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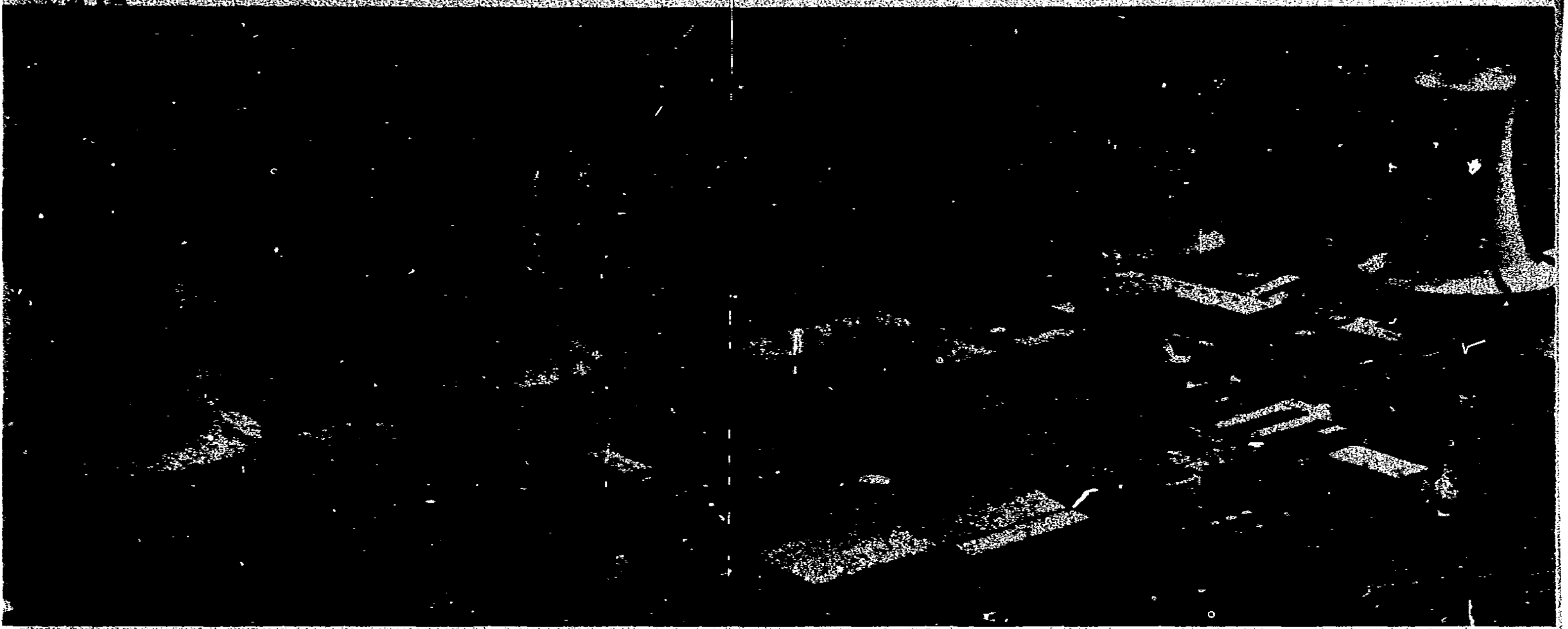

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Preliminary Radioiodine Source-Term and Inventory Assessment for TMI-2

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March 1983

Prepared for the
U.S. Department of Energy
Three Mile Island Operations Office
Under DOE Contract No. DE-AC07-76ID01570

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GEND 028
Distribution Category: UC-78
TMI Supplement

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Prepared for EG&G Idaho, Inc.
Under Subcontract No. K-1810
and the U.S. Department of Energy
Three Mile Island Operations Office
Under DOE Contract No. DE-AC07-76IDO1570

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ABSTRACT

The results of an analysis of radioiodine behavior during and after the March 28, 1979 accident at TMI-2 are presented. All available measurement data for ^{129}I and ^{131}I are summarized. Measurements show that from 17% to 28% of the iodine originally in the fuel could be accounted for outside the core on August 28, 1979 (when the first basement water sample was taken). To fill in gaps where measurements are not available mathematical models are used. Models for the transport of iodine from the fuel to the Reactor Building are developed and benchmarked to measurements. It is estimated that approximately 1% of the iodine originally in the fuel was made airborne during the accident, and the maximum air concentration during the accident resulted from transport of 0.2% of the original core inventory.

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INTRODUCTION

The purpose of this work was to perform a preliminary analysis of the fate of radioiodine during and following the accident at Three Mile Island, Unit 2 (TMI-2). The plant had been operating for approximately 95 effective full power days when it tripped at 0400 on March 28, 1979. Subsequent events lead to reactor coolant system (RCS) liquid being released to containment until approximately 0620 when the block valve (RC-V2) was closed. At this time radiation monitors began recording substantial radiation levels in containment indicating that the loss of coolant had caused fuel damage. Starting at approximately 0713 and lasting until approximately 1700 hours, coolant, as steam and liquid, was released to containment intermittently carrying radioactivity from the damaged core. At 1350 hours, containment sprays were initiated for approximately 5 minutes which reduced concentrations of radionuclides in the containment atmosphere. Containment pressure fell below atmospheric pressure at 1730 hours and remained there, indicating that little or no vapor escaped from the RCS after that time. However, some leakage of RCS liquid continued.[6]

This work summarizes the quantities of iodine in plant systems and compartments from the start of the accident. The systems and compartments included are the reactor coolant system, containment basement, surfaces inside containment, containment atmosphere and the auxiliary building. To the extent possible, measurement results were used in the analysis. For time intervals when data were not available, mathematical models were used to fill in the missing information. Models were adjusted to correspond with measurements made prior to and following such intervals.

Relevant measurement results are summarized in the first part of the report. The second part of the report describes the mathematical models and presents model results.

RADIOIODINE MEASUREMENTS AT TMI-2

The results of radioiodine measurements made at TMI-2 are summarized in this section. The media sampled included reactor coolant, the Reactor Building atmosphere, the Reactor Building basement liquid, liquids in the Auxiliary Building, effluent air discharged from the Auxiliary and Fuel Handling Buildings, charcoal from filter beds in the Auxiliary and Fuel Handling Building discharge lines, and samples of paint from surfaces in the Reactor Building. In addition, iodine concentration on Reactor Building surfaces was estimated from gamma spectra obtained by a collimated detector positioned outside the building. Most of the samples collected soon after the accident were analyzed for ^{131}I by gamma spectrometry. Subsequent samples were analyzed for ^{129}I by x-ray spectrometry and neutron activation analysis. Each major inventory component is treated in a separate subsection. To facilitate comparisons among nuclides and between samples taken at different times, the measured activities have been decay corrected to the time of reactor shutdown, 0400 on March 28, 1979, and expressed as a fraction of the shutdown inventory of the measured radionuclide.

Reactor Coolant System Liquid

Iodine-131 was detected in reactor coolant system (RCS) liquid samples collected before August 15, 1979.[1,2] The concentration of ^{129}I has been measured in two RCS samples.[3-5] The results of the measurements are given in Table 1. In addition to these measurements, the ^{131}I concentration of a sample taken at 0850 on the day of the accident was reported to be 81 μCi/ml.[6] However, the result is questionable because of sampling difficulties and the probable existence of a vapor barrier between the liquid in the core and the liquid being sampled. After 2000 hours on the day of the accident, an in-service reactor coolant pump assured reasonably good mixing in the RCS. Iodine-129, -131, and -133 concentrations were measured in the first RCS sample listed in Table 1. The results for ^{131}I, ^{133}I, and the first ^{129}I measurements are in good agreement and indicate that about 9.2% of the iodine inventory was in the RCS 37 hours after reactor shutdown. The quantity of radioiodine in the

TABLE 1. MEASUREMENTS OF RADIOIODINES IN THE TMI-2 REACTOR COOLANT SYSTEM

Time (hr) After Shutdown (Date)	Radionuclide	Percent of Initial Core Inventory[a]	Reference
37 (3/29/79)	^{131}I	7.6	2
	^{133}I	8.9	2
	^{129}I	11	3
	^{129}I	0.78	3,4
	^{129}I	19[b]	5
316 (4/10/79)	^{131}I	14	1
458 (4/16/79)	^{131}I	12	1
483 (4/17/79)	^{131}I	12	1
522 (4/18/79)	^{131}I	12	1
605 (4/22/79)	^{131}I	9.7	1
676 (4/25/79)	^{131}I	8.9	1
841 (5/2/79)	^{131}I	6.7	1
1040 (5/10/79)	^{131}I	7.5	1
1230 (5/18/79)	^{131}I	8.0	1
1570 (6/1/79)	^{131}I	8.0	1
1710 (6/7/79)	^{131}I	5.2	1
1860 (6/13/79)	^{131}I	4.7	1
2000 (6/19/79)	^{131}I	4.6	1
2190 (6/27/79)	^{131}I	4.2	1
2380 (7/5/79)	^{131}I	4.9	1
2530 (7/11/79)	^{131}I	5.8	1
2670 (7/17/79)	^{131}I	3.6	1
2840 (7/24/79)	^{131}I	5.1	1
3010 (7/31/79)	^{131}I	3.7	1
3200 (8/8/79)	^{131}I	0.43	1
12100 (8/14/80)	^{129}I	1.0	3
	^{129}I	0.5	3,4

---

a. Computed using a reactor coolant system volume of 3.23×10^8 ml and initial inventories of 7.0×10^7 Ci, 1.6×10^8 Ci, and 0.22 Ci for ^{131}I, ^{133}I, and ^{129}I, respectively. The inventories were taken from Reference 21.

b. Possibly not an aliquot of the same sample.[3]

RCS liquid at times prior to 37 hours can only be inferred from calculational models. Samples taken from 37 to 522 hours showed that the RCS liquid iodine increased to 12--14% of the core inventory and then declined to about 1% of the core inventory at 12,100 hours after shutdown. The increase is believed to be due to leaching of iodine from the damaged fuel or from internal surfaces of the primary system, where it may have deposited following release from the fuel. The decrease after about 522 hours is due to leakage of water from the RCS into the Reactor Building Basement. Radioiodine input and output rates to the RCS liquid are similar to radiocesium input and output rates during the same time period.[6]

Reactor Building Basement

The Reactor Building Basement is potentially a large reservoir for radioiodines. The few measurements that have been made are summarized in Table 2. Each value is the sum of radioiodine in the liquid and in filterable solids, when present. Each sample is discussed briefly below.

The first set of samples was taken on August 28, 1979 and analyzed by Oak Ridge National Laboratory (ORNL).[7] The samples were taken through a long tygon tube threaded through a penetration above the basement water level. Although tygon is a notorious "getter" of elemental iodine when used for air sampling, it is doubtful that much plateout of iodine occurred from the liquid stream. This is because the deposition rate is controlled by the mobility of the ion in a liquid and is therefore very slow compared to the sampling rate. The bottom sample, taken three inches above the floor, contained solids which were centrifuged and analyzed separately. The only value for ^{131}I concentration in the basement (0.12 μCi/ml) is from the liquid portions of these samples, and is equivalent to 17 percent of the core inventory at 0400 hours March 28, 1979. This estimate is based on 516,000 gallons of liquid in the reactor building basement on August 28, 1979. The ^{129}I concentration in the liquid fraction of the samples, 0.078 μg/ml, represents 12 percent of the core inventory. The ^{129}I concentration in the solids is reported in terms of the total volume of the bottom sample: 0.07 μg/ml. The calculation of the ^{129}I inventory in basement solids requires an assumption on the

TABLE 2. MEASUREMENTS OF RADIOIODINES IN THE TMI-2 REACTOR BUILDING SUMP

Time (hr) After Shutdown (Date)	Radionuclide	Percent of Initial Core Inventory[a] Liquid	Solids	Total	Reference
3,680(8/28/79)	^{131}I	17[b]	N.M.[c]	>17	7
	^{129}I	12[b]	0.4-7[d]	12-19	7
18,680(5/14/81)	^{129}I	5.5[e]	0.5-5[d]	6-11	8,9
21,870(9/24/81)	^{129}I	25[f]	N.M.	>25	10

---

a. Core inventories at shutdown of ^{129}I and ^{131}I taken as 0.22 Ci and 7.0×10^7 Ci respectively.

b. Based on an estimated basement volume of 5.16×10^5 gal on August 28, 1979 and the average concentration found in three samples of liquid.

c. N.M. = not measured.

d. Varies according to amount of solids estimated. See text.

e. Based on an estimated basement volume of 6.4×10^5 gal on May 14, 1981 and the weighted average of the concentrations found in four samples of the liquid fraction.

f. Based on an estimated basement volume of 6.5×10^5 gal on September 24, 1981.

depth of solids-laden water. The minimum depth is three inches, which corresponds to a volume of 1.69×10^4 gallons. The maximum possible depth is just below the next higher sample (60 inches above the floor), in which no solids were reported, which corresponds to a volume of 3.38×10^5 gallons. The ^{129}I core inventory in the solids thus ranges from 0.4 to 7 percent. The total core inventory in the basement ranges from 12 to 19 percent.

A second set of basement samples was taken during the tenth entry (May 14, 1981) and analyzed by EG&G Idaho, Inc.[8,9] The lowest sample of the four was from the basement floor and contained solid materials. The next sample up was 5 3/8 inches above the floor. It and the top two samples "contained no observable solids".[8,9] The ^{129}I concentration in the liquids decreased toward the bottom of the basement. The fraction of the core inventory in the liquid was 5.5 percent. The ^{129}I concentration in the solids, determined by neutron activation analysis, was 0.065 percent by weight. There was 0.9 mg of solids per milliliter of the sample, so the ^{129}I concentration in the solids was 1.0×10^{-4} µCi/ml of sample as collected. The amount of ^{129}I in the basement solids depends on the depth of solids-bearing water used in the calculation. An upper limit is 5 inches which yields a core fraction of 5 percent. If a 0.5-inch height is assumed as the lower limit, this represents 0.5 percent of the ^{129}I core inventory. The total core inventory represented thus lies between about 6 and 11 percent.

The final basement sample, C101, was collected September 24, 1981, prior to the start of processing of the basement water by the Submerged Demineralizer System.[10] Although the sample was filtered, the only ^{129}I value reported is for the liquid portion and is thus a lower limit. The concentration of 2.2×10^{-5} µCi/ml corresponds to a core-inventory fraction of 25 percent, the highest measured.

The calculations of fractional core inventories of iodine in the basement all share the assumption that each sample is representative of the entire basement at the time of collection. The differences in the results are probably due at least partly to inhomogeneities in the basement contents.

Reactor Building Atmosphere

The first sample of the reactor building atmosphere was collected 75 hours after shutdown on March 31, 1979.[2] Ten more samples were collected and analyzed for ^{131}I.[11-13] The last two ^{131}I measurements were made using an airborne radioiodine species sampler.[14,15] The results of these measurements are shown in Table 3. Also shown in Table 3 are the results of ^{129}I measurements made during 1980, both prior to and after the Reactor Building purge.[16,17]

The average of the two analyses performed on the first sample represents 0.005% of the original core inventory. Subsequent samples represent fractional core inventories which range from a low of 0.001% to a high of 0.03%. The average of all samples is 0.01% of the original core inventory. From June 1979 to May 1980, iodine levels in containment atmosphere steadily declined from 0.03% to 0.0015%. After purging, the level (0.0011%) was not much lower than the level just prior to purging (0.0015%). The reason for the decrease from June 1979 to May 1980 may be that the rate of resuspension of iodine from surfaces decreased over that time period thus changing the iodine equilibrium between surfaces and air. A more detailed discussion of iodine behavior is included in the next section. Suffice it to say here that at 75 hours the measured level of iodine in containment atmosphere was 0.005%.

The radioiodine species sampling results are quite similar for gaseous ^{129}I and ^{131}I. The principal species found in all measurements was organic. The commonly recognized volatile forms of iodine* are elemental

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*References 14 and 15 describe a sampling system in which air is passed serially through four adsorbers which selectively collect different forms of iodine. The first is a particulate filter which collects and retains iodine irreversibly attached to particles. The second adsorber is CdI_2 which retains elemental and passes less reactive forms. The third is IPh which collects a form of iodine called HOI which penetrates the CdI_2 adsorber but does not adsorb CH_3I. The fourth adsorber is impregnated charcoal or silver zeolite which collects all known forms of iodine, and placed after CdI_2 and IPh it collects the least reactive forms of iodine, e.g., organic iodine of which CH_3I is the most likely form. In this report elemental, HOI and organic iodine refer to those forms of iodine identified with the above sampling system.

TABLE 3. MEASUREMENTS OF RADIOIODINES IN THE TMI-2 REACTOR BUILDING ATMOSPHERE

Time (hr) After Shutdown (Date)	Radionuclide	Percent of Initial Core Inventory[a]	Reference
75 (3/31/79)	^{131}I	0.0026	2
	^{131}I	0.0065	11
127 (4/2/79)	^{131}I	0.0012	12
890 (5/4/79)	^{131}I	0.0053	12
1130 (5/14/79)	^{131}I	0.0058	12
1420 (5/26/79)	^{131}I	0.020	12
1450 (5/27/79)	^{131}I	0.019	12
1520 (5/30/79)	^{131}I	0.030	12
1540 (5/31/79)	^{131}I	0.019	12
2040 (6/21/79)	^{131}I	0.014	12
2170 (6/26/79)	^{131}I	0.010[b]	13
2480 (7/9/79)	^{131}I	0.0089[c]	13
9590 (5/1/80)	^{129}I	0.0017[d]	16
9630 (5/3/80)	^{129}I	0.0015[e]	16
11770 (7/31/80)	^{129}I	0.0011[f]	17

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a. Computed using an average net air volume of 5.46×10^{10} cm^3 in the Reactor Building for the ^{131}I samples. For the ^{129}I samples, the net air volume was taken to be 5.37×10^{10} cm^3. The difference is due to the volume taken up by sump water.

b. Radioiodine species: 0.04% of airborne activity associated with particulates, 7.8% I_2, 5.7% HOI, and 86% organic iodides.

c. Radioiodine species: 0.04% of airborne activity associated with particulates, 9.2% I_2, 13% HOI, and 78% organic iodides.

d. Based on average of four samples of total gaseous iodine collected over 2-day period.

e. Gaseous radioiodine species: 19% I_2, 16% HOI, 65% organic iodides. The uncertainties associated with these species fractions are ~30% of the values, much larger than the uncertainties for the other species measurements.

f. Peak activity measured after most of the purge had been completed and airborne activity returned to equilibrium. Gaseous radioiodine species: 2.5% I_2, 7.3% HOI, 90% organic iodides.

(I_2), organic (e.g., CH_3I) and a form which behaves unlike I_2 or organic which is generally called hyporodous acid (HOI). In addition to these forms, iodine can exist in particulate matter (e.g., as CsI) or adsorbed on particulate matter. The iodine on building surfaces could have been transported there in any of these states, either singly or in combination.

Reactor Building Surfaces

The first estimates of the radioiodine concentration on surfaces within the Reactor Building were derived from gamma spectra obtained on June 1, 1979 using a collimated Ge(Li) detector located outside the Reactor Building equipment hatch.[18] The results of those measurements and of direct measurements on samples removed from the Reactor Building at later times are given in Table 4.[7,10,19] The first sample for direct analysis was the cutout of the penetration seal plate obtained on August 28, 1979.[7] The estimates based on analyses of ^{129}I in six paint samples[10,19] average 0.24% of the initial inventory which is lower but comparable to the earlier results for ^{131}I.

Auxiliary and Fuel Handling Buildings

Measurements of ^{131}I in the Auxiliary and Fuel Handling Building liquids have shown that reactor coolant bleed tanks (RCBT) contained the vast majority of the radioiodine.[6,20] Table 5 shows the estimated liquid inventories based on analyses of both ^{131}I and ^{129}I.[3,6,20] The tank samples analyzed for ^{129}I were taken at different times. Sampling dates were December 20, 1979, January 28, 1980, and February 4, 1980 for RCBT-A, RCBT-B, and RCBT-C, respectively. The later estimates of individual tank and total tank inventories agree within about a factor of two. Note that tank capacities were used in the calculation of total activity, and therefore the results represent upper limits. Radioiodine levels in other liquids, in charcoal filter beds and on building surfaces are trivial[6] compared with those in RCBT liquids. At the time of this writing, measurements results of ^{129}I in sludge material taken from makeup and

TABLE 4. MEASUREMENTS OF RADIOIODINE ON SURFACES IN THE TMI-2 REACTOR BUILDING

Time (hr) After Shutdown (Date)	Measurement Type	Percent of Initial Core Inventory[a]	Reference
1560 (6/1/79)	^{131}I, Indirect, by Gamma Spectrometry	0.49	18
3680 (8/28/79)	^{131}I, Direct, Sample of Penetration Seal Plate	0.69	7
17000 (3/7/81)	^{129}I, Direct, Paint From Seal Table Area	0.13	19
17000 (3/7/81)	^{129}I, Direct, Paint From Stairwell Area	0.47	19
17000 (3/7/81)	^{129}I, Direct, Fallen Paint Chip, Found on 305' Level	0.025	19
19600 (6/21/81)	^{129}I, Direct, Paint on EJB,[b] 305' Level	0.36	10
19700 (6/25/81)	^{129}I, Direct, Paint on EJB,[b] 347' Level	0.17	10
19800 (7/1/81)	^{129}I, Direct, Fallen Paint From Dome, Found on 347' Level	0.29	10

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a. Computed using a surface area of 2.2×10^8 cm^2.

b. Electrical Junction box.

TABLE 5. MEASUREMENTS OF RADIOIODINES IN AUXILIARY BUILDING TANKS

Time (hr) After Shutdown (Date)	Radionuclide	Percent of Initial Core Inventory In The Reactor Coolant Bleed Tanks[a]				Reference
		A	B	C	Total	
46--50 (3/30/79)	^{131}I	5×10^{-6}	0.051	0.054	0.10	12
140--150 (4/3/79)	^{131}I	0.077	0.054	ND[b]	--	12
1900 (6/14/79)	^{131}I	0.72	1.1	1.1	2.9	20
6420 to (12/20/79)	^{129}I	1.3	1.1	2.3	4.7	3
7520 (2/4/80)	^{129}I	0.59	0.59	1.1	2.2	3,4

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[a] Computed assuming liquid volumes of 2.92×10^8 ml, which is the capacity of each tank.

[b] No data available.

letdown filters are not available. In another analysis of ^{131}I behavior,[6] it was estimated that the letdown demineralizer may have collected 7% of the radioiodine. However, no measurements are available and therefore demineralizer radioiodine is not included in Table 5.

Summary of Iodine Inventory Evaluation

The observed fractional inventories in the various media are summarized in this section. The first time at which a detailed account of the measurements of iodine inventory can reasonably be attempted is August 28, 1979, 3680 hours after shutdown. Prior to that time, no direct measurements of the basement or Reactor Building surfaces had been made. Table 6 summarizes the results of inventory measurements at 3680 hours (8/28/79). The possible range in the portion of the total inventory contained in the media sampled is from 17 to 28 percent. There is no evidence that any other system or area of the plant held a significant quantity of iodine. (As noted above, the letdown demineralizer inventory could raise the fraction of the total iodine accounted for at that time).

The same accounting procedure (using data from References 1, 3 and 20) identifies approximately 50% of the ^{3}H and ^{137}Cs inventory on the same date. The fractional inventories of these elements in the RCS and Auxiliary Building[3,20] are comparable to those for iodine. The principal difference between the cesium and tritium inventories on the one hand and the iodine inventory on the other lies in the fractions found in Reactor Building Basement.

Potential locations of additional iodine have been identified. These are listed and discussed briefly below:

- relatively undamaged fuel
- reactor vessel internals and RCS piping
- pressurizer relief system including the reactor coolant drain tank
- makeup and letdown system
- solids in the Reactor Building sump and basement
- Reactor Building air cooler surfaces

TABLE 6. SUMMARY OF MEASURED RADIOIODINE INVENTORIES ON AUGUST 28, 1979

Location	Percent of Initial Core Iodine Inventory
Reactor Coolant System	2--3
Reactor Building Basement	12--19
Reactor Building Atmosphere	0.002--0.003
Reactor Building Surfaces	0.5--0.7
Auxiliary Building Liquids	2--5
Total Identified	17--28

Although none of these locations has yet been analyzed for iodine content by direct or indirect measurement techniques, each is a candidate for future measurements that can improve the accounting in Table 6. Because portions of the reactor core remained at relatively low temperatures, undamaged fuel would be expected to contain a large fraction of the original iodine inventory. Radioiodine released from the fuel at higher temperatures may have deposited on internal surfaces, such as the plenum or primary piping, that were exposed for an extended time. Much of the released iodine passed through the pressurizer and its relief lines to the RCDT. The contents of the RCDT have not been sampled; neither have the interior surfaces of the tank and associated piping. Part of the iodine present in the RCS between 0400 and 1900 on March 28 would have been collected on the letdown demineralizer. Radiation survey data show that the demineralizer contains large amounts of radioactivity. The high concentration of ^{129}I in solids present in basement samples suggests that additional iodine may be present in the basement. If the concentrations of solids in samples collected to date under-represent the true mass of solids in the basement, then additional samples may indicate a higher inventory than that in Table 6. The surfaces of the heat exchangers in the five Reactor Building air coolers may have removed iodine from the containment atmosphere. During the period when the concentration of radioiodine was the highest in containment air, it is likely that the surface of the cooling coils was covered with condensate. Therefore, some of the iodine removed by the coolers followed the condensate into the Reactor Building Basement.

Implications of Data Regarding the Chemical Form of Airborne Radioiodine

The purpose of this section is to examine surface measurements of ^{137}Cs, ^{131}I, and ^{129}I to provide insight into the form in which iodine was transported to surfaces. Because of the likely presence of iodine in the form of CsI escaping from the core, it is worthwhile to examine the relative quantities of iodine and cesium measured on surfaces. Core inventories of radioiodine and radiocesium[21] at the time of the accident are shown in Table 7. Also shown are ^{127}I and ^{133}Cs which are stable fis-

TABLE 7. DISTRIBUTION OF IODINE ISOTOPES AS CESIUM IODIDE

Cesium			Atoms of Iodine if All I is Assumed to be CsI						
Isotope	Curies	Atoms		^{129}I	^{131}I	^{133}I	^{135}I	^{127}I	Total
^{134}Cs	1.58(5)	5.53×10^{23}	[0.0056] [a]	3.03×10^{22}	1.45×10^{22}	3.41×10^{21}	1.03×10^{21}	8.45×10^{21}	5.78×10^{22}
^{135}Cs	2.63	1.33×10^{25}	[0.134]	7.30×10^{23}	3.49×10^{23}	8.38×10^{22}	2.47×10^{22}	2.03×10^{23}	1.39×10^{24}
^{137}Cs	8.43(5)	4.28×10^{25}	[0.43]	2.35×10^{24}	1.12×10^{24}	2.70×10^{23}	7.96×10^{22}	6.54×10^{23}	4.47×10^{24}
^{133}Cs	--	4.28×10^{25}	[0.43]	2.35×10^{24}	1.12×10^{24}	2.70×10^{23}	7.96×10^{22}	6.54×10^{23}	4.47×10^{24}
		9.95×10^{25}		5.46×10^{24}	2.60×10^{24}	6.27×10^{23}	1.85×10^{23}	1.52×10^{24}	1.04×10^{25}
				[0.525]	[0.25]	[0.060]	[0.018]	[0.146]	

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a. The numbers in brackets are fraction of total atoms of that isotope.

sion products not included in Reference 21. On the basis of other calculations,[45] ^{133}Cs and ^{137}Cs atoms were assumed to be equal in number, and the number of ^{127}I atoms was taken to be 28% of ^{129}I atoms. As a first approximation, these isotopes are in the same proportion as their fission yields. Table 7 shows there were approximately 10 cesium atoms for each iodine atom.

Table 8 summarizes pertinent measurements and shows the ratio of iodine to cesium atoms on building surfaces. If all the iodine was transported to containment surfaces as cesium iodide, and both the cesium and iodine persisted in the same proportion thereafter, the I/Cs ratio would be 1. If all 10 cesium atoms escaped the core and were transported, deposited and persisted proportionately with each iodine atom, the ratio of iodine to cesium atoms on containment surfaces would be 0.1. Table 8 shows that the measured range was 0.7 to 300. All but one measurement showed a ratio substantially greater than 1. The average ratio on the paint chip samples was approximately 30. The iodine to cesium ratio on the seal plate cutout was 300.

The measurement showing an I/Cs ratio less than 1 was the only one made which included a floor area, in this case the floor in front of the equipment hatch on the 305' level. The difference between the ratio on the floor and wall is the level of cesium-137 present. The iodine levels are nearly the same, 0.49% of the core on the floor and an average of 0.30% on the wall samples. On the other hand, ^{137}Cs measured on the floor was 32 times higher than the average ^{137}Cs measured from wall samples. Apparently, higher levels have persisted on floors. Wall and floor measurements of ^{137}Cs made before the March 1982 Decontamination Experiment showed that floor levels were eight times the levels on nearby walls.[46]

Either of two explanations is consistent with these findings. One is that the iodine was transported, deposited and persisted independently of the cesium. If it were in a gaseous form, it would deposit rather evenly on vertical and horizontal surfaces alike. The cesium on the other hand,

TABLE 8. IODINE AND CESIUM INVENTORIES ON REACTOR BUILDING SURFACES[a]

Time (hr) After Shutdown	Nuclide Measured[b]	Total Cs and I Atoms[c]	Iodine Atoms / Cesium Atoms
1560 (6/1/79) Floor	^{131}I ^{137}Cs	$5.1x10^{22}$ $7.0x10^{22}$	0.73
1560 (6/1/79) Wall	^{131}I ^{137}Cs	$3.1x10^{22}$ $2.2x10^{21}$	14.
3680 (8/28/79)	^{131}I ^{137}Cs	$7.2x10^{22}$ $2.3x10^{20}$	300.
17000 (3/7/81)	^{129}I ^{137}Cs	$1.4x10^{22}$ $6.7x10^{20}$	21.
17000 (3/7/81)	^{129}I ^{137}Cs	$4.9x10^{22}$ $1.0x10^{22}$	4.9
17000 (3/7/81)	^{129}I ^{137}Cs	$2.6x10^{21}$ $1.4x10^{20}$	19.
19600 (6/21/81)	^{129}I ^{137}Cs	$3.7x10^{22}$ $5.2x10^{20}$	71.
19700 (6/25/81)	^{129}I ^{137}Cs	$1.8x10^{22}$ $4.3x10^{20}$	42.
19800 (7/1/81)	^{129}I ^{137}Cs	$3.0x10^{22}$ $3.4x10^{21}$	8.8

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a. Unless otherwise indicated, measurements were made on vertical surfaces.

b. See Table 4 for ^{131}I and ^{129}I levels on surfaces. ^{137}Cs levels were taken from references shown in Table 4.

c. To calculate total iodine and cesium atoms convert ^{131}I or ^{129}I or ^{137}Cs measurements to atoms and divide by the fraction each nuclide is of the total, i.e., 0.25, 0.525 and 0.43 respectively. (See Table 7).

deposited mainly on horizontal surfaces, perhaps by gravitational settling of particles. Another explanation is that the iodine was transported and deposited as CsI, but only the iodine persisted on vertical surfaces, while the cesium was washed off by condensate.

The first alternative best fits all the available information. Experiments have shown floor to wall ratios of deposited particulate matter of 3 to 12.[27] This range is lower than the measured ratio of 32. However, one would expect that containment sprays, which operated at TMI-2, would increase floor concentrations relative to wall concentrations. If iodine deposited as CsI, then from 95% to 99% of the cesium was later washed off surfaces by natural processes. In view of the difficulties encountered in removing cesium during the March 1982 decontamination experiment,[46] such a high removal rate under natural conditions seems unlikely.

Of the known iodine species, the only one with a deposition velocity greater than that of small particles is elemental. The deposition velocities of both organic iodide and HOI are lower than that for small particles. Therefore, if the iodine was present in either of these forms, the I/Cs ratio would have been much lower than measured.

It should be pointed out that the existence of a small fraction of elemental iodine in the containment atmosphere does not preclude the possibility that iodine escaped from the fuel and even the RCS as CsI. Oxidation of CsI in small particles or I^- in small water droplets entrained in steam are potential sources of elemental iodine. Indeed there is direct evidence that the iodine which entered the basement may have been in a form other than volatile iodine. In 1981, a sample of basement liquid was analyzed for volatile iodine (CCl_4 extractable), iodide and iodate.[51] The nuclide measured was ^{129}I. The results were 2% volatile iodine, 96% iodide and 2% iodate. Similar measurements were made in this laboratory[43] of water to which iodine had been added as I_2 and as NaI. Analytical results were reported in terms of volatiles and nonvolatiles. The nonvolatile fraction includes iodide and iodate. If 20% of the core iodine inventory were dissolved in 180,000 gallons of basement water the concentration would be approximately 7.5×10^{-4} mg/ml. (See page 28 for explanation of basement liquid volume). The relevant laboratory test results[43] in this concentration range were as follows. For addition as I_2, 27% and 51% of the total iodine was measured to be in the volatile form at pH 9.3 and 7.0, respectively. For these two experiments, the concentration of total iodine was approximately 10^{-3} mg/ml. In one experiment iodine was added as NaI to a pH 5.2, 25°C solution. Only 1.8% was measured to be in the volatile form. This fraction is obviously closest to the one measured in TMI basement water, and supports the assumption that most of the iodine entered the basement in ionic form. The large uncertainty in applying these results to TMI is the very large difference in times between iodine injection and analysis. In the laboratory experiments, a typical time between iodine injection and analysis was two hours. The comparable time for the TMI sample was two years.

CALCULATIONS OF RADIOIODINE BEHAVIOR AT TMI-2

The previous section illustrates the uncertainty about the fate of iodine during and after the accident at TMI-2. The first reliable measurements of radioiodine in the RCS and containment atmosphere, made 37 and 75 hours after shutdown respectively, provide benchmarks for any models used to compute iodine behavior and distribution. In this section, models of a variety of processes are employed to provide estimates of iodine behavior during the early hours of the accident. The processes treated by calculational models are the following:

- release of iodine from the fuel
- deposition of iodine on surfaces from the gas space within the primary system
- movement of reactor coolant to the RCDT and basement and to the Auxiliary Building
- distribution of iodine in the air, on surfaces and in basement liquids after release to the Reactor Building
- removal of airborne iodine by Reactor Building sprays.

Release of Iodine From the Fuel

The expected release of iodine from the UO_2 fuel matrix during the accident depends upon a number of parameters, but the most important is the temperature distribution within the core as a function of time.[22] Estimates of the temperature history of five sections of the TMI-2 core during the accident have been made using the Transient Reactor Analysis Code (TRAC).[23] These temperature estimates were used together with release rate coefficients for iodine and its precursors antimony and tellurium, to compute the iodine release fraction. The fractional release rate coefficients used were the best estimate values given in Figure 4.3 of Reference 22. These release rate coefficients were derived from experimental measurements of the total release from fuel at the end of a period during which the fuel was at a known temperature. Thus the release rates are less well known than the total release and are considered to be order of magnitude estimates.[22]

The release of iodine from the fuel during the period between 10,000 s and 12,500 s (2.8 to 3.5 hours) after the accident was estimated for each section of the core considered in the TRAC calculation. The estimated fuel temperatures suggest that the lower half of the core did not contribute any of the initial iodine release while virtually all the iodine was released from the hottest region. Most of the release from the fuel is estimated to have occurred in a period of about 10 minutes beginning 3.2 hours after shutdown. The high fuel temperatures were reduced dramatically by the high pressure injection flow about 3.3 hours after shutdown. Later increases in the iodine inventory in the RCS are probably attributable to leaching of iodine from the fuel or from internal surfaces of the vessel, plenum, and primary piping.

The fractional release rates for cesium are comparable to those for iodine at temperatures above 1200°C.[22] Thus the initial cesium release is estimated to have been the same as that for iodine. Subsequent increases in the RCS inventory were also observed for cesium, presumably also due to leaching from the fuel or primary surfaces. Because the initial releases of the two elements and their subsequent additions to the RCS are calculated to be nearly equal, one would expect the basement inventories to be more nearly equal than measurements indicate.

In the transport calculations, three time periods are considered: core voiding and uncovery (2.0--3.2 hr), excessive core temperatures (3.2--3.33 hr), and post high pressure injection (after 3.33 hr). The core average release rate constants used in the calculations are given in Table 9. After 3.33 hr, the release rate constants are assumed to be those in the last column of Table 9. They correspond to a temperature of 1100°C, even though the average temperature of the core was not 1100°C during the period. It was known to be lower because the core was covered with water. However, it is likely that leaching of iodine from fuel occurred after recovery of the core.[6] Also iodine which had plated out on internal surfaces was subject to leaching after recovery. Table 12 shows that 20% of the core iodine inventory is calculated to have escaped to the RCS up to 3.33 hours. After 3.33 hours, approximately 6% is calculated to have escaped to the RCS. The total for the 13.25 hour period is calculated to have been 26%.

Deposition on Reactor Vessel Internals

The reactor vessel internals have a surface to coolant volume ratio of 0.2 cm^{-1}. See Reference 41 for a discussion of the surface to volume ratio used. This relatively large surface to volume ratio, coupled with even a moderate residence time of gases, provides an opportunity for the deposition or condensation of gases. The purpose of this section is to explore the likelihood that deposition on reactor vessel internals was a significant mechanism for reducing releases of radioiodine to the TMI-2 containment.

The following is a simplified model describing the removal process over short time periods:

TABLE 9. CORE AVERAGE RELEASE RATE CONSTANTS USED FOR TRANSPORT CALCULATIONS

Element	Release Rate Constant (s^{-1})		
	2.0--3.2 hr	3.2--3.33 hr	3.33--13.25 hr
I, Xe, Cs	1.7×10^{-5}	3.0×10^{-4}	2.7×10^{-6}
Te	3.3×10^{-6}	1.3×10^{-4}	1.0×10^{-7}
Sb	7.5×10^{-7}	1.8×10^{-5}	1.7×10^{-8}

$$\frac{dq_a}{dt} = I + r\ q_s - v_d(A/V)\ q_a - Q/V\ q_a \qquad (1)$$

$$\frac{dq_s}{dt} = v_d(A/V)q_a - r\ q_s \qquad (2)$$

where

q_a = quantity of iodine in free volume (Ci)
I = iodine input rate to the vessel (Ci/s)
r = resuspension rate from surfaces (s^{-1})
q_s = quantity of iodine on surfaces (Ci)
v_d = deposition velocity of iodine to surfaces (cm/s)
A/V = surface to volume ratio of reactor vessel internals (cm^{-1})
Q = steam and hydrogen flow rate. (cm^3/s)

Deposition measurements[42] for iodine generated as HI onto stainless steel prefilmed with an oxide coating showed that v_d decreased with increasing temperature. At 400°C, the temperature recorded in the hot leg area during the period of fuel damage, v_d was reported to be approximately 0.005 cm/sec.[42] The lower regions of the reactor vessel were probably hotter so that 0.005 cm/sec represents an upper limit. More recent measurements[47] suggest that the deposition velocity for CsI on prefilmed 304 stainless steel also decreases with increasing temperature. The lowest temperature used was 550°C where the deposition velocity was measured to be approximately 0.001 cm/sec. By extrapolating the deposition velocity versus temperature curve (Figure 7 in Reference 47) to 400°C, a deposition velocity of 0.015 cm/sec is obtained. As is discussed below, at concentrations of CsI which may have been present in the TMI-2 core area, condensation would have occurred at a temperature higher than 400°C. This is discussed below.

Resuspension is a major contributor to air concentrations only when the surface activity is large. During the early periods of deposition the surface activity is small and resuspension is relatively unimportant.

Values of r ranging from 10^{-9} to 10^{-5} sec^{-1} have been observed.* In this calculation a value of $r=10^{-6}$ sec^{-1} is used. Using 10^{-5} or 10^{-9} sec^{-1} will produce approximately the same results.

As a first approximation, the vapor flow from the core area is assumed to be through the PORV after the block valve was opened at approximately 3.2 hours. This flow ranged from 30 lb/sec for a short time after the 3.2 hour opening to 15 lb/sec for most of the time thereafter. A value of 20 lb/sec (550 kg/min) is used for this calculation. For the period from 3.2 to 8 hours after the trip, pressure ranged from 2000 psi to 1200 psi. Most of the iodine is calculated to have been released during this period (see Table 12). The corresponding range in specific volume of steam during the period is 0.19 to 0.36 ft^3/lb. For the purposes of this calculation a specific volume of 0.3 ft^3/lb is used for the first five hours. This is equivalent to a flow of 360 cfm and the ratio $Q/V = 0.004$ s^{-1}. With this value of Q/V and $A/V = 0.2$ cm^{-1}, $v_d = 0.005$ cm/s, and $r = 10^{-6}$ s^{-1}, equations 1 and 2 were used to approximate the fraction of the iodine retained by internal reactor surfaces.

The calculation shows that less than 0.4% of the iodine released from the fuel is expected to be retained on interior reactor surfaces. Even if the deposition velocity were 0.015 cm/s, the calculated retention would be only 0.7%. It appears that under average conditions it is not likely that deposition, as thought of in the conventional way on internal surfaces, could have resulted in a significant loss of iodine. During approximately 1/3 of the five hour period, the block valve was closed and Q/V was very small. The remaining 2/3 of the time Q/V was approximately 50% larger. Equations 1 and 2 were used to calculate the deposition during the two time periods assuming that the release rate to the RCS was the same for both periods. A value of $Q/V = 0.00045^{-1}$ was used for the closed block valve case. The results show that even under these non average conditions, conventional deposition analyses show that internal vessel surfaces were probably not sites for significant deposition of iodine.

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*10^{-9} sec^{-1} is derived from ^{129}I measurements at TMI-2. See Table 15. 10^{-5} sec^{-1} is from Reference 35.

Another process by which iodine might be retained on reactor internal surfaces is the condensation of CsI. The following range of conditions are estimated for the early stages of the accident:

Molar ratio of cesium to iodine: ~10

Molar ratio of iodine to water:

Maximum of $\sim 1 \times 10^{-4}$, equivalent to 14 g I leaving the core per minute;

Typically $\sim 4 \times 10^{-5}$, equivalent to 5 g I per minute;

Minimum of $\sim 5 \times 10^{-6}$, equivalent to 0.7 g I per minute.

(If all the water in the primary system (as steam) were mixed with all the iodine in the core the molar ratio would be about 5×10^{-6}. Page 5.4, Reference 22).

Temperature of plenum: ~1100°C at bottom to ~400°C at top.

Pressure: ~1200 to ~2000 psi.

Molar ratio of H to O: 2.0 (reducing).

Under these conditions iodine is present mostly as CsI (Appendix C, Reference 22). (See also References 48 and 49).

The temperatures below which CsI would condense for various I/H_2O ratios (and Cs/I = 10 and p = 14.7 psi) are (Table C.1, Reference 22):

I/H_2O	T_{CsI}
2×10^{-3}	760°C
2×10^{-5}	555°C
2×10^{-7}	418°C

Hence it seems likely some CsI would condense on the cooler part of the plenum at atmospheric pressure. Under the range of accident pressures the vapor pressure of CsI changes only a few percent. (For example, at 738°C the vapor pressure is 1 mm Hg at 14.7 psi, 1.06 mm Hg at 1200 psi, and 1.11 mm Hg at 2000 psi). Thus, higher pressures do not change this conclusion.

If an appreciable fraction of the iodine was in the core area as CsI, it appears that conditions favoring condensation did exist where temperatures approached the hot leg temperature. However, it is difficult and beyond the scope of this work to quantify the likely loss of CsI due to condensation, and we have assumed no loss due to CsI deposition on reactor internal surfaces.

Model for Iodine Transport from RCS to Reactor and Auxiliary Buildings

The transport of fission products from the core to the RCS and subsequently to the Reactor and Auxiliary Buildings is a dynamic process that involves both release from the fuel and discharge of reactor coolant from the RCS as a function of time.

The general rate equation for the inventory of a radioactive nuclide in the reactor coolant for this model is:

$$\frac{dN_c}{dt} = \alpha N_f + F \lambda_{i-1} N_{i-1,c} - \lambda N_c - \beta N_c - \gamma N_c \qquad (3)$$

where

N_c = inventory of nuclide in reactor coolant (atoms)

α = escape rate coefficient (s^{-1}) for nuclide (see Table 9)

N_f = inventory of nuclide in fuel (atoms) (see Table 7)

$F \lambda_{i-1} N_{i-1,c}$ = input (atoms/s) from precursor in coolant which decays to nuclide of interest

λ = decay constant of nuclide (s^{-1})

β = removal coefficient for discharge via letdown (s^{-1}) (see Table 10)

γ = removal coefficient for discharge from the RCS, e.g., through pressurizer (s^{-1}) (see Table 11).

TABLE 10. ESTIMATED LETDOWN FLOW RATES*

Time (hr) After Shutdown	Estimated Letdown Flow Rates (kg/min)
2.0--8.0	265
8.0--9.3	133
9.3--14.0	265
14.0--14.8	151
14.8--15.3	303
15.3--18.5	227
18.5--26.5	0.0
26.5--35.0	95

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* To obtain the removal coefficient, ß, divide flow rate by the mass of reactor coolant, 11500 kg.

TABLE 11. ESTIMATED COOLANT FLOW FROM RCS[1]

Time (hr) After Trip	Estimated Discharge Rate (kg/min)
2.0--2.08	270
2.08--2.33	340
2.33--3.20	110[2]
3.20--3.33	770
3.33--3.42	11600
3.42--3.50	620
3.50--3.51	530
3.51--3.67	110[2]
3.67--3.75	520
3.75--4.00	630
4.00--4.33	560
4.33--4.58	540
4.58--4.83	480
4.83--5.17	500
5.17--5.72	460
5.72--5.83	3100
5.83--6.17	3400
6.17--7.00	3400
7.00--7.50	3300
7.50--7.63	6300
7.63--7.75	760
7.75--7.83	650
7.83--7.92	540
7.92--8.00	440
8.00--8.17	330
8.17--8.50	270
8.50--8.75	200
8.75--9.23	180
9.23--9.24	160
9.24--9.33	110[2]
9.33--9.50	160
9.50--9.80	110[2]
9.80--9.82	380
9.82--10.0	210
10.00--10.33	1700
10.33--10.58	1500
10.58--11.20	170
11.20--12.57	110[2]
12.57--13.25	250

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1. To obtain the removal coefficient λ, divide flow rate by the mass of reactor coolant, 11500 kg.

2. Estimated leakage from relief valves based on pre accident estimates.

Similar equations apply to each nuclide of interest in the coolant and to the same nuclides in the fuel. They form sets of simultaneous differential equations that describe the behavior of each nuclide. The equations were solved numerically using the RADTRAN[50] computer code which also computes the cumulative inventories of activity transported (a) to the Auxiliary Building via the letdown and (b) to the Reactor Building via the discharge through the pressurizer.

The rate constants used in the calculations are provided as input. The estimated average rate constants for transport from fuel to coolant were given in Table 9 for the three time periods of interest. The removal coefficients of fission products via letdown were computed using data on letdown flow rates as a function of time.[24,25] The estimated letdown flow rates for various time periods are given in Table 10.

Discharge of RCS liquid out of the PORV was calculated using the Moody critical flow tables and the RCS hot leg temperatures and pressures. The calculated flow rates out of the PORV are presented in Figure 1. The total release from the PORV was estimated to be 80,000 gallons. The total quantity of water known to have been injected into the RCS up to 0800 on March 30, 1979 was approximately 265,000 gallons, based on borated water storage tank (BWST) level changes and high pressure injection (HPI) system operation.[26] Release rates of coolant were adjusted so that the total coolant mass in the basement at 13 hours was 180,000 gallons with the remaining 85,000 gallons residing in the reactor coolant bleed tanks. Various assumptions were made about the timing of coolant release rates. One assumption was that pressurizer code relief valves continued to leak at the preaccident rate during the accident. Another was that the majority of the unaccounted for coolant in the basement was released from the RCS during periods of highest reactor coolant pressure and when the block valve was opened. Table 11 summarizes the release rates used in the RADTRAN calculation. No attempt is made to identify the source of coolant loss. The assumption of periodic high release rates of coolant lead to higher containment air concentrations than would be the case if it were assumed that the release rate was uniform over the entire period.

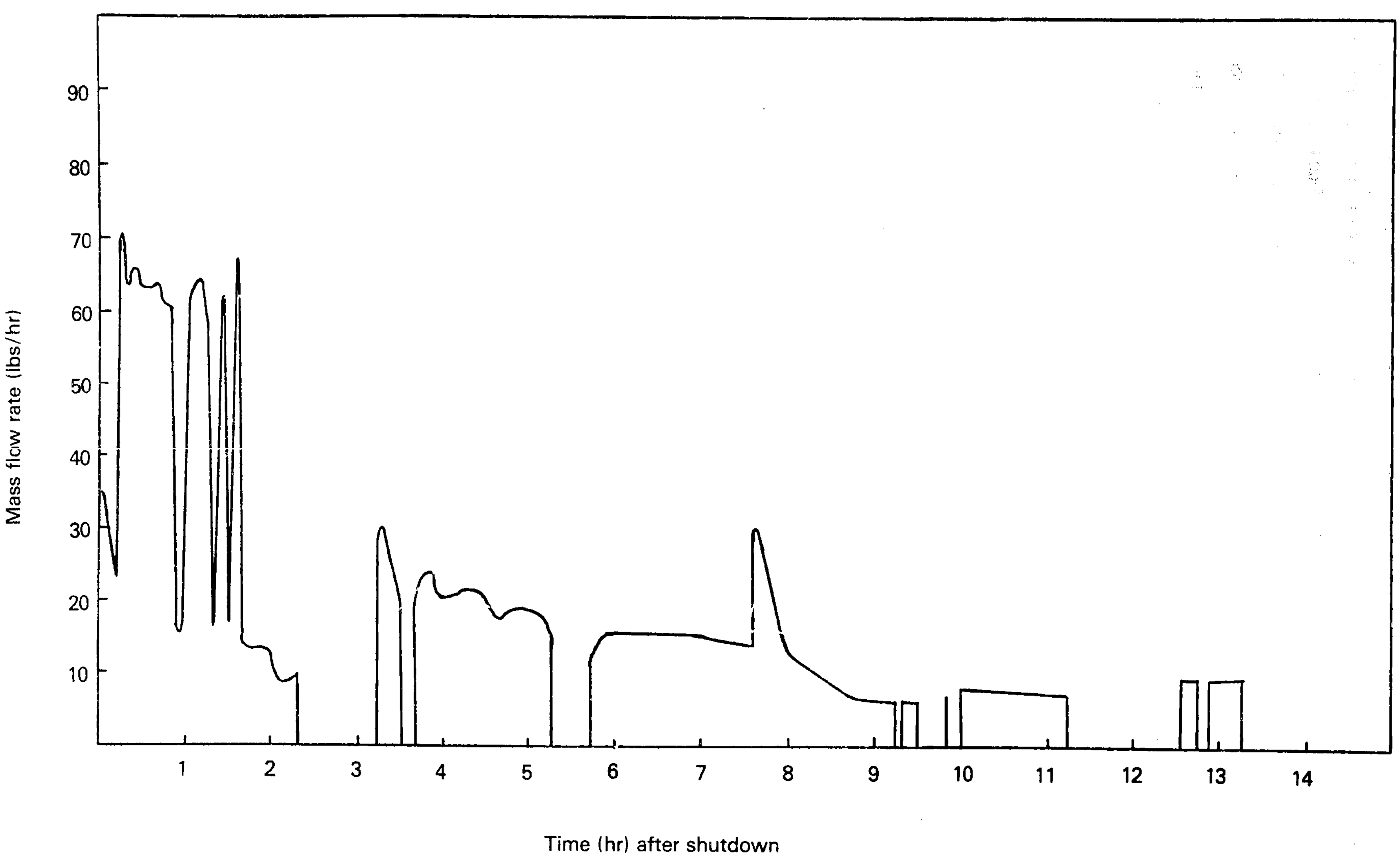

Figure 1. Calculated mass flow from PORV.

Calculated Movement of Iodine from the RCS to the Reactor Building

Initial Transport (2-13 hours)

Early in the accident, iodine from the RCS entered the Reactor Building principally via the Reactor Coolant Drain Tank. The resultant iodine inventory in the Reactor Building, computed using the RADTRAN code as described above, is shown in Table 12. It is assumed that no significant damage to the core had occurred up to 2 hours after shutdown. By the end of the period of block valve operation, (which corresponds to 13.25 hours after shutdown) approximately 17 percent of the iodine inventory was calculated to be in the Reactor Building, carried there in approximately 180,000 gallons of coolant. The calculation assumes that iodine released from the fuel was instantaneously mixed in the coolant and that no significant loss by deposition on the internal surfaces of the vessel, plenum, or piping occurred during transport through the vapor space. Nor is any partitioning of the iodine assumed between the water and the vapor space which separated the core region and the pressurizer.

Transport After 13 Hours

The subsequent movement of iodine to the Reactor Building was computed using measured concentrations of ^{131}I in the reactor coolant[1] and estimated reactor coolant leakage rates.[40] Table 13 shows the cumulative fraction of radioiodine leakage from the RCS (presumably to the basement) after the first coolant sample was taken at hour 37.

Calculations similar to that described above for iodine have also been performed for the cesium inventory using data on ^{137}Cs concentrations in the RCS. Calculations in Reference 40 estimate that 26% of the cesium inventory was discharged from the RCS after 36 hours. Approximately 45% of the cesium inventory was found in the basement on August 28, 1979.[6] That implies that 19% entered the basement prior to 36 hours. As noted earlier, the initial discharge of Cs is believed comparable to that predicted for I which is calculated here to have been 17% during the first 13 hours. Seventeen percent at 13 hours is consistent with the 19% estimate at 36 hours.

TABLE 12. ESTIMATED FRACTION OF ORIGINAL CORE IODINE INVENTORY IN THE RCS, REACTOR BUILDING AND AUXILIARY BUILDING

	Fraction of Core Inventory		
Time (hr) After Shutdown	Reactor Coolant System	Reactor Building	Auxiliary Building
2.08	0.0047	8.7×10^{-6}	8.4×10^{-6}
2.15	0.0087	2.7×10^{-5}	3.0×10^{-5}
2.33	0.019	1.3×10^{-4}	1.4×10^{-4}
3.20	0.067	8.4×10^{-4}	0.0018
3.33	0.19	0.0030	0.0026
3.41	0.16	0.034	0.0033
3.50	0.16	0.036	0.0040
3.67	0.15	0.039	0.0051
3.75	0.15	0.039	0.0056
3.95	0.15	0.042	0.0070
4.33	0.14	0.048	0.0095
4.58	0.14	0.051	0.011
4.83	0.14	0.054	0.013
5.17	0.13	0.058	0.015
5.72	0.13	0.063	0.018
5.83	0.12	0.070	0.019
7.00	0.064	0.13	0.023
7.50	0.051	0.15	0.024
7.63	0.045	0.15	0.025
7.83	0.044	0.16	0.025
7.91	0.045	0.16	0.025
8.17	0.045	0.16	0.026
8.50	0.047	0.16	0.026
8.75	0.048	0.16	0.026
9.33	0.050	0.16	0.027
9.90	0.052	0.16	0.028
10.33	0.049	0.16	0.029
10.58	0.047	0.17	0.030
11.33	0.049	0.17	0.031
12.33	0.053	0.17	0.033
13.25	0.055	0.17	0.035

TABLE 13. RADIOIODINE FROM RCS LEAKAGE AFTER 37 HOURS

Sampling Time (1979)	Hours Since Shutdown	Leakage From RCS (ml)	^{131}I Conc.* (mCi/ml)	Cumulative Fraction of Core Inventory
1700, 3/29/79	37	--	1.45×10^{4}	--
0730, 4/10/79	314	1.46×10^{8}	9.5×10^{4}	0.061
0600, 4/16/79	456	7.47×10^{7}	5.1×10^{3}	0.089
0700, 4/17/79	481	1.31×10^{7}	4.8×10^{3}	0.094
2140, 4/18/79	520	2.03×10^{7}	4.1×10^{3}	0.102
0900, 4/22/79	603	4.03×10^{7}	2.4×10^{3}	0.114
0820, 4/25/79	674	3.45×10^{7}	1.7×10^{3}	0.123
0500, 5/2/79	839	7.97×10^{7}	7.1×10^{2}	0.140
1200, 5/10/79	1038	7.23×10^{7}	3.9×10^{2}	0.157
1100, 5/18/79	1229	6.93×10^{7}	2.1×10^{2}	0.174
1200, 6/1/79	1566	1.22×10^{8}	63	0.204
1400, 6/7/79	1711	5.26×10^{7}	24	0.213
1200, 6/13/79	1854	1.35×10^{7}	13	0.215
1000, 6/19/79	1996	1.34×10^{7}	7.7	0.217
1300, 6/27/79	2191	1.84×10^{7}	3.5	0.219
1230, 7/5/79	2383	1.80×10^{7}	2.03	0.222
1000, 7/11/79	2524	1.33×10^{7}	1.5	0.224
1200, 7/17/79	2670	1.37×10^{7}	0.55	0.226
1200, 7/24/79	2838	1.58×10^{7}	0.43	0.228
1200, 7/31/79	3006	6.78×10^{6}	0.17	0.229
1045, 8/8/79	3197	7.69×10^{6}	0.01	0.229

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* Decay corrected to 0400 on March 28, 1979.

Transport of Iodine Released to the Reactor Building

The model employed to estimate the distribution of iodine following its release to the Reactor Building is shown in Figure 2. The model is a combination of previous modeling approaches that provides for explicit consideration of airborne iodine species. (See footnote on Page 14 for explanation of species). The processes treated explicitly by the model and the important assumptions employed are described below, following a brief discussion of the background of experimental work that led to its formation.

Releases of iodine into reactor containments or closed volumes simulating them have shown that, under either wet or dry conditions, the concentration of airborne activity decreases rapidly with time.[27,28] Within a short period an equilibrium is established and the air concentration remains essentially constant thereafter. (A number of illustrative examples of such behavior are contained in the proceedings of an NSAC workshop on radioiodine.[37]) For conditions when iodine and steam are released simultaneously, a model was developed to account for transport of airborne iodine from the air to the film of moisture on condensing surfaces and thence to the basement.[31] That basic model was used in the Reactor Safety Study[32] to estimate the behavior of inorganic iodine in reactor containment buildings. A somewhat different model, with rate constants derived from the Containment Systems Experiment (CSE) data, has also been described.[33]

Studies of iodine behavior in airborne discharges at operating boiling water reactors led to the development of a simple iodine deposition-resuspension model to explain the relative depletion of short-lived radioiodines during transport through buildings and ventilation systems.[34] Laboratory studies of deposition and resuspension of iodine were conducted to estimate resuspension rate constants for surfaces commonly found in nuclear power plants.[35] Because changes in iodine species were observed in both the in plant and laboratory experiments, the conversion was believed to occur on the surfaces. A more complex model was developed to reflect these observations and species changes that have also been observed within containment buildings of pressurized water reactors.[36,37]

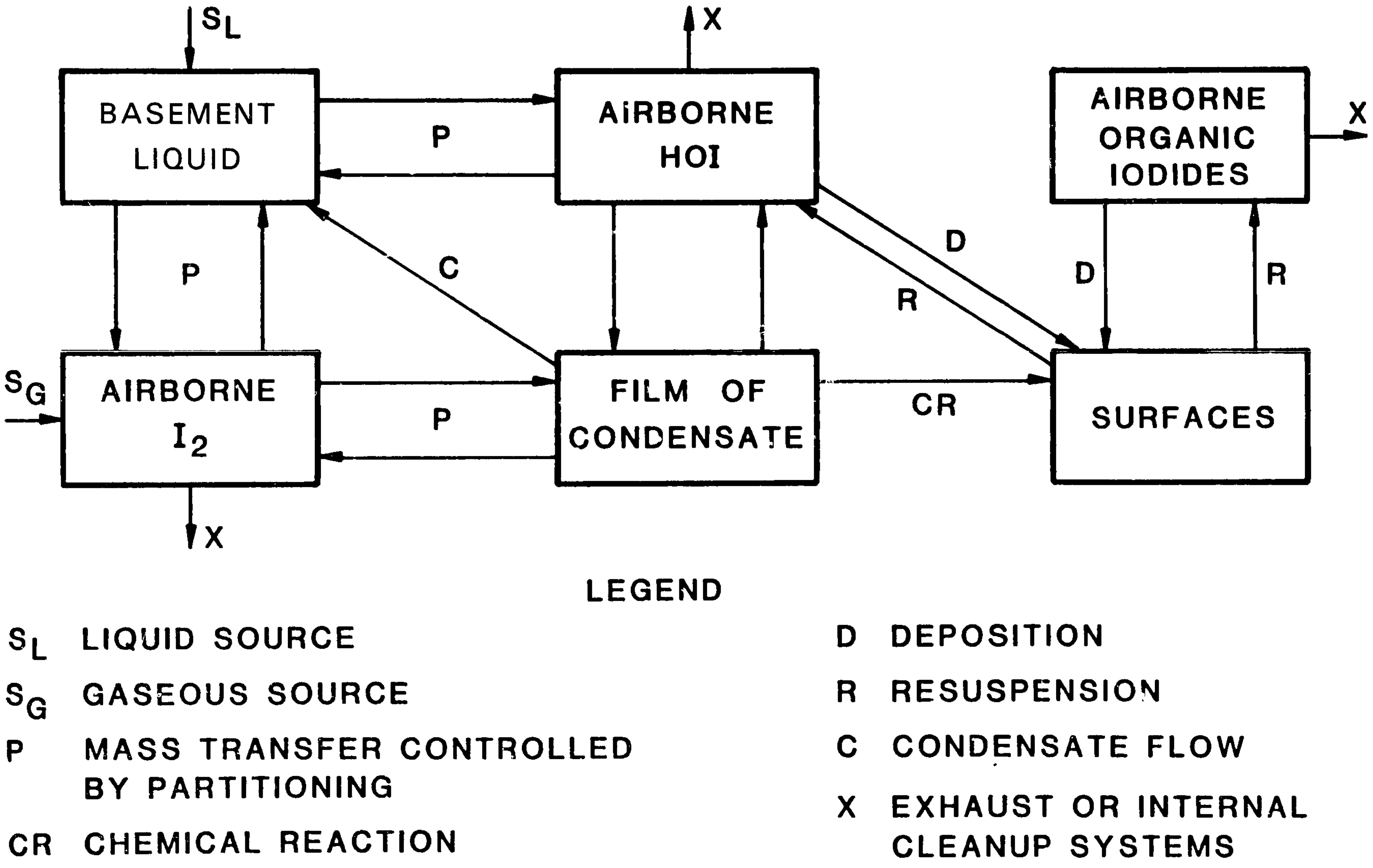

Figure 2. Mathematical model of radioiodine transport.

The model in Figure 2 is a combination of the deposition-resuspension modeling approach and that developed as part of the Containment Systems Experiment (CSE) program.[27] Radioiodine can enter the Reactor Building as a gas or in liquid discharged from the RCS. It is assumed in Figure 2 that iodine was present in the containment atmosphere as a relatively reactive gas such as I_2. As discussed above, measurements of surface concentrations of ^{137}Cs and $^{131,129}I$ support this assumption. Iodine could also have been discharged in coolant after dissolving as CsI or I_2. In this case, it would run directly to the basement or be entrained by escaping steam and transported to the containment atmosphere. The airborne and liquid source terms for the Reactor Building are represented by S_G and S_L, respectively, in Figure 2. Because the pH and temperature of the liquid were both high, the capability to treat both I_2 and HOI as volatile species released from the water is included.[43] However, in this work only I_2 like releases are considered.

The Reactor Building atmosphere is treated as a single volume in the model. The surface-to-volume ratio for the lower portion of the building (below the 305' level) is 0.008 cm^{-1} which is twice that for the whole containment (see Table 14). However, the four coolers which operated throughout the accident provided substantial mixing of the air in the Reactor Building. The ratio of cooler design flow rate to building volume is about 0.07 per minute. Experimental studies have shown that, when there is air recirculation, the mass transport coefficient from air to building walls is approximately twice that for natural convection alone.[38,39] It should be noted that even though the higher deposition rate was applicable at TMI, it was not used here. In this study, the higher the assumed deposition rate the lower the calculated airborne concentrations. Because it is not known what iodine species were present, it was thought best to use the lower deposition rate.

As pointed out above, all studies of iodine behavior in enclosed spaces shows a decrease in concentration to an equilibrium level. In the present model, the equilibrium is assumed to reflect a balance between transport to and from wet and dry surfaces. At equilibrium, the surfaces, and condensate if present, have been found to contain the overwhelming

TABLE 14. SURFACE AREAS AND COMPARTMENT VOLUMES
TMI UNIT 2 REACTOR BUILDING[52,53]

I. RCDT Discharge Compartment	
(A) Surface Area	
Walls	3705 ft^2
Floor	719 ft^2
Ceiling	600 ft^2
(B) Volume	
Compartment	15,458 ft^3
II. Remaining 282'6" to 303'	
(A) Surface Area	
Containment Walls	6257 ft^2
D-Ring Walls	4777 ft^2
Miscellaneous Walls	11595 ft^2
Elevator Cubicle	1558 ft^2
Floor	6373 ft^2
Ceiling	6373 ft^2
(B) Volume	82,633 ft^3
III. Elevation 305' to 345'6"	
(A) Surface Area	
Containment Walls	17,098 ft^2
D-Ring (outside)	12,413 ft^2
Air Coolers	3,138 ft^2
Elevator and Stairs (outside)	3,596 ft^2
Floor	11,230 ft^2
Ceiling (3x Floor)	33,690 ft^2
(B) Volume	306,073 ft^3
IV. 347'6" to Dome	
(A) Surface Area	
Containment Walls	40,943 ft^2
Dome	28,000 ft^2
D-Ring (outside) and top	7,054 ft^2
Steam Generator Cavity (to 281'6")	30812 ft^2
Elevator and Stairs	2150 ft^2

TABLE 14 (continued)

Liner		11,571 ft^2
Floor		16525 ft^2
(B) Volumes		
Steam Generator Cavity		232,980 ft^3
Liner		34,418 ft^3
Floor-Dome		1,407,038 ft^3
V. Totals		
(A) Surface Area:		
280 elevation	36932.2	
305 elevation	81,165	
347 elevation	137,053	
RCDT	5024	
Total	260,174 ft^2	
Volumes		
RCDT discharge compartment		15,458 ft^3
280 elevation		82,633 ft^3
305 elevation		306,073 ft^3
347'el		1,674,436 ft^3
Total		2,078,600 ft^3
Miscellaneous Equipment		- 28,600 ft^3
		2,050,000 ft^3

majority of the initially injected activity. Desorption or resuspension of radioiodine deposited on the surfaces occurs very slowly. In the full vessel Containment Systems Experiment (CSE) tests,[27] for example, approximately 52% of the activity was estimated to be retained by painted surfaces, even after attempted decontamination. Other experimental programs using small samples have shown tenacious retention of iodine by a variety of surfaces.[29,30] Thus, it would be expected that the radioiodine released into the Reactor Building atmosphere would be deposited on condensing and noncondensing surfaces and reach an equilibrium in a short time (hours). The equilibrium between surfaces, basement liquid, and the air would be maintained unless the physical conditions within the containment changed sufficiently. This equilibrium concept is important because it provides the connection between the calculated early behavior of iodine at TMI and later measurements.

Transport of I_2 to and from the basement liquid and condensate film is assumed to depend upon the difference in concentration between the bulk air and the liquid-air interface. Transport of organic iodides to and from surfaces is treated using the deposition-resuspension approach. Transport of HOI can be handled in either way. No mechanistic hypothesis that accounts for species changes on surfaces is proposed. The observations of changes in airborne species are reflected in the resuspension rate constants.

The mathematical relationships describing the processes included in the model are given in Appendix A. The basis for selection of the rate constants used in the calculations is summarized in Table 15. To the extent possible, measurements of iodine behavior in the TMI-2 containment were used to obtain the rate constants.* Fundamental parameters, like the surface to volume ratio given above, were obtained from descriptions of the Reactor Building. The free volume was reduced to account for the presence of basement liquid.

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*Values of the rate constants were determined by fitting the model to the ^{129}I air concentrations during and after the purge.[17]

TABLE 15. RATE CONSTANTS USED IN AIRBORNE IODINE TRANSPORT MODEL

Process	Parameter Value	Basis
A. Air-to-Surface Transport Coefficient (cm/s)		
k_2 (I_2 to film)	3.75×10^{-4} s^{-1}	CSE results[38] without recirculating air
δ_3 (HOI to wall)	0.0012 cm/s	Derived from TMI-2 post-purge ^{129}I air concentration measurements[17] and ^{129}I surface concentration data.
δ_4 (Organic to wall)	0.00029 cm/s	
$\frac{k_1 A_L}{V}$ (I to basement surface)	5.3×10^{-7} s^{-1}	Derived from TMI-2 post-purge ^{85}Kr air concentration measurements.[17]
B. Overall Iodine Partition Coefficient, H	10^4	CSE Natural Response Tests[27] (Data considered inadequate to justify split between I_2 and HOI).
C. Condensation Rate Constant for Film to Basement Flow, k_c	1.36×10^{-3} s^{-1}	Best estimate for conditions during the accident. Film thickness assumed to be 0.1 cm.
D. Resuspension Rate Constants		
r_3 (HOI)	2.1×10^{-9} s^{-1}	Derived from TMI-2 post-purge ^{129}I air concentration measurements[17] and ^{129}I surface concentration data.
r_4 (Organic Iodides)	4.7×10^{-9} s^{-1}	
E. Reaction Rate for I With Paint		
k_p	2.3×10^{-3} s^{-1}	BMI measurements with Phenoline 368 paint.[30]

The model was used to calculate the behavior of iodine when assuming that: (1) all iodine was released from the primary coolant into the basement and (2) all iodine was released from the primary coolant directly to containment atmosphere. Of specific interest in this comparison was the relative contribution to the air and surface concentrations for the two release pathways. Table 16 shows the results. As can be seen, under the conditions assumed, releases to the basement liquid would result in almost no airborne and surface radioiodine activities compared to releasing the activity directly to reactor building air. This conclusion is most dependent on the assumed partition coefficient for iodine in basement water. The value used was 10^4. If the iodine were released as CsI, 10^4 is probably too small. In laboratory tests[43] the ratio of water to air concentration when iodine was added as NaI at 25°C was 2×10^6 and 2×10^7 at pH 5.2 and 6.5 respectively. Although the temperature was higher than 25°C in the TMI-2 basement the pH was also higher. Therefore, for leakage of CsI to the basement, use of a higher value of the partition coefficient would be appropriate. This would lead to air and wall concentrations even smaller than those in Table 16. The overall effect of river water inleakage to containment from the air coolers is not known. However, the inleakage would lower the average temperature and pH of basement water.

The model was run with a source term derived from differentiating the cumulative iodine quantities released to the reactor building, computed using 50 RADTRAN (see Table 12). Table 17 shows the results of the calculation for times out to 1000 hours after shutdown. In the calculation, it is assumed that all iodine is released directly to the reactor building atmosphere.

The checks on the calculated values in Table 17 are the later measured values on the surface and air concentrations of radioiodine. The procedure used to normalize the calculated values with the later measurements was to multiply the release rates in Table 17 by the ratio of the measured to the calculated surface quantity. The maximum measured value, 0.7% was used (see Table 4). The normalizing ratio was 0.07 because the calculated value was 10%, assuming all the iodine was released to Reactor Building air. Figure 3 shows the resulting quantities in air and on surfaces out

TABLE 16. COMPARISON OF CALCULATED REACTOR BUILDING SURFACE AND AIRBORNE RADIOIODINE ACTIVITIES FOR RELEASES TO THE AIR AND TO THE BASEMENT LIQUID

Time (hr) After Shutdown	Fraction of Total Release[a]					
	Airborne		On Surfaces		On Basement	
	Release To Basement[b]	Release To Air[c,d]	Release To Basement[b]	Release To Air[c,d]	Release To Basement[b]	Release To Air[c,d]
1	$1.2x10^{-7}$	0.046	--	0.025	0.083	0.012
2	$3.5x10^{-7}$	0.059	$2.1x10^{-7}$	0.064	0.17	0.038
4	$8.8x10^{-7}$	0.063	$1.2x10^{-6}$	0.17	0.33	0.098
6	$1.4x10^{-6}$	0.063	$3.1x10^{-6}$	0.27	0.50	0.16
8	$2.0x10^{-6}$	0.063	$5.8x10^{-6}$	0.38	0.67	0.22
12	$3.0x10^{-6}$	0.063	$1.4x10^{-5}$	0.59	1.0	0.35
22	$3.3x10^{-6}$	$2.2x10^{-4}$	$4.1x10^{-5}$	0.63	1.0	0.37
42	$3.3x10^{-6}$	$4.8x10^{-4}$	$9.4x10^{-5}$	0.63	1.0	0.37
62	$3.3x10^{-6}$	$7.0x10^{-4}$	$1.5x10^{-4}$	0.63	1.0	0.37
82	$3.3x10^{-6}$	$8.9x10^{-4}$	$2.0x10^{-4}$	0.63	1.0	0.37
112	$3.5x10^{-6}$	$1.1x10^{-3}$	$2.8x10^{-4}$	0.63	1.0	0.37

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a. Release rate was 1 Ci/sec for 12 hours or $4.32x10^{4}$ Ci.

b. All iodine released directly to the basement liquid.

c. All iodine released directly to the air.

d. Does not include condensate film on surface.

TABLE 17. CALCULATED DISTRIBUTION OF RADIOIODINE IN THE REACTOR BUILDING

Time (hr) After Shutdown	Release Rate (Ci/sec)[a]	Cumulative Radioiodine Fraction			
		Air	Condensate Film	Surfaces	Basement Liquid
2.08	51	--	--	--	--
3.33		1.5(-3)[b]	1.5(-4)	9.0(-4)	5.3(-4)
3.41	8400	3.1(-2)	1.4(-3)	1.4(-3)	8.2(-4)
3.5	226	2.6(-2)	2.3(-3)	4.2(-3)	2.3(-3)
3.7		2.2(-2)	2.3(-3)	8.1(-3)	5.0(-3)
4.0		1.7(-2)	1.8(-3)	1.4(-2)	9.5(-3)
4.4		1.3(-2)	1.4(-3)	2.1(-2)	1.2(-2)
5.4		9.2(-3)	9.3(-4)	3.0(-2)	1.3(-2)
5.7		8.9(-3)	9.0(-4)	3.3(-2)	1.7(-2)
5.8	930	1.4(-2)	1.1(-3)	3.5(-2)	2.0(-2)
6.0		1.8(-2)	1.7(-3)	3.5(-2)	2.1(-2)
6.5		2.6(-2)	2.5(-3)	4.2(-2)	2.5(-2)
6.7		2.8(-2)	2.8(-3)	4.7(-2)	2.7(-2)
7.6		3.3(-2)	3.2(-3)	7.0(-2)	4.1(-2)
8.0	37	2.0(-2)	2.3(-3)	8.2(-2)	4.8(-2)
8.4		1.3(-2)	1.4(-3)	8.8(-2)	5.1(-2)
9.6		3.8(-3)	4.0(-4)	9.6(-2)	5.6(-2)
9.9 Before Sprays		3.3(-3)	3.4(-4)	9.6(-2)	5.7(-2)
After Sprays	69	1.6(-4)	3.4(-4)	9.6(-2)	5.9(-2)
10.2		9.8(-4)	8.6(-5)	9.6(-2)	6.0(-2)
10.5		1.5(-3)	1.4(-4)	9.6(-2)	6.0(-2)
10.9		2.0(-3)	1.9(-4)	9.7(-2)	6.0(-2)
11.9		2.5(-3)	2.5(-4)	9.8(-2)	6.1(-2)
12.9		2.5(-3)	2.5(-4)	1.0(-1)	6.2(-2)
13.25		2.6(-3)	2.6(-4)	1.0(-1)	6.3(-2)
20.00	0	2.2(-5)	5.0(-8)	1.0(-1)	6.4(-2)
50.00		7.3(-5)	--	1.0(-1)	6.4(-2)
100.00		4.7(-4)	--	1.0(-1)	6.4(-2)
200.00		7.2(-4)	--	1.0(-1)	6.4(-2)
400.00		9.7(-4)	--	1.0(-1)	6.4(-2)
800.00		1.1(-3)	--	1.0(-1)	6.4(-2)
1000.00		1.1(-3)	--	1.0(-1)	6.4(-2)

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a. Release rates calculated by differentiating cumulative releases to the Reactor Building in Table 12. All iodine is assumed to be released to the Reactor Building atmosphere.

b. Read 1.5(-3) as 1.5×10^{-3}.

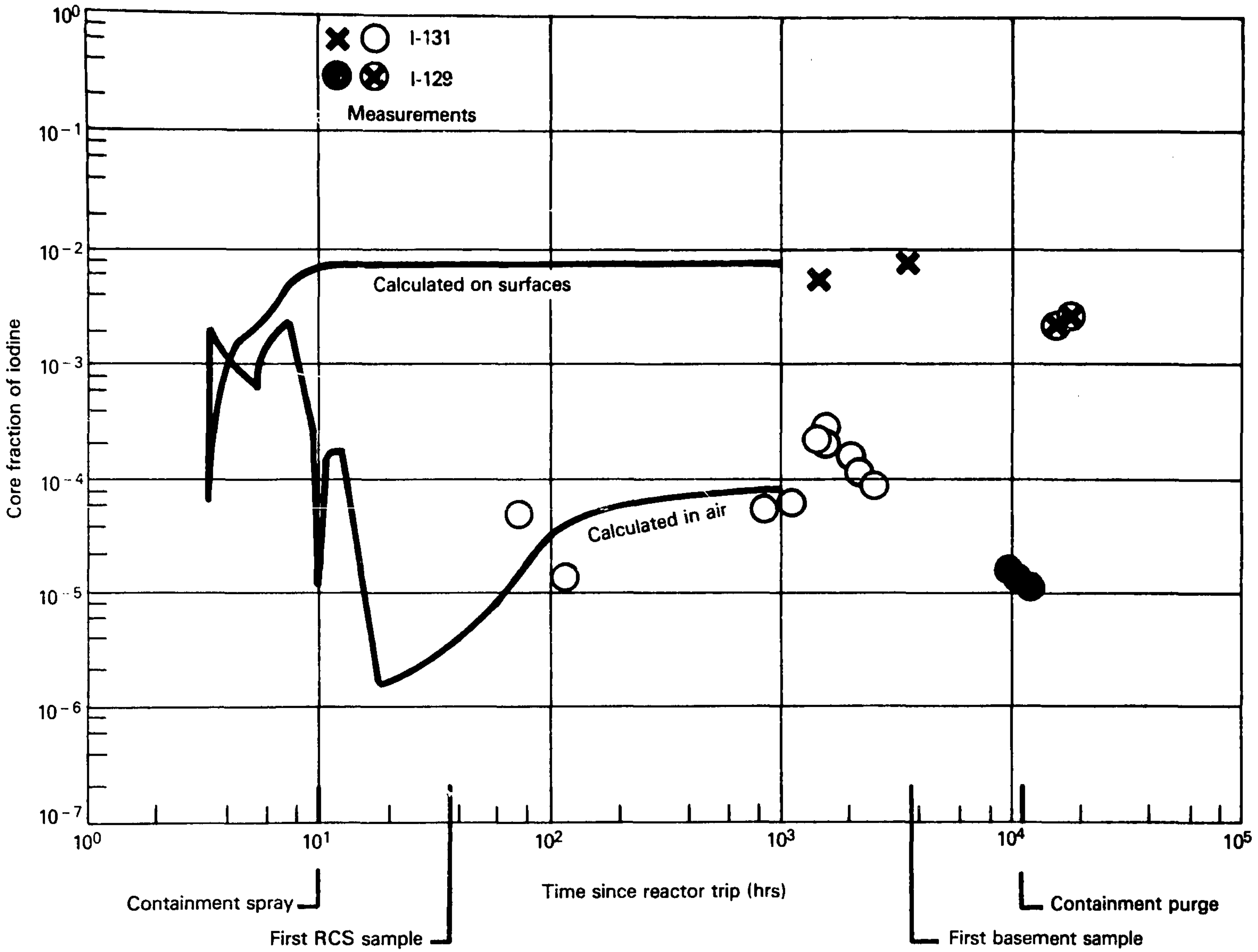

Figure 3. Radioiodine on surfaces and in air of reactor building.

to 1000 hours. Measured values are also shown for comparison. Note that the maximum quantity of radioiodine calculated to have been airborne during the accident was 0.2% of the original core inventory. If none of the radioiodine was held up on primary system surfaces or in the reactor coolant bleed tank (as is assumed in this analysis), then most of it must have been transported directly to the basement liquid during the accident. Table 18 shows the estimated basement quantities assuming that 93% of the surface activity was added directly to the basement. Also included are the estimated quantities which leaked to the basement after the accident. Figure 4 summarizes the calculated and measured radioiodine quantities in the RCS and basement liquids.

Table 18 shows that the calculated quantity in the basement when the first basement sample was taken was 39%. The maximum quantity measured was 19% at that time. (See Table 2). All subsequent measurements have been lower than the calculated values. Two possibilities exist to explain the difference. First, iodine may have been removed from the water by adsorption on already existing particles or by chemical formation of insoluble iodine compounds. Spark-source mass-spectrometric analysis of the August 28, 1979 solutions[7] indicated that enough silver may have been present (up to 0.5 ppm) to precipitate some of the iodine as AgI. Also, more silver than iodine was found in the solids of the sample from August 28, 1979 (up to 8 ppm vs 0.7 ppm[7]) and of the May 14, 1981 sample (16 ppm vs 0.65 ppm ^{129}I[44], or 1.2 ppm total I).

The more compelling reason to believe that the "missing" iodine is in the basement solids and eluding measurement is that ^{137}Cs has been found in the basement liquid in a quantity which is consistent with assumed fuel release rates. However, one cannot rule out the possibility that iodine was retained on reactor vessel internal surfaces or in the reactor coolant drain tank.

TABLE 18. CALCULATED BASEMENT RADIOIODINE

Time (hr) After Shutdown	Calculated Additions During First 13 Hours[a]	Calculated Additions After Accident
3.33	0.0014	--
3.50	0.0062	--
4.0	0.023	--
5.4	0.041	--
6.0	0.054	--
6.5	0.064	--
6.7	0.071	--
7.6	0.106	--
8.0	0.124	--
8.4	0.133	--
9.9	0.148	--
10.9	0.150	--
13.25	0.156	--
314	--	0.061
520	--	0.102
674	--	0.123
1038	--	0.157
1229	--	0.174
1556	--	0.204
2191	--	0.219
2670	--	0.226
3006	--	0.229

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a. Includes 93% of radioiodine calculated to be on surfaces.

b. Calculated from measured RCS concentrations and coolant leakage rates. See Table 13.

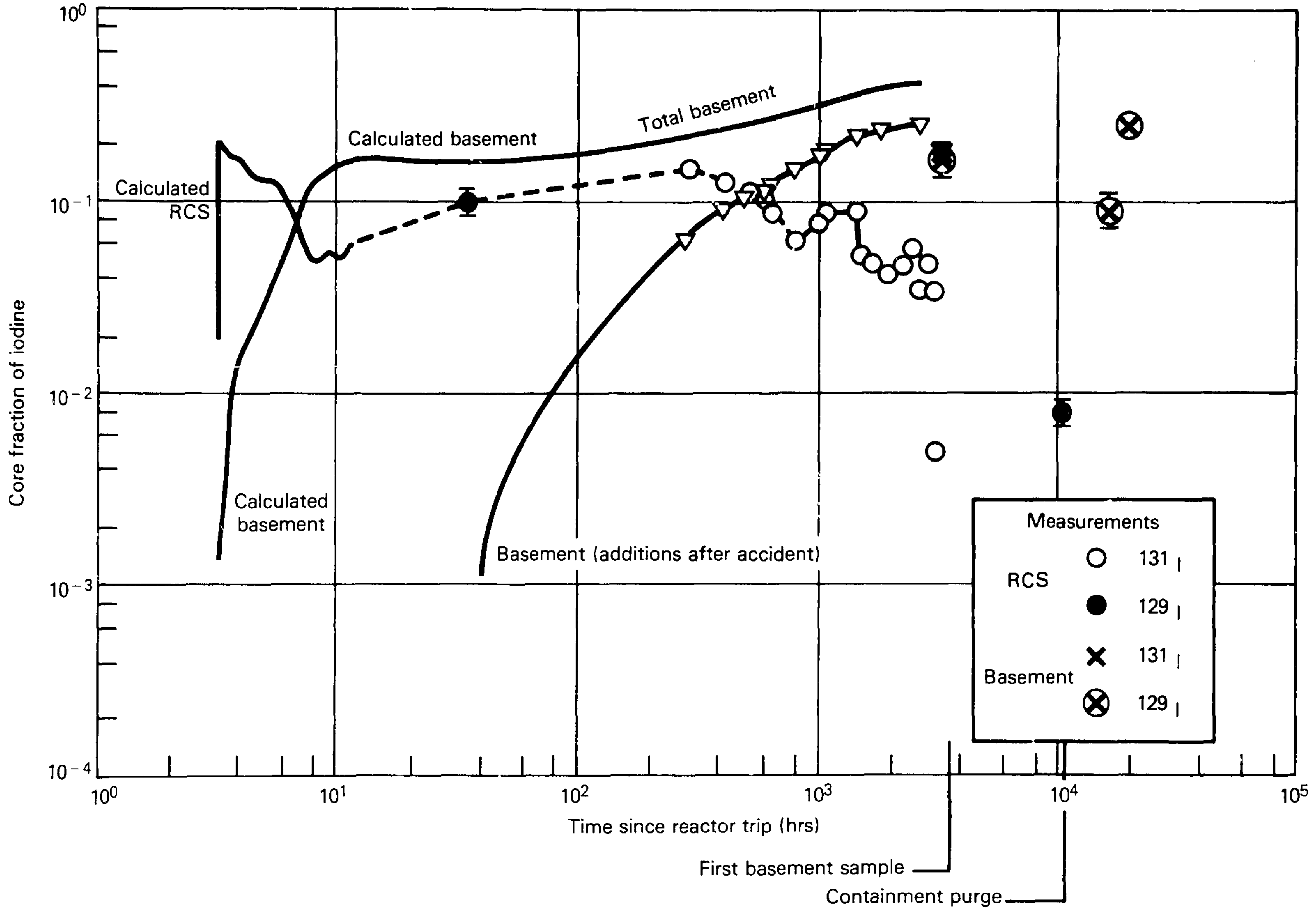

Figure 4. Radioiodine in RCS and reactor building basement liquids.

Calculated Movement of Iodine from the RCS to the Auxiliary Building

Table 12 also contains the estimated cumulative transport of iodine to the Auxiliary Building during the first 13 hours after reactor shutdown. The assumptions mentioned above, good mixing of the iodine with coolant in the reactor vessel and no deposition in transport through the vapor space, also apply to this calculation. The predicted transport agrees with iodine inventories found in the reactor coolant bleed tanks at long times after shutdown, but not with the early measurements (see Table 5). There are a number of possible explanations, none of which provides a completely satisfactory picture:

1. The bulk of the iodine (and the cesium) transported during this period was collected by the letdown demineralizer and did not reach the bleed tanks. At present there are no data on the iodine inventory in the remains of the letdown demineralizer resin. In addition, there is no clear picture of the transport rates of iodine from the RCS to the RCBTs at later times.

2. Transport of activity to the Auxiliary Building may have been partially blocked by the vapor space in the hot leg, with steam condensation providing the only mass transport. If this blockage was effective, it could have operated for an extended period. One A-loop pump was operated only briefly during the period between 3 and 13 hours after shutdown.

If the reported analyses of the RCBT concentrations prior to April 3 are correct, a combination of the two above suggestions seems to be required. Another alternative is:

3. The RCBT concentration data are not valid. It is known that the samples collected were small and that the analysis would have been difficult because of the high concentrations.

These samples may have been diluted prior to analysis. If so it is possible that dilution was overlooked in the calculation of concentration on analyses sheets.

Future radiochemical analyses will provide additional information about the contents of the letdown demineralizer. Additional investigations may shed light on other aspects of this question.

RADTRAN calculations were also carried out for hours 13 to 37. The same release rate constant used from 3.33 to 13 hours was used as the source term throughout the entire period. (See Table 9). At the end of 37 hours the calculation indicated approximately 10% of the original core iodine in the RCS which agrees well with the measured value at 1700 on 3/29/79. However, the calculation also indicated that under the assumed conditions 10% should also have appeared in the auxiliary building. This is higher than the measurements in the RCBTs. However, a significant fraction may have been adsorbed on the letdown demineralizers.

CONCLUSIONS

- o Measurement results show that at the time of the first basement sample (August 28, 1982), from 17% to 28% of the iodine originally in the fuel could be accounted for in the Reactor Building. Using the same accounting procedure, approximately 51% of the ^{137}Cs could be accounted for. The most plausible explanation for the difference is that chemical reactions took place in the basement liquid during and after the accident and iodine was precipitated to the bottom of the Reactor Building Basement. However, retention of iodine in the RCS or RCDT should not be dismissed as an explanation.

- o Measurement results show relatively little iodine on Reactor Building surfaces. The highest quantity measured on surfaces was 0.7% of the original core inventory.

- Air measurements have shown airborne iodine activities ranging from 0.002% to 0.03% of the core inventory. First air measurements, made 75 hours after shutdown, indicated that 0.005% of the core inventory was airborne. Even if all this iodine is assumed to be organic, the observed amount is less than 1/100 that specified in Regulatory Guide 1.4 or WASH-1400 (Reference 32).

- Measurements of ^{137}Cs and radioiodine on Reactor Building surfaces suggest that radioiodine deposited as elemental iodine. This does not preclude the possibility of iodine existing as CsI in the RCS. In fact, basement water measurements suggest that iodine entered the basement as the iodide ion.

- Analyses indicate that the source of surface iodine contamination was iodine that was released directly to containment atmosphere during the accident as opposed to later partitioning from basement water.

- The quantity of iodine on surfaces is related to past air concentrations. Mathematical models, benchmarked with measurement data, show that the maximum quantity of radioiodine in Reactor Building air during the accident was 0.2% of the original core inventory.

RECOMMENDATIONS FOR FUTURE WORK

- Measure the concentration of ^{129}I in the residual material remaining on the 281' level after the basement water has been drained. It is important to know if the missing iodine is on solid material in the basement. It is also necessary to obtain an estimate of the total mass of residual material.

- Measure the concentration of ^{129}I in residuals at the bottom on the RCDT and estimate the total mass of residuals.

- Sample the resin (or what is left of it) in the letdown demineralizer and analyze for ^{129}I. Useful information on the ^{129}I content on the letdown demineralizer may be obtainable by sampling the liquid downstream of the demineralizer. On the assumption that the resin has been destroyed by radiation, the water downstream may contain a significant concentration of ^{129}I.

- Analyze surface samples already taken for ^{129}I. Depending on results it may be worthwhile to collect more samples, particularly at the 281' level.

- Perform sensitivity analysis on the model used in this study. Part of the sensitivity analysis would be to evaluate alternative release and transport scenarios; for example, to evaluate consequences of releasing all iodine to containment air in the form of CsI.

- Analyze surface scrapings from reactor vessel internals for ^{129}I. Also measure the $^{129}I/^{137}Cs$ ratio. If internal surfaces retained a significant quantity of iodine, the $^{129}I/^{137}Cs$ ratio should be relatively high.

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APPENDIX A

MODEL OF IODINE BEHAVIOR IN TMI-2 CONTAINMENT

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MODEL OF IODINE BEHAVIOR IN TMI-2 CONTAINMENT

The model used to predict the behavior of iodine released into the TMI-2 containment is shown in Figure 2 in the main text. The differential equations that describe the movement of iodine are as follows.

$$\frac{dq_L}{dt} = S_L + k_1 q_2 + k_1 q_3 + k_c q_F - k_{12} q_L - k_{13} q_L - \lambda q_L \qquad \text{(A-1)}$$

$$\frac{dq_2}{dt} = S_2 + k_{12} q_L + k_{22} q_F - k_1 q_2 - k_2 q_2 - \lambda_v q_2 - \lambda_v q_2 - f_2 q_2 \qquad \text{(A-2)}$$

$$\frac{dq_3}{dt} = k_{13} q_L - k_{23} q_F + r_3 q_s - k_1 q_3 - k_2 q_3 - \delta_3 q_3 - \lambda_v q_3 - f_3 q_3 - \lambda q_3 \qquad \text{(A-3)}$$

$$\frac{dq_4}{dt} = r_4 q_s - \delta_4 q_4 - \lambda_v q_4 - f_4 q_4 - \lambda q_4 \qquad \text{(A-4)}$$

$$\frac{dq_F}{dt} = k_2 q_2 + k_2 q_3 - k_c q_F - k_{22} q_F - k_{23} q_F - k_p q_F - \lambda q_F \qquad \text{(A-5)}$$

$$\frac{dq_s}{dt} = k_p q_F + \delta_3 q_3 + \delta_4 q_4 - r_3 q_s - r_4 q_4 - \lambda q_s \qquad \text{(A-6)}$$

where

q_L = the radioiodine activity (Ci) in liquid in the basement

q_2 = the airborne radioiodine activity (Ci) present as I_2

q_3 = the airborne radioiodine activity (Ci) present as HOI

q_4 = the airborne radioiodine activity (Ci) present as organic iodides

q_F = the radioiodine activity (Ci) in the film of condensed water

q_S = the radioiodine activity (Ci) on the building surfaces

S_L = the rate (Ci/s) at which radioiodine activity enters the basement liquid from the RCS

S_2 = the rate (Ci/s) at which radioiodine activity enters the airspace as I_2

k_1 = rate constant (s^{-1}) for radioiodine transfer from air to basement liquid

k_{12}, k_{13} = rate constants (s^{-1}) for radioiodine transfer from basement liquid to air for I_2 and HOI, respectively

λ = rate constant (s^{-1}) for radioactive decay

k_2 = rate constant (s^{-1}) for radioiodine transfer from air to surface film

k_{22}, k_{23} = rate constants (s^{-1}) for radioiodine transfer from surface film to air for I_2 and HOI, respectively

λ_v = removal rate constant (s^{-1}) for containment purge systems

f_2, f_3, f_4 = removal rate constants (s^{-1}) for containment cleanup systems for I_2, HOI, and organic iodides, respectively

$\delta_3 \delta_4$ = rate constants (s^{-1}) for deposition of HOI and organic iodides, respectively

$r_3 r_4$ = rate constants (s^{-1}) for resuspension of HOI and organic iodides, respectively

k_c = rate constant (s^{-1}) for transport of condensate from the wall surfaces to the basement

k_p = rate constant (s^{-1}) for radioiodine transport from liquid film to painted wall surface.

The basement-to-air transfer constants, k_{12} and k_{13}, are related to the bulk air-to-basement parameter by

$$k_{12} = \frac{k_1 \, Vb_2}{H_2 \, V_L} \tag{A-7}$$

and

$$k_{13} = \frac{k_1 \, Vb_2}{H_3 \, V_L} \tag{A-8}$$

where

V = the building volume (cm^3)

V_L = the basement volume (cm^3)

H_2, H_3 = the distribution coefficients for I_2 and HOI, respectively

B_2, B_3 = fractions of the liquid phase radioiodine that partition to I_2 and HOI.

Similar relationships are used to relate k_{22} and k_{23} to the bulk air-to-film transfer parameter, k_2.

The deposition rate constants can be written as products of appropriate deposition velocities and the surface-to-volume ratio.

$$\delta_i = \frac{V_{di} A}{V} \tag{A-9}$$

where V_{di} is the deposition velocity (cm/s) of species I, and A is the area (cm^2) of exposed surfaces.