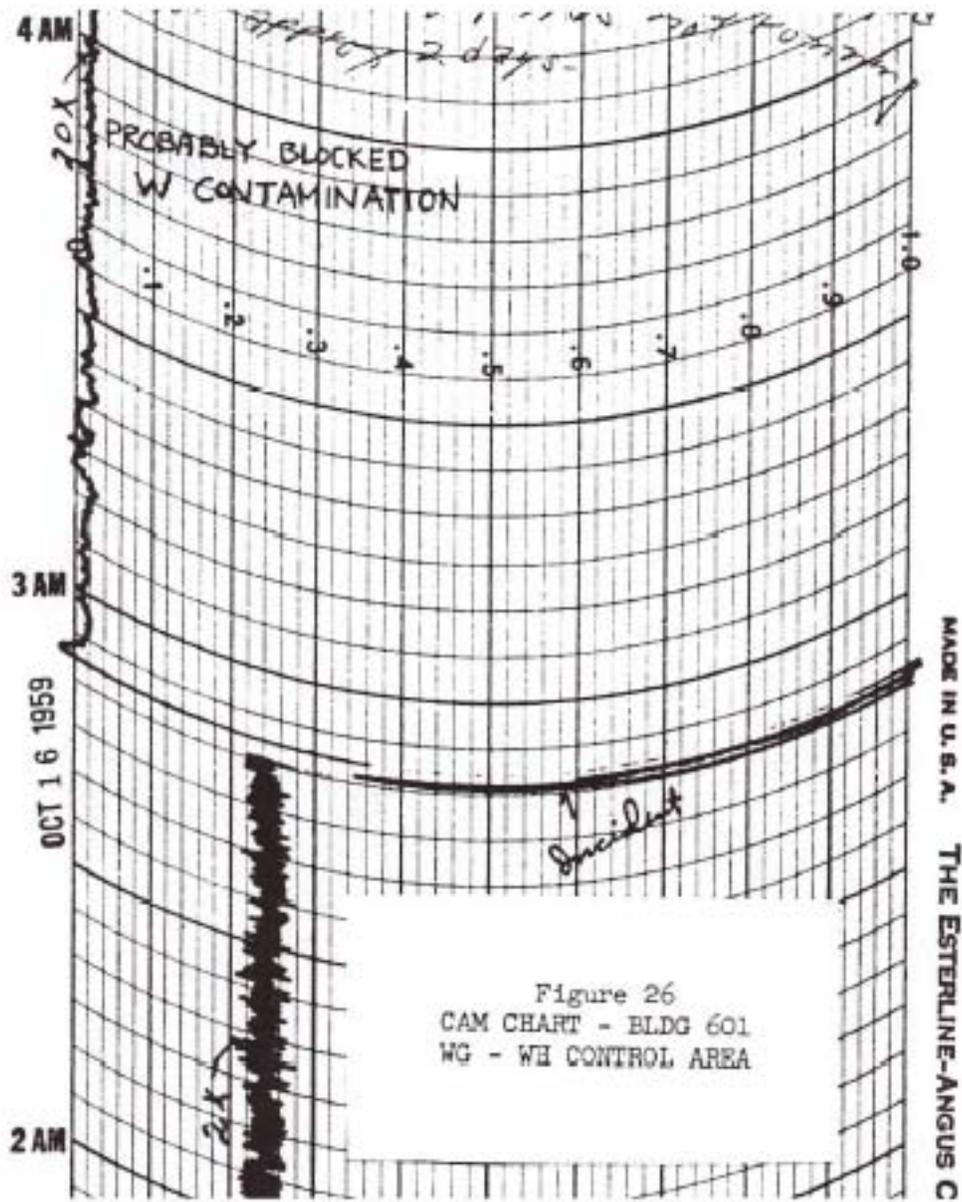


Figure 7-2 Continuous air monitor chart in the storage vessel room  
(Ginkel et al. 1960)



The solution transfers during and directly following the event may be summarized as follows (Ginkel et al. 1960).

Liquid in WH-100 prior to the event	600–800 liters
Liquid containing HEU transferred from Cell B to WH-100	201 liters
Liquid lost by WH-100 by boiling and pressure rise	900 liters
Liquid gained by WG-101 directly post-criticality	600 liters

Therefore, pressurization of WH-100 due to the liberated heat caused it to lose possibly all of its liquid contents, most of which showed up in WG-101. The transfer was affected by a combination of vapor and liquid flow through the steam jet line and back through the “diversion box,” which was evidently reset for flow into WG-101 by the pressure surge. Note that some 300 liters went elsewhere, some to the other two waste tanks, and perhaps the balance as vapor to the VOG line and the stack.

The important feature is that essentially all of the uranium was ultimately accounted for, indicating that the 300 liters of liquid apparently lost during the criticality was mainly vapor. The uranium – and presumably also the Mo-99, Ba-140, and La-140 on which the severity of the event was based – was left behind in the system, ultimately recovered by washing operations. Ginkel et al. (1960) gives the following uranium balance.

HEU in the affected Cell B vessels	88.6 +/- 0.5 kg
HEU from other sources	8.9 +/- 0.3 kg
Total to account for	97.5 +/- 0.8 kg
HEU accounted for	96.7 +/- 0.6 kg
HEU Lost	0.8 +/- 1.4 kg

### *Conclusion Regarding Material Lost*

At most, about 2% of the uranium, and hence the solid fission products, Mo and Ba, upon which the severity of the event was based, was lost.

## **7.2 Estimated Number of Fissions**

### **7.2.1 Radiochemical Analyses Results**

The severity of the event was estimated by counting the amount of key fission products in the final solutions. Ginkel et al. (1960) lists the following criteria for fission product selection.

- (1) Intermediate half-life. Too short a half-life would entail excessive decay correction; too long a half-life would confound the results with other material in the waste tank.
- (2) No volatility, nor any volatility of a prominent precursor.
- (3) Well-known and high yield.

## (4) Existing capability for radiochemical purification.

Molybdenum-99 fits all the criteria, with Ba-140 and La-140 close behind. (However, the mass-140 fission products have 13.6-second xenon precursor.)

A requirement of this method is that all the final solutions be known. The significance of the good material balance on uranium, within 2%, indicates that the non-volatile fission products were completely contained in the final solutions, after the remedial transfers and dilutions with boric acid solution.

After several days of dilutions with boric acid solution and washings to recover as much uranium as possible, all the original solution from vessels WH-100 and WG-101 and the two CFD tanks ended up primarily in WH-100 (14,700 liters), with a smaller amount in Cell D (3,300 liters).

Another key requirement is that the samples from the final vessels be representative of the contents. Unfortunately, only one sample was taken from each of the final two solutions. Since a different degree of dilution occurred in each tank, it is not possible to judge the sampling error based on these two samples.

One sample was taken from WH-100 and one sample was taken from Cell D. The portion of the sample intended for Mo-99 analysis was subdivided into four aliquots from each vessel. Molybdenum was chemically purified from each of the eight aliquots and was read by  $\beta$ -counting in a calibrated geometry. Barium-140 and La-140 were similarly treated with a fewer number of aliquots. The total number of fissions determined from the Mo-99, Ba-140 and La-140 analyses are summarized below in [Table 7-1](#).

**Table 7-1 Total number of fissions determined from the molybdenum-99, barium-140, and lanthanum-140 analyses**

Mo-99, Ba-140 and La-140 analyses	Number of fissions
<b>Mo-99 analyses</b>	
WT-100 sample from 14,700 liters	$3.69 \times 10^{19}$
Sample from Cell D, 3,300 liters *	$3.16 \times 10^{18}$
Total fissions	$4.01 \times 10^{19}$
<b>Ba-140 analyses</b>	
WT-100 sample from 14,700 liters	$3.72 \times 10^{19}$
Sample from Cell D, 3,300 liters *	$4.03 \times 10^{18}$
Total fissions	$4.12 \times 10^{19}$
<b>La-140 analyses</b>	
WT-100 sample from 14,700 liters	$2.59 \times 10^{19}$
Sample from Cell D, 3,300 liters *	$4.36 \times 10^{18}$
Total fissions	$3.01 \times 10^{19}$

\* Listed as 3,000 liters in [Ginkel et al. \(1960\)](#), a typographical error.

Since the most aliquots were taken for the Mo-99 determinations,  $4 \times 10^{19}$  fissions appears to be the best estimate, with the Ba-140 and La-140 results as corroboration.

### ***7.2.2 Possible Errors in Estimating the Severity of the Event***

#### *Sampling Error*

Large errors could occur in sampling a large tank in which density stratification may occur. Despite this realization, only one sample was drawn from each of the two vessels containing the formerly critical solution. As a result, there is no available estimate of the sampling error.

Exhibit E in [Vance et al. \(1959\)](#) indicates that the sampling protocol for PEW vessels like WH-100 began with at least 30 minutes of agitation by means of air sparging.<sup>1</sup> Nevertheless, there is no way of knowing how effective this was in gaining a representative sample.

#### *Radiochemical Analysis Error*

The four duplicate Mo-99 analyses conducted on the sample drawn from WH-100 showed an average deviation from the mean of 4.5%. The difference between the Mo-99 and the Ba-140-based results was 2.5%. Therefore, the chemical analysis error was on the order of 2%-5%.

#### *Lost Solution Error*

As noted above, at most 2% of the uranium was not accounted for. Therefore, 2% is the maximum possible error caused by failure to sample all of the formerly critical solution.

#### *Decay Chain and Yield Errors*

The Mo-99 formation/decay chain is simple with only short half-life, non-volatile precursors. The mass-99 yield value used is the same as in a standard reference ([Voigt 1976](#)).

## **7.3 Leakage Pathways from the Idaho Chemical Processing Plant Building**

Referring to [Figure 7-3](#), undoubtedly the open 1-inch VOG vent line from each of the four waste tanks in the waste tank room was a major escape route for airborne activity. As shown in [Figure 2-11](#), the line leads to a VOG header in which air is drawn by blower in the Waste Disposal Building, CPP-604, at a rate of about 1,000 cfm. The flow may (or may not) pass through a filter in CPP-604 on its way to the stack. The delay time between entering the VOG system and emission from the stack is short, probably less than one minute.

Emissions from the VOG system as a result of the criticality event should be expected to peak at certain times. The VOG system undoubtedly provided pressure relief for the initial pressurization of the waste vessel system due to boiling in WH-100. In addition, each sampling

---

<sup>1</sup>Such sparging was an effective means for evaporating volatiles from the liquid and moving them through the VOG line. Sampling activities continued at least through October 18. Therefore, iodine and noble gas release can be expected to have continued at least through October 18.

operation, preceded by 30 minutes of air sparging, should have caused a peak in the release, as well as liquid transfers impelled by air or steam pressure between vessels. Such liquid transfer and sampling operations continued at least through October 18.

In addition, radioactivity probably entered the cell air system as evidenced by traces recorded by the continuous air monitors (CAMs) located throughout the building (Ginkel et al. 1960). As seen in Figure 2-11, the building air is drawn by a blower in building CPP-631 directly to the stack at a rate of approximately 100,000 cfm. Lag time between radioactivity appearance in the cell air system and emission from the stack should be quite short.

However, several of the CAMs were probably responding to direct emissions from the critical vessel and not to air contamination. Other CAMs may have been activated by radioactivity in air from the stack, drawn into the building. So the CAM records do not present a clear picture. The only route for building air contamination appears to have been via the piping leading to the open floor sump system.

## 7.4 The Stack Monitor Record Following the Accident

### 7.4.1 The Iodine-131 and Iodine-132 Record

Table 7-2 shows the daily release record of I-131 and I-132 (Stack Monitor Datasheets (SMDs), or Hayden Notegrams) from October 13, 1959, the date of the start of RaLa Run 34 and three days prior to the criticality, through October 23, 8 days following the criticality.

**Table 7-2 Daily release record of iodine-131 and iodine-132 from October 13–23, 1959**  
(Stack Monitor Datasheets)

Date	I-131 Ci/day	I-132 Ci/day
October 13*	0.06	44.1
October 14	0.57	10.6
October 15	0.4	5.0
October 16†	3.73	9,780‡
October 17	2.44	85.1
October 18	0.97	5.9
October 19	1.01	4.8
October 20	1.15	6.5
October 21	0.94	5.6
October 22	0.87	0.5
October 23	0.18	0.7

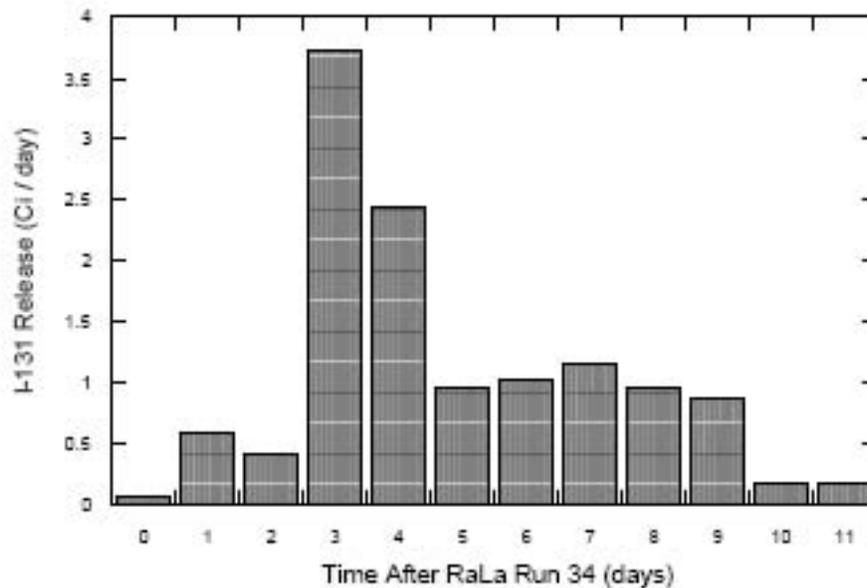
\* Start of RaLa Run 34.

† Date of criticality (2:50 am).

‡ Clearly an error. The 44.1 Ci RaLa release on October 13 must also be suspect. Post-criticality daily releases of I-132 will be calculated based on a model.

It is seen by comparison with the isotope production in the incident from an ORIGEN calculation (Section 7.5) that the high I-132 reading on the date of the criticality is an obvious error. Until the cause of the error is identified, all of the I-132 readings are suspect.

Figure 7-4 is a bar graph of the daily I-131 releases beginning with October 13, the start of RaLa Run 34, extending through the criticality date, Date 3 in the figure, and the following 10 days. Figure 7-4 suggests that releases from the criticality continued for at least 7 days.



**Figure 7-4 Daily releases of iodine-131 following RaLa Run 34, October 13, 1959**  
 Criticality occurred on Day 3, October 16.  
 ([Stack Monitor Datasheets](#))

#### 7.4.2 Search for $\gamma$ -Interferences with the Iodine-132 Reading

The most likely cause of the erroneously high I-132 reading was a contaminant in the stack monitor sampler solution emitting at the  $\gamma$ -energy peak used for the analysis. A search was conducted for a possible contaminant, using ORIGEN2. None has been found.

## 7.5 Radionuclide Production and Decay

### 7.5.1 Radioiodine Production and Decay

In view of the short release times, more nuclides than were considered for RaLa routine releases come into play. The following reasonable but arbitrary selection criteria will be adopted: all nuclides with half-lives greater than 10 minutes and with quantity greater than 1 curie at 1 hour will be taken into account. (This rule will be stretched to include 3.0-minute Br-85 and 3.07-minute Kr-89 because of their high curie yield.)

Therefore, the following radioiodines, given in [Table 7-3](#), will be considered.

**Table 7-3 Summary of radioiodines considered in this study**

Radioiodine	Half-life	Maximum level, Ci
I-131	8.04 days	26
I-132	78.2 hours*	102.8
I-133	20.8 hours	588
I-134	52.6 minutes	763.9
I-135	6.61 hours	1930

\* Half-life of precursor Te-132

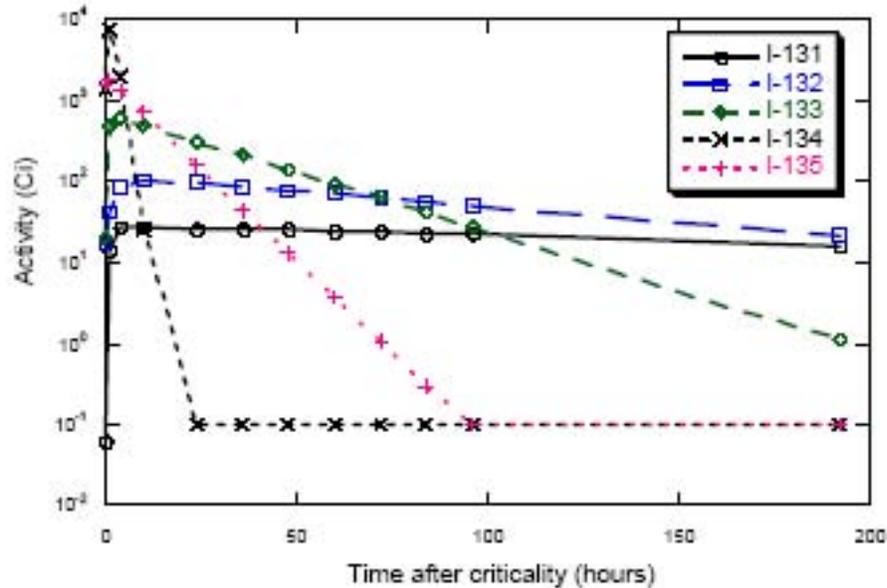
The maximum curie levels, obtained from ORIGEN2, do not always occur at 0-decay time due to precursor effects. Note that the effective half-life of I-132 is listed, i.e., the half-life of its long-lived precursor, Te-132.

The curie levels for these radioiodines are shown in [Figure 7-5](#) as a function of decay time after the criticality. They were calculated using ORIGEN2, assuming  $4 \times 10^{19}$  thermal fissions in U-235. [Appendix D](#) lists the ORIGEN output for all nuclides produced in the event, from which [Figure 7-5](#) was extracted.

Note that the maximum I-132 level in the event was about 100 Ci. Therefore, the 9,780 Ci release given in the [SMDs](#) for October 16 could not have happened.

[Figure 7-5](#) shows that I-134 levels reach nearly 10,000 Ci before falling to negligible levels at the end of the first day. I-135 peaks at about 2,000 Ci and requires about three days before falling to 1 Ci. Iodine-133 peaks at about 900 Ci before falling to 1 Ci after 8 days.

The curie levels, shown in [Figure 7-5](#), are the totals for each nuclide at a particular decay time. As such they include the unreleased quantities still in vessels plus the amount that has been released to the air. The source term, i.e., the release rate as a function of time, must be based on a model that predicts the release rate from the quantity remaining in the tank as a function of time.



**Figure 7-5 Inventory of radioiodines produced during the criticality and its evolution with time for 8 days after the criticality**

### 7.5.2 Radiobromine Production and Decay

Bromine is chemically similar to iodine and hence is evolved from acid solutions like the waste tank at the same rate as iodine. The radiobromines are short-lived and generally have negligible impact compared with the radioiodines, even with short decay times, like the 2-day decay of the RaLa elements. However, the rapidity of a criticality accident may alter these generalities.

Radiobromine production and decay after the criticality is shown in [Figure 7-6](#). Note that several radiobromines peak quite high and rapidly decay. However, 31.8-minute half-life Br-84 is fairly persistent, showing a significant presence out to about 10 hours.

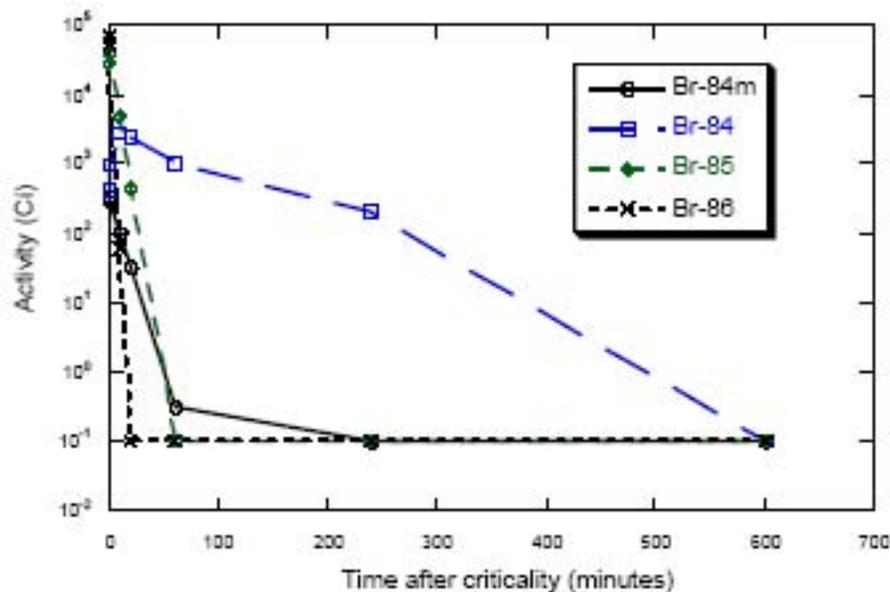


Figure 7-6 Inventory of radiobromines as a function of time after criticality

## 7.6 The Post-Criticality Iodine Source Term

### 7.6.1 Iodine Release Model

The approach taken is to use the I-131 releases recorded by the stack monitor for October 16, 1959, to determine a release rate constant,  $K$ , which will then be used to determine release rates for the other radioiodines.

The post-criticality release rate will be assumed to be proportional to the quantity of the isotope remaining in the liquid as follows:

$$S(t) = K I_L(t) \quad \text{Eq. 7.1}$$

Where:

- $S(t)$  curies/minute = the instantaneous source term
- $K$  fraction/minute = the release rate constant
- $I_L(t)$  = the curies of an iodine isotope remaining in the liquid at time  $t$ -minutes following the criticality

In general, the expression for  $I_L(t)$  is quite complex, since it must account for production due to precursor decay, radioactive decay, and evaporation at prior times. However, for cases where the precursors are fairly short-lived, losses from the liquid would be due to the first order processes of evaporation ( $K$ ) and decay ( $\lambda$ ), leading to the expression,

$$I_L(t) = I_{L0} \exp(-(K+\lambda)t) \quad \text{Eq. 7.2}$$

where  $I_{L0}$  is the initial curies in the liquid, which is also the initial total curies since evaporation has not yet occurred.<sup>2</sup> Therefore,  $I_{L0}$  may be obtained from an isotope code such as ORIGEN2.

The total release over all time is obtained by integrating  $S(t)$  from zero to infinite time, using Eq. 7.2 for the quantity in the liquid.

$$\text{Total Release (Ci)} = K I_{L0}/(K + \lambda) \quad \text{Eq. 7.3}$$

Note that Eq. 7.3 predicts that the total release of long-lived nuclides (small  $\lambda$  in comparison to  $K$ ) equals  $I_{L0}$ , in accordance with intuition. At the other extreme, short-lived nuclides will release only fraction  $K/\lambda$  of  $I_{L0}$  because of rapid radioactive decay.

It will be seen below that the INEL Historic Dose Evaluation Study (DOE 1991) assumed total release of all xenons and kryptons, including some with quite short half-lives. Accounting for decay prior to release by the factor  $K/(K+\lambda)$  from Eq. 7.3 would have a significant effect for some of the nuclides. Daily releases may be obtained by integrating Eq. 7.1 over each day:

$$\text{REL} - i = \int_{t_1}^{t_2} S(t) dt = \int_{t_1}^{t_2} K I_L(t) dt \quad \text{Eq. 7.4}$$

REL-i is the curie release for the i-th day, counting days from midnight to midnight in accordance with the stack monitor sampling schedule. Note that REL-1, the time span for the first day, extends from  $t_1 = 0$  to  $t_2 = 21$  hours, since the criticality occurred at 3:00 AM. Integrating Eq. 7.4 results in the following:

$$\text{REL} - i = \left( \frac{K I_{L0}}{K + \lambda} \right) \left( \exp(-(K + \lambda)t_1) - \exp(-(K + \lambda)t_2) \right) \quad \text{Eq. 7.5}$$

Where  $t_1$  and  $t_2$  are the initial and final times of the particular period, respectively, in consistent units.

Equation 7.5 may be used to evaluate  $K$  by applying it to the measured I-131 for the first day. For this case:

$$\text{REL-1} = 3.73 \text{ Ci}, I_{L0} = 26 \text{ Ci}, t_1 = 0, t_2 = 17 \text{ hr}^3 \quad \text{Eq. 7.6}$$

<sup>2</sup>Accuracy may be improved by adding terms to account for production from precursor decay.

<sup>3</sup>The final time,  $t_2$  is taken as 17 hours instead of 21, because the maximum in  $I_L$  occurred at approximately 4 hours due to relatively slow production from its precursor.

which leads to the following:

$$K = 1.52 \times 10^{-4} \text{ fraction/min} \quad \text{Eq. 7.7}$$

### 7.6.2 Measured Iodine-131 Releases Due to the Criticality

Releases of I-131 for 8 days following the event as given by the SMDs or the Hayden Notegrams will be attributed to the criticality. Some of this release was undoubtedly due to RaLa Run 34, which began on October 13, 1959. However, Figure 7-4 shows that I-131 releases following Run 34 (starting on day number 0 in the chart) were small compared with the jump occurring on Day 3, October 16, 1959, due to the criticality.

The span of time of 8 days attributed to the criticality may also be based on the observation that significant amounts of I-131 and I-132 produced in the criticality remained in the liquid after 8 days due to relatively slow release and decay.

Iodine-131 releases from the SMDs are listed in Table 7-4. A portion of the releases shown in the table may be attributed to RaLa Run 34, which began on October 13. However, the trace shown in Figure 7-4 indicates that the portion due to RaLa was fairly small.

**Table 7-4 Iodine-131 releases due to the criticality accident**  
(Stack Monitor Data)

Date	I-131 release, Ci
October 16*	3.73
October 17	2.44
October 18	0.97
October 19	1.01
October 20	1.15
October 21	0.94
October 22	0.87
October 23	0.18
<b>TOTAL</b>	<b>11.3</b>

\* Criticality began at about 2:55 AM

### 7.6.3 Releases of the Other Radioiodines

Releases per day for I-132, I-133, I-134, and I-135 may be obtained from Eq. 7.5 using the initial quantities, the loss rate coefficient, K, the decay constant and the time interval, as given in Section 7.6.1. The results are given in Table 7-5.

**Table 7-5 Releases per day (Ci) of iodine-132, iodine-133, iodine-134, and iodine-135 following the Idaho Chemical Processing Plant criticality of October 16, 1959**

Date	I-132	I-133	I-134	I-135
October 16*	16.4	74.4	87	140
October 17	12.5	33.1	0	13.2
October 18	8.13	12.0	0	0.86
October 19	5.28	4.32	0	0.1
October 20	3.43	1.56	0	0
October 21	2.23	0.56	0	0
October 22	1.45	0.20	0	0
October 23	0.94	0.1	0	0
<b>Totals</b>	50.4	126	87	154
<b>Total, all radioiodines for the criticality event = 429 Ci<sup>†</sup></b>				

\* 21 hour day for the event beginning 3:00 AM

<sup>†</sup> Includes 11.3 Ci I-131 from [Table 7-2](#)

## 7.7 Radiobromine Release Following the Criticality

Three radiobromines fall into the nuclide selection criteria ([Table 7-6](#)).

**Table 7-6 Summary of radiobromine considered in this study**

Radiobromine	Half-life	Initial curies
<b>Br-83</b>	2.39 hours	33.8
<b>Br-84</b>	31.8 minutes	2,697
<b>Br-85</b>	3.0 minutes	5,024

The selection criterion was bent to include 3.0 minute Br-85, due to its large initial curie level.

Applying [Eq. 7.5](#) to the radiobromines yields the daily releases given in [Table 7-7](#).

**Table 7-7 Radiobromine releases (Ci) following the criticality**

Date	Br-83	Br-84	Br-85
October 16*	10.3	18.7	3.3
October 17	0	0	0
October 18	0	0	0
<b>Totals</b>	10.3	18.7	3.3
<b>Total, all radiobromines for the criticality event = 32.3 Ci</b>			

\* 21-hour day for the event beginning 3:00 AM.

## 7.8 Xenon Release Following the Criticality

The nuclide selection criteria includes the following five xenon nuclides (Table 7-8).

**Table 7-8 Summary of radioxenons considered in this study**

Radioxenon	Half-life	Initial activity (Ci)
Xe-133	5.29 days	76
Xe-135m	15.6 minutes	1,414
Xe-135	9.08 hours	634
Xe-137	3.83 minutes	1.508 x 10 <sup>5</sup>
Xe-138	14.1 minutes	5.234 x 10 <sup>4</sup>

This selection rule is again bent for Xe-137 due to the high initial curies.

The three most persistent xenon nuclides produced in the criticality, Xe-133, Xe-135m, and Xe-135, are shown in Figure 7-7, as taken from Appendix D data. These are total amounts, including the liquid and air at any particular time. Two intermediate lived xenons are shown in Figure 7-8, Xe-137 and Xe-138.

Note that Xe-133 production and decay is not well fit by the assumption of Eq. 7.3 due to slow buildup from its precursors. Therefore, the Day 1 release estimate for Xe-133 should be higher than actually incurred. Subsequent days should be underestimated, approximately compensating for the error.

The release rates for the xenons (and kryptons) should be higher than for the iodines due to their chemical inertness; i.e., the rate of release factor, K, should be higher. There is no solid way of knowing how high it should be. There is a slight clue from the Ba-140 determinations in the post-critical liquid. As noted above, essentially all of the Ba-140 was retained despite its earlier life as 13.6-second xenon. This indicates that K for xenon was less than about 0.03 fraction/minute, assuming 99% retention of Xe-140.<sup>4</sup>

An arbitrary selection is made of a K that is 10 times higher for xenons and kryptons than determined for iodine, as possibly representative of the higher noble gas release rate.

$$\begin{aligned}
 K(\text{xenons, kryptons}) &= 10 \times K(\text{iodines bromines}) \\
 &= 1.5 \times 10^{-3} \text{ (fraction/minute)}
 \end{aligned}
 \tag{Eq. 7.8}$$

Applying the method as used for the radioiodines with the higher release rate constant leads to the daily releases given in Table 7-9.

<sup>4</sup>A better determination of K could have been obtained by sampling the end solution for cesium-138, the 32.2 min. daughter of 14.1 min. Xe-138. In the extreme of highly rapid xenon evolution, no Cs-138 should have been detected.

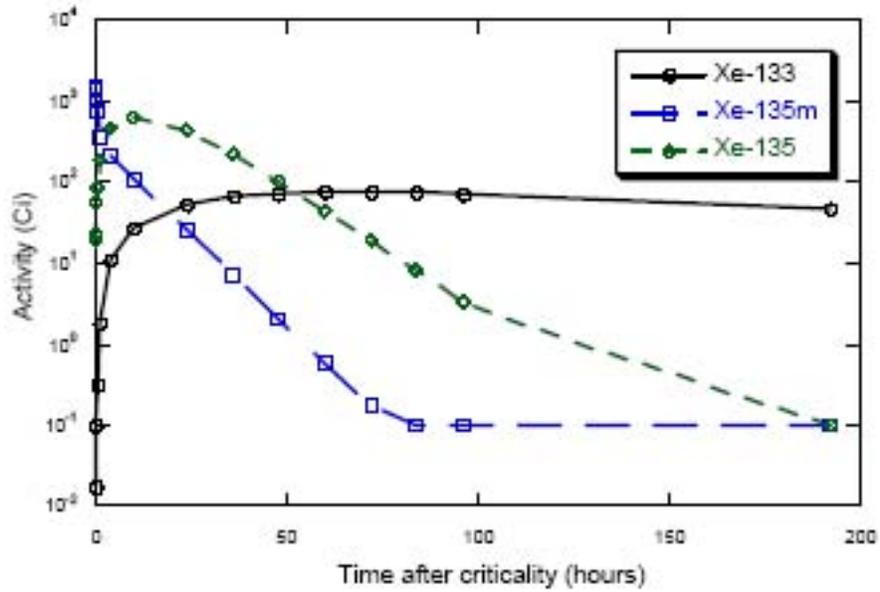


Figure 7-7 Inventory of long-lived xenons as a function of time after the criticality

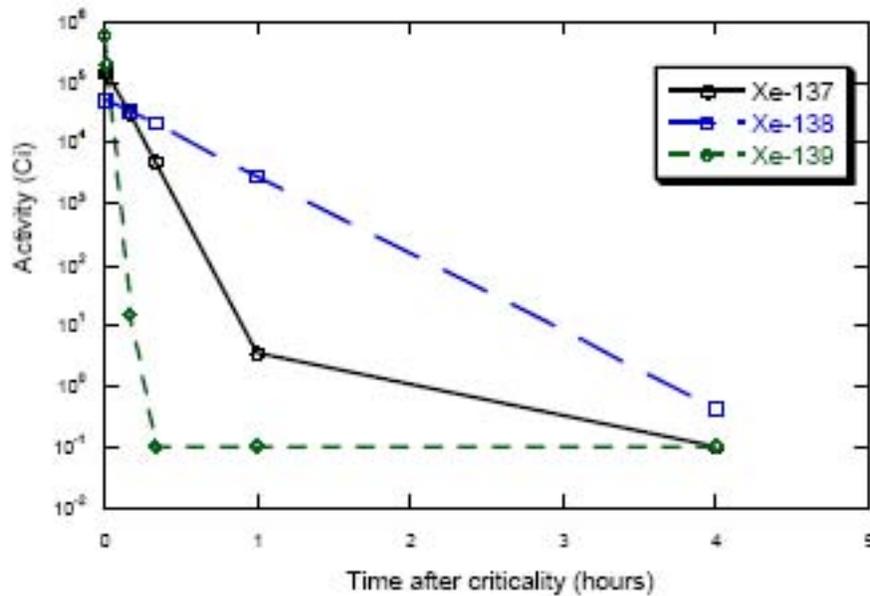
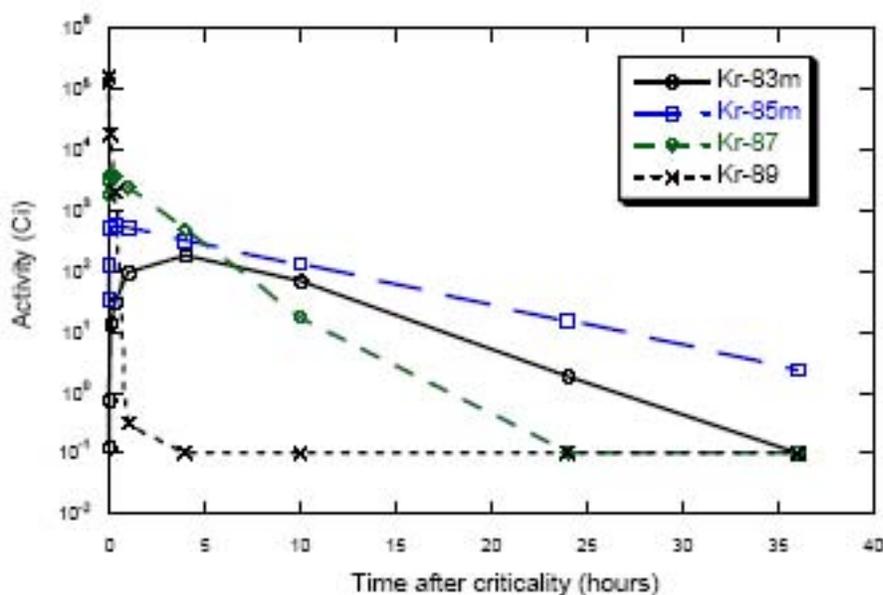


Figure 7-8 Inventory of intermediate-lived xenons as a function of time after the criticality



**Figure 7-9 Inventory of radiokryptons as a function of time after the criticality**

**Table 7-9 Releases per day (Ci) of xenon-133, xenon-135m, xenon-135, xenon-137, and xenon-138 following the Idaho Chemical Processing Plant criticality of October 16, 1959**

Date	Xe-133	Xe-135m	Xe-135	Xe-137	Xe-138
October 16*	62.0	46.7	335	1250	1570
October 17	8.48	0	10.1	0	0
October 18	0.84	0	0.2	0	0
October 19	0.1	0	0	0	0
October 20	0	0	0	0	0
October 21	0	0	0	0	0
October 22	0	0	0	0	0
October 23	0	0	0	0	0
<b>Total</b>	<b>71.4</b>	<b>46.7</b>	<b>345</b>	<b>1250</b>	<b>1570</b>

**Total, all radioxenons for the criticality event = 3,290 Ci**

\* 21 hour day for the event beginning 3:00 AM

Radioxenons have essentially zero dose factor for breathing or ingestion. In view of the rapid release during a criticality, their major impact was probably as a contributor to occupational dose due to immersion. [Table 7-10](#) lists several short-lived xenons that may have had this effect.

**Table 7-10 Short-lived xenons contributing to an occupational immersion dose**

Short-lived xenons	Half-life	Curies at zero decay time
<b>Xe-135m</b>	15.6 minutes	$1.41 \times 10^3$
<b>Xe-137</b>	3.83 minutes	$1.41 \times 10^5$
<b>Xe-138</b>	14.1 minutes	$5.23 \times 10^5$
<b>Xe-139</b>	39.5 seconds	$5.75 \times 10^5$
<b>Xe-140</b>	13.6 seconds	$5.80 \times 10^5$
<b>Xe-141</b>	1.73 seconds	$1.39 \times 10^5$
<b>Xe-142</b>	1.22 seconds	$3.97 \times 10^4$

## 7.9 Krypton Release Following the Criticality

The radiokryptons present to an extent greater than 1 curie after 10 minutes are shown in [Figure 7-9](#). Using the same method as for the xenons leads to the daily releases of kryptons following the event given in [Table 7-11](#).

**Table 7-11 Releases per day (Ci) of krypton-83m, krypton-85m, krypton-87, krypton-88, and krypton-89 following the Idaho Chemical Processing Plant criticality of October 16, 1959**

Date	Kr-83m	Kr-85m	Kr-87	Kr-88	Kr-89
<b>October 16*</b>	36.9	207	544	683	1030
<b>October 17</b>	0	1.12	0	0.6	0
<b>October 18</b>	0	0	0	0	0
<b>October 19</b>	0	0	0	0	0
<b>Totals</b>	36.9	208	544	684	1030
<b>Total, all kryptons for the criticality event = 2,500 Ci</b>					

\* 21-hour day for the event beginning 3:00 AM.

As for the xenons, several short-term kryptons may have had an impact on occupational dose. All those present after 1 minute to an extent greater than 100 Ci are listed in [Table 7-12](#).

**Table 7-12 Short-lived radiokryptons, greater than 100 Ci at 1 minute**

Radiokrypton	Half-life	Curies at 0-decay time
<b>Kr-87</b>	76.4 minute	$1.77 \times 10^3$
<b>Kr-89</b>	3.07 minute	$1.54 \times 10^5$
<b>Kr-90</b>	32.3 second	$6.19 \times 10^5$
<b>Kr-91</b>	8.6 second	$5.54 \times 10^5$

---

## 7.10 Comparison of Criticality Accident Releases with the Idaho National Engineering Laboratory Historic Dose Evaluation

### 7.10.1 Similarities/Differences in Approach

#### *Nuclide Selection*

This study selected all iodines, bromines, xenons, and kryptons with half-lives in excess of 10 minutes and with a greater than 1 curie presence 1 hour after the event. This rule was bent to include 3-minute Br-85 and 4-minute Xe-137 due to their high curie yields.

The DOE (1991) selection of iodines was identical to this study. DOE (1991), however, omitted 2.4-hour Br-83 initially present to a 103 Ci extent, and short-lived Br-85 initially present to a 5,024 Ci extent.

DOE (1991) also omitted 5-day Xe-133 (76 Ci) and short-lived Xe-137 (151,000 Ci).

#### *Source Term Definition*

This study defined the source term as the quantity of radioactive material in curies released from the stack.

DOE (1991) defined the source term for this event as the quantity released from the stack, and decayed by 0.52 hours, “the time calculated for the plume to reach the INEL site boundary.”

#### *Inclusion of Non-Volatiles*

This study excluded non-volatile materials in the source term, because all significant non-volatiles have quite short half-life volatile precursors. Therefore, they should not have vaporized from the critical solution.

DOE (1991) included the following non-volatile daughters of kryptons in the source term: Rb-89, Sr-89, Sr-90, Sr-91, Sr-92, Y-91, Y-92, and Y-93. Presumably, this was based on 100% escape of the Kr precursors and birth in the atmosphere. This is not appropriate in a source term estimate for the following reasons.

- All of these non-volatiles have short-lived precursors. Very little of the 32-second Kr-90, 8.6 second Kr-91, or 1.8-second Kr-92 escaped.
- The birth of radioactive daughters in the atmosphere properly belongs in the atmospheric dispersion model, which would also determine the distribution and deposition of the daughters along the transport path.

DOE (1991) also included the following non-volatile daughters of the xenons: Cs-137, Cs-138, Ba-139, Ba-140, Ba-141, Ba-142, La-141, and La-142. The same comments made for the krypton daughters apply. Several are born from extremely short-lived xenons, and would not

have completely escaped. (Note that essentially all of the Ba-140 was found in the system by sampling.)

### *Release Model*

DOE (1991) assumed the I-131 release for the event was that recorded for October 16 in the Hayden Notegrams,<sup>5</sup> decayed by 0.52 hours. However, examination of the I-131 release record indicates that releases continued for about 7 days at a progressively diminishing rate.

Both the present study and DOE (1991) recognized that the I-132 releases reading in the SMDs (or the Hayden Notegrams) for October 16 were obviously erroneously high.

DOE (1991) assumed that releases of I-132, I-133, I-134, and I-135 were “the same fraction of the total produced in the criticality as measured for I-131.” Presumably, each was decayed by 0.52 hours. No mention is made of the release assumption for bromine.

This study adopts a release rate model based on the I-131 data for October 16. Applying this method to the other radioiodines leads to a variation in the fraction released due to their different decay rates. The same model was applied to the radiobromines.

DOE (1991) assumed that all the kryptons and xenons with 10-minute or more half-life produced in the event were completely and immediately released. As noted in Section 7.6.1, short-lived nuclides decayed to a greater or lesser extent depending on the relative magnitudes of the decay and release rates.

### **7.10.2 Comparison of Releases**

Table 7-13 lists the total bromine, krypton, iodine, and xenon total releases (i.e., for all days) from this study compared with DOE (1991, Table A.41).

The differences in estimates may be explained by differences in release assumptions. Some examples are the following:

- The higher I-131 release of this study is due to counting releases from 7 days following the event, compared with 1 day for DOE (1991). Iodine-131 should persist in the solution due to its 8.04-day half-life and relatively slow evolution rate.
- The higher I-132 release of this study is due to its persistence over several days due to the long-lived precursor as compared to a 1-day fractional release in DOE (1991).
- The difference in the Br-84 release appears to be largely due to the decay to the site boundary assumed by DOE (1991).

---

<sup>5</sup>DOE (1991) did not reference the Hayden Notegrams or the SMDs as the source of the I-131 release, but it must have been the case.

- The difference in Xe-138 release is due to decay before evolution dictated by the assumed release model, which overcomes the 0.53-hour decay time to site boundary assumed by DOE (1991).

**Table 7-13 Total (all days) halogen and noble gas release comparison: this study versus DOE (1991)**

Nuclides	Curies released	
	This Study	DOE (1991)
<b>Br-83</b>	10.3	*
<b>Br-84</b>	18.7	4.93
<b>Br-85</b>	3.3	*
<b>Kr-83m</b>	36.9	*
<b>Kr-85m</b>	208	162
<b>Kr-87</b>	544	782
<b>Kr-88</b>	684	930 <sup>†</sup>
<b>Kr-89</b>	1,030	*
<b>I-131</b>	11.3	3.72
<b>I-132</b>	50.4	11.8
<b>I-133</b>	126	78.3
<b>I-134</b>	87	584
<b>I-135</b>	154	254
<b>Xe-133</b>	71.4	*
<b>Xe-135m</b>	46.7	318
<b>Xe-135</b>	345	61.8
<b>Xe-137</b>	1,250	*
<b>Xe-138</b>	1,570	7,710

\* Not listed

† Includes all daughters

## 8.0 UNCERTAINTY ANALYSIS FOR THE ESTIMATED RELEASES OF IODINE FROM THE IDAHO CHEMICAL PROCESSING PLANT

An evaluation of the reliability of the stack monitoring system indicated that the main sources of uncertainty in the estimated releases of iodine are the efficiency of the liquid sampler, the losses of iodine due to deposition in the sampling line, and, for the short-lived I-132, the decay during the 24-hour collection time (Section 6.10).

These sources of uncertainty affect all normal Radioactive Lanthanum (RaLa) runs. During the October 16, 1959, accident, iodine was released from the RaLa vessels where the criticality occurred, was captured by the RaLa off-gas system and by the building ventilation system, and released through the Idaho Chemical Processing Plant (ICPP) stack where the sampler of the monitoring system was located. Since the monitoring system seems to have worked properly during the criticality accident, the sources of uncertainty in the readings of the monitoring system during routine RaLa releases are applicable to the releases during the criticality accident as well.

The sampler efficiency is a source uncertainty that brings a very small bias towards reducing the deterministic release estimates (Eq. 6.5, Section 6.10). On the other hand, the deposition in the sampling line introduces a bias towards increasing the deterministic releases by an uncertain factor with a central value of 15%.<sup>1</sup> Similarly, the decay correction necessary for I-132 increases the deterministic I-132 releases by an uncertain factor with a central value of 3.8 (Section 6.10).

The uncertainties in the sampler efficiency and in the deposition in the sampling line reflect the lack of knowledge about the true value of these parameters, and they are applied correlated for each day of release. The decay correction for I-132 is independent for each day of release, since a peak release of I-132 in each day (e.g., due to movement of the liquid from one vessel to another) could have occurred at any time during the 24-hour collection time, and at different times in different days. Thus, the uncertainty in the decay correction factor was applied as an independent random variable for each day of the release. Latin Hypercube sampling methods were used to propagate the uncertainties.

For I-131 and I-133, an overall uncertain bias correction factor can be defined as  $[(0.5/EFF) \times (\text{Triangular}(1.0, 1.0, 1.5))]$ , where (0.5/EFF) is the sampler efficiency correction (Eq. 6.5) and the triangular probability distribution function is the bias correction due to the deposition in the sampling line (Section 6.10). The unitless bias correction factor has a central value of 1.14 and a 95% confidence interval of 0.8 to 1.8. This factor multiplies the deterministic estimates of the I-131 and I-133 daily releases and is applied correlated for each day of releases.

---

<sup>1</sup> In Section 7.10, the correction of the deposition in the sampling line is defined as triangular probability distribution function with a minimum value and the mode of 1.0, and the maximum value of 1.5 (Triangular(1.0,1.0,1.5)). This distribution has a 50<sup>th</sup> percentile of 1.15 (or 15% increase) and a mean value of 1.17 (or an increase of 18%).

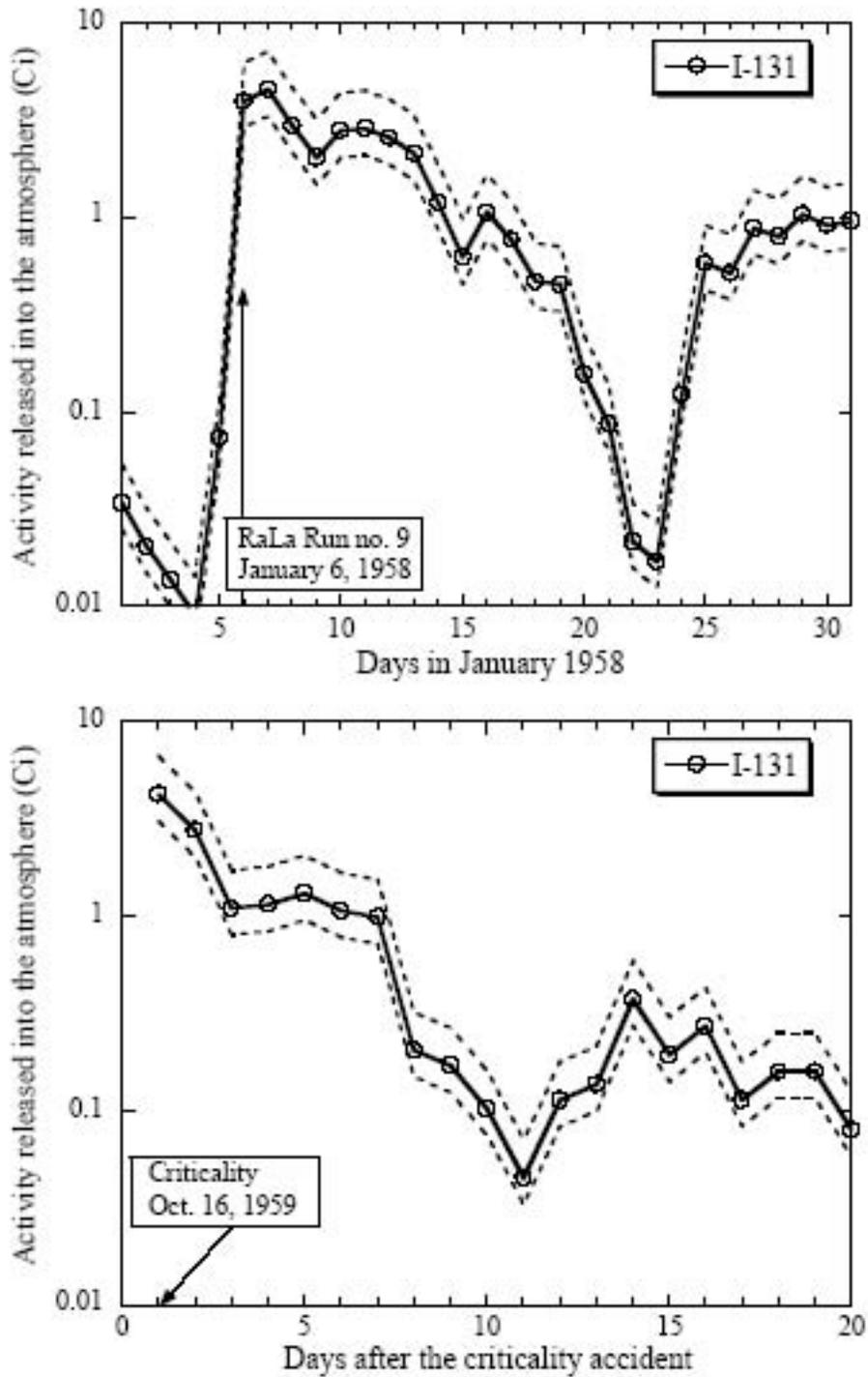
For I-132, an overall uncertain bias correction factor can be defined as  $[(0.5/EFF) \times (\text{Triangular}(1.0,1.0,1.5)) \times \text{Triangular}(0.5,2.0,10.0)]$ , where the first two terms are discussed above and the third term represents the uncertainty due to the decay of I-132 during the 24-hr collection time (Section 6.10.2). The bias correction factor for I-132 has a central value of 4.4 and a 95% confidence interval of 1.1 to 12.2, and it multiplies the deterministic estimates of the daily releases for I-132. It is applied so that the uncertainties in the sampler efficiency and in the deposition in the sampling line are correlated for each day of release, while the I-132 decay correction is independent for each day of release.

The total emissions of I-131, I-132 and I-133 from ICPP, adjusted by the uncertain bias correction factors presented above, are presented in Table 8-1 for the routine releases from RaLa (including the 1959 criticality accident) and for the criticality accident by itself. Figure 8-1 shows an example of the daily emissions of I-131 (adjusted by the uncertain bias correction factor) for the month of January 1958 and for the days after the criticality accident.

**Table 8-1 Estimated atmospheric releases of radioactive iodine from the Idaho Chemical Processing Plant during 1957–1959 (Ci)**

Isotope	Routine Releases (including October 16, 1959, criticality accident)		
	95% Confidence Interval		
	Lower bound	Central Estimate*	Upper Bound
I-131	2,400	3,200	5,100
I-131	25,000	37,000	58,000
I-131	340	470	730
October 16, 1959, criticality accident			
I-131	9.4	13	20
I-131	130	230	450
I-131	110	140	230

\* The 50<sup>th</sup> percentile of the probability distribution describing the uncertainty in the estimated releases. All values are rounded to two significant digits.



**Figure 8-1 Daily releases of iodine-131 during January 1958 and after the 1959 criticality accident, adjusted by the uncertain bias correction factor**

The solid line is the 50<sup>th</sup> percentile and dotted lines represent the 95% confidence interval.

---

## REFERENCES

- AEC. Annual Report of the Health & Safety Division. IDO-12014, Doc MC 52155; 1959.
- Anderson EL, Weech ME. Development of the RaLa Process for Installation in the ICPP. IDO-14307; July 8, 1954.
- Anderson EL, Weech ME. Status of Development of the RaLa Process as of April, 1954. IDO-14300; 1954.
- Anderson EL et al. Laboratory Development of a Process for Separating Ba-140 from MTR Fuel. IDO-14445; March 27, 1959.
- Ayers AL, Burns P. Waste Management at the ICPP. ANS Winter Meeting, San Francisco; December 12, 1960.
- Beahm EC. Te Behavior in Containment under LWR Accident Conditions. NUREG/ CR4338 and ORNL-TM-9726; February 1986.
- Beahm EC et al. Organic Iodide Formation Following Nuclear Reactor Accidents. NUREG/CR-4327; April 1985.
- Blanco RE. Laboratory Development of the MTR-RaLa Process for the Production of Barium-140. ORNL-1148; August 19, 1952.
- Bower JR, Buckham JA. Control of Fission Product Activity During Short-Cooled Fuel Processing Connected with the ICPP RALA Process. IAEA Panel on Reprocessing of Highly Irradiated Fuels; Vienna; May 27-30, 1969.
- Bower JR et al. Evaluation of Dissolver Off-Gas Treatment. PTR-261 EXPURGATED; November 27, 1957.
- Bradshaw RL, Cottrell WD. A Study of the Contribution of the RALA Process to Atmospheric Contamination at ORNL. ORNL CF 54-11-186; 1954.
- Briesmeister JF (ed). MCNP-A; General Monte Carlo N-Particle Transport Code 4C. LA-13709-M; 2000.
- Cederberg GK, Bower JR. Minimizing Gaseous and Particulate Activity in the ICPP Off-Gas. TID-7593; 1959.
- Cederberg GK, MacQueen DK. Containment of I-131 Released by the RaLa Process. R&D Report, AEC; IDO-14566; 1961.
- Chamberlain HV. Safety Analysis for Modifications of ICPP Dissolver and Vessel Off-Gas Systems. ICP-1011; October 1972.

---

Chizhikov DM, Shchastlivyi VP. Tellurium and the Tellurides (translated by E.M. Elkin). Collet's Publishers, London; 1970.

Cooper WC. Tellurium. Van Nostrand Reinhold, New York; 1971.

Coykendall JW. A portion of IDO Notebook #356 (pp. 1-41); January–June 1957.

CRC. Handbook of Chemistry and Physics (Weast, R.C. Ed.). CRC Press, Cleveland, Ohio; 1974.

Directory. Directory of Nuclear Reactors: Vol. II, Research, Test and Experimental Reactors. IAEA, Vienna; 1959.

Department of Energy (DOE). Idaho National Engineering Laboratory Historical Dose Evaluation. DOE/ID-12119 (Vol. 2: Appendices: Section A.2.2.4); 1991.

Doc MC 60312. Handwritten, unauthored, stack monitor data and calculation sheets, February 1957–May 1956. Evidently copied from three logbooks; 1957-1956.

Dyer FF. Personal communication to RP Wichner; 2001.

Eckerman KF et al. Limiting Values of Radionuclide Intake and Air Concentrations and Dose Conversion Factors for Inhalation Submersion and Ingestion. EPA—520/1-88-020; 1988.

Ginkel et al. Nuclear Incident at the ICPP on October 16, 1959. IDO-10035; February 15, 1960.

Girton RC et al. The Stack Monitoring System at the ICPP. ICP-1034; September 1973.

Green TE, Turley PR. Selenium and Tellurium in: Treatise on Analytical Chemistry, Part II, Vol. 7 (Kolthoff, I.M. and Elving, P.J. Eds.). John Wiley and Sons, New York; 1961.

Greenwood NN, Earnshaw A. Chemistry of the Elements. Pergamon, Oxford; 1984.

Handbook. Materials Testing Handbook in: USAEC Research Reactor Handbook. 1955.

Hanford Works Monthly Report. HW-13561-DEL; May 1949.

Hayden Notegrams. 1957-1961. Activity Discharged to the Atmosphere. Notegrams from R.E. Hayden. Doc MC 60111 or Doc MC 55203; February 1957- May 1961.

Health and Safety Division (H&SD). Annual Report of the Health and Safety Division, 1959. IDO-12014, October 1960; Doc MC 52155; 1960.

Holladay DW. A Literature Survey: Methods for the Removal of Iodine Species from the Off-Gases and Liquid Waste Streams of Nuclear power Plants and Nuclear Fuel Processing Facilities, with Emphasis on Solid Sorbents. ORNL/TM-6350; 1979.

- 
- Holm CH. Retention of Iodine in Process Solutions by Mercury Salts. HW-21103; June 1951.
- ICPP. Progress Report for July-September 1954. Technical Branch, ICPP. IDO-14337; 1954.
- Klienberg J, Cowan GA. The Radiochemistry of Fluorine, Chlorine, and Iodine. National Academy of Sciences, National Research Council. NAS-NS 3005; 1960.
- Kocher DC. Radioactive Decay Data Tables. DOE/TIC-11026; 1981.
- Kudryavtsev AA. The Chemistry and Technology of Selenium and Tellurium (translated from the 2<sup>nd</sup> edition and revised by E.M. Elkin). Collet's Publishers, London; 1974.
- Leddiccotte GW. The Radiochemistry of Tellurium. Nuclear Science Series, Subcommittee on Radiochemistry, National Academy of Sciences-National Research Council; Washington, DC; NAS-NS 3038; July 1961.
- Legler BM, Chase WL, Martin MD, Paige DM, Wrigley FK. Pilot Plant Development of a Process for Separating Barium-140 from MTR Fuel. AEC Research and Development Report, Phillips Petroleum Co., Atomic Energy Division, Contract No. AT (10-1)-205, Idaho Operations Office, U.S. Atomic Energy Commission; IDO-14344; September 1, 1955.
- Legler BM et al. Startup Operation of a Production Facility for Separating Ba-140 from MTR Fuel. IDO-14414; September 1, 1957. Doc MC-50197; 1957.
- MacFarlane RE, Muir DW. The NJOY Nuclear Data Processing System. LA-12740-M; 1994
- Merck. The Merck Index, Eighth Edition (Stecher, P.G. Ed.). Merck and Co., Rahway, NJ; 1968.
- MTR Progress Reports. 1957-1960. Material Testing Reactor Progress Reports in: Doc MC 50001. (Sanitized versions of classified documents of the "MTR Progress Reports," which were used in the analyses. Note that the reports were implemented by Phillips Petroleum Co, under Contract No. AT (10-1)-205, for the Idaho Operations Office, U.S. Atomic Energy Commission.)
- IDO-16312; Cycle No. 75, September 4 to 24, 1956.
  - IDO-16315; Cycle No. 76, September 25 to October 14, 1956.
  - IDO-16319; Cycle No. 77, October 15 to November 4, 1956.
  - IDO-16320; Cycle No. 78, November 5 to November 25, 1956.
  - IDO-16321; Cycle No. 79, November 26 to December 16, 1956.
  - IDO-16323; Cycle No. 80, December 17, 1956 through January 6, 1957.
  - IDO-16324; Cycle No. 81, January 7 to 28, 1957.
  - IDO-16328; Cycle No. 82, January 29 to February 18, 1957.
  - IDO-16336; Cycle No. 83, February 19 through March 10, 1957.
  - IDO-16369; Cycle No. 84, March 11 thru March 31, 1957.
  - IDO-16377; Cycle No. 85, March 11 thru April 21, 1957.
  - IDO-16381; Cycle No. 86, April 22, thru May 12, 1957.
  - IDO-16385; Cycle No. 87, May 12 through June 2, 1957.
  - IDO-16392; Cycle No. 88, June 3, thru 23, 1957.
-

IDO-16400; Cycle No. 89, June 24, thru July 15, 1957.  
IDO-16410; Cycle No. 90, July 16, thru August 5, 1957.  
IDO-16412; Cycle No. 91, August 6 through 25, 1957.  
IDO-16417; Cycle No. 92, August 26 through September 15, 1957.  
IDO-16424; Cycle No. 93, September 16, through October 5, 1957.  
IDO-16427; Cycle No. 94, October 6 through 28, 1957.  
IDO-16433; Cycle No. 95 and 96, October 29 through December 9, 1957.  
IDO-16438; Cycle No. 97, December 10 through 29, 1957.  
IDO-16439; Cycle No. 98, December 30, 1957 thru January 20, 1958.  
IDO-16444; Cycle No. 99, January 21, thru February 10, 1958.  
IDO-16447; Cycle No. 100, February 11, thru March 3, 1958.  
IDO-16450; Cycle No. 101, March 4 thru 24, 1958.  
IDO-16451; Cycle No. 102, March 25, thru April 14, 1958.  
IDO-16458; Cycle No. 103, April 15 thru may, 1958.  
IDO-16469; Cycle No. 104, May 6 thru 26, 1958.  
IDO-16476; Cycle No. 105, May 27 thru June 16, 1958.  
IDO-16478; Cycle No. 106, June 17 thru July 7, 1958.  
IDO-16495; Cycle No. 107, July 7 thru August 3, 1958.  
IDO-16496; Cycle No. 108 and 109, August 4 thru September 8, 1958.  
IDO-16500; Cycle No. 110, September 9 thru 29, 1958.  
IDO-16499; Cycle No. 111, September 29 through October 20, 1958.  
IDO-16509; Cycle No. 112, October 20 - November 10, 1958.  
IDO-16511; Cycle No. 113, November 10 - December 1, 1958.  
IDO-16515; Cycle No. 114, December 1 through December 21, 1958.  
IDO-16518; Cycle No. 115; December 21, 1958 - January 10, 1959.  
IDO-16521; Cycle No. 116; January 10 - February 2, 1959.  
IDO-16523; Cycle No. 117, February 2 through February 24, 1959.  
IDO-16529, Cycle No. 118, February 24 through March 15, 1959.  
IDO-16530, Cycle No. 119, March 15 through April 6, 1959.  
IDO-16533, Cycle No. 120, April 6 thru April 27, 1959.  
IDO-16541, Cycle No. 121, April 27 through May 18, 1959.  
IDO-16544, Cycle No. 122, May 18 through June 8, 1959.  
IDO-16546, Cycle No. 123, June 8 - June 28, 1959.  
IDO-16548; Cycle No. 124, June 29 through July 20, 1959.  
IDO-16556; Cycle No. 125, July 20 - August 9, 1959.  
IDO-16564; Cycle No. 126, August 8 through August 31, 1959.  
IDO-16565; Cycle No. 127, August 31 - September 20, 1959.

NAS-NA. The Radiochemistry of Tellurium. NAS-NS 3038. 1961.

Paige DM et al. Two Gas Cleaning Problems at ICPP Site. 5<sup>th</sup> AEC Air Cleaning Conference, Harvard Air Cleaning Laboratory; June 1958; TID-7551; 1958.

Pourbaix M. Atlas of Electrochemical Equilibria in Aqueous Solutions (translated by F.A. Franklin). Pergamon, Oxford; 1966.

Price W J. Nuclear Radiation Detection. McGraw-Hill Series in: Nuclear Engineering; 1958.

Project Letters. 1957a. A series of project letters dealing with early iodine releases and monitoring with various authors both in the Technical and the Health Physics Branches of the ICPP. Doc MC 60105.

Project Letters. Doc MC 60106. Some duplication with Doc MC 60105; 1957b.

Renier J-P, Groff AC. ORIGEN2. 1-A revised and updated version of the Oak Ridge Isotope Generation and Depletion Code. ORNL-5621; 1980.

Renier J-P, Alexander CW. An Advanced Version of the Oak Ridge Isotope Generation and Depletion Code with Fission Product Yields for 34 Actinides. ORNL-TM; To be published.

Quarterly Progress Reports of the Technical Section of the ICPP (QPRs ICPP).

IDO-14314; September 1954.  
IDO-14324; Second Quarter, 1954.  
IDO-14337; April 1, 1955.  
IDO-14350; August 12, 1955.  
IDO-14362; February. 6, 1956.  
IDO-14364; May 1, 1956.  
IDO-14383; September 1, 1956.  
IDO-14385; October 15, 1956.  
IDO-14391; January 15, 1957.  
IDO-14400; March 13, 1957.  
IDO-14410; July 18, 1957.  
IDO-14419; September 25, 1957.  
IDO-14422; December 31, 1957.  
IDO-14430; February 14, 1958.  
IDO-14453; June 1958.  
IDO-14444; (Supplement) July 10, 1958.  
IDO-14464; (Supplement) April 27, 1959.  
IDO-14471; March 1959.  
IDO-14488; (Supplement) September 21, 1959.  
IDO-14488; (Supplement) March 7, 1960.  
IDO-14509; September 1959.  
IDO-14512; June 20, 1960.

Rich BL. Letter to AL Ayers, June 10, 1957. File: BLR-8-57A, in: Doc MC 60106; 1957.

Shaffer VP. Design and Operations Manual for RaLa Processing L Cell. IDO-18004; 1955.

Shchastlivyi. Translated by E.M. Elkin, Collet's Publishers, London; 1970.

Smith DL, Bradford DJ. Characterization of the RaLa Off-Gas Cell, CP-631. WM-F181-010; May 1981. Also in: Doc MC-95195.

Stack Monitor Datasheets. Doc MC 60312; 1957-1967.

---

Stevenson CE. Supplemental Technical Report from January–March 1958, ICPP. IDO-14444; in: Doc MC 50078; 1958.

Till JE et al. Identification and Prioritization of Radionuclide Releases from the Idaho National Engineering and Environmental Laboratory. RAC Report 3-CDC, Task Order 5-2000 DRAFT.

Toth LM et al. The Chemical Behavior of Iodine in Aqueous Solutions up to 150°C, I. An Experimental Study of Non-Redox Conditions. NUREG/CR-3514; 1984.

US Department of Energy (DOE). Idaho National Engineering Laboratory Historical Dose Evaluation. DOE/ID-12119, Vol 2: Appendices, Section A.2.2.4; 1991.

Vance FP et al. Action of the ad hoc Accident Review Committee in the Event at the CPP of 0300 Hours October 16, 1959. Va-111-59A; 1959.

Voigt AF. Table of Fission Product Nuclides. Prepared for U.S. ERDA. IS-4052; 1976.

Weigel. Uranium in: The Chemistry of the Actinide Elements. Vol. 1 (Katz JJ, Seaborg GT, Morss LR, Eds.). Chapman and Hall, London; 1986.

Weinberg AM et al. The Materials Testing Reactor and Related Research Reactors in: Proc. of the International Conference on the Peaceful Uses of Atomic Energy, Vol. 2. Geneva; August 1955; United Nations; 1956.

Wichner RP. E-mail to Mauro J, Cohen S re: Tellurium Problem Statement. August 9, 2002.

Wichner RP. Personal communication. 2002b.

Wong MA, Roberts RS. Description of the Stack Monitoring System and Effluent Measuring Techniques at the ICPP. ENICO-1005; August 1979.

Wymer RG et al. Caustic Dissolution: Preliminary Studies. ORNL-1782; January 28, 1955.

## APPENDIX A

### REFERENCES: RALA PROCESS DESIGN AND OPERATION

#### A.1 Primary References

Note: No primary document on process operation and modifications was found after September 1957, including MTR irradiation data.

Anderson EL, Weech ME. Status of Development of the RaLa Process as of April 1954. IDO-14300; 1954.

Anderson EL, Weech ME. Development of the RaLa Process for Installation in the ICPP. IDO-14307; July 8, 1954.

Anderson EL et al. Laboratory Development of a Process for Separating Ba-140 from MTR Fuel. IDO-14445. March 27, 1959. (This work was probably performed before 1957. Publication was delayed.)

Bower JR et al. Evaluation of the Dissolver Off-Gas Treatment. PTR-261; November 27, 1957.

Chamberlain HV. Safety Analysis of the ICPP Dissolver and Vessel Off-Gas System. ICP-1011; 1972.

Girton RC et al. The Stack Monitoring System at the ICPP. ICP-1034; September 1973.

Hayden Notegrams. Daily releases of I-131, I-132 from the Health Physics Section of the ICPP. Doc MC 60111 or Doc MC 55203; March 1958–May 1961.

Legler BM et al. Pilot Plant Development of a Process for the Separation of Ba-140 from MTR Fuel. IDO-14344; also Doc MC-73478; September 1, 1955.

Legler BM et al. Startup Operation of a Production Facility for Separating Ba-140 from MTR Fuel. IDO-14414; also Doc MC-50197; September 1, 1957.

Project Letters. A series of project letters dealing with early iodine releases and monitoring with various authors both in the Technical and the Health Physics Branches of the ICPP. Doc MC 60105; 1957a.

Project Letters. A series of project letters dealing with early iodine releases and monitoring with various authors both in the Technical and the Health Physics Branches of the ICPP. Doc MC 60106. Some duplication with Doc MC 60105; 1957b.

QPRs ICPP, Quarterly Progress Reports of the Technical Section of the ICPP. These are of limited value for source term estimation. There is essentially no design or operational information. The following were acquired:

IDO-14314; September 1954.  
IDO-14324; Second Quarter, 1954.  
IDO-14337; April 1, 1955.  
IDO-14350; August 12, 1955.  
IDO-14362; February 6, 1956.  
IDO-14364; May 1, 1956.  
IDO-14383; September 1, 1956.  
IDO-14385; October 15, 1956.  
IDO-14391; January 15, 1957.  
IDO-14400; March 13, 1957.  
IDO-14410; July 18, 1957.  
IDO-14419; September 25, 1957.  
IDO-14422; December 31, 1957.  
IDO-14430; February 14, 1958.  
IDO-14453; June 1958.  
IDO-14444; (Supplement) July 10, 1958.  
IDO-14464; (Supplement) April 27, 1959.  
IDO-14471; March 1959.  
IDO-14488; (Supplement) September 21, 1959.  
IDO-14488; (Supplement) March 7, 1960.  
IDO-14509; September 1959.  
IDO-14512; June 20, 1960.

Shaffer VP. Design and Operations Manual for RaLa Processing L Cell. IDO-18004; 1955.

Smith DL, Bradford DJ. Characterization of the RaLa Off-Gas Cell, CP-631. WM-F181-010; May 1981.

Stack Monitor Datasheets. Doc MC 60312; 1957–1967.

Wong MA, Roberts RS. Description of the Stack Monitoring System and Effluent Measuring Techniques at the ICPP. ENICO-1005; August 1979.

## **A.2 Selected Secondary References**

Early development work at Oak Ridge reported by Blanco (1952) and early dissolver tests at Oak Ridge (Wymer, et. al., 1955) provide technical background to the Idaho RaLa work.

The dates and durations of Radioactive Lanthanum (RaLa) Runs 1 to 37 from February 1, 1957, to December 15, 1959, are given without reference in the Health and Safety Division Annual Report for 1959 (H&SD 1960). Since only dates are given, run times are accurate to +/- 12 hours. Also, the date for Run 21 is listed the same as Run 20, possibly a numbering error. This report also cites the first run in which the charcoal filters were added as an iodine control device. The information is not referenced.

A much-cited paper is [Cederberg and MacQueen \(1961\)](#), evidently intended as a conference paper. The paper provides general information on iodine retention in process solutions, process and scrubber solution changes, equipment modifications for improved iodine retention, and a broad description of the charcoal absorber. This is a highly useful paper, but dates are approximate, quantities are unverifiable, and key quantitative information is not referenced.

[Cederberg and Bower \(1959\)](#) reiterate much of the information in [Cederberg and MacQueen \(1961\)](#), but provide a rough sketch of the off-gas system. The diagram shows that the off-gas from all RaLa process vessels, including the dissolver and the centrifuges, may be routed to the dissolver off-gas (DOG), which passed through either the off-gas scrubber or into the vessel off-gas system (VOG), which bypasses the scrubber. The off-gas flow scheme is an important consideration, as discussed in this report.

[Bower and Buckham \(1969\)](#) show a crude flow sheet, which may or may not be completely correct, and a line diagram of the scrubber–charcoal absorber system. The paper gives, without reference, the curies of I-131 fed to the dissolver as averages for groups of five runs. The source of this information was not cited in the reference and we were unable to uncover additional information of this type. This information, however, would be useful to this project. They also cite Run 24 (March 18, 1959) as the first run in which the scrubber solution was changed to the acid–mercury composition, again without reference.

[Ayers and Burns \(1960\)](#) provide an informative discussion of the RaLa off-gas system.

[Smith and Bradford \(1981\)](#) describe the RaLa off-gas cell prior to decommissioning.

[Bower et al. \(1957\)](#) could be a useful primary document, but the available version is extensively expurgated.

[Wong and Roberts \(1979\)](#) describe a much later version of the stack monitoring system, but provide some insight to the 1957–1959 system.

[Chamberlain \(1972\)](#) describes process modifications in 1972, thereby providing some information regarding the earlier system.

**APPENDIX B****SAMPLE ORIGEN2 OUTPUT,  
MATERIALS TESTING REACTOR IRRADIATION  
FOR RALA RUN 2, 2-DAY COOLING TIME**

The following is a portion of ORIGEN2 output listing curies of fission products and activation products in the fuel element at discharge and at cooling times 6 hours, 12 hours, 18 hours, 1 day, 1.5 days, 2.0 days, 2.5 days, 3.0 days, 3.5 days, 4.0 days, 6.0 days, 8.0 days, 14 days, 18 days, and 22 days.

This is calculated for fuel element P-146, discharged from the Materials Testing Reactor (MTR) on February 18, 1957, and charged 2 days later in the dissolver for Radioactive Lanthanum (RaLa) Run 2. Thus, the RaLa charge corresponds to the 2.0D cooling column.









OUTPUT UNIT = 11 PAGE 59

ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 11/07/2002 at 00:22:10

\* Decay after Irradiation for MTR Case : ic02\_i34\_82c\_15 RECYCLE # 0 FISSION PRODUCTS

POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES

MTR Fuel Element

	Discharge	6.0HR	12.0HR	18.0HR	1.0D	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	6.0D	8.0D	10.0D	14.0D	18.0D	22.0D
NB 93M	9.91E-06	1.00E-05	1.01E-05	1.02E-05	1.03E-05	1.06E-05	1.08E-05	1.10E-05	1.12E-05	1.14E-05	1.16E-05	1.25E-05	1.33E-05	1.42E-05	1.59E-05	1.76E-05	1.93E-05
BR 94	3.55E-03	0.00E+00															
KR 94	3.56E+01	0.00E+00															
RB 94	1.42E+04	0.00E+00															
SR 94	6.63E+04	0.00E+00															
Y 94	7.11E+04	1.61E-01	3.40E-07	7.21E-13	1.53E-18	6.86E-30	0.00E+00										
ZR 94	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 94	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09	9.59E-09
NB 94M	1.18E-03	5.76E-21	0.00E+00														
BR 95	7.40E-05	0.00E+00															
KR 95	2.00E+01	0.00E+00															
RB 95	1.27E+03	0.00E+00															
SR 95	5.70E+04	0.00E+00															
Y 95	7.07E+04	3.50E+06	1.67E-16	7.97E-27	0.00E+00												
ZR 95	2.28E+04	2.28E+04	2.27E+04	2.26E+04	2.26E+04	2.25E+04	2.23E+04	2.22E+04	2.21E+04	2.20E+04	2.19E+04	2.14E+04	2.09E+04	2.05E+04	1.96E+04	1.88E+04	1.80E+04
NB 95	8.36E+03	8.43E+03	8.50E+03	8.57E+03	8.64E+03	8.77E+03	8.91E+03	9.04E+03	9.17E+03	9.29E+03	9.42E+03	9.89E+03	1.03E+04	1.07E+04	1.14E+04	1.20E+04	1.25E+04
NB 95M	1.41E+02	1.42E+02	1.43E+02	1.44E+02	1.44E+02	1.46E+02	1.47E+02	1.47E+02	1.48E+02	1.49E+02	1.49E+02	1.50E+02	1.49E+02	1.48E+02	1.48E+02	1.38E+02	1.33E+02
MO 95	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BR 96	5.41E-06	0.00E+00															
KR 96	8.66E+01	0.00E+00															
RB 96	2.16E+02	0.00E+00															
SR 96	3.51E+04	0.00E+00															
Y 96	6.62E+04	0.00E+00															
ZR 96	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 96	8.77E+01	7.34E+01	6.14E+01	5.14E+01	4.30E+01	3.01E+01	2.11E+01	1.48E+01	1.04E+01	7.25E+00	5.08E+00	1.22E+00	2.94E-01	7.06E-02	4.09E-03	2.37E-04	1.37E-05
MO 96	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
KR 97	3.25E-03	0.00E+00															
RB 97	7.13E+00	0.00E+00															
SR 97	6.34E+02	0.00E+00															
Y 97	3.12E+04	0.00E+00															
ZR 97	6.56E+04	5.13E+04	4.01E+04	3.13E+04	2.45E+04	1.50E+04	9.15E+03	5.60E+03	3.42E+03	2.09E+03	1.28E+03	1.79E+02	2.49E+01	3.48E+00	6.79E-02	1.32E-03	2.58E-05
NB 97	6.55E+04	5.49E+04	4.30E+04	3.36E+04	2.63E+04	1.51E+04	9.21E+03	5.63E+03	3.44E+03	2.11E+03	1.29E+03	1.79E+02	2.51E+01	3.50E+00	6.82E-02	1.33E-03	2.59E-05
NB 97M	6.21E+04	4.85E+04	3.80E+04	2.97E+04	2.32E+04	1.42E+04	8.67E+03	5.30E+03	3.24E+03	1.98E+03	1.21E+03	1.69E+02	2.36E+01	3.30E+00	6.43E-02	1.25E-03	2.44E-05
MO 97	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
KR 98	8.19E-01	0.00E+00															
RB 98	2.15E+00	0.00E+00															
SR 98	3.98E+03	0.00E+00															
Y 98	8.05E+03	0.00E+00															
ZR 98	6.15E+04	0.00E+00															
NB 98	6.34E+04	0.00E+00															
NB 98M	4.26E+02	3.35E+00	2.64E-02	2.07E-04	1.63E-06	1.01E-10	6.24E-15	3.86E-19	2.39E-23	1.48E-27	9.14E-32	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
MO 98	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TC 98	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09	2.19E-09
RB 99	6.35E-05	0.00E+00															
SR 99	4.28E+02	0.00E+00															
Y 99	1.01E+04	0.00E+00															
ZR 99	5.00E+04	0.00E+00															
NB 99	6.18E+04	0.00E+00															
NB 99M	4.49E+03	0.00E+00															
MO 99	6.45E+04	6.06E+04	5.69E+04	5.34E+04	5.01E+04	4.42E+04	3.90E+04	3.43E+04	3.03E+04	2.67E+04	2.35E+04	1.42E+04	8.59E				













OUTPUT UNIT = 11 PAGE 66

ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 11/07/2002 at 00:22:10

\* Decay after Irradiation for MTR Case : ic02\_i34\_82c\_15 RECYCLE # 0 FISSION PRODUCTS

POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDES TABLE: RADIOACTIVITY, CURIES

MTR Fuel Element

	Discharge	6.0HR	12.0HR	18.0HR	1.0D	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	6.0D	8.0D	10.0D	14.0D	18.0D	22.0D
I130M	1.06E+02	9.65E-11	8.77E-23	8.79E-35	0.00E+00												
XE130	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CD131	4.56E-01	0.00E+00															
IN131	3.70E+01	0.00E+00															
SN131	9.89E+03	0.00E+00															
SB131	2.83E+04	5.59E-01	1.09E-05	2.11E-10	4.09E-15	1.54E-24	5.85E-34	0.00E+00									
TE131	2.49E+04	8.88E+02	7.65E+02	6.66E+02	5.80E+02	4.39E+02	3.33E+02	2.52E+02	1.91E+02	1.45E+02	1.10E+02	3.62E+01	1.20E+01	3.94E+00	4.29E-01	4.67E-02	5.08E-03
TE131M	4.46E+03	3.90E+03	3.40E+03	2.96E+03	2.57E+03	1.95E+03	1.48E+03	1.12E+03	8.49E+02	6.44E+02	4.88E+02	1.61E+02	5.31E+01	1.75E+01	1.91E+00	2.07E-01	2.26E-02
I131	2.69E+04	2.65E+04	2.60E+04	2.55E+04	2.50E+04	2.41E+04	2.31E+04	2.22E+04	2.13E+04	2.05E+04	1.96E+04	1.66E+04	1.39E+04	1.17E+04	8.32E+03	5.89E+03	4.17E+03
XE131	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE131M	2.22E+02	2.23E+02	2.24E+02	2.25E+02	2.25E+02	2.27E+02	2.28E+02	2.28E+02	2.29E+02	2.29E+02	2.29E+02	2.26E+02	2.19E+02	2.11E+02	1.90E+02	1.66E+02	1.43E+02
CD132	4.32E-06	0.00E+00															
IN132	2.14E-01	0.00E+00															
SN132	6.51E+03	0.00E+00															
SB132	2.10E+04	0.00E+00															
SB132M	9.52E+03	0.00E+00															
TE132	4.50E+04	4.27E+04	4.05E+04	3.84E+04	3.64E+04	3.27E+04	2.94E+04	2.65E+04	2.38E+04	2.14E+04	1.92E+04	1.26E+04	8.21E+03	5.37E+03	2.29E+03	9.78E+02	4.18E+02
I132	4.55E+04	4.39E+04	4.17E+04	3.96E+04	3.75E+04	3.37E+04	3.03E+04	2.73E+04	2.45E+04	2.20E+04	1.98E+04	1.29E+04	8.46E+03	5.53E+03	2.36E+03	1.01E+03	4.30E+02
XE132	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS132	1.98E+00	1.93E+00	1.88E+00	1.83E+00	1.78E+00	1.69E+00	1.60E+00	1.52E+00	1.44E+00	1.36E+00	1.29E+00	1.04E+00	8.43E-01	6.80E-01	4.43E-01	2.89E-01	1.88E-01
BA132	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN133	4.36E-03	0.00E+00															
SN133	9.55E+02	0.00E+00															
SB133	2.64E+04	0.00E+00															
TE133	4.31E+04	6.25E+01	6.91E-01	7.65E-03	8.46E-05	1.04E-08	1.27E-12	1.55E-16	1.90E-20	2.32E-24	2.84E-28	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE133M	3.37E+04	3.73E+02	4.12E+00	4.56E-02	5.05E-04	6.17E-08	7.56E-12	9.25E-16	1.13E-19	1.39E-23	1.70E-27	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I133	7.33E+04	6.17E+04	5.05E+04	4.14E+04	3.39E+04	2.27E+04	1.52E+04	1.02E+04	6.84E+03	4.59E+03	3.08E+03	6.21E+02	1.25E+02	2.53E+01	1.03E+00	4.22E-02	1.72E-03
I133M	8.49E+02	0.00E+00															
XE133	6.31E+04	6.32E+04	6.30E+04	6.25E+04	6.17E+04	5.96E+04	5.71E+04	5.43E+04	5.14E+04	4.85E+04	4.57E+04	3.56E+04	2.75E+04	2.12E+04	1.25E+04	7.39E+03	4.36E+03
XE133M	2.07E+03	2.06E+03	2.02E+03	1.97E+03	1.90E+03	1.74E+03	1.56E+03	1.39E+03	1.22E+03	1.07E+03	9.25E+02	5.10E+02	2.75E+02	1.47E+02	4.15E+01	1.17E+01	3.30E+00
CS133	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA133	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN134	5.21E-06	0.00E+00															
SN134	8.64E+01	0.00E+00															
SB134	3.91E+03	0.00E+00															
SB134M	3.72E+03	0.00E+00															
TE134	7.70E+04	1.97E+02	5.03E-01	1.28E-03	3.27E-06	2.14E-11	1.40E-16	9.11E-22	5.95E-27	3.88E-32	0.00E+00						
I134	8.65E+04	2.59E+03	2.72E+01	2.49E-01	2.20E-03	1.67E-07	1.27E-11	9.61E-16	7.28E-20	5.52E-24	4.18E-28	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I134M	4.02E+03	0.00E+00															
XE134	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE134M	2.51E+01	0.00E+00															
CS134	7.31E+01	7.32E+01	7.32E+01	7.32E+01	7.31E+01	7.31E+01	7.31E+01	7.30E+01	7.30E+01	7.30E+01	7.29E+01	7.28E+01	7.27E+01	7.25E+01	7.23E+01	7.20E+01	7.17E+01
CS134M	5.32E+02	1.27E+02	3.02E+01	7.20E+00	1.72E+00	9.75E-02	5.54E-03	3.15E-04	1.79E-05	1.02E-06	5.77E-08	6.00E-13	6.25E-18	6.50E-23	7.05E-33	0.00E+00	0.00E+00
BA134	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN135	6.46E-01	0.00E+00															
SB135	1.07E+03	0.00E+00															
TE135	3.58E+04	0.00E+00															
I135	6.96E+04	3.71E+04	1.98E+04	1.05E+04	5.62E+03	1.60E+03	4.54E+02	1.29E+02	3.67E+01	1.04E+01	2.96E+00	1.93E-02	1.26E-04	8.21E-07	3.49E-11	1.49E-15	6.32E-20
XE135	2.23E+03	2.00E+04	2.26E+04	1.96E+04	1.52E+04	7.84E+03	3.64E+03	1.60E+03	6.80E+02	2.84E+02	1.17E+02	3.16E+00	8.22E-02	2.12E-03	1.40E-06	9.28E-10	6.14E-13
XE135M	1.30E+04	5.94E+03	3.17E+03	1.69E+03	9.00E+02	2.56E+02	7.27E+01	2.07E+01	5.87E+00	1.67E+00	4.74E-01	3.09E-03	2.02E-05	1.32E-07	5.59E-12	2.38E-16	1.01E-20</

OUTPUT UNIT = 11 PAGE 67

ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 11/07/2002 at 00:22:10

\* Decay after Irradiation for MTR Case : ic02\_i34\_82c\_15 RECYCLE # 0 FISSION PRODUCTS

POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES

MTR Fuel Element

	Discharge	6.0HR	12.0HR	18.0HR	1.0D	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	6.0D	8.0D	10.0D	14.0D	18.0D	22.0D
CS135M	1.22E+02	1.10E+00	9.96E-03	8.98E-05	8.10E-07	6.60E-11	5.37E-15	4.37E-19	3.56E-23	2.89E-27	2.35E-31	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA135	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA135M	1.07E-02	9.22E-03	7.98E-03	6.90E-03	5.97E-03	4.47E-03	3.34E-03	2.50E-03	1.87E-03	1.40E-03	1.05E-03	3.29E-04	1.03E-04	3.24E-05	3.18E-06	3.13E-07	3.08E-08
SN136	3.32E-02	0.00E+00															
SB136	6.46E+00	0.00E+00															
TE136	1.43E+04	0.00E+00															
I136	2.92E+04	0.00E+00															
I136M	1.36E+04	0.00E+00															
XE136	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS136	1.11E+02	1.10E+02	1.08E+02	1.07E+02	1.05E+02	1.03E+02	9.98E+01	9.72E+01	9.46E+01	9.22E+01	8.98E+01	8.08E+01	7.26E+01	6.54E+01	5.29E+01	4.28E+01	3.46E+01
BA136	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA136M	1.83E+01	1.80E+01	1.78E+01	1.76E+01	1.73E+01	1.69E+01	1.64E+01	1.60E+01	1.56E+01	1.52E+01	1.48E+01	1.33E+01	1.20E+01	1.08E+01	8.72E+00	7.05E+00	5.71E+00
SB137	7.13E+01	0.00E+00															
TE137	4.29E+03	0.00E+00															
I137	3.33E+04	0.00E+00															
XE137	6.79E+04	0.00E+00															
CS137	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.62E+02	1.61E+02	1.61E+02
BA137	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA137M	1.54E+02	1.53E+02															
SB138	2.19E-03	0.00E+00															
TE138	4.81E+02	0.00E+00															
I138	1.48E+04	0.00E+00															
XE138	7.01E+04	1.58E-03	3.55E-11	8.00E-19	1.80E-26	0.00E+00											
CS138	6.90E+04	5.36E+01	2.31E-02	9.96E-06	4.29E-09	7.97E-16	1.48E-22	2.75E-29	8.19E-36	0.00E+00							
CS138M	2.46E+03	0.00E+00															
BA138	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
LA138	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13	1.68E-13
SB139	2.89E-04	0.00E+00															
TE139	1.44E+01	0.00E+00															
I139	6.45E+03	0.00E+00															
XE139	5.52E+04	0.00E+00															
CS139	7.06E+04	2.21E-07	6.55E-19	1.94E-30	0.00E+00												
BA139	6.90E+04	3.84E+03	1.88E+02	9.20E+00	4.50E-01	1.08E-03	2.58E-06	6.18E-09	1.48E-11	3.53E-14	2.36E-16	7.75E-27	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
LA139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CE139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PR139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE140	7.43E+01	0.00E+00															
I140	8.25E+02	0.00E+00															
XE140	3.81E+04	0.00E+00															
CS140	6.27E+04	0.00E+00															
BA140	5.16E+04	5.09E+04	5.03E+04	4.96E+04	4.89E+04	4.76E+04	4.63E+04	4.51E+04	4.39E+04	4.27E+04	4.16E+04	3.73E+04	3.35E+04	3.00E+04	2.42E+04	1.95E+04	1.57E+04
LA140	4.99E+04	5.00E+04	5.01E+04	5.01E+04	5.00E+04	4.97E+04	4.92E+04	4.85E+04	4.78E+04	4.69E+04	4.60E+04	4.21E+04	3.82E+04	3.44E+04	2.78E+04	2.24E+04	1.80E+04
CE140	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PR140	1.22E-01	0.00E+00															
TE141	1.97E-02	0.00E+00															
I141	7.98E+01	0.00E+00															
XE141	9.35E+03	0.00E+00															
CS141	4.53E+04	0.00E+00															
BA141	6.47E+04	7.69E-02	8.98E-08	1.05E-13	1.22E-19	1.67E-31	0.00E+00										
LA141	6.35E+04	2.39E+04	8.31E+03	2.89E+03	1.00E+03	1.21E+02	1.45E+01	1.75E+00	2.11E-01	2.54E-02	3.06E-03	6.46E-07	1.36E-10	2.87E-14	1.28E-21	5.66E-29	2.24E-36









OUTPUT UNIT = 11 PAGE 72

ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 11/07/2002 at 00:22:10

\* Decay after Irradiation for MTR Case : ic02\_i34\_82c\_15 RECYCLE # 0 FISSION PRODUCTS

POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES

MTR Fuel Element

	Discharge	6.0HR	12.0HR	18.0HR	1.0D	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	6.0D	8.0D	10.0D	14.0D	18.0D	22.0D
TM169	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
YB169	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ER170	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TM170	3.34E-08	3.33E-08	3.33E-08	3.32E-08	3.32E-08	3.31E-08	3.30E-08	3.29E-08	3.28E-08	3.27E-08	3.26E-08	3.23E-08	3.19E-08	3.16E-08	3.09E-08	3.03E-08	2.96E-08
TM170M	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
YB170	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ER171	7.65E-14	4.40E-14	2.54E-14	1.46E-14	8.40E-15	2.76E-15	9.45E-16	3.24E-16	9.06E-17	4.58E-17	0.00E+00						
TM171	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.71E-10	1.70E-10	1.70E-10	1.70E-10	1.69E-10	1.68E-10	1.68E-10
YB171	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ER172	1.36E-16	1.24E-16	1.14E-16	1.04E-16	9.53E-17	7.91E-17	6.57E-17	5.45E-17	4.53E-17	3.76E-17	3.76E-17	2.07E-17	6.66E-18	6.66E-18	0.00E+00	0.00E+00	0.00E+00
TM172	7.98E-11	7.48E-11	7.01E-11	6.56E-11	6.15E-11	5.39E-11	4.73E-11	4.15E-11	3.64E-11	3.20E-11	2.80E-11	1.66E-11	9.85E-12	5.84E-12	2.05E-12	7.20E-13	2.53E-13
YB172	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TOTAL	5.63E+06	1.26E+06	1.03E+06	8.98E+05	8.10E+05	6.93E+05	6.19E+05	5.65E+05	5.25E+05	4.92E+05	4.65E+05	3.86E+05	3.33E+05	2.95E+05	2.42E+05	2.06E+05	1.81E+05

## APPENDIX C

### BEHAVIOR OF TELLURIUM IN THE ALKALINE AND ACID DISSOLVERS<sup>1</sup>

#### C.1 Introduction

The Materials Testing Reactor (MTR) fuel assemblies treated by the Radioactive Lanthanum (RaLa) process to separate barium-140 contained one to two grams of tellurium, a fission product that was decaying to iodine, primarily Te-132 ([Wichner 2002a](#)). The two essential questions addressed in this report are (1) the nature of tellurium in each basic step of the process and (2) the potential for release of tellurium to the environment during processing that would, on decay, contribute to the quantity of radioiodine released.

Tellurium (Te) was produced in the fuel element during uranium fission and was present as the metal intimately mixed with uranium metal in the fuel elements. The presence of aluminum telluride ( $\text{Al}_2\text{Te}_3$ ) at the interface of the uranium fuel and aluminum cladding or the presence of one of the tellurides of uranium ( $\text{U}_x\text{Te}_y$ , x from 1 to 4 and y from 1 to 9) within the fuel is not likely. Aluminum telluride, a very unstable material, is produced by melting aluminum and tellurium in the presence of nitrogen ([Kudryavtsev 1974](#)). Uranium tellurides are produced in a similar fashion in sealed tubes at controlled temperatures ([Weigel 1986](#)).

The proposed fate of tellurium discussed in this appendix is based on process steps described in “Pilot Plant Development of a Process for Separating Barium-140 from MTR Fuel” by [Legler et al. \(1955\)](#). The essential steps of the RaLa process described in the report consisted of the following:

- (1) Dissolution of the aluminum cladding surrounding the uranium fuel element with strong alkaline solution
- (2) Dissolution of the uranium fuel in nitric acid
- (3) Concentration of the uranium solution by heating and precipitation of barium and strontium nitrate with fuming nitric acid, followed by separation of barium from strontium by chromate precipitation
- (4) Conversion of barium chromate to barium nitrate, with precipitation, purification, and drying of the final barium nitrate

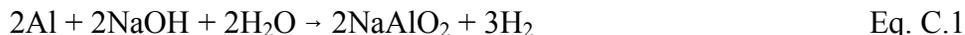
The behavior and fate of tellurium in each of these basic steps will be considered in the following sections.

---

<sup>1</sup>Appendix C was written by Dr. William S. Richardson, III, SC&A, Inc.

## C.2 Step 1: Dissolution of the Aluminum Cladding Surrounding the Uranium Fuel Element with Strong Alkaline Solution

In the first process step, fuel assemblies were added to an aqueous solution of 5 M sodium hydroxide (NaOH). A solution of 5 M sodium hydroxide is a very alkaline solution with a pH of almost 15. The system was then purged with nitrogen, and the mixture was heated to dissolve the aluminum cladding. Aluminum is converted to a solution of sodium aluminate (NaAlO<sub>2</sub>) in strong alkali with the release of hydrogen gas:



Off-gassing of hydrogen was controlled by temperature regulation. Sodium aluminate is very soluble in hot water and is considered soluble in cold water (Weast 1974).

During this process step, metallic tellurium was not lost by sublimation from the alkali solution: tellurium has a melting point of 452°C (Cooper 1971) and a vapor pressure of about 10<sup>-9</sup> torr at 90°C (363°K) (Cooper 1971). As long as the solution was not boiling, loss of tellurium was not expected in acid or base solution (Leddicotte 1961). Formation of gaseous tellurium hydride (TeH<sub>2</sub>) and subsequent loss during the first step is not a consideration since the hydride cannot be made by direct combination of the elements hydrogen and tellurium (Cooper 1971), even at a temperature of 650°C, when it is formed in poor yields (Bagnall 1966). Greenwood and Earnshaw (1984) write, “The hydride is also unstable above 0°C and decomposes in moist air and on exposure to light.” Even if a trace quantity were produced and survived the process step, the acid would dissolve in the strong alkali solution to produce sodium telluride (Na<sub>2</sub>Te) (Cooper 1971).

The behavior of cesium and iodine under similar reaction conditions is an important consideration in determining the behavior of tellurium in the first process step. It is known that cesium dissolves during a similar alkali-treatment process of uranium fuel elements.

“Also of interest was knowledge of the distribution of fission product cesium between the aluminate solution and the undissolved U residue. Radiochemical analysis of these two fractions showed that more than 93% of the initial cesium was released to the solution during caustic dissolving.” (ICPP 1954).

In another, similar process, 90% of the iodine was found in the alkali solution: “About 10% of the iodine is found in the precipitate and the remainder in the supernatant, with essentially no volatilization at this point” (Cederberg and MacQueen 1959). Release of cesium metal and most of the iodine in the alkali process step, as well as the powdered nature of the uranium remaining after alkali processing (Wichner 2002b), suggests that tellurium metal in the fuel element was exposed to the alkali solution, and most of the metal subsequently dissolved in the sodium hydroxide solution. According to Pourbaix (1966), “. . . it (tellurium) can be reduced in alkaline solution to give ditelluride ions Te<sub>2</sub><sup>2-</sup> and possibly telluride ions Te<sup>2-</sup>.” Pourbaix continues, “In actual fact aqueous solutions free from oxidizing agents have no action on tellurium unless they are very alkaline, . . .” Beahm (1986) is clear in pointing to this behavior of tellurium in strong alkali in his report. In addition, Kudryavtsev (1974) indicates that tellurium disproportionates

(undergoes both oxidation and reduction) on heating in strong alkali to produce tellurite ions ( $\text{TeO}_3^{-2}$ ) and telluride ions ( $\text{Te}^{-2}$ ) and that the reaction reverses on cooling according to the following equation:

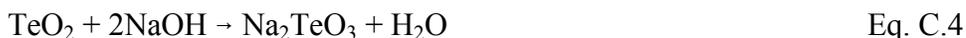


Chizhikov and Shchastlivyi (1970) state that, “The solubility of tellurium in alkalis depends on the temperature. Increasing the temperature shifts the reaction to the right and cooling reverses the reaction.” Tellurite in an alkaline solution can also be oxidized by gamma irradiation to tellurate ( $\text{TeO}_4^{-2}$ ) on recoil (Cooper 1971). These observations strongly suggest that tellurium is present in the hot alkaline solution as soluble sodium telluride ( $\text{Na}_2\text{Te}$ ), sodium ditelluride ( $\text{Na}_2\text{Te}_2$ ), and sodium tellurite ( $\text{Na}_2\text{TeO}_3$ ), with the possibility of some sodium tellurate ( $\text{Na}_2\text{TeO}_4$ ) also present. In this form, they would follow the aqueous solution of sodium aluminate to a waste receiver tank.

On cooling, the dissolution step will reverse and regenerate metallic tellurium. Soluble tellurates, in contrast, are stable, and they are reduced to tellurites with difficulty (Pourbaix 1966). In the aqueous solution, tellurium will then begin to react slowly with water to form tellurium dioxide ( $\text{TeO}_2$ ), “even under normal conditions,” (Nazarenko and Ermakov 1972) as follows:



Considering the small, original amount of tellurium present and the slow rate of reaction, the volume of hydrogen produced would be negligible and go unnoticed. Since the amount of sodium hydroxide used in the reaction mixture was about twice that required to dissolve the aluminum present, the solution remaining after dissolution of the cladding was still basic, more than 2 M (Legler et al. 1955). Therefore, the small amount of tellurium dioxide formed during the slow process dissolved in the alkali solution to form sodium tellurite (Nazarenko and Ermakov 1972). Kudryavtsev (1974) refers to the ease of formation of tellurites in caustic alkaline solutions in the following expression:



The waste aluminate solution was subsequently mixed with waste nitric acid solution several days after it was collected; however, the alkali solution was neutralized and the medium was rendered a dilute nitric acid solution (Wichner 2002b). Under these conditions, the tellurium metal present reacted with the dilute nitric acid to produce tellurous acid (Chizhikov and Shchastlivyi 1970) in the following reaction:



The small amount of sodium tellurite that formed in the first few days was also converted by nitric acid into tellurous acid (Cooper 1971):



Tellurous acid is the hydrated form of tellurium dioxide as illustrated by the following:



Tellurium dioxide is very stable in water (Pourbaix 1966) and sparingly soluble in water, approximately  $4 \times 10^{-5} \text{ M}$  (Kudryavtsev 1974). In contrast, tellurous acid is less stable, and the solubility is about one-tenth that of tellurium dioxide,  $3 \times 10^{-6} \text{ M}$  (Kudryavtsev 1974). These are probably the reasons that Pourbaix (1966) suggests that tellurium dioxide is likely the predominate form in a dilute acid solution.

Considering the solubility of tellurium dioxide and the mass of tellurium present in the initial fuel element (1-2 g), at least 480 liters of water would be needed to completely dissolve the tellurium dioxide present. In contrast to Pourbaix's suggestion, Cooper (1971) writes that at low temperatures (room temperature), tellurous acid might not convert to tellurium dioxide. If this were the case, tellurium would be present as tellurous acid, and the acid would require approximately 6,400 liters of water to dissolve it. The RaLa process used less than 425 liters of water per fuel assembly in any one step of the process (Legler et al. 1955), so tellurium was present in the dilute nitric-acid aluminate waste solution as an insoluble form, tellurium dioxide or tellurous acid, and a soluble form, sodium tellurate, produced in the initial alkaline treatment process. Note that tellurium dioxide would not dissolve in the dilute nitric acid ( $\text{HNO}_3$ ) solution as a nitric acid addition compound ( $2\text{TeO}_2 \cdot \text{HNO}_3$ ) (see Section C.4, Step 3), because the acid solution in Step 1 was not concentrated nitric acid (Kudryavtsev 1974). Concentrated nitric acid is 15.9 M (Merck 1968).

### C.3 Step 2: Dissolution of the Uranium Fuel in Nitric Acid

The solid product cake from the sodium hydroxide dissolution step contained uranium, silica, and metal fission-product particulates along with residual sodium aluminate solution (Legler et al. 1955). Some metallic tellurium was probably present because of incomplete availability of the metal during alkali treatment (ICPP 1954; Cederberg and MacQueen 1959). In the second basic step of the RaLa process, the cake, primarily uranium, was dissolved in 6 M nitric acid. Silica did not dissolve during this process, and the supernatant was separated from the insoluble silica and recovered for additional treatment. The metallic tellurium, which remained in the residual aluminate solution after the solution cooled and before it aged or was neutralized with nitric acid, also reacted with nitric acid to produce tellurous acid, as cited in Section C.2 (Chizhikov and Shchastlivyi 1970).



As stated above, as long as the solution is not boiling, loss of tellurium is not expected in either acid or base solution (Leddicotte 1961). Therefore, tellurium was present after nitric acid treatment in the same forms found in the nitric acid-treated waste solution from Step 1: primarily as an insoluble form, tellurium dioxide or tellurous acid, and a trace of soluble sodium tellurate remaining from the residue solution. As in Step 1, tellurium dioxide would not dissolve in 6 M nitric acid as a nitric acid addition compound (see Step 3), because the acid solution in Step 2 is not concentrated.

#### **C.4 Step 3: Concentration of the Uranium Solution by Heating and Precipitation of Barium and Strontium Nitrate with Fuming Nitric Acid (Concentrated Nitric Acid with Gaseous Nitrogen Dioxide Added), followed by Separation of Barium from Strontium by Chromate Precipitation**

The third basic step of the RaLa process consisted of the addition of carrier barium to the nitric acid solution and evaporation of the solution to a concentrated solution. Fuming nitric acid was added to precipitate barium nitrate  $[\text{Ba}(\text{NO}_3)_2]$  and strontium nitrate  $[\text{Sr}(\text{NO}_3)_2]$ . Tellurium dioxide formed a soluble nitrate addition compound  $(2\text{TeO}_2 \cdot \text{HNO}_3)$  in the concentrated acid (Green and Turley 1961). Each time the nitric acid solution was separated from the precipitated nitrates, this soluble form of tellurium would follow the acid solution to a waste tank. Addition of water to the wet precipitate would release trace quantities of tellurium dioxide left behind with the nitrate precipitates. It would probably remain in solution since a trace amount would not exceed the limits of solubility of the dioxide. There is no evidence in the literature that tellurium precipitates or co-precipitates under these chemical conditions.

#### **C.5 Step 4: Conversion of Barium Chromate to Barium Nitrate, with Precipitation, Purification, and Drying of the Final Barium Nitrate**

After alternate steps of precipitating barium nitrate with fuming nitric acid or precipitating barium chromate ( $\text{BaCrO}_4$ ), followed by dissolution of the nitrate precipitate (on removal of nitric acid and adding water) or treating the chromate precipitate with fuming nitric acid to convert it back to insoluble barium nitrate (in fuming nitric acid), the trace amount of tellurium present would alternate between the more soluble nitrate addition compound and tellurium dioxide. In either case, tellurium would be in solution and be removed to a waste tank each time the supernatant was separated from a precipitate.

#### **C.6 Summary**

Tellurium was in the following forms in each step of the RaLa process, either as a soluble ion or as suspension or insoluble compound in the aqueous reaction mixture:

Step 1 Tellurium was present as soluble sodium salts of tellurite, telluride, and ditelluride in the hot, strongly alkaline solution; but on cooling and neutralization and acidification with nitric acid, as insoluble tellurium dioxide or tellurous acid and as soluble sodium tellurate from the initial alkaline process.

Step 2 Tellurium was present predominately as insoluble tellurium dioxide or tellurous acid and traces of soluble sodium tellurate, all in greatly reduced quantities relative to total tellurium in all forms found in Step 1.

### Steps 3 and 4

Tellurium was present as small quantities of the soluble nitrate adduct of tellurium dioxide, which was removed with fuming the nitric acid solution, but as small quantities of insoluble tellurium dioxide in the aqueous wash of the precipitates.

There is no evidence for the formation in any process step of a volatile compound that could have been lost to the environment by sublimation, volatilization, or as a gaseous product, or for the loss of any form of tellurium in an aerosol from a boiling solution.

## C.7 References

Beahm EC. Te Behavior in Containment under LWR Accident Conditions, NUREG/CR-4338 and ORNL-TM-9726; February 1986.

Cederberg GK, MacQueen DK. Containment of I-131 Released by the RaLa Process. Research and Development Report, Atomic Energy Commission, IDO-14566; 1961.

Chizhikov DM, Shchastlivyi VP. Tellurium and the Tellurides (translated by E.M. Elkin). Collet's Publishers, London; 1970.

Cooper WC. Tellurium. Van Nostrand Reinhold, New York; 1971.

CRC Press. Handbook of Chemistry and Physics (R.C. Weast, ed.). CRC Press; Cleveland, Ohio; 1974.

Green TE, Turley PR. Selenium and Tellurium in: Treatise on Analytical Chemistry, Part II, Vol. 7. IM Kolthoff and PJ Elving, eds. John Wiley and Sons, New York; 1961.

Greenwood NN, Earnshaw A. Chemistry of the Elements. Pergamon, Oxford; 1984.

ICPP. Progress Report for July-September, 1954. Technical Branch, ICPP; IDO-14337; 1954.

Kudryavtsev AA. The Chemistry and Technology of Selenium and Tellurium. Translated from the 2<sup>nd</sup> edition and revised by E.M. Elkin. Collet's Publishers, London; 1974.

Leddicotte GW. The Radiochemistry of Tellurium. Nuclear Science Series, Subcommittee on Radiochemistry, National Academy of Sciences-National Research Council, Washington, D.C.; NAS-NS 3038; July, 1961.

Legler BM, Chase WL, Martin MD, Paige DM, Wrigley FK. Pilot Plant Development of a Process for Separating Barium-140 from MTR Fuel. AEC Research and Development Report, Phillips Petroleum Co., Atomic Energy Division, Contract No. at (10-1)-205; Idaho Operations Office, U.S. Atomic Energy Commission; IDO-14344; September 1, 1955.

Merck. The Merck Index (Eighth Ed.). PG Stecher, ed. Merck and Co., Rahway, NJ; 1968.

Pourbaix M. Atlas of Electrochemical Equilibria in Aqueous Solutions (translated by F.A. Franklin). Pergamon, Oxford; 1966.

Weigel. Uranium in: The Chemistry of the Actinide Elements, Vol. 1. JJ Katz, GT Seaborg, LR Morss, eds. Chapman and Hall, London; 1986.

Wichner RP. E-mail message to Mauro J and Cohen S; Subject: Tellurium Problem Statement; August 9, 2002.

Wichner RP. Personal Communication to Richardson, III W; 2002b.

September 20, 2002

Dr. Robert P. Wichner  
SENES Oak Ridge, Inc.  
102 Donner Drive  
Oak Ridge, Tennessee 37830

Dear Dr. Wichner:

Please find attached to this letter my report entitled, "Chemical Behavior of Tellurium in the RaLa Process." It addresses the questions you raised in our initial contact by email concerning the chemical form of tellurium in each of the basic steps of the RaLa process and the potential for loss of tellurium to the environment during the process steps.

I want to take this opportunity to thank you for providing several documents referenced in the report and for additional information about the RaLa process that you provided in conversations and by email correspondence. In addition, I want to thank you for the stimulating conversations we had about tellurium chemistry and the RaLa process during the preparation of the report.

If I can be of further service, please do not hesitate to call on me. I hope we have the opportunity to work together again.

Sincerely yours,

William S. Richardson, III, PhD.  
Associate

cc Dr. John Mauro

attachment

## APPENDIX D

### NOBLE GAS, IODINE, AND TELLURIUM PRODUCTION IN THE IDAHO CHEMICAL PROCESSING PLANT CRITICALITY ACCIDENT OF OCTOBER 16, 1959

The following is a portion of the ORIGEN2 output for the criticality accident of October 16, 1959, discussed in [Chapter 7](#). The assumption is  $4 \times 10^{19}$  thermal fissions in U-235, occurring rapidly in a single event, i.e., no re-criticalities. This portion includes krypton, bromine, iodine, and tellurium production in curies and decay to 24 hours. The complete record includes all nuclides in grams and curies decayed to 16 days.







OUTPUT UNIT = 11 PAGE 177  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

## 7 NUCLIDE TABLE: RADIOACTIVITY, CURIES

10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
GE 88	2.64E-02	0.00E+00															
AS 88	3.59E+01	0.00E+00															
SE 88	4.13E+04	3.77E-08	0.00E+00														
BR 88	2.80E+05	2.21E+04	2.36E-06	1.96E-17	0.00E+00												
KR 88	2.12E+03	2.52E+03	2.46E+03	2.36E+03	2.01E+03	9.64E+02	2.23E+02	7.30E+00	7.30E+00	3.90E-01	2.08E-02	1.11E-03	5.94E-05	3.17E-06	1.69E-07	1.12E-17	0.00E+00
RB 88	1.91E+02	2.75E+02	9.33E+02	1.41E+03	1.98E+03	1.08E+03	2.49E+02	8.15E+00	8.15E+00	4.35E-01	2.33E-02	1.24E-03	6.63E-05	3.54E-06	1.89E-07	1.25E-17	0.00E+00
SR 88	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AS 89	4.35E-02	0.00E+00															
SE 89	1.13E+03	0.00E+00															
BR 89	1.62E+05	1.57E+01	0.00E+00														
KR 89	1.54E+05	1.27E+05	1.77E+04	1.99E+03	3.17E-01	2.55E-18	0.00E+00										
RB 89	5.36E+03	1.14E+04	2.51E+04	1.83E+04	3.04E+03	8.29E-01	6.15E-08	1.42E-24	1.42E-24	0.00E+00							
SR 89	5.03E-02	1.32E-01	2.03E+00	4.13E+00	7.43E+00	8.06E+00	8.03E+00	7.97E+00	7.97E+00	7.91E+00	7.86E+00	7.80E+00	7.75E+00	7.70E+00	7.64E+00	7.24E+00	6.48E+00
Y 89	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Y 89M	2.95E-02	2.21E-03	1.67E-13	9.49E-25	0.00E+00												
AS 90	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SE 90	5.01E+02	0.00E+00															
BR 90	6.03E+04	3.13E-07	0.00E+00														
KR 90	6.19E+05	1.72E+05	1.60E+00	4.14E-06	0.00E+00												
RB 90	9.14E+04	1.41E+05	1.62E+04	1.17E+03	2.68E-01	6.15E-14	0.00E+00										
RB 90M	2.61E+04	2.84E+04	7.37E+03	1.47E+03	2.32E+00	5.82E-13	0.00E+00										
SR 90	2.78E-03	9.70E-03	4.39E-02	4.81E-02	4.87E-02												
Y 90	1.45E-04	1.47E-04	2.04E-04	2.93E-04	6.59E-04	2.25E-03	5.22E-03	1.14E-02	1.14E-02	1.59E-02	1.99E-02	2.34E-02	2.65E-02	2.92E-02	3.16E-02	4.26E-02	4.79E-02
Y 90M	2.98E-03	2.97E-03	2.87E-03	2.76E-03	2.38E-03	1.22E-03	3.18E-04	1.39E-05	1.39E-05	9.51E-07	6.50E-08	4.44E-09	3.04E-10	2.08E-11	1.42E-12	6.75E-22	0.00E+00
ZR 90	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ZR 90M	1.36E-05	0.00E+00															
SE 91	1.27E+00	0.00E+00															
BR 91	9.94E+03	0.00E+00															
KR 91	5.45E+05	4.58E+03	9.47E-16	0.00E+00													
RB 91	4.61E+05	2.72E+05	4.38E+02	3.45E-01	1.33E-13	0.00E+00											
SR 91	3.47E+02	8.06E+02	1.26E+03	1.24E+03	1.18E+03	9.50E+02	6.13E+02	2.21E+02	2.21E+02	9.20E+01	3.83E+01	1.60E+01	6.66E+00	2.77E+00	1.16E+00	1.05E-03	8.64E-10
Y 91	6.83E-04	2.78E-03	4.32E-02	9.71E-02	3.62E-01	1.83E+00	4.21E+00	6.97E+00	6.97E+00	7.85E+00	8.18E+00	8.29E+00	8.31E+00	8.29E+00	8.25E+00	7.88E+00	7.17E+00
Y 91M	1.43E+00	6.21E+00	8.72E+01	1.70E+02	3.97E+02	5.75E+02	3.90E+02	1.40E+02	1.40E+02	5.85E+01	2.44E+01	1.02E+01	4.23E+00	1.76E+00	7.34E-01	6.66E-04	5.49E-10
ZR 91	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 91	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SE 92	2.55E-01	0.00E+00															
BR 92	2.95E+02	0.00E+00															
KR 92	1.91E+05	2.92E-05	0.00E+00														
RB 92	7.56E+05	8.29E+01	0.00E+00														
SR 92	4.14E+03	4.51E+03	4.34E+03	4.16E+03	3.51E+03	1.63E+03	3.51E+02	9.78E+00	9.78E+00	4.55E-01	2.11E-02	9.81E-04	4.56E-05	2.12E-06	9.83E-08	2.13E-18	0.00E+00
Y 92	4.86E+01	6.31E+01	1.90E+02	3.20E+02	7.49E+02	1.47E+03	9.49E+02	1.03E+02	1.03E+02	1.14E+01	1.16E+00	1.14E-01	1.10E-02	1.06E-03	1.01E-04	6.92E-13	3.22E-29
ZR 92	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 92	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SE 93	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BR 93	8.59E+00	0.00E+00															
KR 93	4.52E+04	2.73E-10	0.00E+00														
RB 93	5.57E+05	4.38E+02	0.00E+00														
SR 93	9.14E+04	9.01E+04	3.92E+04	1.56E+04	3.86E+02	2.30E-05	8.17E-20	0.00E+00									
Y 93	7.69E+01	1.85E+02	8.09E+02	1.09E+03	1.22E+03	9.99E+02	6.62E+02	2.53E+02	2.53E+02	1.11E+02	4.88E+01	2.14E+01	9.40E+00	4.12E+00	1.81E+00	2.50E-03	4.74E-09
ZR 93	4.72E-11	1.61E-10	4.35E-09	1.27E-08	5.42E-08	2.26E-07	4.80E-07	7.88E-07	7.88E-07	8.95E-07	9.42E-07	9.63E-07	9.72E-07	9.76E-07	9.78E-07	9.79E-07	9.79E-07
NB 93	0.00E+00	0.0															

OUTPUT UNIT = 11 PAGE 178

ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

## 7 NUCLIDE TABLE: RADIOACTIVITY, CURIES

10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
NB 93M	1.47E-11	1.47E-11	1.47E-11	1.47E-11	1.48E-11	1.72E-11	2.92E-11	8.01E-11	8.01E-11	1.36E-10	1.98E-10	2.61E-10	3.25E-10	3.89E-10	4.54E-10	9.73E-10	2.01E-09
BR 94	1.64E-02	0.00E+00															
KR 94	2.88E+02	0.00E+00															
RB 94	2.17E+05	4.20E-02	0.00E+00														
SR 94	4.47E+05	2.62E+05	1.85E+03	7.56E+00	2.10E-09	0.00E+00											
Y 94	1.16E+04	2.36E+04	3.02E+04	2.11E+04	4.95E+03	7.19E+00	1.52E-05	8.78E-19	8.78E-19	3.95E-30	0.00E+00						
ZR 94	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 94	1.76E-12	1.87E-12	2.50E-12	2.74E-12	2.86E-12												
NB 94M	1.89E-03	1.69E-03	6.24E-04	2.06E-04	2.46E-06	5.43E-15	2.65E-32	0.00E+00									
BR 95	3.60E-04	0.00E+00															
KR 95	2.41E+02	0.00E+00															
RB 95	1.38E+04	0.00E+00															
SR 95	7.30E+05	1.48E+05	8.26E-02	9.33E-09	0.00E+00												
Y 95	4.40E+04	6.43E+04	3.90E+04	2.02E+04	1.44E+03	9.94E-03	4.74E-13	0.00E+00									
ZR 95	3.12E-01	7.43E-01	4.35E+00	6.50E+00	8.63E+00	8.78E+00	8.76E+00	8.70E+00	8.66E+00	8.61E+00	8.56E+00	8.52E+00	8.47E+00	8.42E+00	8.07E+00	7.40E+00	7.40E+00
NB 95	2.61E-04	2.68E-04	6.02E-04	1.36E-03	5.70E-03	2.72E-02	6.99E-02	1.68E-01	1.68E-01	2.51E-01	3.33E-01	4.14E-01	4.93E-01	5.71E-01	6.48E-01	1.22E+00	2.17E+00
NB 95M	5.91E-04	5.92E-04	6.14E-04	6.65E-04	9.59E-04	2.40E-03	5.16E-03	1.11E-02	1.11E-02	1.56E-02	1.97E-02	2.34E-02	2.68E-02	2.98E-02	3.24E-02	4.59E-02	5.19E-02
MO 95	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BR 96	3.00E-05	0.00E+00															
KR 96	1.01E+03	0.00E+00															
RB 96	2.32E+03	0.00E+00															
SR 96	5.47E+05	1.67E+01	0.00E+00														
Y 96	2.64E+05	2.08E+05	1.38E+04	6.77E+02	3.94E-03	0.00E+00											
ZR 96	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB 96	4.81E-02	4.81E-02	4.79E-02	4.76E-02	4.67E-02	4.27E-02	3.57E-02	2.36E-02	2.36E-02	1.65E-02	1.16E-02	8.10E-03	5.67E-03	3.97E-03	2.78E-03	1.61E-04	5.38E-07
MO 96	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
KR 97	1.90E-02	0.00E+00															
RB 97	4.92E+01	0.00E+00															
SR 97	4.99E+03	0.00E+00															
Y 97	4.43E+05	2.39E-11	0.00E+00														
ZR 97	7.15E+02	7.23E+02	7.18E+02	7.13E+02	6.94E+02	6.14E+02	4.80E+02	2.70E+02	2.70E+02	1.65E+02	1.01E+02	6.17E+01	3.77E+01	2.31E+01	1.41E+01	2.75E-01	1.05E-04
NB 97	2.05E+01	2.54E+01	8.12E+01	1.39E+02	3.18E+02	5.82E+02	5.13E+02	2.72E+02	2.72E+02	1.66E+02	1.02E+02	6.21E+01	3.80E+01	2.32E+01	1.42E+01	2.76E-01	1.13E-04
NB 97M	4.09E+02	5.46E+02	6.80E+02	6.76E+02	6.57E+02	5.81E+02	4.54E+02	2.56E+02	2.56E+02	1.56E+02	9.56E+01	5.85E+01	3.57E+01	2.18E+01	1.34E+01	2.60E-01	9.90E-05
MO 97	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
KR 98	6.94E+00	0.00E+00															
RB 98	1.83E+01	0.00E+00															
SR 98	5.41E+04	3.08E-17	0.00E+00														
Y 98	1.03E+05	4.73E-17	0.00E+00														
ZR 98	7.28E+05	1.91E+05	1.09E+00	1.63E-06	0.00E+00												
NB 98	7.55E+05	2.10E+05	1.20E+00	1.79E-06	0.00E+00												
NB 98M	9.20E+01	9.07E+01	8.04E+01	7.03E+01	4.10E+01	3.64E+00	2.86E-02	3.52E-07	3.52E-07	2.18E-11	1.35E-15	8.33E-20	5.15E-24	3.19E-28	1.97E-32	0.00E+00	0.00E+00
MO 98	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TC 98	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23	1.10E-23
RB 99	5.18E-04	0.00E+00															
SR 99	5.32E+03	0.00E+00															
Y 99	1.38E+05	0.00E+00															
ZR 99	7.69E+05	2.50E-02	0.00E+00														
NB 99	8.87E+05	5.74E+04	2.46E-07	5.77E-20	0.00E+00												
NB 99M	1.69E+04	1.30E+04	1.18E+03	8.19E+01	1.91E-03	2.76E-24	0.00E+00										
MO 99	1.17E+02	1.79E+02	1.90E+02	1.91E+02	1.89E+02	1.84E+02	1.72E+02	1.49E+02	1.49E+02	1.31E+02	1.16E+02	1.02E+02	8.99E+01	7.92E+01	6.98E+01	2.55E+01	3.39E+00
TC 99</																	



OUTPUT UNIT = 11 PAGE 180  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
MO105	8.67E+04	4.07E+04	3.97E+01	1.80E-02	7.50E-16	0.00E+00											
TC105	5.00E+03	9.65E+03	6.78E+03	2.85E+03	8.92E+01	1.50E-05	4.27E-19	0.00E+00									
RU105	5.05E+00	2.50E+01	2.62E+02	3.72E+02	4.12E+02	2.59E+02	1.02E+02	1.14E+01	1.14E+01	1.75E+00	2.69E-01	4.13E-02	6.35E-03	9.74E-04	1.50E-04	4.62E-11	4.40E-24
RH105	3.56E-04	4.08E-03	4.58E-01	1.51E+00	6.84E+00	2.53E+01	4.10E+01	4.07E+01	4.07E+01	3.32E+01	2.64E+01	2.09E+01	1.65E+01	1.30E+01	1.03E+01	1.57E+00	3.64E-02
RH105M	3.03E-01	2.75E+00	7.33E+01	1.04E+02	1.16E+02	7.28E+01	2.85E+01	3.21E+00	3.21E+00	4.92E-01	7.56E-02	1.16E-02	1.78E-03	2.74E-04	4.20E-05	1.30E-11	1.24E-24
PD105	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Y106	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ZR106	1.36E-01	5.10E-20	0.00E+00														
NB106	5.89E+02	1.12E-19	0.00E+00														
MO106	6.06E+04	5.97E+02	5.18E-16	0.00E+00													
TC106	4.22E+04	1.98E+04	8.10E-01	1.06E-05	3.17E-25	0.00E+00											
RU106	2.77E-02	7.07E-02	9.39E-02	9.39E-02	9.39E-02	9.39E-02	9.38E-02	9.37E-02	9.37E-02	9.36E-02	9.35E-02	9.35E-02	9.34E-02	9.33E-02	9.32E-02	9.25E-02	9.11E-02
RH106	9.17E-03	4.49E-02	9.39E-02	9.39E-02	9.39E-02	9.39E-02	9.38E-02	9.37E-02	9.37E-02	9.36E-02	9.35E-02	9.35E-02	9.34E-02	9.33E-02	9.32E-02	9.25E-02	9.11E-02
RH106M	2.03E-08	2.02E-08	1.93E-08	1.83E-08	1.49E-08	5.77E-09	8.71E-10	1.06E-11	1.06E-11	2.41E-13	5.57E-15	5.05E-17	7.53E-17	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PD106	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG106	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Y107	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ZR107	9.73E-04	0.00E+00															
NB107	1.19E+02	0.00E+00															
MO107	1.94E+04	2.88E+01	0.00E+00														
TC107	1.85E+04	5.69E+03	1.42E-02	8.37E-09	0.00E+00												
RU107	1.60E+03	3.13E+03	8.77E-02	1.68E+02	2.29E-01	2.87E-14	0.00E+00										
RH107	1.75E+01	9.82E+01	5.60E+02	5.20E+02	1.56E+02	4.97E-01	5.03E-06	1.12E-17	1.12E-17	1.15E-27	0.00E+00						
PD107	8.95E-13	1.19E-11	7.59E-10	1.89E-09	4.40E-09	5.39E-09	5.40E-09										
PD107M	1.11E-13	3.53E-14	8.24E-22	2.73E-30	0.00E+00												
AG107	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ZR108	1.01E-05	0.00E+00															
NB108	3.79E-01	0.00E+00															
MO108	3.10E+03	2.84E-09	0.00E+00														
TC108	8.86E+03	3.40E+00	0.00E+00														
RU108	1.21E+03	1.20E+03	3.00E+02	6.43E+01	1.35E-01	1.23E-13	0.00E+00										
RH108	7.79E+02	1.22E+03	3.20E+02	6.85E+01	1.45E-01	1.31E-13	0.00E+00										
RH108M	7.47E-12	6.65E-12	2.31E-12	7.15E-13	5.00E-15	3.27E-24	0.00E+00										
PD108	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG108	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG108M	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CD108	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ZR109	1.66E-04	0.00E+00															
NB109	4.62E+00	0.00E+00															
MO109	1.28E+03	4.24E-15	0.00E+00														
TC109	2.88E+03	1.29E+03	8.35E-01	2.40E-04	1.64E-18	0.00E+00											
RU109	1.57E+03	1.75E+03	2.61E+00	7.64E-04	5.21E-18	0.00E+00											
RH109	1.67E+02	6.15E+02	1.04E+02	1.17E+00	1.11E-08	0.00E+00											
RH109M	2.24E+02	6.17E+02	8.14E+00	4.91E-03	8.11E-17	0.00E+00											
PD109	2.19E-02	2.02E-01	1.02E+01	1.21E+01	1.22E+01	1.05E+01	7.67E+00	3.73E+00	3.73E+00	2.01E+00	1.09E+00	5.85E-01	3.15E-01	1.70E-01	9.16E-02	6.53E-04	3.32E-08
PD109M	3.53E+00	3.03E+01	3.98E+02	9.68E+01	2.63E-01	7.36E-13	0.00E+00										
AG109	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG109M	4.35E-03	7.26E-02	1.02E+01	1.21E+01	1.22E+01	1.05E+01	7.68E+00	3.73E+00	3.73E+00	2.01E+00	1.09E+00	5.85E-01	3.15E-01	1.70E-01	9.16E-02	6.53E-04	3.32E-08
CD109	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
NB110	3.60E-03	0.00E+00															
MO110	4.49E+02	1.28E-07	0.00E+														





OUTPUT UNIT = 11 PAGE 183  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
RU120	3.39E-06	0.00E+00															
RH120	2.49E-03	0.00E+00															
PD120	3.99E+02	2.36E-02	0.00E+00														
AG120	5.36E+02	3.25E-02	0.00E+00														
CD120	1.15E+03	5.29E+02	3.34E-01	9.30E-05	5.58E-19	0.00E+00											
IN120	2.78E+02	3.43E+02	9.42E-01	3.36E-04	2.21E-18	0.00E+00											
IN120M	6.08E+02	2.82E+02	1.78E-01	4.95E-05	2.97E-19	0.00E+00											
SN120	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
RH121	6.44E-05	0.00E+00															
PD121	2.21E+00	0.00E+00															
AG121	3.57E+02	3.41E-04	0.00E+00														
CD121	1.58E+03	6.57E+01	1.31E-11	1.02E-25	0.00E+00												
IN121	1.22E+03	4.95E+02	8.46E-04	3.00E-10	0.00E+00												
IN121M	5.44E+01	6.03E+01	9.23E+00	1.13E+00	2.54E-04	9.65E-21	0.00E+00										
SN121	3.17E-01	7.30E-01	9.84E-01	9.96E-01	9.82E-01	9.08E-01	7.78E-01	5.42E-01	5.42E-01	3.97E-01	2.91E-01	2.13E-01	1.57E-01	1.15E-01	8.41E-02	7.02E-03	4.90E-05
SN121M	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.32E-07	1.31E-07	1.31E-07
SB121	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
RH122	2.64E-08	0.00E+00															
PD122	4.46E-01	2.69E-15	0.00E+00														
AG122	5.17E-01	2.92E-15	0.00E+00														
CD122	1.93E+03	1.01E+00	0.00E+00														
IN122	2.21E+03	7.02E+01	3.96E-15	3.16E-33	0.00E+00												
IN122M	8.56E+01	7.83E-11	0.00E+00														
SN122	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SB122	6.40E-06	7.32E-06	1.13E-05	1.22E-05	1.24E-05	1.20E-05	1.12E-05	9.67E-06	9.67E-06	8.50E-06	7.48E-06	6.58E-06	5.78E-06	5.09E-06	4.48E-06	1.60E-06	2.06E-07
SB122M	5.64E-03	4.78E-03	1.08E-03	2.08E-04	2.82E-07	3.54E-20	0.00E+00										
TE122	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
RH123	4.25E-09	0.00E+00															
PD123	4.87E-03	0.00E+00															
AG123	1.87E+01	2.18E-20	0.00E+00														
CD123	1.67E+03	1.19E+01	5.39E-19	0.00E+00													
IN123	2.02E+03	3.34E+01	1.43E-18	0.00E+00													
IN123M	2.39E+02	1.34E+02	5.53E-02	9.55E-06	8.48E-21	0.00E+00											
SN123	1.09E-03	1.56E-03	1.85E-03	1.85E-03	1.85E-03	1.84E-03	1.84E-03	1.84E-03	1.84E-03	1.83E-03	1.83E-03	1.82E-03	1.82E-03	1.81E-03	1.81E-03	1.77E-03	1.69E-03
SN123M	3.07E+01	4.07E+01	3.61E+01	3.04E+01	1.52E+01	6.76E-01	1.34E-03	6.57E-10	6.56E-10	2.57E-15	1.01E-20	3.94E-26	1.54E-31	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SB123	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE123	1.21E-23	1.21E-23	1.21E-23	1.21E-23	1.21E-23	1.21E-23	1.22E-23	1.23E-23	1.23E-23	1.25E-23	1.26E-23	1.27E-23	1.28E-23	1.29E-23	1.30E-23	1.39E-23	1.57E-23
TE123M	1.24E-09	1.24E-09	1.24E-09	1.24E-09	1.24E-09	1.24E-09	1.23E-09	1.23E-09	1.23E-09	1.23E-09	1.22E-09	1.22E-09	1.22E-09	1.21E-09	1.21E-09	1.18E-09	1.13E-09
PD124	1.09E-02	0.00E+00															
AG124	4.36E+00	0.00E+00															
CD124	1.96E+03	1.74E+02	5.94E-08	1.80E-18	0.00E+00												
IN124	2.52E+03	2.14E+02	7.30E-08	2.21E-18	0.00E+00												
SN124	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SB124	1.29E-05	1.33E-05	1.40E-05	1.40E-05	1.40E-05	1.40E-05	1.39E-05	1.38E-05	1.38E-05	1.38E-05	1.37E-05	1.36E-05	1.35E-05	1.34E-05	1.32E-05	1.16E-05	1.16E-05
SB124M	7.73E-02	4.94E-02	8.83E-04	1.01E-05	1.72E-13	0.00E+00											
TE124	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PD125	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG125	5.57E-04	0.00E+00															
CD125	5.92E+02	4.35E-09	0.00E+00														
IN125	1.24E+03	3.88E-05	0.00E+00														
IN125M	1.04E+03	3.33E+01	9.46														

OUTPUT UNIT = 11 PAGE 184  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
SN125	1.01E-01	1.04E-01	1.04E-01	1.04E-01	1.04E-01	1.03E-01	1.01E-01	9.66E-02	9.66E-02	9.32E-02	8.99E-02	8.67E-02	8.37E-02	8.07E-02	7.79E-02	5.84E-02	3.29E-02
SN125M	2.57E+02	2.62E+02	1.37E+02	6.59E+01	3.58E+00	7.28E-06	3.01E-17	0.00E+00									
SB125	6.37E-05	1.91E-04	1.02E-03	1.48E-03	1.89E-03	1.92E-03	1.94E-03	1.98E-03	1.98E-03	2.01E-03	2.04E-03	2.07E-03	2.10E-03	2.12E-03	2.15E-03	2.33E-03	2.56E-03
TE125	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE125M	1.21E-10	3.64E-10	1.15E-08	3.58E-08	1.71E-07	8.28E-07	2.15E-06	5.26E-06	5.26E-06	7.96E-06	1.07E-05	1.34E-05	1.62E-05	1.90E-05	2.18E-05	4.49E-05	9.23E-05
PD126	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG126	3.91E-07	0.00E+00															
CD126	1.16E+03	1.85E-02	0.00E+00														
IN126	1.72E+03	3.12E-02	0.00E+00														
SN126	1.30E-07	1.32E-07															
SB126	1.05E-02	1.05E-02	1.07E-02	1.09E-02	1.12E-02	1.12E-02	1.11E-02	1.07E-02	1.07E-02	1.04E-02	1.01E-02	9.84E-03	9.57E-03	9.30E-03	9.05E-03	7.23E-03	4.62E-03
SB126M	5.50E+00	5.30E+00	3.82E+00	2.65E+00	6.16E-01	8.68E-04	1.34E-07	1.32E-07									
TE126	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG127	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE126	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CD127	4.08E+02	0.00E+00															
IN127	5.46E+03	5.19E-06	0.00E+00														
IN127M	1.55E+03	1.74E-02	0.00E+00														
SN127	1.37E+02	1.39E+02	1.32E+02	1.25E+02	1.00E+02	3.73E+01	5.14E+00	5.06E-02	5.06E-02	9.64E-04	1.84E-05	3.50E-07	6.66E-09	1.27E-10	2.42E-12	4.19E-26	0.00E+00
SN127M	2.18E+02	1.84E+02	4.07E+01	7.61E+00	9.28E-03	7.21E-16	0.00E+00										
SB127	1.82E-01	2.24E-01	4.83E-01	6.68E-01	1.23E+00	2.62E+00	3.21E+00	3.00E+00	3.00E+00	2.74E+00	2.51E+00	2.29E+00	2.09E+00	1.91E+00	1.75E+00	8.50E-01	2.01E-01
TE127	9.84E-05	3.15E-04	3.81E-03	9.90E-03	4.94E-02	3.95E-01	1.20E+00	2.16E+00	2.16E+00	2.33E+00	2.28E+00	2.15E+00	1.99E+00	1.83E+00	1.68E+00	8.28E-01	2.08E-01
TE127M	7.47E-08	2.00E-07	2.23E-06	5.79E-06	2.94E-05	2.55E-04	9.24E-04	2.53E-03	2.54E-03	3.79E-03	4.94E-03	5.98E-03	6.92E-03	7.79E-03	8.57E-03	1.27E-02	1.52E-02
I127	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE127	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
AG128	7.07E-10	0.00E+00															
CD128	3.31E+02	3.32E-12	0.00E+00														
IN128	4.35E+03	5.95E-02	0.00E+00														
SN128	6.89E+02	6.86E+02	6.17E+02	5.48E+02	3.43E+02	4.14E+01	6.02E-01	3.12E-05	3.12E-05	6.61E-09	1.40E-12	2.97E-16	6.30E-20	1.34E-23	2.83E-27	0.00E+00	0.00E+00
SB128	2.45E+00	2.45E+00	2.42E+00	2.39E+00	2.27E+00	1.80E+00	1.14E+00	3.87E-01	3.87E-01	1.54E-01	6.10E-02	2.43E-02	9.64E-03	3.83E-03	1.52E-03	9.44E-07	3.64E-13
SB128M	9.67E+01	1.35E+02	3.66E+02	4.69E+02	4.02E+02	5.02E+01	7.31E-01	3.79E-05	3.78E-05	8.03E-09	1.70E-12	3.61E-16	7.65E-20	1.62E-23	3.44E-27	0.00E+00	0.00E+00
TE128	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I128	5.08E-03	4.94E-03	3.85E-03	2.92E-03	9.62E-04	6.52E-06	3.00E-10	2.27E-20	2.27E-20	4.79E-29	0.00E+00						
XE128	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CD129	1.09E-02	0.00E+00															
IN129	3.32E+03	0.00E+00															
SN129	4.06E+03	3.71E+03	1.61E+03	6.40E+02	1.59E+01	9.46E-07	3.36E-21	0.00E+00									
SN129M	9.61E+03	7.29E+03	6.01E+02	3.76E+01	5.73E-04	1.21E-25	0.00E+00										
SB129	5.03E+01	8.30E+01	2.04E+02	2.32E+02	2.25E+02	1.40E+02	5.33E+01	5.64E+00	5.63E+00	8.22E-01	1.20E-01	1.75E-02	2.55E-03	3.71E-04	5.41E-05	1.10E-11	4.59E-25
TE129	2.07E-01	7.84E-01	1.27E+01	2.98E+01	8.65E+01	1.36E+02	6.26E+01	6.83E+00	6.83E+00	1.10E+00	2.58E-01	1.35E-01	1.16E-01	1.12E-01	1.10E-01	1.01E-01	8.59E-02
TE129M	7.85E-05	2.04E-04	2.90E-03	7.05E-03	2.45E-02	8.48E-02	1.45E-01	1.76E-01	1.76E-01	1.78E-01	1.76E-01	1.74E-01	1.73E-01	1.71E-01	1.69E-01	1.56E-01	1.32E-01
I129	5.65E-15	4.53E-14	4.65E-12	2.25E-11	2.25E-10	2.17E-09	5.14E-09	6.92E-09	6.92E-09	7.11E-09	7.15E-09	7.16E-09	7.18E-09	7.19E-09	7.20E-09	7.27E-09	7.41E-09
XE129	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE129M	8.61E-17	8.61E-17	8.61E-17	8.61E-17	8.61E-17	8.61E-17	8.42E-17	8.00E-17	8.00E-17	7.65E-17	7.32E-17	7.00E-17	6.70E-17	6.41E-17	6.13E-17	4.37E-17	2.15E-17
CD130	3.17E+03	0.00E+00															
IN130	8.63E+03	0.00E+00															
SN130	3.56E+04	2.95E+04	5.52E+03	8.57E+02	4.97E-01	1.35E-15	0.00E+00										
SB130	6.60E+02	6.49E+02	5.55E+02	4.67E+02	2.33E+02	1.03E+01	2.01E-02	9.60E-09	9.59E-09	3.66E-14	1.40E-19	5.33E-25	2.03E-30	6.59E-36	0.00E+00	0.00E+00	0.00E+00
SB130M	8.66E+03	1.11E+04	1.20E+04	5.41E+03	8.07E+01	2.04E-07	1.28E-24	0.00E+00									
TE130	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I130	2.62E-02	2.69E-02	3.07E-0														

OUTPUT UNIT = 11 PAGE 185  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
I130M	8.81E-01	8.16E-01	4.08E-01	1.89E-01	8.67E-03	8.27E-09	7.52E-21	0.00E+00									
XE130	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CD131	2.29E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN131	3.70E+02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN131	7.70E+04	3.98E+04	1.05E+02	1.43E-01	4.87E-13	0.00E+00											
SB131	1.01E+04	1.15E+04	1.02E+04	7.54E+03	2.26E+03	9.95E+00	1.93E-04	1.96E-15	1.96E-15	7.40E-25	2.75E-34	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE131	6.03E+02	8.64E+02	3.01E+03	4.25E+03	3.86E+03	8.99E+01	5.04E+00	3.64E+00	3.64E+00	2.76E+00	2.09E+00	1.58E+00	1.20E+00	9.10E-01	6.89E-01	7.50E-02	8.88E-04
TE131M	1.62E+01	1.64E+01	1.91E+01	2.13E+01	2.55E+01	2.56E+01	2.23E+01	1.62E+01	1.62E+01	1.23E+01	9.28E+00	7.03E+00	5.33E+00	4.04E+00	3.06E+00	3.33E-01	3.95E-03
I131	5.97E-02	1.04E-01	1.20E+00	3.43E+00	1.41E+01	2.64E+01	2.66E+01	2.62E+01	2.62E+01	2.57E+01	2.51E+01	2.43E+01	2.36E+01	2.28E+01	2.20E+01	1.59E+01	8.01E+00
XE131	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE131M	2.53E-06	2.56E-06	4.80E-06	1.49E-05	1.74E-04	2.06E-03	6.30E-03	1.59E-02	1.59E-02	2.37E-02	3.11E-02	3.81E-02	4.46E-02	5.07E-02	5.64E-02	8.79E-02	1.02E-01
CD132	1.52E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN132	1.08E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN132	6.77E+04	2.39E+04	2.07E+00	6.31E-05	5.47E-23	0.00E+00											
SB132	6.02E+04	5.60E+04	6.84E+03	5.76E+02	2.88E-02	1.28E-21	0.00E+00										
SB132M	2.34E+04	1.98E+04	4.49E+03	8.62E+02	1.17E+00	1.47E-13	0.00E+00										
TE132	4.68E+01	5.87E+01	1.06E+02	1.13E+02	1.13E+02	1.10E+02	1.05E+02	9.25E+01	9.25E+01	8.32E+01	7.48E+01	6.72E+01	6.04E+01	5.43E+01	4.89E+01	2.09E+01	3.80E+00
I132	1.65E+01	1.67E+01	1.99E+01	2.44E+01	4.06E+01	8.29E+01	1.03E+02	9.52E+01	9.52E+01	8.57E+01	7.70E+01	6.93E+01	6.23E+01	5.60E+01	5.03E+01	2.15E+01	3.92E+00
XE132	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS132	1.20E-15	1.20E-15	1.20E-15	1.20E-15	1.19E-15	1.18E-15	1.15E-15	1.08E-15	1.08E-15	1.02E-15	9.64E-16	9.12E-16	8.63E-16	8.17E-16	7.73E-16	5.03E-16	2.15E-16
BA132	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN133	2.08E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN133	1.40E+04	7.25E-09	0.00E+00														
SB133	1.07E+05	8.01E+04	5.96E+03	3.32E+02	3.19E-03	0.00E+00											
TE133	1.43E+04	1.85E+04	2.15E+04	1.33E+04	1.89E+03	5.64E+01	6.23E-01	1.70E-05	1.70E-05	2.08E-09	2.54E-13	3.11E-17	3.81E-21	4.66E-25	5.71E-29	0.00E+00	0.00E+00
TE133M	6.66E+03	6.60E+03	5.96E+03	5.27E+03	3.19E+03	3.36E+02	3.72E+00	1.01E-04	1.01E-04	1.24E-08	1.52E-12	1.86E-16	2.27E-20	2.78E-24	3.40E-28	0.00E+00	0.00E+00
I133	2.03E+01	3.42E+01	1.75E+02	2.96E+02	4.94E+02	5.82E+02	4.90E+02	3.07E+02	3.07E+02	2.06E+02	1.38E+02	9.26E+01	6.21E+01	4.16E+01	2.79E+01	1.14E+00	1.90E-03
I133M	1.33E+04	1.31E+02	1.14E-16	0.00E+00													
XE133	1.17E-02	1.42E-02	9.78E-02	3.11E-01	1.80E+00	1.09E+01	2.75E+01	5.38E+01	5.38E+01	6.63E+01	7.29E+01	7.55E+01	7.57E+01	7.42E+01	7.18E+01	4.60E+01	1.63E+01
XE133M	7.43E-02	7.45E-02	8.03E-02	9.52E-02	2.00E-01	8.30E-01	1.95E+00	3.51E+00	3.51E+00	4.06E+00	4.18E+00	4.04E+00	3.77E+00	3.43E+00	3.07E+00	9.94E-01	8.07E-02
CS133	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA133	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
IN134	3.97E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN134	1.17E+03	4.91E-19	0.00E+00														
SB134	6.09E+04	1.39E+03	2.32E-12	8.83E-29	0.00E+00												
SB134M	5.80E+04	1.19E+03	7.64E-13	1.01E-29	0.00E+00												
TE134	2.00E+04	2.01E+04	1.74E+04	1.47E+04	7.57E+03	3.83E+02	9.77E-01	8.72E-07	8.71E-07	5.69E-12	3.72E-17	2.43E-22	1.58E-27	1.03E-32	0.00E+00	0.00E+00	0.00E+00
I134	1.38E+03	1.76E+03	4.13E+03	5.68E+03	7.64E+03	1.97E+03	2.62E+01	4.64E-04	4.64E-04	3.54E-08	2.68E-12	2.03E-16	1.54E-20	1.17E-24	8.84E-29	0.00E+00	0.00E+00
I134M	1.11E+04	9.18E+03	1.70E+03	2.61E+02	1.45E-01	3.30E-16	0.00E+00										
XE134	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE134M	2.44E+02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS134	4.41E-07	4.42E-07	4.58E-07	4.74E-07	5.34E-07	7.11E-07	8.39E-07	8.77E-07	8.77E-07	8.78E-07	8.78E-07	8.78E-07	8.77E-07	8.77E-07	8.76E-07	8.73E-07	8.67E-07
CS134M	2.74E-03	2.72E-03	2.63E-03	2.53E-03	2.15E-03	1.05E-03	2.51E-04	8.83E-06	8.82E-06	5.01E-07	2.85E-08	1.62E-09	9.19E-11	5.22E-12	2.96E-13	3.21E-23	0.00E+00
BA134	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SN135	6.32E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SB135	1.59E+04	3.79E-07	0.00E+00														
TE135	5.08E+05	5.84E+04	2.00E-04	7.82E-14	0.00E+00												
I135	1.55E+03	1.91E+03	1.93E+03	1.90E+03	1.77E+03	1.29E+03	6.88E+02	1.59E+02	1.59E+02	4.51E+01	1.28E+01	3.64E+00	1.03E+00	2.94E-01	8.35E-02	3.55E-06	6.43E-15
XE135	1.95E+01	2.31E+01	5.49E+01	8.56E+01	1.83E+02	4.57E+02	6.34E+02	4.27E+02	4.27E+02	2.20E+02	1.02E+02	4.49E+01	1.91E+01	7.98E+00	3.29E+00	2.31E-03	1.02E-09
XE135M	1.41E+03	1.36E+03	1.01E+03	7.47E+02	3.56E+02	2.07E+02	1.10E+02	2.54E+01	2.54E+01	7.22E+00	2.05E+00	5.83E-01	1.66E-01	4.71E-02	1.34E-02	5.69E-07	1.03E-15
CS135	3.09E-11	4.34E-11</															

OUTPUT UNIT = 11 PAGE 186  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER=1.00000E+00 MW, BURNUP=1.00000E+00 MWD, FLUX=1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
CS135M	5.69E-01	5.61E-01	4.99E-01	4.38E-01	2.60E-01	2.47E-02	2.22E-04	3.77E-09	3.76E-09	3.07E-13	2.50E-17	2.03E-21	1.65E-25	1.35E-29	1.09E-33	0.00E+00	0.00E+00
BA135	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA135M	2.75E-06	2.75E-06	2.74E-06	2.73E-06	2.68E-06	2.50E-06	2.16E-06	1.54E-06	1.54E-06	1.15E-06	8.62E-07	6.45E-07	4.83E-07	3.61E-07	2.70E-07	2.66E-08	2.57E-10
SN136	3.73E-01	0.00E+00															
SB136	5.57E+01	0.00E+00															
TE136	1.97E+05	2.72E+04	4.95E-04	1.24E-12	0.00E+00												
I136	1.50E+05	1.22E+05	1.44E+03	9.60E+00	1.90E-08	0.00E+00											
I136M	1.31E+05	5.29E+04	1.55E+01	1.83E-03	3.61E-19	0.00E+00											
XE136	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS136	3.64E-02	3.64E-02	3.64E-02	3.64E-02	3.63E-02	3.61E-02	3.56E-02	3.45E-02	3.45E-02	3.36E-02	3.27E-02	3.19E-02	3.10E-02	3.02E-02	2.94E-02	2.38E-02	1.56E-02
BA136	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA136M	5.99E-03	5.99E-03	5.99E-03	5.99E-03	5.98E-03	5.94E-03	5.86E-03	5.69E-03	5.69E-03	5.54E-03	5.39E-03	5.25E-03	5.12E-03	4.98E-03	4.85E-03	3.93E-03	2.57E-03
SB137	6.91E+02	0.00E+00															
TE137	6.65E+04	4.61E-01	0.00E+00														
I137	4.31E+05	8.16E+04	2.01E-02	9.15E-10	0.00E+00												
XE137	1.41E+05	1.51E+05	3.14E+04	5.14E+03	3.69E+00	2.63E-14	0.00E+00										
CS137	3.51E-03	1.02E-02	4.14E-02	4.78E-02	4.90E-02												
BA137	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
BA137M	5.62E+00	4.29E+00	4.02E-01	6.78E-02	4.64E-02	4.64E-02	4.64E-02	4.64E-02	4.64E-02	4.64E-02	4.63E-02						
SB138	1.18E-02	0.00E+00															
TE138	7.11E+03	6.92E-08	0.00E+00														
I138	2.33E+05	3.54E+02	0.00E+00														
XE138	5.23E+04	5.15E+04	3.32E+04	2.03E+04	2.88E+03	4.31E-01	9.70E-09	1.39E-26	1.39E-26	0.00E+00							
CS138	1.57E+03	2.77E+03	9.92E+03	1.31E+04	1.00E+04	2.55E+02	1.10E-01	1.54E-09	1.54E-09	2.87E-16	5.32E-23	9.88E-30	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CS138M	8.43E+03	6.64E+03	7.73E+02	7.08E+01	4.99E-03	1.03E-21	0.00E+00										
BA138	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
LA138	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17	5.55E-17
SB139	1.98E-03	0.00E+00															
TE139	1.65E+02	0.00E+00															
I139	9.82E+04	2.94E-03	0.00E+00														
XE139	5.75E+05	2.03E+05	1.55E+01	4.16E-04	2.13E-22	0.00E+00											
CS139	4.21E+04	6.45E+04	4.11E+04	1.96E+04	1.03E+03	1.77E-03	5.24E-15	0.00E+00									
BA139	2.55E+02	7.17E+02	4.88E+03	6.81E+03	6.54E+03	1.48E+03	7.22E+01	6.33E-02	6.32E-02	1.52E-04	3.63E-07	8.68E-10	2.08E-12	5.03E-15	0.00E+00	0.00E+00	0.00E+00
LA139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
CE139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PR139	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE140	9.90E+02	0.00E+00															
I140	1.14E+04	1.84E-17	0.00E+00														
XE140	5.80E+05	2.73E+04	3.04E-08	1.60E-21	0.00E+00												
CS140	4.06E+05	2.86E+05	8.32E+02	1.23E+00	5.83E-12	0.00E+00											
BA140	1.10E+01	2.47E+01	4.16E+01	4.16E+01	4.13E+01	4.07E+01	3.95E+01	3.95E+01	3.84E+01	3.74E+01	3.64E+01	3.54E+01	3.45E+01	3.36E+01	2.70E+01	1.75E+01	
LA140	2.69E-01	2.74E-01	3.74E-01	4.92E-01	9.62E-01	3.00E+00	6.74E+00	1.39E+01	1.39E+01	1.86E+01	2.22E+01	2.49E+01	2.70E+01	2.85E+01	2.95E+01	2.93E+01	2.01E+01
CE140	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PR140	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE141	1.70E-01	0.00E+00															
I141	8.96E+02	0.00E+00															
XE141	1.39E+05	4.41E-06	0.00E+00														
CS141	5.84E+05	1.13E+05	3.54E-02	2.11E-09	0.00E+00												
BA141	2.57E+04	3.54E+04	2.70E+04	1.85E+04	4.05E+03	4.38E+00	5.11E-06	7.34E-20	7.34E-20	1.00E-31	0.00E+00						
LA141	4.32E+01	1.37E+02	9.74E+02	1.60E+03	2.46E+03	1.65E+03	5.73E+02	4.85E+01	4.85E+01	5.84E+01	7.04E-01	8.48E-02	1.02E-02	1.23E-03	1.48E-04	6.59E-12	1.30E-26
CE141	2.41E-04	1.54E-03	7.90E-02														

OUTPUT UNIT = 11 PAGE 187  
 ORIGEN2 V2.5 (10-15-2002) for PC Linux, Run on 01/19/2003 at 13:30:55

\* Decay after HEU Accident of 4.0E19 Fissions RECYCLE # 0 FISSION PRODUCTS  
 POWER= 1.00000E+00 MW, BURNUP= 1.00000E+00 MWD, FLUX= 1.00E+00 N/CM\*\*2-SEC

7 NUCLIDE TABLE: RADIOACTIVITY, CURIES  
 10 kilograms HEU

	DISCHARGE	1.0MI	10.0MI	20.0MI	60.0MI	4.0HR	10.0HR	24.0HR	24.0HR	1.5D	2.0D	2.5D	3.0D	3.5D	4.0D	8.0D	16.0D
PR141	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ND141	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
TE142	6.76E-02	0.00E+00															
I142	1.47E+01	0.00E+00															
XE142	3.97E+04	6.25E-11	0.00E+00														
CS142	3.18E+05	9.97E-06	0.00E+00														
BA142	6.34E+04	6.03E+04	3.36E+04	1.76E+04	1.32E+03	1.14E-02	8.47E-13	0.00E+00									
LA142	3.75E+02	8.36E+02	3.74E+03	5.25E+03	5.43E+03	1.46E+03	9.87E+01	1.85E-01	1.85E-01	8.49E-04	3.90E-06	1.79E-08	8.20E-11	3.77E-13	1.68E-15	3.30E-34	0.00E+00
CE142	2.30E-15	9.94E-15	2.88E-13	8.66E-13	3.73E-12	1.07E-11	1.30E-11	1.31E-11									
PR142	2.55E-07	2.66E-07	3.45E-07	4.00E-07	4.70E-07	4.34E-07	3.49E-07	2.10E-07	2.10E-07	1.36E-07	8.81E-08	5.71E-08	3.69E-08	2.39E-08	1.55E-08	4.78E-10	4.55E-13
PR142M	1.91E-05	1.82E-05	1.19E-05	7.40E-06	1.11E-06	2.15E-10	8.14E-18	3.61E-35	3.61E-35	0.00E+00							
ND142	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I143	2.74E-01	0.00E+00															
XE143	5.79E+02	0.00E+00															
CS143	1.60E+05	3.81E-06	0.00E+00														
BA143	8.90E+05	4.29E+04	4.78E-08	2.51E-21	0.00E+00												
LA143	3.69E+04	4.87E+04	3.16E+04	1.93E+04	2.66E+03	3.59E-01	6.52E-09	5.65E-27	5.65E-27	0.00E+00							
CE143	7.40E+00	2.35E+01	1.49E+02	2.35E+02	3.49E+02	3.45E+02	3.04E+02	2.27E+02	2.27E+02	1.76E+02	1.37E+02	1.06E+02	8.27E+01	6.43E+01	5.00E+01	6.65E+00	1.18E-01
PR143	9.54E-05	6.33E-04	2.96E-02	9.91E-02	5.39E-01	2.80E+00	6.88E+00	1.44E+01	1.44E+01	1.91E+01	2.26E+01	2.50E+01	2.68E+01	2.79E+01	2.87E+01	2.72E+01	1.86E+01
ND143	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I144	5.19E-04	0.00E+00															
XE144	4.69E+02	4.10E-16	0.00E+00														
CS144	3.37E+04	9.23E-14	0.00E+00														
BA144	7.13E+05	1.64E+04	2.73E-11	1.04E-27	0.00E+00												
LA144	5.36E+05	2.79E+05	2.47E+01	7.52E-04	6.53E-22	0.00E+00											
CE144	4.70E-01	1.20E+00	1.66E+00	1.65E+00	1.65E+00	1.65E+00	1.65E+00	1.63E+00	1.60E+00								
PR144	2.05E-02	6.17E-02	5.81E-01	9.54E-01	1.52E+00	1.66E+00	1.66E+00	1.66E+00	1.66E+00	1.66E+00	1.66E+00	1.65E+00	1.65E+00	1.65E+00	1.65E+00	1.63E+00	1.60E+00
PR144M	2.10E-01	1.91E-01	9.21E-02	4.75E-02	2.06E-02	2.00E-02	2.00E-02	1.99E-02	1.99E-02	1.99E-02	1.99E-02	1.99E-02	1.98E-02	1.98E-02	1.98E-02	1.96E-02	1.92E-02
ND144	6.31E-24	3.08E-23	1.95E-21	6.86E-21	3.99E-20	2.26E-19	6.02E-19	1.48E-18	1.48E-18	2.23E-18	2.98E-18	3.73E-18	4.47E-18	5.22E-18	5.96E-18	1.19E-17	2.36E-17
I145	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE145	5.18E+00	4.45E-20	0.00E+00														
CS145	3.03E+03	1.18E-19	0.00E+00														
BA145	3.04E+05	3.72E+02	0.00E+00														
LA145	4.97E+05	1.38E+05	3.43E-01	2.03E-07	0.00E+00												
CE145	6.64E+04	1.12E+05	1.73E+04	1.72E+03	1.67E-01	1.45E-19	0.00E+00										
PR145	4.80E+01	2.36E+02	1.24E+03	1.34E+03	1.25E+03	8.85E+02	4.42E+02	8.72E+01	8.72E+01	2.17E+01	5.40E+00	1.34E+00	3.35E-01	8.33E-02	2.07E-02	3.05E-07	6.61E-17
ND145	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PM145	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
SM145	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE146	7.84E-01	4.22E-20	0.00E+00														
CS146	1.76E+01	5.30E-20	0.00E+00														
BA146	1.13E+05	6.97E-04	0.00E+00														
LA146	3.92E+05	2.89E+03	7.51E-17	0.00E+00													
CE146	2.15E+04	2.45E+04	1.58E+04	9.72E+03	1.38E+03	2.11E-01	4.92E-09	7.66E-27	7.66E-27	0.00E+00							
PR146	2.98E+02	9.76E+02	5.23E+03	7.01E+03	4.66E+03	3.79E+01	1.27E-03	4.51E-14	4.51E-14	5.00E-23	5.53E-32	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
ND146	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
PM146	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19	2.54E-19
SM146	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
XE147	1.33E-02	0.00E+00															
CS147	8.92E+01	0.00E+00															
BA147	3.03E+04	2.36E-04	0.00E+00	0.0													

## APPENDIX E

### THE HAYDEN DAILY RELEASE NOTEGRAMS

[Appendix E](#) gives a sampling of the record of the radioiodine release reports compiled by R.E. Hayden of the Health Physics Department of the Idaho Chemical Processing Plant (ICPP) ([Doc MC 6011](#); also Doc MC 55203, a duplicate) from February 1957 through December 1959. They were compiled from the [Stack Monitor Datasheets \(Doc MC 60312\)](#), as shown by the spot checks outlined in [Chapter 5](#).

The [Hayden Notegrams](#) were the source of the Health and Safety Division release summaries (e.g., [IDO-12014](#)), which were tabulated as releases per Radioactive Lanthanum (RaLa) run. In turn, these iodine release tables were reproduced in the [DOE Report \(DOE 1991\)](#) directly from the Health and Safety Division Annuals.

[Appendix E](#) contains a sampling of the record for illustration. Although the Notegrams begin for the first week of February 1957, [Appendix E](#) displays the first easily legible entry, that for the weeks of April 2–6, April 7–13, April 14–20, and April 21–27, 1957. Note that only weekly totals of I-133 and ( $\beta$ -I) are given, obtained by summing the daily releases given in the Stack Monitor Datasheets.

This format continued until July 7, 1957, when daily releases began to be listed. The Notegram from this initial date is the second entry in this appendix.

On May 25, 1958, the format changed to include daily releases of I-132. The May 25, 1958, report is shown in this appendix. Note that [Chapter 5](#) cited March 1, 1958, as the initial date of I-132. Thus, 14 days of I-132 releases are missing from the Notegrams. The changed format was retained through December 30, 1959, shown as the last entry in this appendix.

INTER-OFFICE CORRESPONDENCE

**PHILLIPS PETROLEUM COMPANY**

SUBJECT

Idaho Falls, Idaho

January 5, 1960

Activity Discharged to Atmosphere  
Ha-4-60A-N

## NOTEGRAM

To: A. L. Ayers

From: R. E. Hayden:wt/DKJ 

According to calculations based on chemical analysis of samples of scrubber solution from the Stack-Gas-Monitor, activity was discharged to the atmosphere via the CPP Stack during the week of December 27, 1959 through January 2, 1960, as follows:

<u>Date</u>	<u>Day</u>	<u>Approximate</u> <u><math>^{132}\text{I}</math> mc</u>	<u>Approximate</u> <u><math>^{131}\text{I}</math> mc</u>	<u>Gross Beta emitters</u> <u>other than Iodine</u> <u>(<math>\beta</math>-I) mc</u>	<u>a mc</u>
12-27-59	S	600	10	240	0.68
12-28-59	M	0	10	80	0
12-29-59	T	0	10	50	0
12-30-59	W	0	20	80	0
12-31-59	T	0	0	90	0
1-1-60	F	0	10	90	0
1-2-60	S	0	0	50	0
	total (curies)				
		0.6	0.06	0.68	$6.8(10^{-4})$

cc: R. B. Lemon  
S. F. Fairbourne  
R. E. Commander  
I. J. Wells  
D. K. Jenson  
R. E. Hayden  
File

OKIT

Idaho Falls, Idaho

October 21, 1957

Activity Discharged to Atmosphere  
Ha-216-57A-8

## MEMORANDUM

To: E. G. Reid  
From: R. E. Hayden/ELR

According to calculations, based on chemical analyses of the Stack Wash-Water Scrubber solution samples, activity was discharged to the atmosphere via the CPP stack during the week of October 13, 1957 through October 19, 1957 as follows:

<u>Date</u>	<u>Day</u>	<u>mc of I-131 Released</u>	<u>mc of Particulate <math>\beta</math> Emitters Other Than I (<math>\beta</math>-I) Released</u>
10/13/57	S	6,200	808
10/14/57	M	9,580	772
10/15/57	T	7,590	573
10/16/57	W	13,680	523
10/17/57	Th	7,500	562
10/18/57	F	2,660	612
10/19/57	S	1,900	338
Total (curies)		49.1	4.2

cc: A. L. Ayers  
E. L. Sexton  
S. F. Fairbourne  
E. L. Rich  
R. E. Hayden  
MF File

Original signed by  
R. E. Hayden

Idaho Falls, Idaho

December 2, 1957

OMIT?

Activity Discharged to Atmosphere  
Ha-241-57

**MEMORANDUM**

To: D. G. Reid  
From: R. E. Hayden:hw/BLR

According to calculations, based on chemical analyses of samples of the filter solution from the Stack-Gas-Monitor, activity was discharged to the atmosphere via the CPP stack during the week of November 24, 1957 through November 30, 1957 as follows:

<u>Date</u>	<u>Day</u>	<u>1131 mc</u>	<u>Particulate <math>\beta</math> Emitters Other Than I (<math>\beta</math>-I) mc</u>
11/24/57	S	937	624
11/25/57	M	396	423
11/26/57	T	288	326
11/27/57	W	254	787
11/28/57	Th	300	620
11/29/57	F	176	835
11/30/57	S	125	844
Total	(series)	2.5	4.7

cc: A. L. Ayers  
E. L. Sexton  
S. F. Fairbourne  
B. L. Rich  
R. E. Hayden  
HP file

Original signed by  
R. E. Hayden

Idaho Falls, Idaho

April 30, 1957

Activity Discharged to Atmosphere  
Ha-83-57A-N

## NOTEGRAM

To: D. G. Reid  
From: E. E. Hayden:rb

According to calculations, based on chemical analyses of the Stack-Gas-Monitor scrubber solution samples, activity was discharged to the atmosphere as follows:

Time Interval	Curies of I <sup>131</sup> Released	Curies of B emitters other than I <sup>131</sup> (B-I <sup>131</sup> ) released
Week of 4-2-57 - 4-6-57	3.5	0.3
Week of 4-7-57 - 4-13-57	8.9	0.3
Week of 4-14-57 - 4-20-57	36.1	0.5
Week of 4-21-57 - 4-27-57	<u>20.3</u>	<u>0.1</u>
Total Activity Released:	68.8	1.2

The last report covered the period through March 30, 1957. Due to modifications being made on the sampling system, no samples were taken on March 31, 1957 and April 1, 1957.

cc: A. L. Ayers  
E. L. Rich  
E. E. Hayden  
EP File

Idaho Falls, Idaho

May 13, 1957

Activity Discharged to Atmosphere  
Id-96-574-B

**NOTEGRAM**

Re: B. G. Reid  
From: R. E. Hayden:la/ER

According to calculations, based on chemical analyses of the Stack-Gas-Weather Scrubber solution samples, 2.7 curies of  $I^{131}$  activity and 0.1 curies of beta emitters other than Iodine were discharged to the atmosphere via the CFP stack during the week of May 5 through May 11, 1957.

cc: A. L. Ayers  
R. B. Lemon  
R. L. Rich  
R. E. Hayden

ER-File

Original signed by  
R. E. Hayden

Idaho Falls, Idaho

May 26, 1958

Activity Discharged to Atmosphere  
Ka-95-58A-B**MEMORANDUM**To: D. C. Reid  
From: R. E. Hayden:hr/MLB

According to calculations, based on chemical analysis of samples of scrubber solution from the Stack Gas Monitor, activity was discharged to the atmosphere via the GPP stack during the week of May 18, 1958 through May 24, 1958 as follows:

<u>Date</u>	<u>Day</u>	<u>131 mc</u>	<u>Particulate Beta Emitters Other Than I (S-I) mc</u>
5/18/58	S	7,840	580
5/19/58	M	6,000	657
5/20/58	T	5,480	885
5/21/58	W	2,900	990
5/22/58	Th	3,140	1,200
5/23/58	F	1,780	925
5/24/58	S	1,500	658
Totals (curies)		28.6	5.9

cc: A. L. Ayers  
E. L. Sexton  
S. F. Fairbourne  
B. L. Rich  
R. E. Hayden  
MF FileOriginal signed by  
R E Hayden

Idaho Falls, Idaho

June 3, 1958

Activity Discharged to Atmosphere  
IN-100-52A-B**NOTEGRAM**To: D. C. Reid  
From: E. E. Hayden:hw/ELR

According to calculations, based on chemical analysis of samples of scrubber solution from the Stack Gas Monitor, activity was discharged to the atmosphere via the CPP stack during the week of May 25, 1958 through May 31, 1958 as follows:

Date	Day	Approximate <u><math>^{132}</math> mc</u>	<u><math>^{131}</math> mc</u>	Approximate Particulate Beta Emitters Other Than I ( $\beta$ -I) mc
5/25/58	S	0	1,300	330
5/26/58	M	0	3,320	390
5/27/58	T	0	2,870	355
5/28/58	W	835,000*	49,500*	17,500*
5/29/58	Th	50,000	33,400	6,330
5/30/58	F	26,000	14,000	2,860
5/31/58	S	<u>13,000</u>	<u>6,350</u>	<u>1,160</u>
Totals (curies)		924	110.7	29.0

\* Data run No. 158P

cc: A. L. Ayers  
E. L. Sexton  
S. F. Fairbourne  
B. L. Rich  
E. E. Hayden  
HP FileOriginal signed by  
E. E. Hayden

Idaho Falls, Idaho

July 18, 1957

Activity Discharged to Atmosphere  
EM-141-57A-B

**MEMORANDUM**

To: D. G. Reid  
 From: R. E. Hayden:hw/ELB

According to calculations, based on chemical analyses of the Stack-Top-Monitor Scrubber solution samples, activity was discharged to the atmosphere via the GPP stack during the week of July 7, 1957 through July 13, 1957 as follows:

<u>Date</u>	<u>Day</u>	<u>mc of I<sup>131</sup> Released</u>	<u>mc of <math>\beta</math> Emitters other than I (<math>\beta</math>-I) Released</u>
7/7/57	Sunday	812	101
7/8/57	Monday	576	62
7/9/57	Tuesday	372	60
7/10/57	Wednesday	304	73
7/11/57	Thursday	254	56
7/12/57	Friday	186	57
7/13/57	Saturday	155	45
	Totals (curies)	2.7	0.9

PM

cc: A. L. Ayers  
 E. L. Sexton  
 H. F. Fairbourne  
 B. L. Kish  
 R. E. Hayden  
 HW File

Original signed by  
 R. E. Hayden

Idaho Falls, Idaho

August 18, 1958

Activity Discharged to Atmosphere  
EA-147-53A-B

**NOTICE**

To: D. G. Reid  
 From: R. E. Hayden/ELR

According to calculations, based on chemical analysis of samples of gaseous solution from the Stack Gas Monitor, activity was discharged to the atmosphere via the GEF Stack during the week of August 10, 1958 through August 16, 1958 as follows:

<u>Date</u>	<u>Day</u>	<u>Approximate</u> <u>133 mc</u>	<u>131 mc</u>	<u>Approximate Particulate Beta</u> <u>Emitters Other Than I (S-I) mc</u>
8/10/58	S	0	150	1000
8/11/58	M	5600	12800	4020
8/12/58	T	800	13600	1570
8/13/58	W	3700	8300	2370
8/14/58	Th	0	6100	3000
8/15/58	F	3700	4670	2100
8/16/58	S	0	3190	2300
Total (curies)		13.8	48.8	16.4

cc: A. L. Ayers  
 H. L. Sexton  
 S. F. Fairbourne  
 H. L. Rich  
 R. E. Hayden  
 SF File

Original signed by  
 R. E. Hayden

---

## APPENDIX F

### DAILY RELEASES OF IODINE-131, IODINE-132, AND IODINE-133 FROM FEBRUARY 1, 1957, TO DECEMBER 30, 1959— DETERMINISTIC VALUES

[Appendix F](#) lists tables of I-131, I-132, and I-133 daily releases from February 1, 1957, the first day of Radioactive Lanthanum (RaLa) operation, through December 30, 1959, the last day of the period of interest. RaLa run numbers, from Run 1 through Run 37, are given adjacent to the appropriate dates.

The [Appendix F](#) tables were prepared as follows:

- (1) The [Stack Monitor Datasheets \(Doc MC 60312\)](#) were used as the primary source.
- (2) Spot-checking the [Hayden Notegrams \(Doc MC 60111](#) or its duplicate, Doc MC 55203), the first documentation of the releases, indicated they faithfully recorded releases from the datasheets. However, there were several significant omissions from the Notegrams as follows.
  - Daily reports for I-131 did not begin until June 7, 1957; earlier Notegrams, February 1 to June 7, consisted of weekly summaries.
  - Numerous Notegrams are missing from the record.
  - The Notegrams did not record daily releases of I-132 until May 28, 1958, whereas consistent daily readings began May 1, 1958.
- (3) Omissions of I-131 releases in the Stack Monitor Datasheets were filled in per the method described in Section 5.3. The most significant omissions occurred in the first two months of operation, February and March 1957.
- (4) The record of I-132 daily releases was extended back from May 1, 1958, the first entry in the datasheets, to the beginning, February 1, 1957, using a method developed in [Section 6.3](#).
- (5) The daily release record for I-133 was calculated from the correlation developed in [Section 6.4](#).
- (6) The daily releases for October 1959 include I-131, I-132, and I-133 releases from the Idaho Chemical Processing Plant criticality event on October 16. As described in [Chapter 7](#), the listed I-131 releases were those recorded by the Stack Monitor Datasheets. Iodine-132 and I-133 releases were estimated from the criticality release model described in [Chapter 7](#).

**Daily Releases of I-131, I-132, I-133 (mCi)**

Draft 1 Version; January, 2003

Day	run #	Feb-57			run #	Mar-57		
		I-131	I-132	I-133		I-131	I-132	I-133
1	<b>run 1</b>	71,200	1,008,192	42,744		4,840	4,961	10
2	<b>run 1</b>	56,100	351,545	16,494	(No runs	15,540	8,000	15
3	<b>run 1</b>	32,800	66,122	4,723	in	13,200	0	6
4		9,590	1,711	676	March)	10,820	0	3
5		24,800	0	856		2,050	0	0
6		27,100	4,027	458		1,360	0	0
7		18,400	13,498	152		1,910	0	0
8		20,000	24,532	81		1,850	941	0
9		44,500	64,813	88		1,235	1,124	0
10		28,900	39,725	28		824	579	0
11		26,400	27,060	13		1,360	0	0
12		6,600	3,398	2		777	0	0
13		1,900	0	0		10,900	0	0
14		2,100	0	0		6,950	0	0
15		2,100	0	0		5,175	0	0
16		2,300	0	0		5,175	0	0
17		2,410	0	0		3,400	0	0
18		2,000	1,017	0		1,270	0	0
19		2,200	2,002	0		1,200	0	0
20	<b>run 2</b>	7,000	99,120	4,202		523	0	0
21	<b>run 2</b>	51,600	323,346	15,171		705	0	0
22		10,700	21,570	1,541		412	0	0
23		10,700	1,909	754		470	0	0
24		10,700	0	369		1,330	0	0
25		96,600	14,354	1,634		2,190	0	0
26		46,800	34,332	388		399	0	0
27		27,300	33,486	111		227	0	0
28		18,300	26,653	36		227	0	0
29						227	0	0
30						227	0	0
31						227	0	0
<b>TOTAL (mCi)</b>		<b>661,100</b>	<b>2,162,412</b>	<b>90,522</b>	<b>TOTAL (mCi)</b>	<b>97,000</b>	<b>15,604</b>	<b>34</b>
<b>TOTAL (Ci)</b>		<b>661</b>	<b>2162</b>	<b>91</b>	<b>TOTAL (Ci)</b>	<b>97</b>	<b>16</b>	<b>0.034</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Apr-57			run #	May-57		
		I-131	I-132	I-133		I-131	I-132	I-133
1		54	0	0		1,100	0	0
2		28	0	0		1,770	0	0
3		31	0	0		1,325	0	0
4		909	0	0		537	0	0
5	run 3	1,787	25,304	1,073		619	0	0
6	run 3	920	5,765	270		565	0	0
7		671	1,353	97		514	0	0
8		378	67	27		304	0	0
9		4,170	0	144		294	0	0
10		529	79	9		200	0	0
11		465	341	4		146	0	0
12		1,080	1,325	4		384	0	0
13		4,770	6,947	9		512	0	0
14		3,180	4,371	3		541	0	0
15		4,910	5,033	2		250	0	0
16		4,460	2,296	1		475	0	0
17		13,130	0	2		377	0	0
18		4,470	0	0		236	0	0
19		3,440	0	0	run 4	654	9,261	393
20		6,980	0	0	run 4	7,334	45,958	2,156
21		7,270	0	0		2,436	4,911	351
22		3,210	1,632	0		8,561	1,527	604
23		1,215	1,106	0		5,300	0	183
24		291	204	0		1,675	249	28
25		455	0	0		995	730	8
26		843	0	0		2,210	2,711	9
27		808	0	0		1,217	1,773	2
28		773	0	0		1,410	1,938	1
29		2,828	0	0		825	846	0
30		1,670	0	0		800	412	0
31						783	0	0
<b>TOTAL (mCi)</b>		<b>75,725</b>	<b>55,823</b>	<b>1,646</b>	<b>TOTAL (mCi)</b>	<b>44,349</b>	<b>70,314</b>	<b>3,736</b>
<b>TOTAL (Ci)</b>		<b>76</b>	<b>56</b>	<b>2</b>	<b>TOTAL (Ci)</b>	<b>44</b>	<b>70</b>	<b>3.7</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Jun-57			run #	Jul-57		
		I-131	I-132	I-133		I-131	I-132	I-133
1		685	0	0		4,470	5,483	18
2		650	0	0	(no runs in July)	3,195	4,653	6
3		603	0	0		2,130	2,928	2
4		527	0	0		4,040	4,141	2
5		447	227	0		1,250	643	0
6		1,130	1,028	0		632	0	0
7		650	457	0	812	0	0	
8		225	0	0	576	0	0	
9		168	0	0	372	0	0	
10		95	0	0	304	0	0	
11		182	0	0	254	129	0	
12		104	0	0	186	169	0	
13		73	0	0	155	109	0	
14		620	0	0	132	0	0	
15		285	0	0	205	0	0	
16		285	0	0	3,745	0	0	
17		289	0	0	108	0	0	
18		249	0	0	52	0	0	
19		309	0	0	60	0	0	
20		109	0	0	61	0	0	
21		113	0	0	113	0	0	
22		33	0	0	75	0	0	
23		28	0	0	68	0	0	
24	run 5	2,070	29,311	1,243	74	0	0	
25	run 5	3,350	20,992	985	76	0	0	
26		264	532	38	51	0	0	
27		813	145	57	23	0	0	
28		86,000	0	2,970	16	0	0	
29		33,200	4,933	561	16	0	0	
30		9,460	6,940	78	9	0	0	
31					16	0	0	
<b>TOTAL (mCi)</b>		<b>143,016</b>	<b>64,566</b>	<b>5,933</b>	<b>TOTAL (mCi)</b>	<b>23,276</b>	<b>18,256</b>	<b>29</b>
<b>TOTAL (Ci)</b>		<b>143</b>	<b>65</b>	<b>6</b>	<b>TOTAL (Ci)</b>	<b>23</b>	<b>18</b>	<b>0.029</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Aug-57			run #	Sep-57		
		I-131	I-132	I-133		I-131	I-132	I-133
1		1	0	0		24	0	0
2	(no runs	1	0	0		14	0	0
3	in	9	0	0		17	0	0
4	August)	7	0	0		17	0	0
5		3	0	0		2	0	0
6		8	0	0		21	0	0
7		6	0	0		17	0	0
8		6	0	0		22	0	0
9		14	0	0		4	0	0
10		3	0	0		29	0	0
11		4	0	0	run 6	3,963	56,116	2,379
12		6	0	0	(a 1-day	4,200	26,319	1,235
13		5	0	0	run)	2,560	5,161	369
14		3	0	0		2,215	395	156
15		2	0	0		3,690	0	127
16		0	0	0		1,810	269	31
17		8	0	0		1,730	1,269	14
18		8	0	0		966	1,185	4
19		8	0	0		628	915	1
20		9	0	0		953	1,310	1
21		3	0	0		685	702	0
22		0	0	0		1,123	578	0
23		0	0	0		1,550	0	0
24		0	0	0		1,020	0	0
25		5	0	0		1,450	0	0
26		7	0	0		1,530	0	0
27		3	0	0		327	0	0
28		1	0	0		1,150	585	0
29		0	0	0		1,023	931	0
30		6	0	0		913	641	0
31		8	0	0				
<b>TOTAL (mCi)</b>		<b>144</b>	<b>0</b>	<b>0</b>	<b>TOTAL (mCi)</b>	<b>33,653</b>	<b>96,376</b>	<b>4,318</b>
<b>TOTAL (Ci)</b>		<b>0</b>	<b>0</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>34</b>	<b>96</b>	<b>4.3</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Oct-57			run #	Nov-57		
		I-131	I-132	I-133		I-131	I-132	I-133
1		942	0	0		15,600	8,031	4
2		898	0	0	(no runs	4,320	0	0
3		19,100	0	0	in Nov.)	3,520	0	0
4		19,840	0	0		8,830	0	0
5		9,900	0	0		7,350	0	0
6		19,200	0	0		10,100	0	0
7	<b>run 7</b>	40,600	574,896	24,374		16,200	8,239	0
8	(1-day	12,400	77,703	3,646		31,400	28,578	0
9	run)	9,720	19,595	1,400		19,600	13,769	0
10		4,070	726	287		17,600	0	0
11		3,740	0	129		23,200	0	0
12		5,110	759	86		11,150	0	0
13		6,200	4,548	51		14,370	0	0
14		9,580	11,751	39		7,300	0	0
15		7,550	10,996	15		6,560	0	0
16		13,680	18,804	13		8,360	0	0
17		7,500	7,687	4		4,530	0	0
18		2,660	1,369	1		4,080	0	0
19		1,900	0	0		2,430	0	0
20		1,450	0	0		3,240	0	0
21	<b>run 8</b>	1,535	21,736	922		1,420	0	0
22	(1-day	13,800	86,476	4,057		1,055	0	0
23	run)	25,800	52,011	3,715		1,150	0	0
24		12,800	2,284	903		937	0	0
25		7,050	0	243		396	0	0
26		2,850	423	48		288	0	0
27		2,050	1,504	17		254	0	0
28		2,610	3,201	11		300	0	0
29		3,390	4,937	7		176	0	0
30		3,600	4,948	4		125	0	0
31		6,220	6,375	3				
<b>TOTAL (mCi)</b>		<b>277,745</b>	<b>912,732</b>	<b>39,973</b>	<b>TOTAL (mCi)</b>	<b>225,841</b>	<b>58,616</b>	<b>5</b>
<b>TOTAL (Ci)</b>		<b>278</b>	<b>913</b>	<b>40</b>	<b>TOTAL (Ci)</b>	<b>226</b>	<b>59</b>	<b>0.005</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Dec-57			run #	Jan-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1		155	0	0		30	0	0
2	(no runs	87	0	0		18	0	0
3	in	269	0	0		12	0	0
4	Dec.)	151	0	0		8	0	0
5		171	0	0		65	0	0
6		109	0	0	<b>run 9</b>	3,500	49,560	2,101
7		97	0	0	(1-day	4,000	25,066	1,176
8		98	0	0	run)	2,600	5,241	374
9		131	0	0		1,780	318	126
10		339	0	0		2,460	0	85
11		199	0	0		2,520	374	43
12		121	0	0		2,260	1,658	19
13		94	0	0		1,880	2,306	8
14		52	0	0		1,045	1,522	2
15		68	0	0		550	756	1
16		59	0	0		930	953	0
17		41	0	0		677	349	0
18		42	0	0		413	0	0
19		29	0	0		398	0	0
20		9	0	0		138	0	0
21		31	0	0		76	0	0
22		22	0	0		19	0	0
23		12	0	0		15	8	0
24		30	0	0		108	98	0
25		13	0	0		512	360	0
26		15	0	0		456	0	0
27		9	0	0		773	0	0
28		13	0	0		700	0	0
29		5	0	0		913	0	0
30		41	0	0		805	0	0
31		0	0	0		845	0	0
<b>TOTAL (mCi)</b>		<b>2,512</b>	<b>0</b>	<b>0</b>	<b>TOTAL (mCi)</b>	<b>30,506</b>	<b>88,568</b>	<b>3,934</b>
<b>TOTAL (Ci)</b>		<b>3</b>	<b>0</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>31</b>	<b>89</b>	<b>3.9</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Feb-58			run #	Mar-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1		601	0	0		48,200	24,512	0
2		511	0	0		14,700	13,379	0
3		726	0	0		11,030	7,748	0
4		446	0	0		3,030	0	0
5		470	0	0		2,000	0	0
6		292	0	0		927	0	0
7		290	0	0		1,340	0	0
8		293	0	0		1,000	0	0
9		376	0	0		740	0	0
10		457	0	0		557	0	0
11		352	0	0		258	0	0
12	<b>run 10</b>	3,000	42,480	1,801		258	0	0
13	(1-day	7,430	46,559	2,184	<b>run 11</b>	1,290	18,266	774
14	run)	5,540	11,168	798	<b>run 11</b>	7,400	46,371	2,176
15		6,170	1,101	435		2,960	5,967	426
16		5,010	0	173		2,480	442	175
17		2,330	346	39		1,540	0	53
18		1,470	1,078	12		1,510	224	26
19		1,580	1,938	6		2,350	1,724	19
20		15,280	22,255	30		4,220	5,176	17
21		694	954	1		3,200	4,661	6
22		657	673	0		1,700	2,337	2
23		627	323	0		2,520	2,583	1
24		630	0	0		2,060	1,060	0
25		472	0	0		1,750	0	0
26		392	0	0		1,880	0	0
27		331	0	0		2,260	0	0
28		18,900	0	0		2,420	0	0
29						1,310	0	0
30						1,230	646	0
31						1,590	1,447	0
<b>TOTAL (mCi)</b>		<b>75,327</b>	<b>128,876</b>	<b>5,481</b>	<b>TOTAL (mCi)</b>	<b>129,710</b>	<b>136,545</b>	<b>3,677</b>
<b>TOTAL (Ci)</b>		<b>75</b>	<b>129</b>	<b>5</b>	<b>TOTAL (Ci)</b>	<b>130</b>	<b>137</b>	<b>3.7</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Apr-58			run #	May-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1		1,725	1,212		<b>run 13</b>	5,850	488,000	1,720
2		2,020	0			3,180	11,500	458
3		755	0			1,830	7,500	129
4		595	0			1,980	20,800	68
5		486	0			2,250	20,400	38
6		1,120	0			3,620	11,200	30
7		683	0			1,960	8,000	8
8		823	0			1,720	22,200	3
9		312	0			1,750	7,300	2
10		363	0			678	4,000	0
11		280	0			567	600	0
12		260	0			18,600	7,800	2
13		170	0			39,300	3,900	2
14		195	0			20,500	0	1
15		150	0			12,900	0	0
16	<b>run 12</b>	3,375	47,790	2,026		10,800	4,040	0
17	(1-day	6,560	41,108	1,929		8,780	600	0
18	run)	7,120	14,353	1,025		7,840	160	0
19		3,420	610	241		6,000	430	0
20		2,490	0	86		5,480	1,760	0
21		2,380	354	40		2,900	0	0
22		995	730	8		3,140	0	0
23		1,410	1,729	6		1,780	0	0
24		1,700	2,476	3		1,500	0	0
25		1,260	1,732	1		1,300	0	0
26		993	1,018	0		3,320	0	0
27		956	492	0		2,870	0	0
28		2,000	0	0	<b>run 14</b>	49,500	835,000	29,717
29		276	0	0	(1-day	33,400	50,000	9,820
30	<b>run 13</b>	4,220	59,755	2,533	run)	14,000	26,000	2,016
31						6,350	13,000	448
<b>TOTAL (mCi)</b>		<b>49,092</b>	<b>173,359</b>	<b>7,900</b>	<b>TOTAL (mCi)</b>	<b>275,645</b>	<b>1,544,190</b>	<b>44,462</b>
<b>TOTAL (Ci)</b>		<b>49</b>	<b>173</b>	<b>8</b>	<b>TOTAL (Ci)</b>	<b>276</b>	<b>1544</b>	<b>44.5</b>

**Daily Releases of I-131, I-132, I-133 (mCi)**

Draft 1 Version; January, 2003

Day	run #	Jun-58			run #	Jul-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1		7,900	11,000	273		54	0	0
2	<b>run 15</b>	7,100	49,000	4,262	(no runs	67	0	0
3	91- day	6,300	7,500	1,852	in	567	0	0
4	run)	6,200	23,000	893	July)	17	476	0
5		11,700	2,500	825		42	0	0
6		9,000	4,000	311		67	400	0
7		6,500	13,000	110		49	200	0
8		2,400	2,600	20		58	300	0
9		18,400	4,800	75		96	0	0
10		34,200	7,000	68		38	0	0
11		32,500	900	32		52	0	0
12		25,500	900	12		37	0	0
13		13,400	700	3		24	0	0
14		12,700	0	1		16	200	0
15		6,600	300	0		54	0	0
16		5,300	0	0		32	0	0
17		1,400	0	0		38	0	0
18		1,500	800	0		47	300	0
19		1,000	0	0		19	0	0
20		630	0	0		4	0	0
21		110	0	0		26	0	0
22		216	100	0		31	0	0
23		190	0	0		12	0	0
24		300	0	0		3	0	0
25		440	0	0		8	0	0
26		225	0	0		9	0	0
27		260	0	0		6	200	0
28		76	0	0		0	200	0
29		76	0	0		3	0	0
30		50	0	0		7	0	0
31						12	0	0
<b>TOTAL (mCi)</b>		<b>212,173</b>	<b>128,100</b>	<b>8,737</b>	<b>TOTAL (mCi)</b>	<b>1,495</b>	<b>2,276</b>	<b>0</b>
<b>TOTAL (Ci)</b>		<b>212</b>	<b>128</b>	<b>9</b>	<b>TOTAL (Ci)</b>	<b>1</b>	<b>2</b>	<b>0.0</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Aug-58			run #	Sep-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1	(first	12	0	0		1,170	0	0
2	month	8	0	0	(no runs	1,000	1,300	0
3	with	9	0	0	in Sept.)	470	0	0
4	charcoal	9	0	0		2,000	200	0
5	absorber)	6	100	0		3,730	0	0
6	<b>run 16</b>	270	2,800	162		3,150	0	0
7	(1- day	310	0	91		1,850	0	0
8	run)	290	0	42		2,010	300	0
9		240	700	17		460	0	0
10		150	0	5		1,950	1,100	0
11		12,800	5,600	216		840	0	0
12		13,600	800	113		890	0	0
13	<b>run 17</b>	8,300	3,700	4,983		320	0	0
14	(1-day	6,100	0	1,793		100	100	0
15	run)	4,670	3,700	672		550	800	0
16		3,190	0	225		570	0	0
17		2,770	400	96		630	900	0
18		1,090	600	18		1,060	0	0
19		1,450	200	12		970	100	0
20		820	0	3		250	400	0
21		990	300	2		150	200	0
22		1,000	400	1		100	100	0
23		730	400	0		30	200	0
24		260	0	0		10	100	0
25		480	300	0		40	500	0
26		170	200	0		54	500	0
27		300	400	0		16	100	0
28		3,740	0	0		63	0	0
29		12,600	0	0		46	600	0
30		1,170	300	0		13	100	0
31		990	200	0				
<b>TOTAL (mCi)</b>		<b>78,524</b>	<b>21,100</b>	<b>8,453</b>	<b>TOTAL (mCi)</b>	<b>24,492</b>	<b>7,600</b>	<b>0</b>
<b>TOTAL (Ci)</b>		<b>79</b>	<b>21</b>	<b>8</b>	<b>TOTAL (Ci)</b>	<b>24</b>	<b>8</b>	<b>0.0</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Oct-58			run #	Nov-58		
		I-131	I-132	I-133		I-131	I-132	I-133
1	<b>run 18</b>	16,300	258,000	9,786		450	3,200	0
2	(1-day	10,000	36,900	2,940		560	300	0
3	run)	5,100	4,400	734		450	100	0
4		2,240	3,600	158		540	300	0
5		750	200	26		160	100	0
6		1,590	1,900	27	(no run	310	0	0
7		820	1,200	7	20,	830	200	0
8		720	700	3	numb'g	1,010	200	0
9		560	500	1	error)	900	100	0
10		1,210	200	1		860	0	0
11		630	200	0		460	900	0
12		900	600	0	<b>run 21</b>	3,510	68,900	2,107
13		1,700	0	0	(1-day	4,080	11,600	1,200
14		800	200	0	run)	9,540	25,900	1,374
15		8,690	0	0		3,800	0	268
16		7,000	0	0		990	200	34
17		4,570	0	0		1,160	1,000	20
18		2,460	1,600	0		420	400	3
19		1,650	400	0		1,530	1,300	6
20		1,760	0	0		1,030	100	2
21		1,120	0	0		730	300	1
22	<b>run 19</b>	4,390	419,000	2,635		250	1,400	0
23	(1-day	10,200	60,000	2,999		180	200	0
24	run)	8,100	6,000	1,166		220	0	0
25		4,230	4,900	298		190	300	0
26		2,330	600	80		40	100	0
27		4,660	600	79		80	0	0
28		2,110	800	17		110	100	0
29		1,300	700	5		480	200	0
30		1,170	400	2		160	0	0
31		620	400	1				
<b>TOTAL (mCi)</b>		<b>109,680</b>	<b>804,000</b>	<b>20,968</b>	<b>TOTAL (mCi)</b>	<b>35,030</b>	<b>117,400</b>	<b>5,015</b>
<b>TOTAL (Ci)</b>		<b>110</b>	<b>804</b>	<b>21</b>	<b>TOTAL (Ci)</b>	<b>35</b>	<b>117</b>	<b>5.0</b>

**Daily Releases of I-131, I-132, I-133 (mCi)**

Draft 1 Version; January, 2003

Day	run #	Dec-58			run #	Jan-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		190	200	0		6	0	0
2	(no runs in Dec.)	140	0	0	(no runs in Jan.)	1	0	0
3		180	300	0		1	0	0
4		130	0	0		4	0	0
5		130	200	0		10	200	0
6		100	200	0		10	0	0
7	90	100	0	13	0	0		
8	90	300	0	2	100	0		
9	70	0	0	8	0	0		
10	770	2,100	0	11	100	0		
11	170	0	0	16	0	0		
12	60	0	0	18	0	0		
13	30	400	0	53	0	0		
14	30	100	0	0	400	0		
15	30	0	0	0	300	0		
16	150	200	0	9	0	0		
17	20	300	0	2	0	0		
18	10	0	0	8	0	0		
19	160	0	0	0	200	0		
20	160	0	0	12	0	0		
21	110	0	0	1	0	0		
22	80	0	0	9	0	0		
23	250	200	0	5	0	0		
24	110	0	0	7	0	0		
25	130	200	0	8	0	0		
26	60	100	0	5	0	0		
27	190	100	0	0	0	0		
28	30	300	0	13	0	0		
29	140	1,900	0	3	0	0		
30	0	100	0	10	0	0		
31	5	0	0	3	0	0		
<b>TOTAL (mCi)</b>		<b>3,815</b>	<b>7,300</b>	<b>0</b>	<b>TOTAL (mCi)</b>	<b>248</b>	<b>1,300</b>	<b>0</b>
<b>TOTAL (Ci)</b>		<b>4</b>	<b>7</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>0</b>	<b>1</b>	<b>0.0</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Feb-59			run #	Mar-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		6	0	0		420	0	15
2		4	0	0		250	200	4
3		10	0	0		160	0	1
4	<b>run 22</b>	6,440	88,500	3,866		430	0	2
5	(1-day	9,570	18,000	2,814		320	0	1
6	run)	31,500	50,000	4,535		190	0	0
7		20,300	9,600	1,431		130	100	0
8		7,830	0	270		100	300	0
9		2,250	500	38		250	0	0
10		2,220	200	18		420	700	0
11		1,420	2,500	6		1,120	300	0
12		1,860	200	4		1,090	500	0
13		1,750	0	2		1,200	400	0
14		370	0	0		560	100	0
15		1,930	0	0		330	0	0
16		160	500	0		400	200	0
17		1,350	500	0		150	0	0
18		7,540	1,200	0	<b>run 24</b>	460	23,500	276
19		5,940	0	0	(1-day	13,000	19,100	3,822
20		1,360	0	0	run)	12,600	9,500	1,814
21		70	0	0		4,700	2,800	331
22		460	0	0		1,650	600	57
23		280	0	0		1,390	500	24
24		180	100	0		1,550	0	13
25	<b>run 23</b>	450	5,000	270		3,090	1,700	13
26	(1-day	2,490	800	732		360	700	1
27	run)	2,230	8,900	321		210	200	0
28		1,680	1,300	118		250	200	0
29						100	400	0
30						190	0	0
31						140	400	0
<b>TOTAL (mCi)</b>		<b>111,650</b>	<b>187,800</b>	<b>14,427</b>	<b>TOTAL (mCi)</b>	<b>47,210</b>	<b>62,400</b>	<b>6,374</b>
<b>TOTAL (Ci)</b>		<b>112</b>	<b>188</b>	<b>14</b>	<b>TOTAL (Ci)</b>	<b>47</b>	<b>62</b>	<b>6.4</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Apr-59			run #	May-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		210	100	0		320	5,400	46
2		170	200	0		120	3,700	8
3		80	100	0		120	4,600	4
4		90	0	0		60	4,700	1
5		70	0	0		100	1,700	1
6		90	0	0		40	2,600	0
7		90	0	0		50	2,600	0
8	run 25 (1-day run)	150	8,500	90		60	1,100	0
9		140	1,600	41		30	400	0
10		140	1,800	20		30	400	0
11		190	1,800	13		20	300	0
12		80	1,200	3		20	700	0
13		100	600	2		30	400	0
14		50	700	0		10	500	0
15	320	3,900	1		40	1,000	0	
16	1,520	0	3		60	600	0	
17	760	100	1		30	100	0	
18	250	0	0		20	30	0	
19	310	0	0	run 27 (1-day run)	40	20	24	
20	100	600	0		910	6,800	268	
21	180	700	0		310	2,000	45	
22	50	2,000	0		110	2,900	8	
23	60	1,300	0		90	0	3	
24	90	0	0		100	0	2	
25	90	0	0		60	0	0	
26	60	0	0	50	0	0		
27	40	0	0	70	0	0		
28	60	0	0	80	200	0		
29	run 26 (1-day run)	1,030	87,500	618	50	400	0	
30		1,200	11,600	353	20	200	0	
31					40	300	0	
<b>TOTAL (mCi)</b>		<b>7,770</b>	<b>124,300</b>	<b>1,146</b>	<b>TOTAL (mCi)</b>	<b>3,090</b>	<b>43,650</b>	<b>411</b>
<b>TOTAL (Ci)</b>		<b>8</b>	<b>124</b>	<b>1</b>	<b>TOTAL (Ci)</b>	<b>3</b>	<b>44</b>	<b>0.41</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Jun-59			run #	Jul-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		30	100	0		0	0	0
2		0	700	0		40	900	0
3		20	0	0		10	100	0
4		250	0	0		30	200	0
5		50	0	0		10	100	0
6		20	0	0	run 29	250	7,300	150
7		0	400	0	run 29	2,560	64,500	753
8		20	0	0		1,180	24,500	170
9		0	300	0		280	20,200	20
10	run 28	650	22,400	390		160	7,300	6
11	(1-day	240	6,700	71		60	300	1
12	run)	140	8,700	20		30	600	0
13		40	6,900	3		10	900	0
14		50	5,700	2		90	300	0
15		20	3,000	0		220	400	0
16		10	10,000	0		190	0	0
17		250	0	1		80	0	0
18		200	0	0		70	0	0
19		50	0	0		40	0	0
20		30	0	0		90	0	0
21		20	0	0	run 30	1,100	11,300	0
22		20	0	0	run 30	750	133,400	0
23		40	0	0		750	16,100	0
24		60	0	0		480	5,000	0
25		50	0	0		1,270	8,500	0
26		30	200	0		120	3,900	0
27		60	0	0		60	500	0
28		20	0	0		30	400	0
29		20	0	0		30	900	0
30		20	0	0		40	800	0
31						40	300	0
<b>TOTAL (mCi)</b>		<b>2,410</b>	<b>65,100</b>	<b>487</b>	<b>TOTAL (mCi)</b>	<b>10,070</b>	<b>308,700</b>	<b>1,100</b>
<b>TOTAL (Ci)</b>		<b>2</b>	<b>65</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>10</b>	<b>309</b>	<b>1.1</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Aug-59			run #	Sep-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		60	300	0	<b>run 32</b>	420	9,800	252
2		30	100	0	<b>run 32</b>	280	6,200	82
3		430	0	0		200	4,800	29
4		340	300	0		100	3,100	7
5		340	1,100	0		20	1,100	1
6		180	100	0		40	0	1
7		60	500	0		30	0	0
8		100	0	0		340	800	1
9		80	0	0		690	1,900	1
10		70	0	0		960	600	1
11	<b>run 31</b>	500	40,000	300		320	700	0
12	<b>run 31</b>	190	4,100	56		130	0	0
13		180	5,000	26		90	0	0
14		180	0	13		70	0	0
15		40	3,900	1		30	200	0
16		80	3,100	1		80	0	0
17		80	1,700	1		280	200	0
18		110	0	0		140	0	0
19		90	0	0		160	0	0
20		20	500	0		80	100	0
21		30	100	0		30	300	0
22		10	500	0	<b>run 33</b>	270	10,300	162
23		30	0	0	<b>run 33</b>	190	4,000	56
24		290	700	0		130	5,100	19
25		80	600	0		100	1,100	7
26		90	0	0		90	1,000	3
27		20	900	0		690	800	12
28		50	0	0		960	1,700	8
29		10	100	0		850	200	3
30		40	0	0		300	1,200	1
31		10	200	0				
<b>TOTAL (mCi)</b>		<b>3,820</b>	<b>63,800</b>	<b>399</b>	<b>TOTAL (mCi)</b>	<b>8,070</b>	<b>55,200</b>	<b>646</b>
<b>TOTAL (Ci)</b>		<b>4</b>	<b>64</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>8</b>	<b>55</b>	<b>0.65</b>

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Oct-59			run #	Nov-59		
		I-131	I-132	I-133		I-131	I-132	I-133
1		200	600	0		100	100	0
2		90	1,400	0		140	100	0
3		40	100	0		140	1,700	0
4		20	400	0		70	0	0
5		30	100	0	run 35	800	91,000	480
6		460	2,600	0	run 35	1,830	7,300	538
7		180	400	0		4,470	3,500	644
8		200	500	0		160	600	11
9		50	5,100	0		270	1,200	9
10		70	0	0		300	100	5
11		70	0	0		490	300	4
12		60	600	0		60	1,800	0
13	run 34	60	44,100	36		930	100	2
14	run 34	570	10,600	168		150	1,900	0
15		400	5,000	58		550	1,600	0
16	ICPP	3,730	16,400	74,400		330	800	0
17	Criticality	2,440	12,500	33,100		320	400	0
18	Accident	970	8,130	12,000		390	0	0
19	(*)	1,010	5,280	4,320		270	100	0
20		1,150	3,430	1,560		120	900	0
21		940	2,230	560		300	400	0
22		870	1,450	200		310	0	0
23		180	940	100		540	0	0
24		150	500	0		160	0	0
25		90	500	0		70	0	0
26		40	400	0		20	100	0
27		100	500	0		30	0	0
28		120	400	0		10	0	0
29		330	0	0		10	100	0
30		170	600	0	run 36	20	0	12
31		240	0	0				
<b>TOTAL (mCi)</b>		<b>15,030</b>	<b>124,760</b>	<b>126,501</b>	<b>TOTAL (mCi)</b>	<b>13,360</b>	<b>114,100</b>	<b>1,706</b>
<b>TOTAL (Ci)</b>		<b>15</b>	<b>125</b>	<b>127</b>	<b>TOTAL (Ci)</b>	<b>13</b>	<b>114</b>	<b>1.7</b>

(\*) Releases of I-132 and I-133 estimated from criticality release model, Chapter 8.

## Daily Releases of I-131, I-132, I-133 (mCi)

Draft 1 Version; January, 2003

Day	run #	Dec-59			run #	Jan-60		
		I-131	I-132	I-133		I-131	I-132	I-133
1	<b>run 36</b>	60	600	18				
2		110	0	16				
3		50	0	4				
4		50	0	2				
5		20	0	0				
6		20	0	0				
7		10	0	0				
8		20	0	0				
9		10	0	0				
10		10	0	0				
11		10	0	0				
12		10	0	0				
13		20	200	0				
14		20	0	0				
15	<b>run 37</b>	200	2,900	120				
16	<b>run 37</b>	240	16,800	71				
17		160	1,900	23				
18		210	2,100	15				
19		140	900	5				
20		100	400	2				
21		60	400	0				
22		70	300	0				
23		60	600	0				
24		40	200	0				
25		40	0	0				
26		30	0	0				
27		10	600	0				
28		10	0	0				
29		10	0	0				
30		20	0	0				
31		0	0	0				
<b>TOTAL (mCi)</b>		<b>1,820</b>	<b>27,900</b>	<b>275</b>	<b>TOTAL (mCi)</b>	<b>0</b>	<b>0</b>	<b>0</b>
<b>TOTAL (Ci)</b>		<b>2</b>	<b>28</b>	<b>0</b>	<b>TOTAL (Ci)</b>	<b>0</b>	<b>0</b>	<b>0.0</b>