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RADIONUCLIDE MASS BALANCE FOR THE TMI-2 ACCIDENT:  
DATA THROUGH 1979 AND PRELIMINARY ASSESSMENT OF UNCERTAINTIES

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## EXECUTIVE SUMMARY

A systematic data base of available information needed to calculate mass balances of key radionuclides arising from the Three-Mile Island Unit 2 (TMI-2) accident as a function of time has been assembled. The sample and analysis data represent the likely major sinks except for the solids remaining in the primary system. Surfaces in the primary system are represented only by preliminary data pertaining to cesium deposition on plenum surfaces. TMI-2 component description data are included for the reactor coolant, makeup and purification, and liquid waste systems and the reactor building. The chronology of liquid transfers through the end of 1979 is included, as well as references for all of the data and footnote information to explain special cases.

A mass transfer model has been developed that enables a user to set up flow models of variable complexity by the lumping of components (e.g., by treating the three reactor coolant bleed tanks as a component), to calculate the liquid contents of the components at a chosen mass balance time, to sum volume-concentration products, and to normalize the calculated inventories to the total TMI-2 inventories for each key isotope at the chosen mass balance time. This software also includes utility routines to facilitate regression and isotope ratio analyses.

The current mass balance status can be summarized as follows:

<u>Element</u>	<u>Inventory Fraction Accounted</u>
Tritium	0.53
Krypton	0.47
Xenon	0.35
Iodine	0.22
Cesium	0.51
Strontium	0.022
Uranium	$<10^{-4}$
Plutonium	$<10^{-5}$



The data for the noble gases are very uncertain due to the large uncertainty in the releases during the first several days.

These results indicate a distribution of key elements as follows:

Location	Inventory Fraction			
	H-3	I	Cs	Sr
Reactor coolant	0.02	0.01	0.07	0.01
Reactor building	0.47	0.18 <sup>a</sup>	0.41	0.01
Balance of plant	0.04	0.03	0.03	10 <sup>-3</sup>
Release to river	--	10 <sup>-6</sup>	10 <sup>-9</sup>	--
Release to atmosphere	--	10 <sup>-6</sup>	--	--
Not found	0.47	0.78	0.54	0.98

<sup>a</sup>Analysis of the results indicate that more than 18 percent may have gone to the reactor building and relatively less remained in solids and surfaces in the primary system.

It is concluded that tritium and cesium released into the reactor coolant traveled with the reactor coolant without losses to other phases during transit and storage. The data also suggest that tritium and cesium were not leached from primary solids and surfaces after the accident, although strontium has gradually leached from the primary solids and surfaces over a long period. Much of the iodine transferred to the reactor building sump/basement is suspected of having transferred to surfaces or solids from the sump/basement water and was therefore not found in basement water samples.

It is recommended that:

1. The current data base be maintained and extended as new data become available.

2. The data base and mass transfer methodology be applied to verify assumptions in the data base in respect to accident chronology.
3. The data base and mass transfer methodology be applied to verify or disprove behavior of key nuclides.
4. The calculation methodology should be developed to facilitate incorporation of the inventory in the primary system as new data become available.

## 1.0 INTRODUCTION

Section 1.1 presents a summary of results and recommendations. The background for the mass-balance studies is briefly summarized in Section 1.2; the objectives of the current work are presented in Section 1.3. The mass-balance data base including the FY 1983 additions to it are outlined in Section 1.4; the data base is presented in detail in Appendix C. The mass-transfer model to facilitate mass-balance determinations from the data is described briefly in Section 1.5 and presented in detail in Appendix B.

### 1.1 RESULTS AND RECOMMENDATIONS

The results of work performed to date can be put into four categories:

1. A mass balance based on data for essentially all significant "sinks" (i.e., locations of radioactivity in concentrated amount) outside the reactor coolant system solids and surfaces.
2. A presentation of the trends in the inventories in major sinks through 1979.
3. A preliminary assessment of the uncertainty in the preliminary mass balances.
4. A chronology of liquid transfers through 1979.

The mass balances generated from the current studies are shown in Table 1-1.

Table 1-1. Overall Mass Balances  
(as of December 31, 1979)

Element	Inventory Fraction Accounted
Tritium	0.53
Krypton (Kr-85)	0.47
Xenon (Xe-133)	0.35
Iodine (I-129)	0.22
Cesium (Cs-137)	0.51
Strontium (Avg. of Sr-80 & Sr-90)	0.02
Uranium (U-238)	$<10^{-4}$
Plutonium Pu-239)	$<10^{-5}$

The total inventories calculated from EPICOR II and the submerged demineralizer system (SDS) influent batch concentrations and volumes agreed well with those calculated from corresponding system component liquid volumes and concentrations. Inventories of major sinks were calculated as a function of time; the time behavior of these calculated inventories shows wide variations in some cases where the inventories are small. The time behavior of the reactor coolant inventory (expressed as a fraction of the total core inventory, which includes the time dependence due to decay) consists of a reasonably narrow range of variations from a smooth function for tritium, strontium, cesium, and iodine; these data suggest that the tritium, iodine, and cesium inventories decrease approximately as expected as a consequence of dilution due to leakage of reactor coolant and makeup with clean water. Strontium inventories in the reactor coolant increase rapidly over the first few days and more slowly over the time period to the end of 1979, indicating leaching from solids or surfaces in the primary systems. A more detailed analysis is needed to correct the observed

strontium buildup for dilution due to leakage and makeup and hence to estimate the rate of leaching.

A large fraction of the tritium, cesium, iodine, and strontium that has been accounted for was found in three major liquid sinks: (1) the reactor building basement water, (2) the reactor coolant, and (3) the reactor-coolant bleed tanks. The uncertainties in the mass balances are discussed in detail in Chapter 3.

Some general observations on the data follow.

- o The mass balance accounts for only about half of the total noble gases, but this account is very uncertain because of the uncertainty in the noble gas release during the first month.
- o Tritium and cesium (Cs-137) are about half accounted for in the quantified sinks. These two isotopes are found in the major sinks in about the same proportions to each other. The time behavior of tritium and cesium concentrations in the reactor coolant is approximately as expected from effects of the dilution by makeup water.
- o About one quarter of the iodine is accounted for in identified sinks. Iodine inventory fraction in the reactor building sump is (relative to cesium and tritium) much smaller in the reactor coolant.

The time behavior of iodine concentration in the reactor coolant is approximately as expected from dilution due to makeup water.

- o About 2 percent of the strontium has been found in identified sinks (outside the core surfaces and solids). The concentration of strontium in the reactor coolant continually increased over the period to September 1981

when SDS processing began, although the rate of increase gradually declined with time.

Suggested future work based on the data base and model includes the following:

- o Analyze the tritium, cesium, and iodine concentrations in the reactor coolant as a function of time using the chronology of makeup and leakage flows to determine if the concentration versus time behaviors are well explained by dilution or if other processes are indicated.
- o Analyze the strontium concentration in the reactor coolant as a function of time using the makeup/leakage chronology in order to determine the rate of strontium leaching.
- o Re-estimate the amount of iodine transported to the reactor building basement using the reactor coolant inventory data and the liquid transfer chronology. The form of the leaching/precipitation terms would have to be postulated; the decay and leakage terms are known.

## 1.2 BACKGROUND

Following the accident at TMI-2 in March 1979, GEND, the combined organization of GPU, the Electric Power Research Institute (EPRI), the U.S. Nuclear Regulatory Commission (NRC), and the U.S. Department of Energy (DOE) stated its intention to support an effort to determine as accurately as possible the mass balances of significantly toxic species released or relocated as a result of the accident and recovery and cleanup procedures.<sup>(1)</sup>

During FY 1982, a computerized data base was established that permits the calculation of mass balances for key radionuclides at any time of interest.<sup>(2)</sup> The data base includes:

1. Calculated total inventories of key radionuclides in the whole TMI-2 system as a function of time
2. System component volumes and chronology of liquid transfers
3. Sampling and radiochemical analysis data
4. References that support each data record
5. Footnote information to document special cases

The data entered during FY 1982 included data for 11 isotopes (i.e., Kr-85, Xe-133, I-129, I-131, Cs-134, Cs-137, Sr-89, Sr-90, U-238, Pu-239, and H-3). The sample analysis included all data available through April 30, 1979, plus the data readily available. The system descriptions and chronology were based on a simplified model of the reactor coolant system (RCS), makeup and purification (MUP) system, and liquid waste (WDL) system. The chronology of liquid transfers was represented for the period through April 30, 1979.

Also during FY 1982, a modular computerized mass-transfer calculational scheme to use the data base and to calculate mass balances as a function of time was outlined and partially developed.

### 1.3 OBJECTIVES

The objectives of the FY 1983 work have been to enlarge the data base, improve the preliminary mass balance, and assess the uncertainty in the mass balance. The objectives in terms of enlargement of the data base included the following:

1. Chronology extension for liquid and gas transfers for all of 1979 for the RCS, MUP, WDL, chemical addition, reactor

building spray, nuclear services river water, and reactor purge and recirculation systems.

2. Component volume data to support the chronology extension, including piping, makeup and purification, system components, and EPICOR II.
3. Analyses of radiochemical samples of interest for components of systems itemized under item 1.
4. Analyses of available EPICOR II, and SDS influent samples.
5. Analyses of other samples of interest (e.g., solids and reactor building and reactor plenum surface samples).

The improvements in the preliminary mass balance consisted of the following:

1. The inclusion of the estimated inventories in as many sinks as possible
2. The estimation of inventories in major sinks as a function of time
3. The inclusion of inventories by means of mass transfer calculations for sinks not sampled

The assessment of the uncertainty in the mass balances was expected to:

1. Focus on the validity of the estimated inventories in the major sinks, since the uncertainty in the inventories in other sinks do not significantly affect the overall uncertainty in the mass balance.



2. Consider errors due to the following:

- a. Sampling (i.e., the uncertainty with which a sample represents the volume sampled)
- b. Analysis (i.e., the uncertainty in radiochemical determination of nuclide concentration)
- c. Volume (i.e., the uncertainty in component capacities and contained liquid volumes)
- d. Total inventories (i.e., the uncertainty in the calculated total inventories of the nuclides in the whole TMI-2 system).

1.4 DATA BASE

The data base is presented in Appendix C along with a description of the format of the tables and definitions of symbols. The data base has been expanded in FY 1983 to approximately double its size in FY 1982 as shown on Table 1-2.

The isotope table now includes Te-129m, Ru-106, and Ce-144. The first two of these nuclides was added because they are important in terms of potential health impacts from a reactor accident. Ce-144 was added as an indication of isotopes of high-melting compounds and because several analyses include values of this isotope.

Sample data include liquid and gas samples taken through 1979 from the RCS, WDL, reactor building atmosphere, and station vent. They include other and/or later samples from the reactor building sump/basement water; EPICOR and SDS batch influents; charcoal removed from the heating, ventilation, and air conditioning (HVAC) system; reactor building venting; reactor building surfaces (samples from the 305-foot and above elevations; the hydrogen recombiner spool piece and gamma spectrophotometric viewing of the

Table 1-2. Enhancement of the Data Base in FY 1983

Data Base Table	Number of Records	
	In FY 1982	In FY 1983
Isotope	11	14
Sample - liquid, gases, and solids	653	917
Sample - surfaces	0	89
Analysis	1973	3259
System	219	326
Chronology	331	1269
Event	293	995
Note	67	158
References	429	670
Totals	3976	7697

347-foot operating deck); reactor vessel plenum surfaces (which show cesium deposition); MUP system filters and one demineralizer, sludge from reactor-coolant bleed tanks; and effluent to the Susquehanna River. A new sample table was developed for surface area data to allow identification of the location of reactor building surface samples in terms of cylindrical coordinates.

The systems table was enhanced to include piping volumes within the MUP and WDL systems, volumes and areas of components in the MUP system, EPICOR II batch volumes, reactor building surface areas, surface areas of the plenum region of the reactor vessel, and reactor coolant pressure-temperature data. These data (except reactor building surface areas) were developed from primary sources such as drawings and operator logs.

The chronology table now presents a record through 1979. This represents a major step forward in the quantification of the sequence of events at TMI-2 since prior chronology studies provided a detailed record only through the end of April 1979. The development

is based on primary sources including GPU shift foreman and rad-waste engineer logs and information from EG&G-TIO staff. The chronology development used volume inflow and outflow balancing for the reactor coolant system, the makeup tank, and the auxiliary building sump.

The flows into the reactor coolant system were seal injection and makeup; the out-flows were seal return, letdown, and leakage. The flows into the makeup tank included batch inputs from the reactor coolant bleed tanks, the boric acid mix tank, and demineralized water storage tank as well as the seal return and letdown flows from the reactor coolant system. The flows out of the makeup tank were the seal injection and makeup to the reactor coolant and leakage to the auxiliary building sump. The auxiliary building sump received water due to the leakage from the makeup tank plus other miscellaneous leakage; outflows were batch volumes to the auxiliary building sump tank and miscellaneous waste storage tank.

Interconsistency of this chronology is demonstrated by reasonable liquid volume versus time behavior for the key volumes, although there are some minor anomalies (indicated by periods during which the calculated liquid volumes exceed the tank capacities). The chronology table also includes records of transfers to EPICOR II and the tank farm.

### 1.5 MASS TRANSFER

A mathematical model was developed that facilitates the calculation of a mass balance using accounting arithmetic. The liquid and/or gaseous volumes at a specified time are multiplied by concentration (either measured or calculated from known concentrations of source components and mass transfer chronology) and normalized to the LOR2 total TMI-2 inventories to obtain inventory fraction for isotopes in key components. Summing over all components provides a mass balance for the specified time. The model also facilitates regres-

sion analyses of any chosen set of data and the calculation of isotopic fractions.

The model consists of five modules that are executed sequentially. The Introductory Text Module introduces the user to the system and allows the user to design a flowpath model; a set of state-specific model components are identified. The user may lump components; for example, RCS may include the liquid volume throughout the reactor coolant system components; the three reactor coolant bleed tanks may be considered one lumped component, etc.

The Problem Initialization Module assists the user in initializing the problem by linking the system table of the data base to the established flowpath model. For each model component, the module identifies and sums initial (at start of accident) liquid volumes and design capacities for all of the model components. It provides a list of model (lumped) components, total model component capacities and areas, and the model component liquid volumes at the time of initiation of the accident.

The Volume Calculation Module supports subsequent modules of the system by linking the chronology table of the data base to the system file of model components and identifying mass transfers from the start of the accident to the mass-balance time. The module prompts for one user-specified input parameter during execution of this module, the mass-balance time. This module serves two specific functions in the determination of radionuclide mass balances: (1) it determines the liquid volumes and the net increase or decrease for each of the model components at the mass-balance time; (2) to support cases in which model component in-flow occurred near the mass-balance time and concentrations are known at times other than the mass-balance time, the code identifies for each model component all additions and withdrawals, as well as their associated flow rates and the other components involved in the transfer.

## 2.0 CALCULATION OF MASS BALANCE

The mass-balance calculation outlined in this chapter is based on data found in the Three Mile Island Unit 2 (TMI-2) radionuclide mass-balance data base. The mass balance is the accounting of the radionuclides at any time following the accident on March 28, 1979. Component inventory fractions were calculated by computing the component's activity and normalizing the result to the predicted core inventory, provided by the B&W LOR2 code:<sup>(3)</sup>

$IF_{i,j,k}(t)$  = component inventory fraction of component  $i$ , state  $j$ , and isotope  $k$  at time  $t$

$$IF_{i,j,k}(t) = V_{i,j}(t) \cdot C_{i,j,k}(t) / A_k(t)$$

where

$V_{i,j}(t)$  = volume-state  $j$ , in component  $i$ , at time  $t$

$C_{i,j,k}(t)$  = concentration-state  $j$ , in component  $i$ , isotope  $k$ , at time  $t$

$A_k(t)$  = predicted total inventory of isotope  $k$  at time  $t$

The mass balance was accomplished by summing the inventory fractions for each isotope over all components and mass states.

$TIF_k(t)$  = total inventory fraction of isotope  $K$  at time  $t$

$$TIF_k(t) = \sum_{i=1}^I \sum_{j=1}^J IF_{i,j,k}(t)$$

where  $I$  and  $J$  are total number of components and mass states, respectively, that are considered.

The mass balance presented in this section made use of the calculational tools discussed in Chapter 3.

## 2.1 IDENTIFICATION OF MAJOR AND MINOR SINKS

Following the accident at TMI-2, many plant components contained radionuclide inventories. Using the large amount of radiochemical data in the TMI-2 mass-balance data base, components were sorted into major sinks, containing significant inventory fractions, and minor sinks, which separately are insignificant in terms of mass balance.

The minor sinks have been defined as components that retained inventory fractions less than 0.01 before cleanup. These minor sinks are identified in Table 2-1. Inventory fractions are shown for each component where appropriate data were available. The inventory fractions in all the minor sinks are on the order of 1 percent for cesium, tritium, and iodine, with strontium and xenon considerably lower.

The inventory fractions shown on Table 2-1 have been calculated from data in the TMI-2 mass-balance data base. The neutralizer tanks, miscellaneous waste holdup tank, contaminated drain tanks, and the auxiliary building sump liquid inventory fractions' ranges span the values calculated from the data base samples. The liquid volumes are assumed to be at capacity where flow chronology was unknown. The makeup tank and makeup and purification system (MUP) piping inventory fractions are based on representative reactor coolant system (RCS) liquid samples; the liquid waste system (WDL) piping inventory fractions are based on representative reactor coolant bleed tank liquid samples. The auxiliary and fuel handling building charcoal gas, solids, and surfaces have been documented. (4, 5, 6).

Figure 2-1 provides an approximate distribution of material as found at TMI-2 on December 31, 1979, according to data in the TMI-2

Table 2-1  
Inventory Fractions in Minor Sinks

State	Components	Notes	Inventory Fraction						
			H-3	Sr-89	Sr-90	I-131	Xe-133	Cs-134	Cs-137
Liquid	Makeup Tank	1,2	0.0010-0.0080	0.0002-0.0006	0.0003-0.0006	0.0016-0.0056		0.0016-0.0090	0.0015-0.0073
	Neutralizer Tanks	3				2 E-10-0.0002	7 E-7-0.0001	2 E-7-0.0002	4 E-8-0.0002
	Misc. Waste Holdup Tank	3	0.0002-0.0009			0.0005-0.0032		0.0002-0.0045	0.0002-0.0035
	Contaminated Drain Tanks	1,3	5 E-8-8 E-7			3 E-11-2 E-5	7 E-10-5 E-9	7 E-10-1 E-5	6 E-12-2 E-6
	Aux. Bldg. Sump	1,3		8 E-6-7 E-5	8 E-6-5 E-5	0.0002-0.0004	5 E-5-0.0002	0.0003-0.0005	0.0003-0.0005
	Aux. Bldg. Sump Tank	1,4		4 E-6-3 E-5	4 E-6-2 E-5	9 E-5-0.0002	2 E-5-0.0001	0.0001-0.0002	0.0001-0.0002
	MUP Piping	1,2	0.0001-0.0009	2 E-5-8 E-5	3 E-5-8 E-5	0.0002-0.0008		1 E-5-0.0012	1 E-5-0.0010
	WDL Piping	1,5	4 E-6-4 E-5		7 E-8-9 E-7	1 E-7-3 E-5		1 E-7-4 E-5	8 E-6-3 E-5
	All Liquids		0.0013-0.0098	0.0002-0.0008	0.0003-0.0008	0.0026-0.0008	7 E-5-0.0004	0.0022-0.0157	0.0021-0.0127
	Gas	Aux. Bldg. & FH. Bldg. Charcoal	6				2 E-7-3 E-7		
RCS		3				2 E-8-3 E-7			
All Gases						2 E-7-3 E-7			
Solid	MUP Filters	7						6 E-6-0.0040	5 E-6-0.0030
	All Solids							6 E-6-0.0040	5 E-6-0.0030
Surface	Reactor Building	8		4 E-6-7 E-6	4 E-6-7 E-6			1 E-5-0.0003	7 E-6-0.0002
	Minor Sink Inventory Fraction		0.0013-0.0098	0.0002-0.0008	0.0003-0.0008	0.0026-0.0105	0.0001-0.0004	0.0022-0.0200	0.0021-0.0159

1. Component is assumed to be filled to capacity in lieu of other data from the TMI-2 mass-balance data base.
2. Based on representative reactor coolant system samples in the TMI-2 mass-balance data base.
3. Based on component's available samples in the TMI-2 mass-balance data base.
4. Based on representative auxiliary building sump samples in the TMI-2 mass-balance data base.
5. Based on representative reactor coolant bleed tank samples in the TMI-2 mass-balance data base.
6. Based on estimated particulate and gaseous iodine in the filters/absorbers analyzed<sup>(4)</sup>.
7. Based on Oct 29, 1982, Sept 29, 1981, Jan 6, 1982, MU-F-5B sample analyses<sup>(5)</sup>.
8. See reference 6.

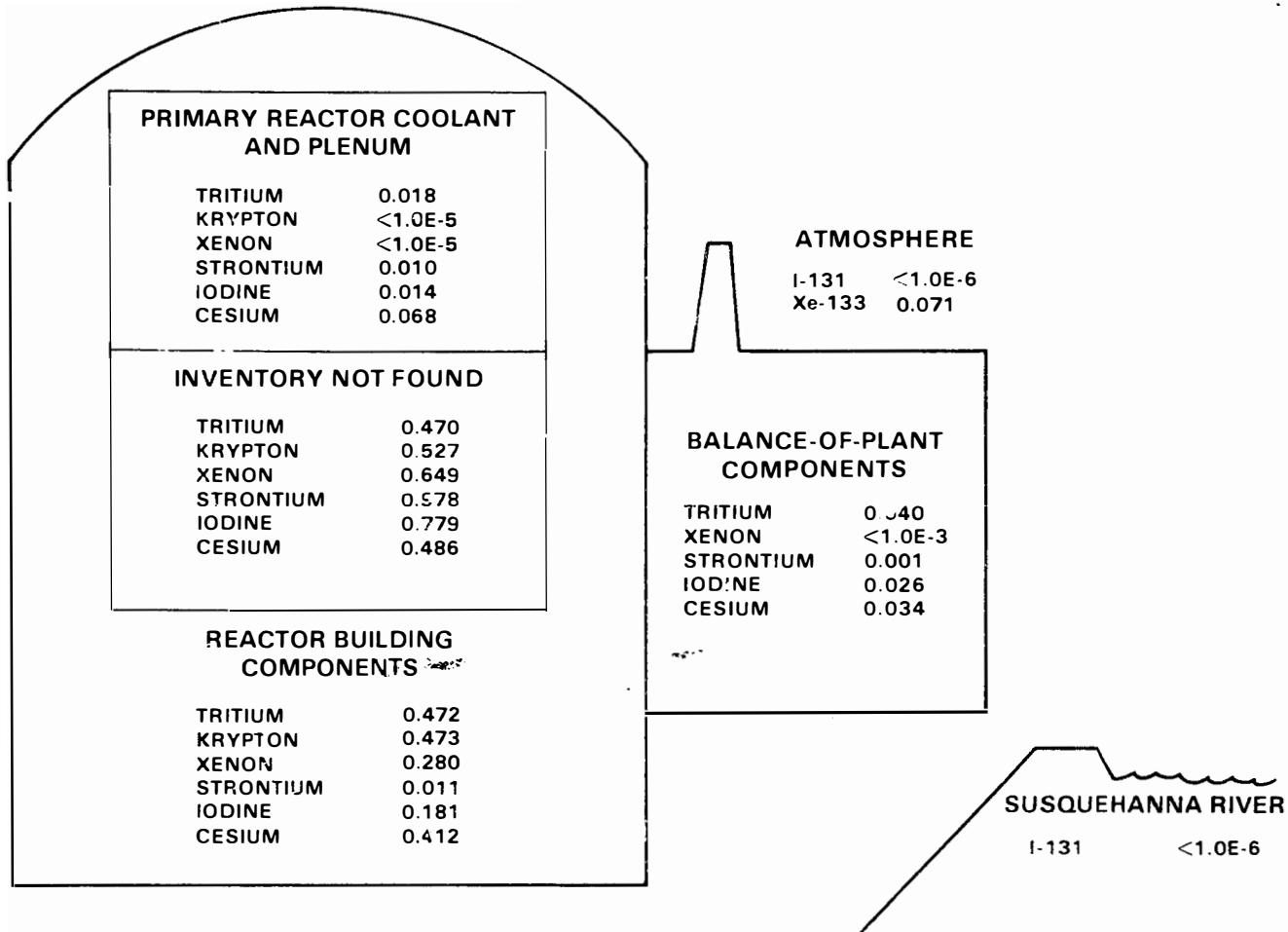


FIGURE 2-1  
 DISTRIBUTION OF KEY RADIONUCLIDES RESULTING FROM A  
 PRELIMINARY MASS BALANCE ON  
 DECEMBER 31, 1979  
 AT 2400 HOURS



mass-balance data base. The calculation of these inventory fractions is presented in the following sections.

From a survey of data from the TMI-2 mass-balance data base, the major radionuclide sinks have been defined as plant components that retained inventory fractions of 0.01 or greater before cleanup. The major sinks presently identified include the following:

1. Reactor building basement (and sump)
2. Reactor coolant system
3. Reactor coolant drain tank
4. Reactor coolant bleed tanks (A, B, C)
5. Reactor building air
6. Atmosphere
7. Reactor vessel plenum and internals surface
8. Makeup and purification demineralizers (A and B)

Sections 2.2, 2.3, and 2.4 present the mass-balance discussion for the identified major sinks in the gaseous, liquid, and surface/solid phases, respectively. Inventory fractions based on data from the TMI-2 mass balance data base are presented for each of the major sinks. Also, any apparent trends are identified and briefly discussed. The results of the mass balance are presented in Section 2.5, along with a discussion of inventory fractions yet to be found.

## 2.2 GASEOUS INVENTORIES AND RELEASES

During the accident at TMI-2, radioactive fission gases, including krypton, iodine, and xenon isotopes were, released from the damaged core to the reactor coolant. These gases traveled throughout the plant via various pathways, evolving from liquids and venting through various systems. The identified gas flowpaths are shown in Figure 2-2. Such evolved gases are normally vented to the waste-gas decay tanks for storage and subsequent release. However, gaseous inventories in the waste-gas decay tanks were vented into the containment on April 6, 1979. Direct release to the station vent occurred through relief valves. A major release pathway came via leaks in the waste-gas compressors into the heating, ventilation and air conditioning (HVAC) system. The reactor containment building air had been sequestered since building isolation at 7:56 a.m. on March 28, 1979. Mass transfer between surfaces and liquids within the reactor building is likely to have occurred, but these processes were not within the scope of this phase of the study. The reactor containment building remained isolated until purging began on June 28, 1980.

### 2.2.1 Gaseous Inventories

Isotopic inventory fractions were calculated for postaccident gases in the reactor building from samples taken during 1979. Inherent in the calculation of these inventory fractions is the assumption of a constant containment air volume of  $2.12 \times 10^6$  cubic feet. Figures 2-3 to 2-7 present the inventory fractions calculated for Kr-85, Xe-133, I-131, Cs-134, and Cs-137, respectively. Standard temperature and pressure were assumed.

Krypton-85 was sampled in the containment building air starting about 7 weeks following the accident. Because of a relatively long half-life and beta decay, this isotope was measured more easily after short-lived isotopes decayed away. Krypton-85 is an indicator of noble gas inventory evolved from the RCS during the

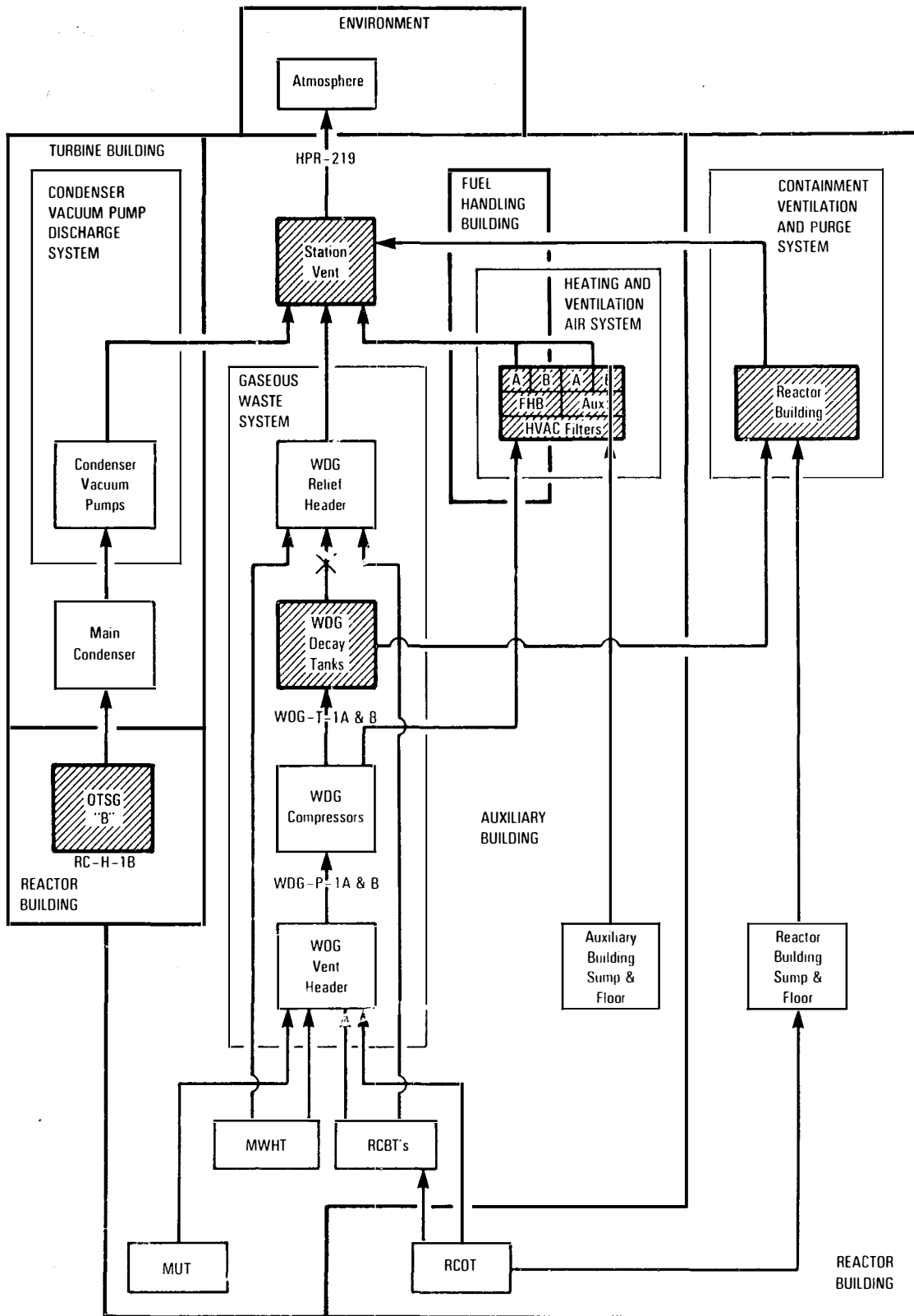


Figure 2-2. MASS TRANSFER FLOW PATHS — GASES

FIGURE 2-3

TMI-2 CONTAINMENT AIR KR-85 INVENTORY

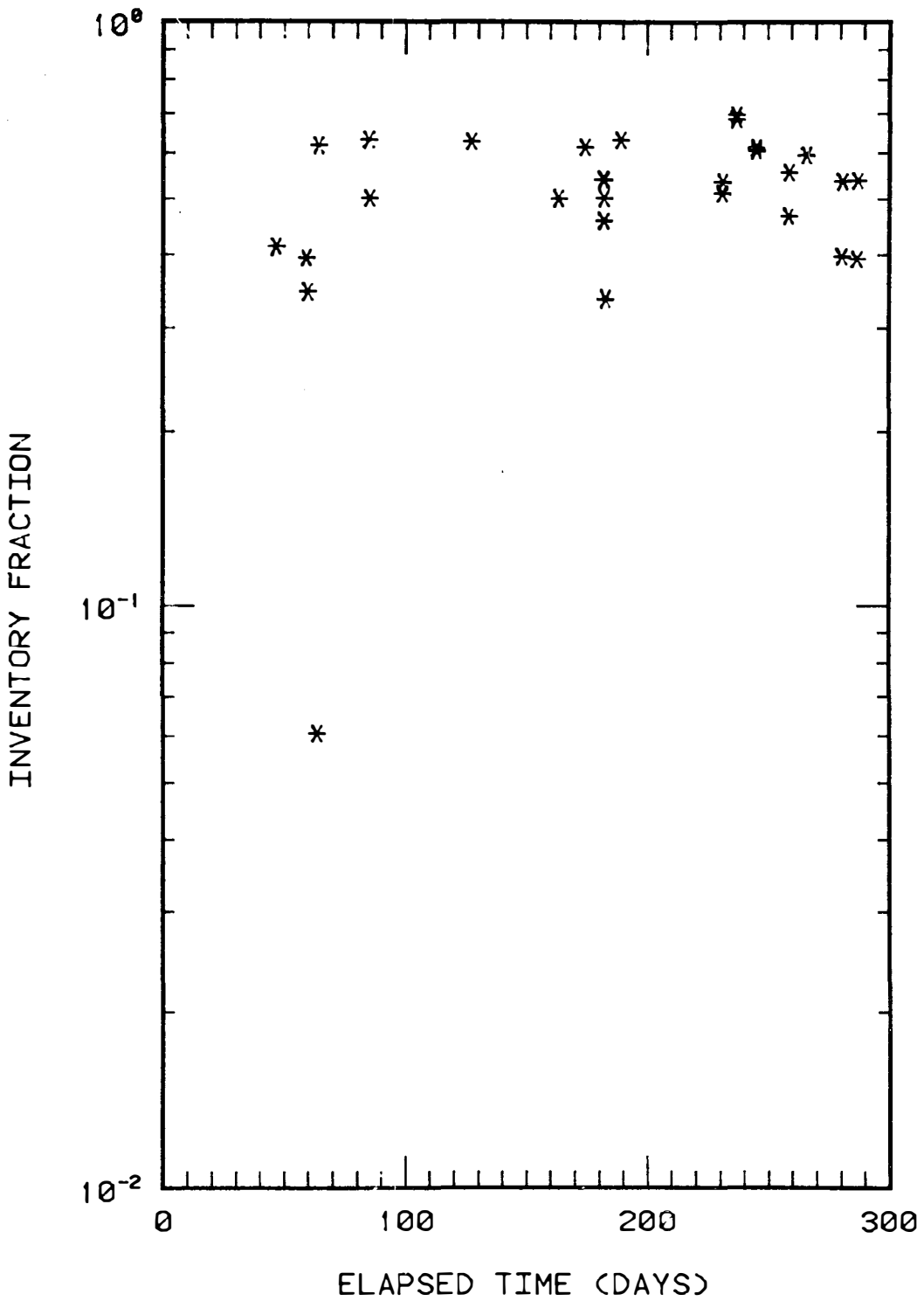


FIGURE 2-4

TMI-2 CONTAINMENT AIR XE-133 INVENTORY

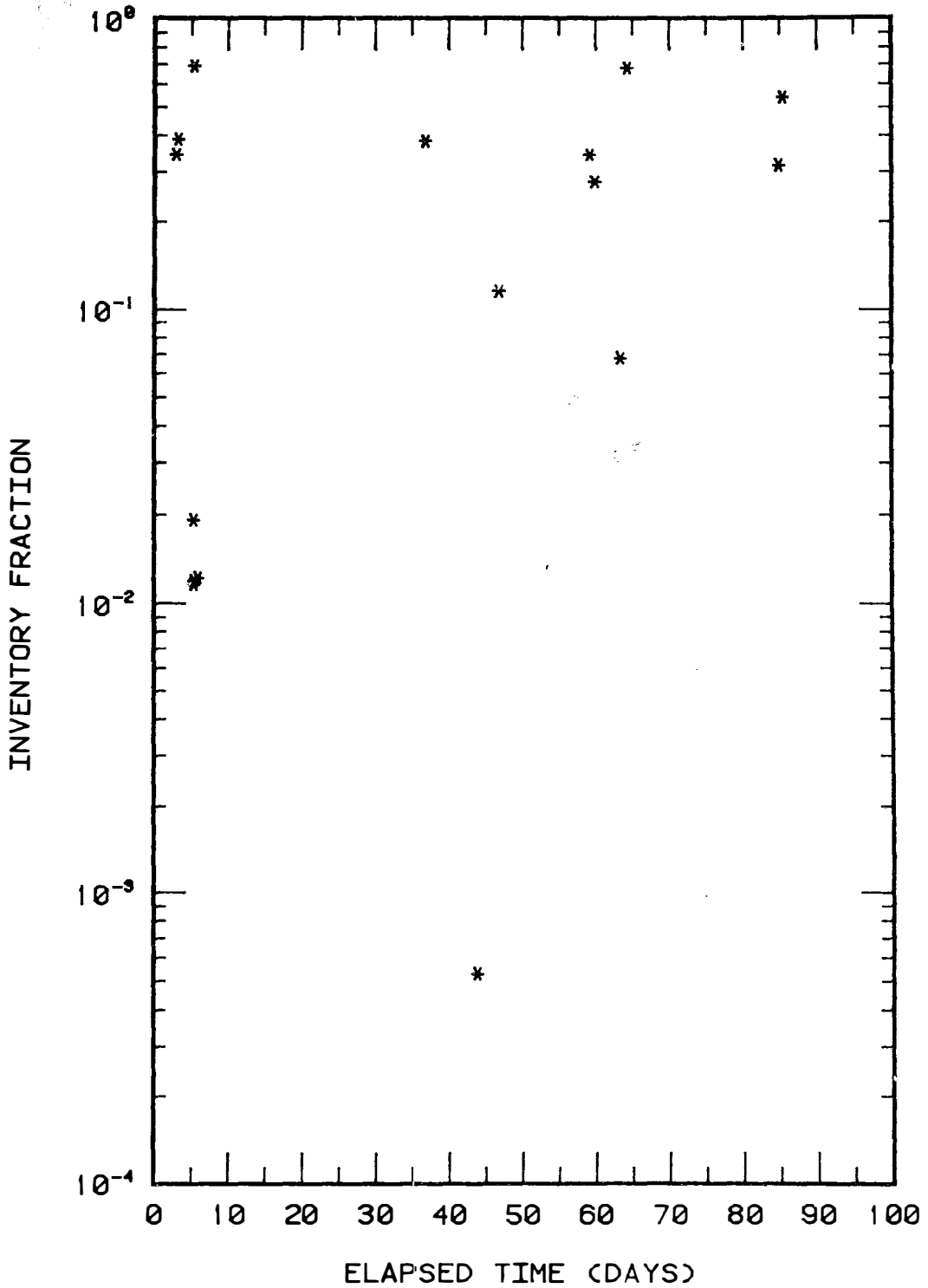


FIGURE 2-5

TMI-2 CONTAINMENT AIR I-131 INVENTORY

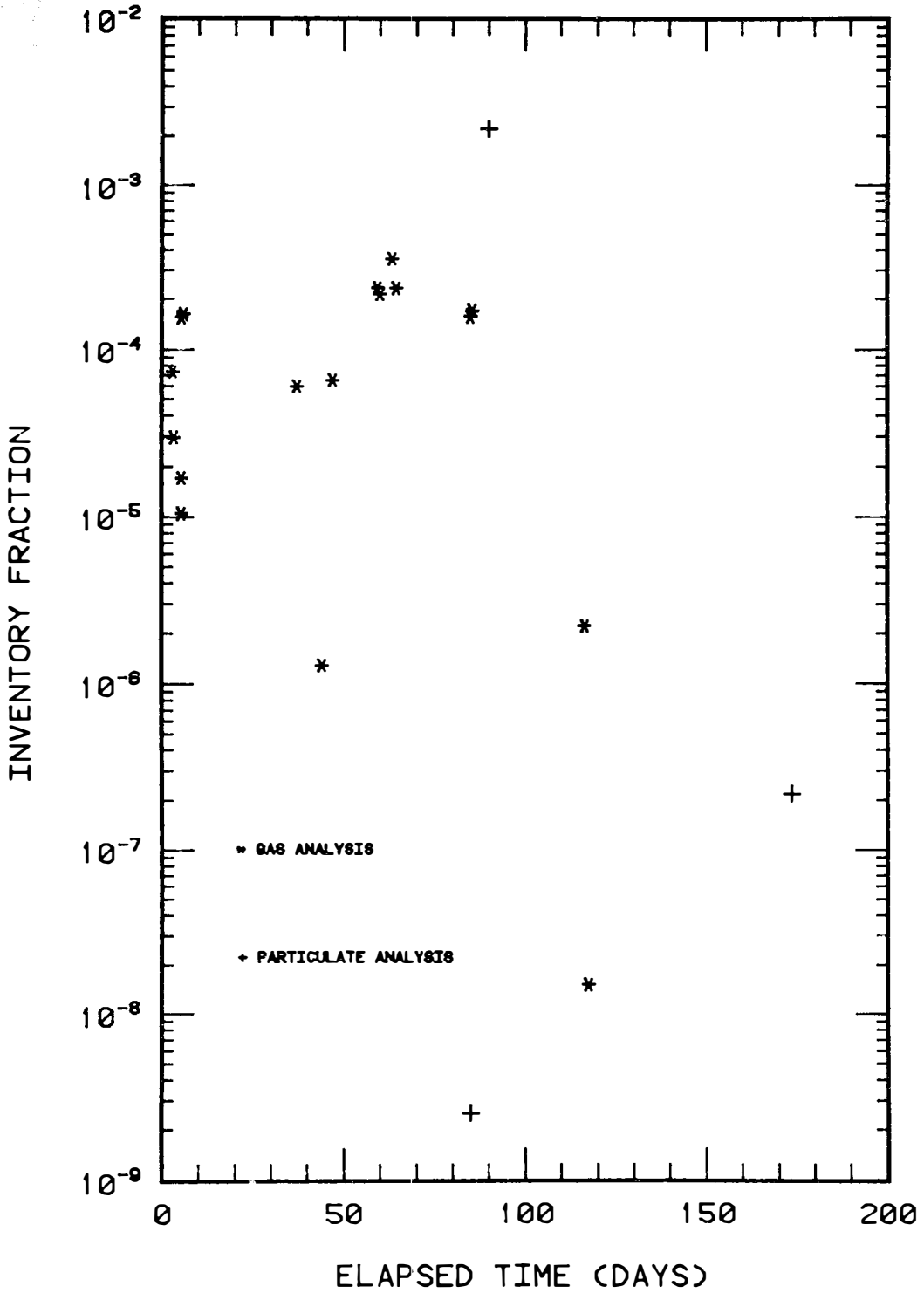


FIGURE 2-6

TMI-2 CONTAINMENT AIR CS-134 INVENTORY

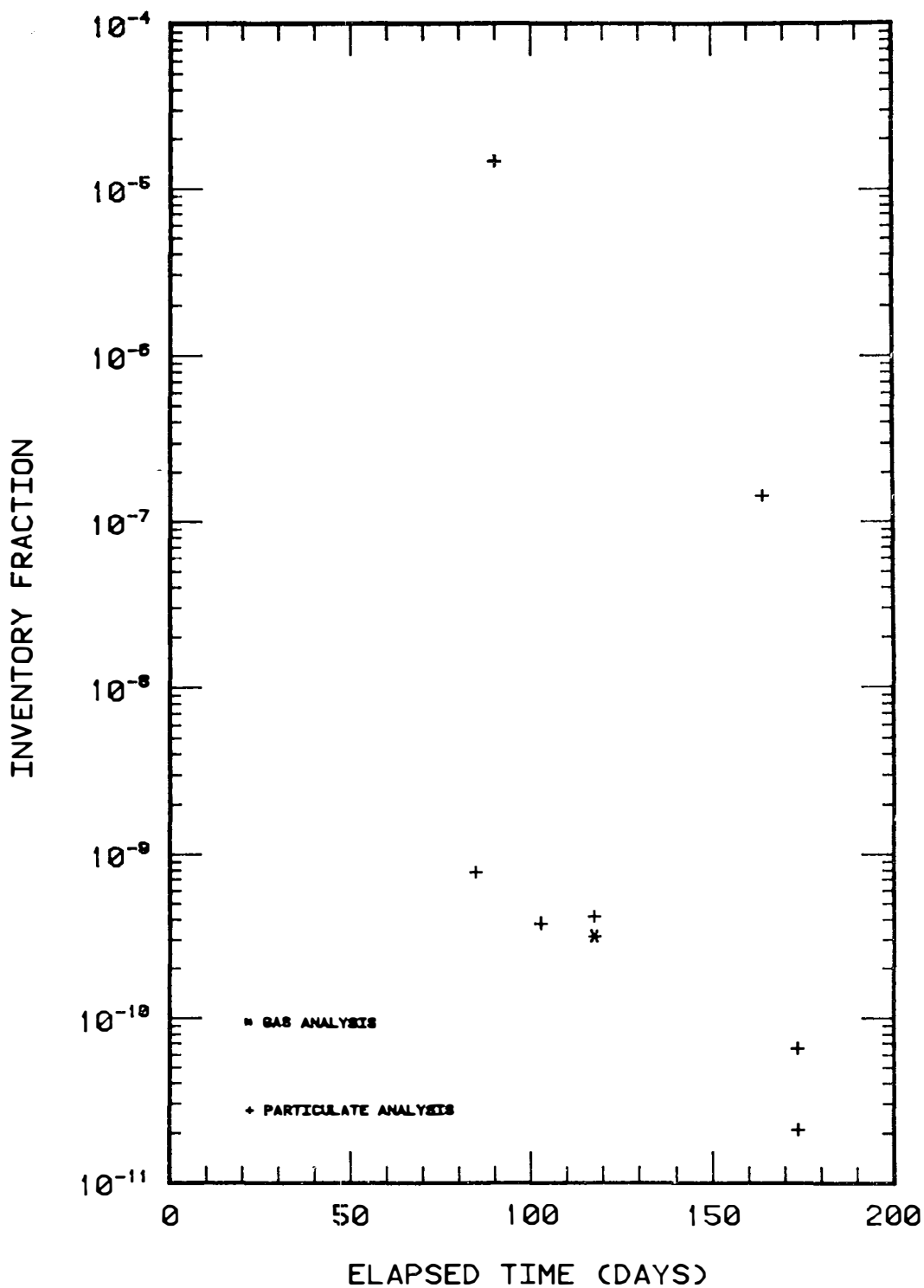
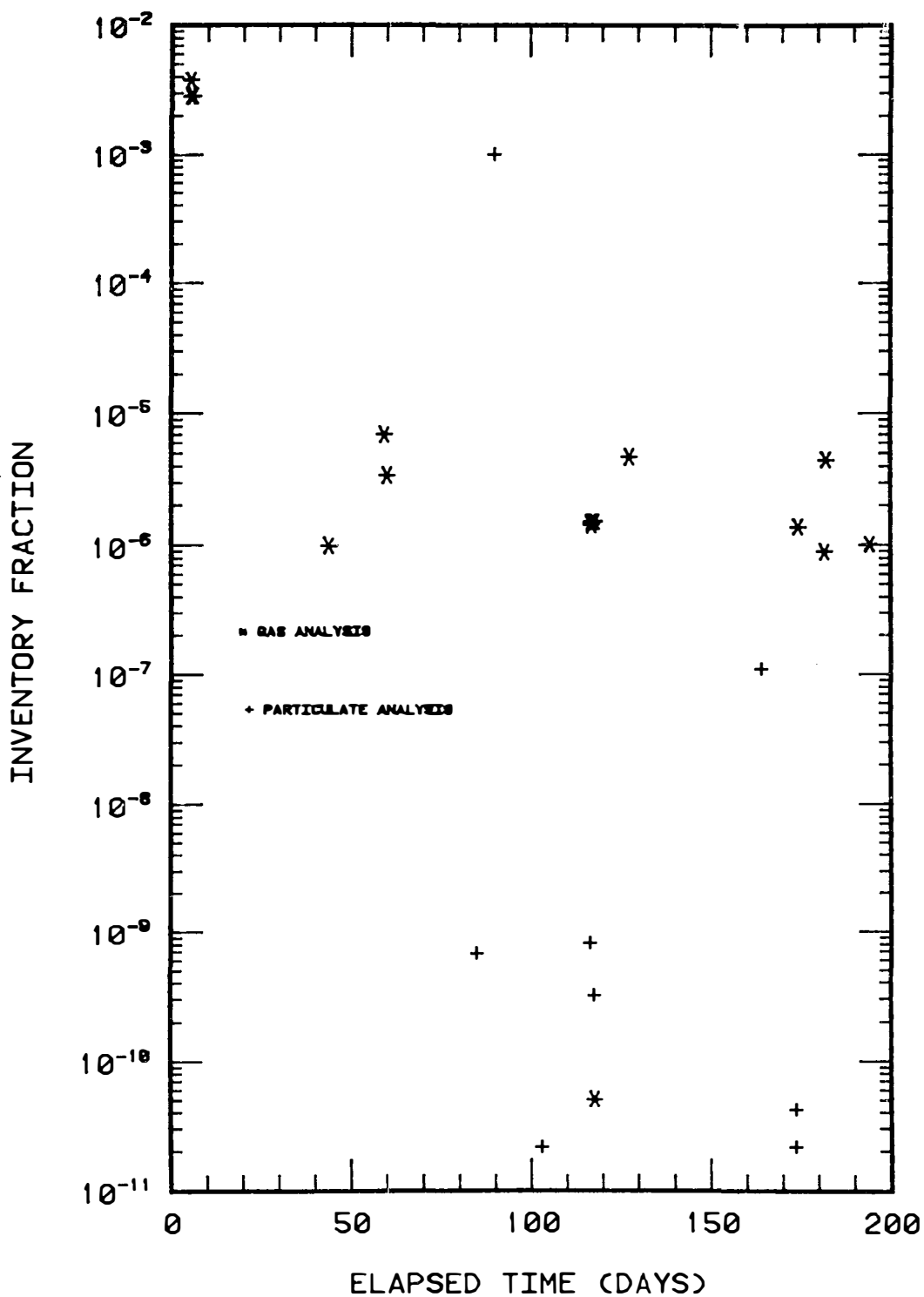


FIGURE 2-7

TMI-2 CONTAINMENT AIR CS-137 INVENTORY





accident and remaining in the reactor containment until venting. Figure 2-3 shows the calculated Kr-85 inventory fractions. Ignoring the one sample taken about 45 days after the accident, which appears to be low by an order of magnitude, the samples taken indicate that the Kr-85 inventory fraction in the reactor building air was constant and within 0.33 to 0.70.

Another indication of noble gas inventories in the reactor containment building is found in Figure 2-4. The Xe-133 samples provide data to approximately 3 months after the accident, due to the short, 5-day half-life. Again ignoring the sample taken about 45 days after the accident and several of the early samples which appear to be low by an order of magnitude, the samples indicate that the Xe-133 inventory fraction in the reactor building is constant and between 0.07 and 0.70.

In addition to the sampling of nobles gases in the reactor building air, numerous I-131 samples were taken prior to the rapid decay of this isotope.

The radioiodine inventory in the TMI-2 containment building air has been determined based on available I-131 samples. The results appear in Figure 2-5. There is a considerable spread in the values shown, probably attributable to the use of different air-sampling techniques. The gas samples, ignoring data taken during the first week and after 90 days, indicate that the inventory fraction of I-131 in the reactor containment building air was between 0.00005 and 0.0004.

It can be concluded that the I-131 inventory fraction in the reactor containment building air was on the order of  $10^{-4}$ , and thus is insignificant from a mass-balance viewpoint. This inventory fraction is lower than those of the noble gases by a factor of several thousand. This observation is also supported by plant radiogas-release estimates discussed later in this chapter.

Inventory fraction estimates of particulate cesium in the TMI-2 containment building are presented in Figures 2-6 and 2-7, for Cs-134 and Cs-137, respectively. The data confirm that cesium inventory fractions in the reactor containment building air are insignificant, from a mass balance point of view, below  $10^{-5}$ .

### 2.2.2 Gaseous Releases

Gaseous releases to the atmosphere followed several flowpaths to the station vent as indicated in Figure 2-2. Uncontrolled releases took place through the waste gas disposal systems (WDG) relief valve vent header, and the HVAC exhaust. Controlled releases were made by purge of the containment building prior to entry in June 1980. The suspected primary-to-secondary coolant leak may have caused a small release of fission gas via the condenser vacuum pumps. Due to the absence of adequate supporting chronology data, the gaseous releases to the environment, for some 34 days following the accident, are adopted as published.

An estimate<sup>(8)</sup> of noble gases released offsite for the first 34 days following the accident has been made. Table 2-2 provides a summary of the estimated releases of Xe-133. The results indicate that 7.1 percent of the noble gases were released to the atmosphere by uncontrolled release. A similar accounting of Kr-85 venting<sup>(7)</sup> from the containment building during purging found 44,000 curies or 49.3 percent of this noble gas to be released. This value agrees well with the containment building inventory of Kr-85 reported in Section 2.2.1.

The release of gaseous radioiodine to the environment has been estimated to have been 14 curies<sup>(8)</sup> based on station vent measurements and 26.8 curies<sup>(4)</sup> based on HVAC filter/adsorber examination. An estimated 191 curies were transferred to the filters/adsorbers. This value produces an inventory fraction released to atmosphere on the order of  $10^{-5}$ . Again, this inventory fraction is lower than the noble gas inventory fraction by a factor of many thousands,

Table 2-2

TMI-2 Xenon-133 Release Estimate<sup>8</sup>

Time Period - 1979	Estimated Release (Ci)	Estimated Inventory Fraction
March 28 - 0700 to March 29 - 1600	$4.9 \times 10^6$	0.035
March 29 - 1600 to March 31 - 1600	$2.1 \times 10^6$	0.017
March 31 - 1600 to April 3 - 1500	$1.1 \times 10^6$	0.012
April 3 - 1500 to April 6 - 1400	$2.7 \times 10^5$	0.004
April 6 - 1400 to April 30 - 2400	<u><math>1.5 \times 10^4</math></u>	<u>0.003</u>
Total	$8.4 \times 10^6$	0.071

which is consistent with the differences in containment gaseous inventory estimates.

During the controlled containment building purge, 1.3 curies of tritium, 5.5 micro curies of Cs-137, and  $5.72 \times 10^{-3}$  micro curies of Sr-90 were estimated to be released.<sup>(7)</sup> These releases account for inventory fractions  $4 \times 10^{-4}$ ,  $7 \times 10^{-12}$ , and  $8 \times 10^{-10}$ , respectively.

## 2.3 LIQUID INVENTORY AND RELEASES

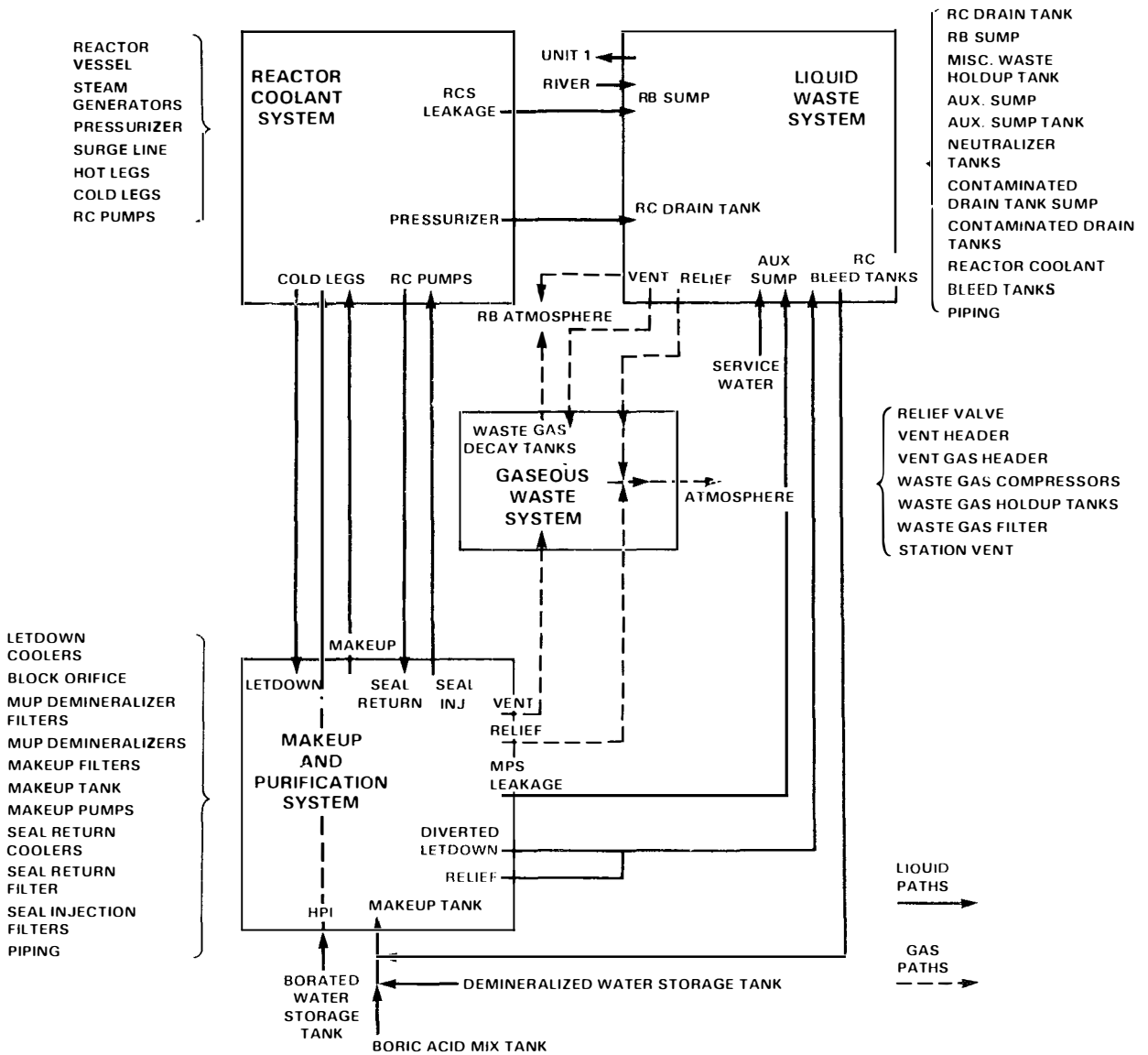
From the mass-balance data<sup>(2)</sup> it is clear that most of the radionuclide inventories, except for noble gases, have been found to date in liquids originating in the RCS. No release of such liquid to the environment occurred at TMI-2. Figure 2-8 provides an illustration of the mass flowpaths considered in the scope of this study. A more detailed description of component liquid chronology follows in the next section for components of interest.

### 2.3.1 Liquid Inventories

The transfer of radionuclides in reactor coolant to various components in TMI-2 has been previously discussed.<sup>(2, 7)</sup> The mass-balance results indicate to date that most of the radionuclide inventories (outside the core and core debris) reside in the major sinks identified in Section 2-1. In this section the inventories associated with the reactor building basement, reactor coolant drain tank, the reactor coolant system, and the reactor coolant bleed tanks are presented. The inventories were calculated based on the liquid mass transfer chronology and the sample analyses in the TMI-2 mass-balance data base.

#### 2.3.1.1 Reactor Building Basement

The largest volume of reactor coolant went to the reactor building basement. This water was approximately 8-1/2 feet deep in the



**FIGURE 2-8  
PRELIMINARY MASS BALANCE  
FLOW PATHS**



basement in September 1981, before the start of processing by the submerged demineralizer system (SDS). Figure 2-9 provides the liquid volume history for the reactor building basement as modeled by the chronology data found the TMI-2 mass-balance data base. A total of 529,000 gallons was added to the basement in 1979. Table 2-3 provides a breakdown of the total volume. This table also indicates that 63 percent of the water was once reactor coolant.<sup>(17)</sup> If the RCS leakage is extrapolated to September 1, 1981, the basement volume would be 643,000 gallons, which is comparable to the 641,000 gallons reported by others.<sup>(15)</sup>

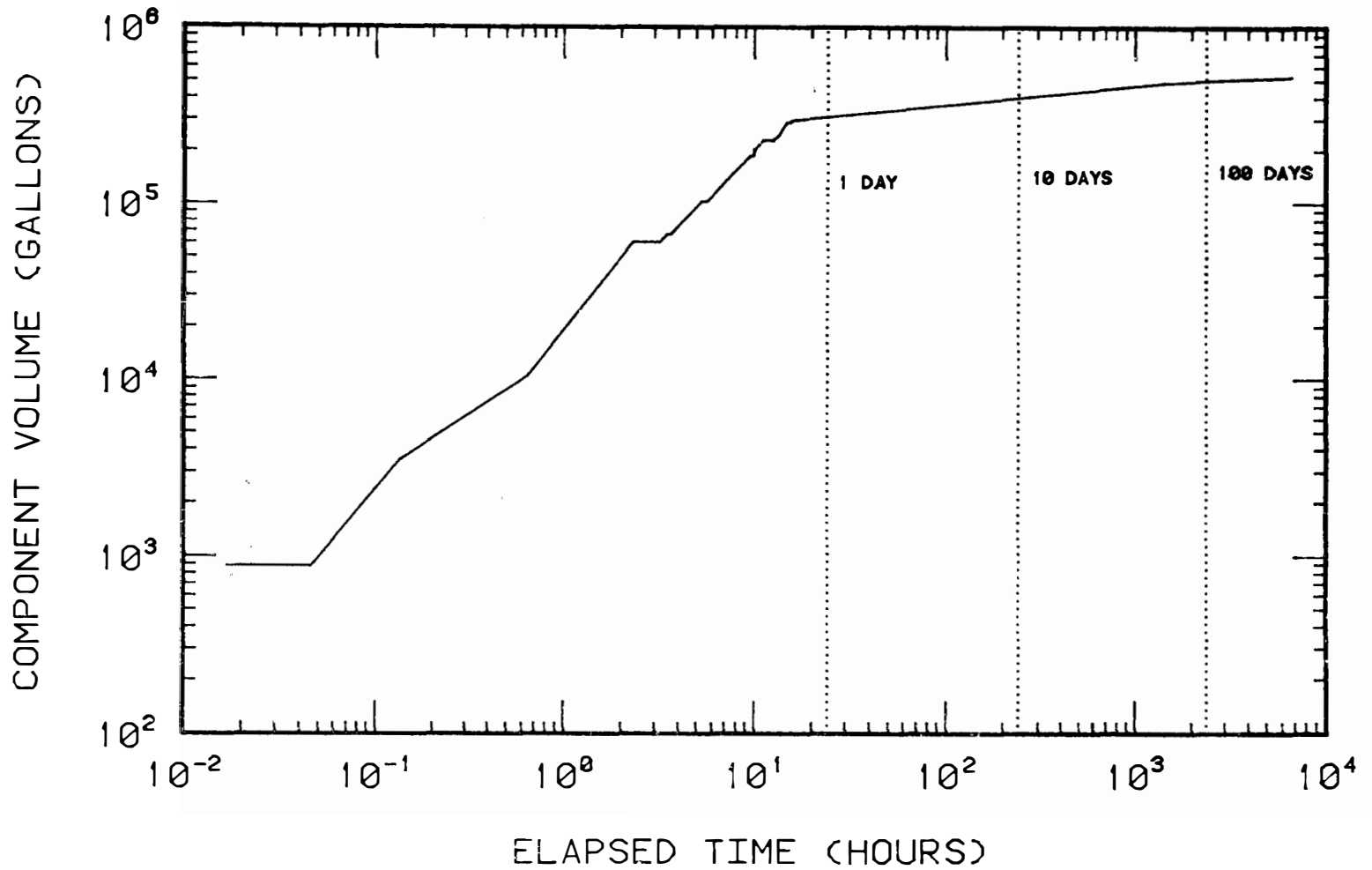
Since most of the water in the reactor building basement was reactor coolant, the high radiation levels hindered sampling work. This large volume of water accounted for significant inventory fractions; however, few data points are available. Using the available radiochemical data and calculated volumes, the inventory fractions were calculated and are presented in Figures 2-10 to 2-13.

Figure 2-10 presents the cesium inventory fractions for Cs-134 and Cs-137 in the reactor building basement water. Due to the exponential nature of various growth and decay functions, least-squares exponential fits of data were made and are shown. The inventory for Cs-134 is approximately 20 to 30 percent higher than for Cs-137. This difference is seen throughout all inventory data presented, and is discussed further in the next chapter.

The cesium inventory fractions in the reactor building sump are the largest outside the reactor vessel. The data shown in Figure 2-10 indicate that approximately 40 percent of the cesium was located in the reactor building basement water. This slight increase in inventory is attributed to continued reactor coolant leakage into the basement. The SDS processed reactor building basement water in batches numbered 1-45 containing a Cs-137 inventory fraction of 0.424.<sup>(9)</sup> This inventory fraction is nearly identical to that calculated herein.

FIGURE 2-9

TMI-2 REACTOR BUILDING SUMP LIQUID VOLUME



2-19

Table 2-3

Reactor Building Basement  
Liquid Volume

Source of Water	Volume (Gal.)	% of Total Volume
RCS ( PORV) March 28, 1979	278,000	53
RCS (leakage) March 28, 1979 to December 31, 1979	52,000	10
Reactor Building Sprays March 28, 1979	17,000	3
Reactor Building Air Coolers (leakage) March 28, 1979 to May 26, 1979	182,000	34
Total December 31, 1979	529,000	100



FIGURE 2-10

TMI-2 RB BASEMENT CESIUM INVENTORY

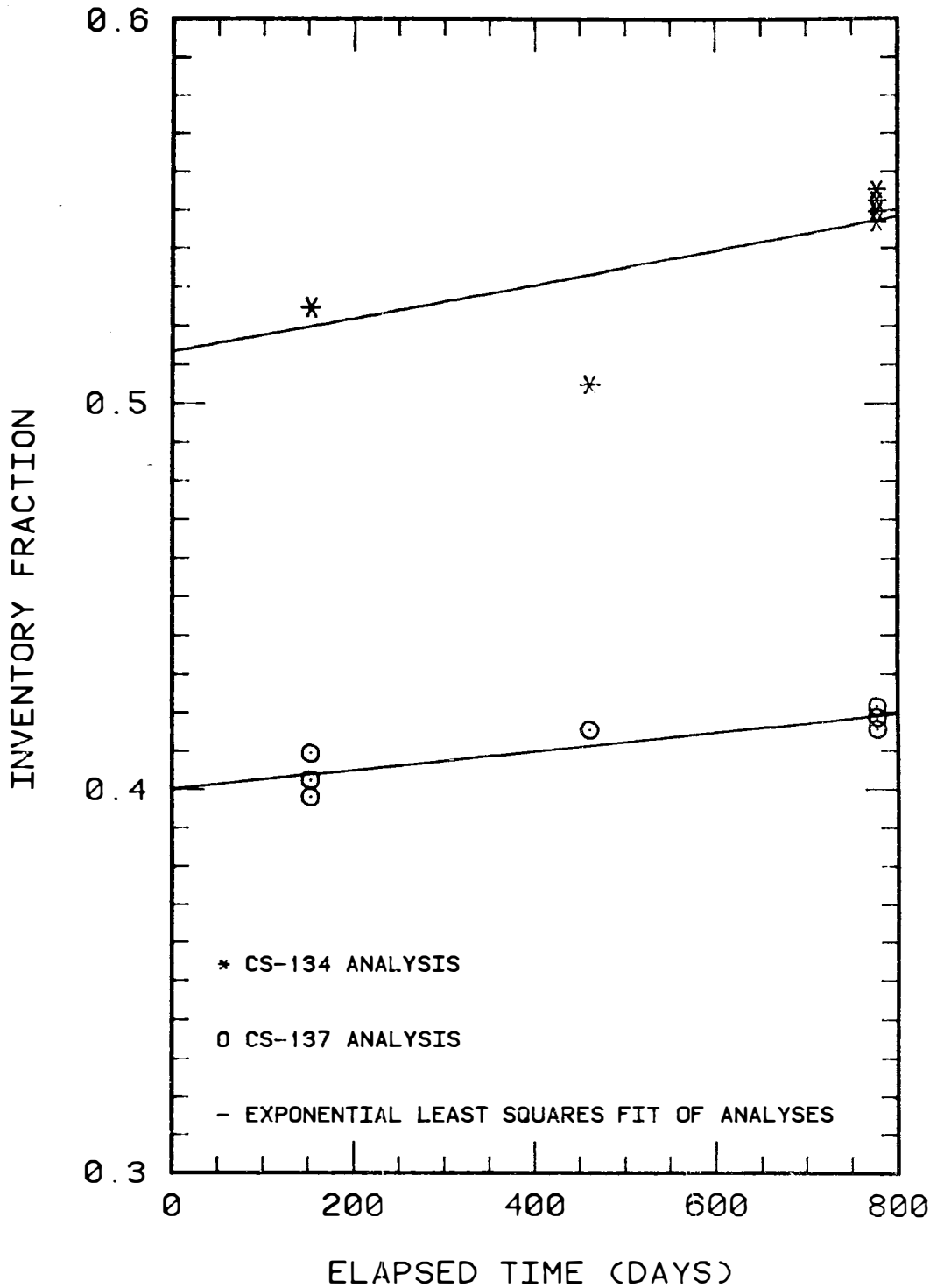


FIGURE 2-11

TMI-2 RB BASEMENT IODINE INVENTORY

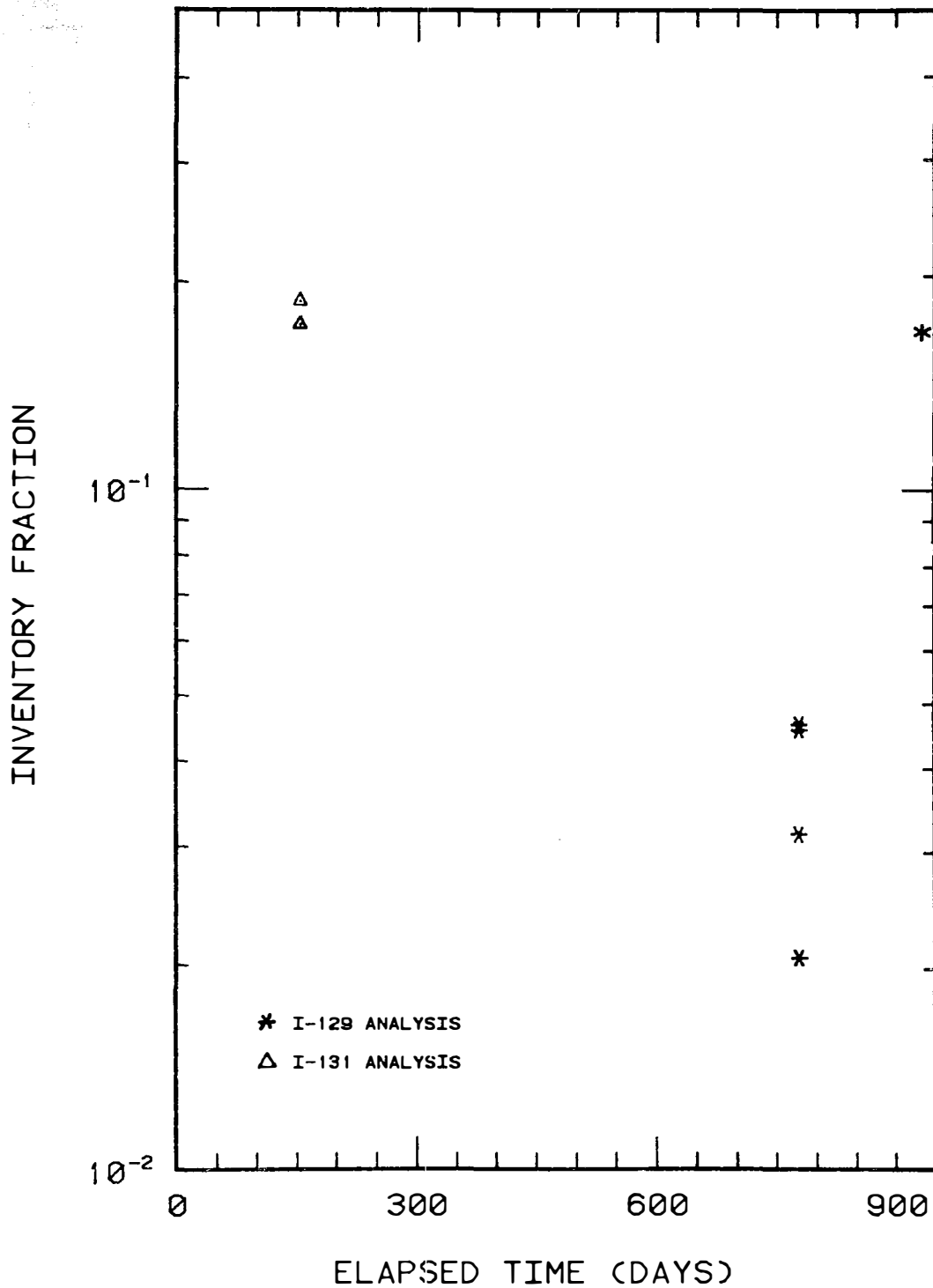


FIGURE 2-12

TMI-2 RB BASEMENT INVENTORIES

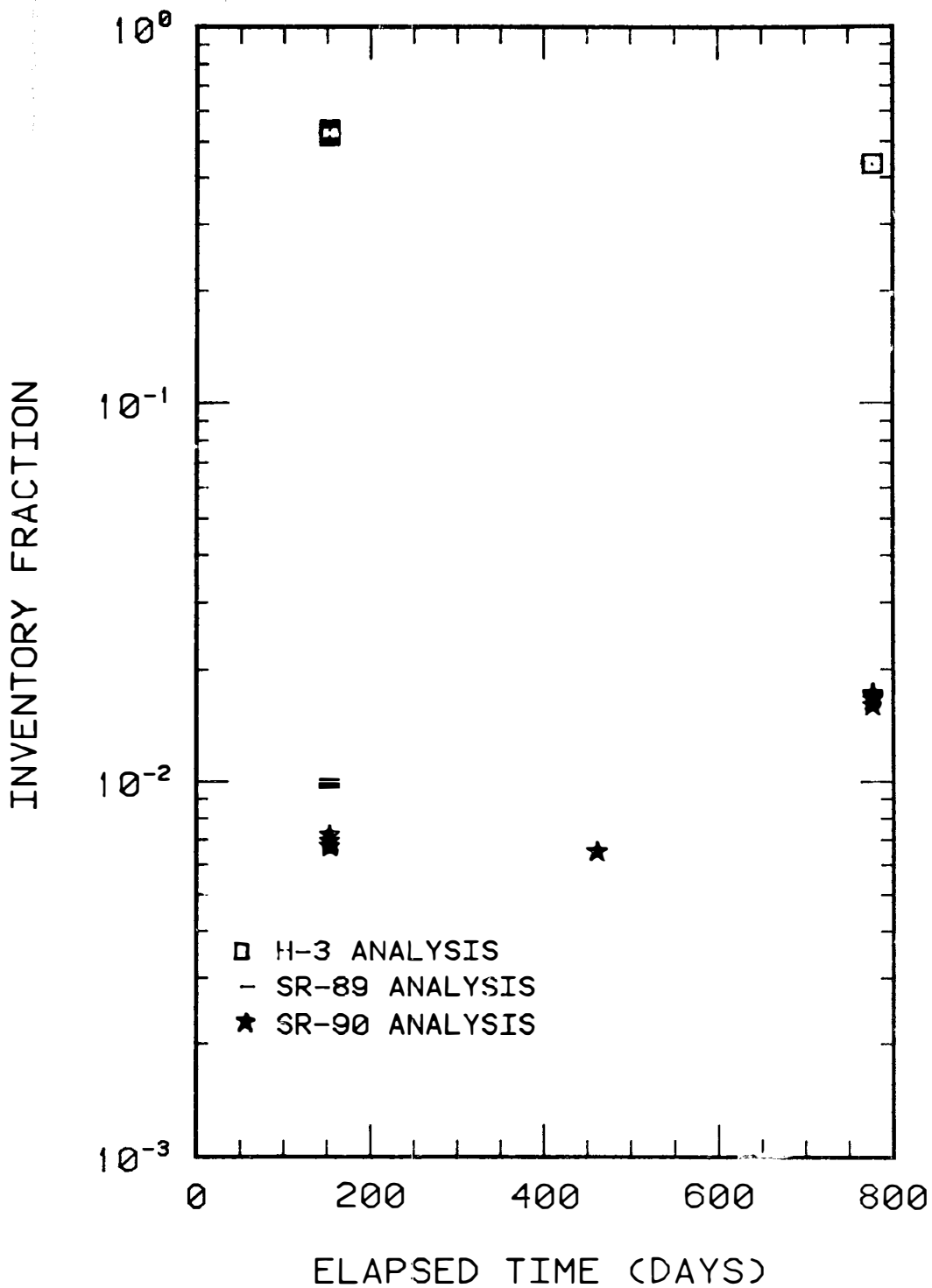


FIGURE 2-13

TMI-2 RB BASEMENT U-238 AND PU-239 INVENTORIES

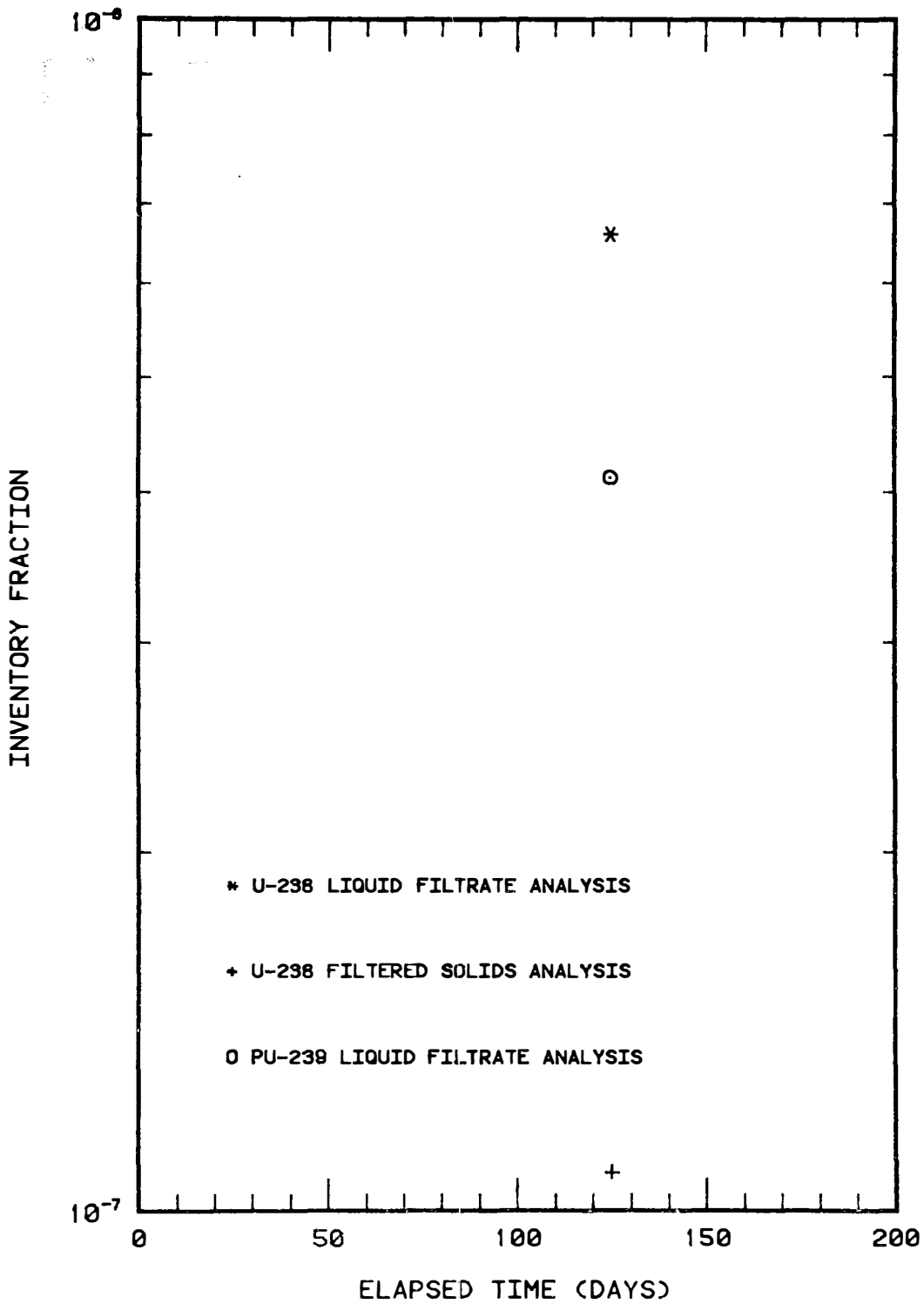


Figure 2-11 provides the radioiodine inventory fractions for the reactor building basement. Only the first basement samples, taken during August 1979, provide data on I-131 because of its rapid decay. These samples yield inventory fractions of 0.17 to 0.19. This is the largest inventory of radioiodine found outside the reactor vessel. Other basement water samples taken over two years after the accident were analyzed for I-129. These samples yield inventory fractions of 0.03 to 0.07.

Figure 2-12 contains inventory fractions for tritium and strontium. The samples yield tritium inventory fractions of 0.52 to 0.42 and strontium inventory fractions of 0.007 to 0.017 in the basement water. The liquid samples indicate that about half of the tritium was found in the reactor basement liquid. This was the largest inventory of tritium found. This is consistent with the relative concentrations of the reactor coolant and reactor coolant basement water; that is about 330,000 gallons of reactor coolant was transferred to the basement (Table 2-3). This is 3.7 times the full volume (88,600 gallons) of the reactor coolant system, and indeed, the basement inventory of 0.42 to 0.52 (on Figure 2-12) is about 3.7 times the earliest measured tritium inventory in the reactor coolant of about 0.12 (shown in Figure 2-16).

The liquid samples indicate that only about 1 percent of the strontium was found in the reactor basement liquid. The Sr-89 and Sr-90 data indicate that the strontium inventory is rising with the addition of reactor coolant leakage. This small rise in strontium inventory occurs faster than the rise in cesium inventory in the basement water because of the increasing strontium inventory and decreasing cesium inventory in the reactor coolant liquid. (The reactor coolant liquid is discussed in the next section.)

Figure 2-13 provides the fuel isotopic inventories. The results indicate that  $10^{-6}$  of the fuel material found its way to the basement water. Because of the lack of data, no trends could be analyzed for U-238 or Pu-239.

### 2.3.1.2 Reactor Coolant System

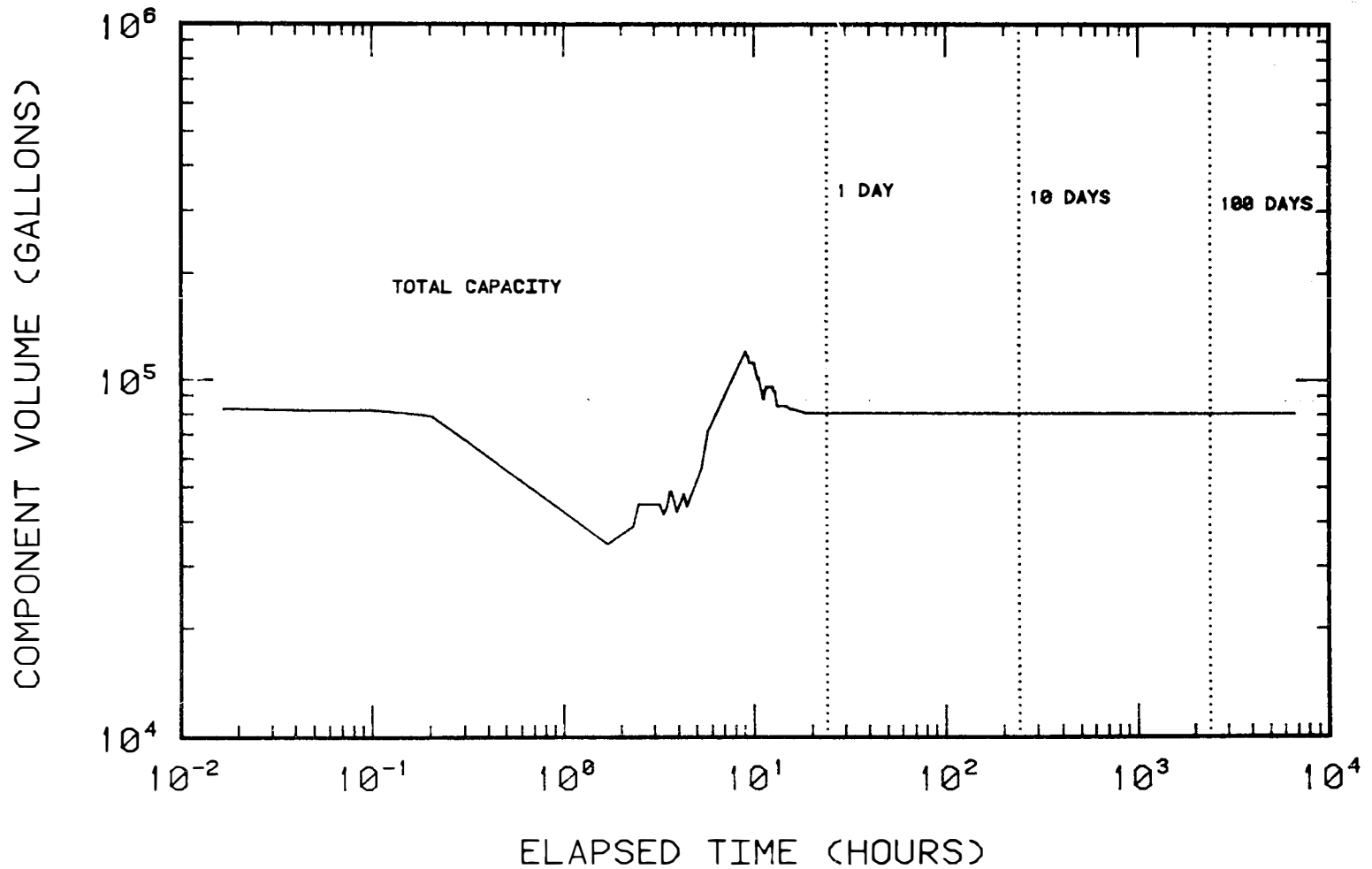
The reactor coolant system liquid transferred released-fission products from the fuel debris to various other components. From the numerous radiochemical samples taken, a good set of data is available to predict inventory fractions in the reactor coolant system liquid.

A chronology for events affecting the reactor coolant system has been previously developed<sup>(2)</sup> based on available sequence-of-event information. The TMI-2 mass-balance data base was used to predict an RCS liquid volume based on these data as shown in Figure 2-14. The chronology assumes that the RCS liquid volume is constant after March 28, 1979 at 2400 hours, that is, makeup flow is equal to let-down flow plus leakage. The makeup tank acts as a surge and supply tank keeping the RCS in a regulated state under normal operating conditions. Although the volume of the makeup tank and the entire MUP system are small, these components were a pathway for much reactor coolant transfer, and it is important to track the movement of the reactor coolant and radionuclides to various plant components. Figure 2-15 shows the makeup tank liquid volume.

Certain irregularities in the chronology in the present mass-balance data base cause a component's liquid volume to exceed the capacity. The RCS liquid volume for the day of the accident shows some irregularity due to the chronology developed to date. Although the decrease in RCS liquid volume is noted for the first 5 hours after reactor scram as expected, a subsequent sudden increase over the volume capacity is indicated. This overprediction, shown on Figure 2-14, results from estimate of average flow rates used for high pressure injection (HPI) addition and discharge rates through the open pilot-operated relief valve (PORV). The average values are used in lieu of actual flowrates which are unavailable. The correction of liquid-volume overprediction requires special analyses to determine HPI and PORV flowrates as a function of time, which could not be performed in the current work phase.

FIGURE 2-14

TMI-2 REACTOR COOLANT SYSTEM LIQUID VOLUME



Similarly, the liquid volume of the makeup tank is overpredicted (Figure 2-15). The correction of this overprediction will require further refinement of the MUP system leakage chronology. Such chronology extension would have a minor effect on the calculated inventory fractions in the major sinks.

The calculated inventory fractions for the reactor coolant system are shown in Figures 2-16 to 2-23.

Figure 2-16 presents the calculated tritium inventory fractions in the RCS. The inventory fraction appears to decline from approximately 0.10 to 0.02 over the first few months, then remains level at about 0.02.

The RCS tritium inventory has been represented by a smooth curve in Figure 2-16. It is anticipated that the tritium inventory would fall due to leakage and subsequent addition of nontritiated water, the correlation of the observed reduction in tritium concentration with the chronology of dilution by makeup for leakage has not been made.

Figures 2-17 and 2-18 present the strontium-89 and -90 inventory in the RCS, respectively. The Sr-89 and Sr-90 inventory fractions range from 0.005 to 0.01. Both isotopes analyzed show similar trends, especially when data in the first several days are excluded.

The RCS strontium inventory has been represented by an exponential fit in Figures 2-17 and 2-18. The same curve fits both sets of data well, and indicates that an exponential buildup of strontium was occurring in the RCS, which was probably due to continued leaching from the core debris. This leaching was greater than the loss of strontium in leakage to the reactor building basement water. This leaching mechanism apparently was responsible for the small, but increasing, inventory fractions in the RCS and basement water,



FIGURE 2-15

TMI-2 MAKEUP TANK LIQUID VOLUME

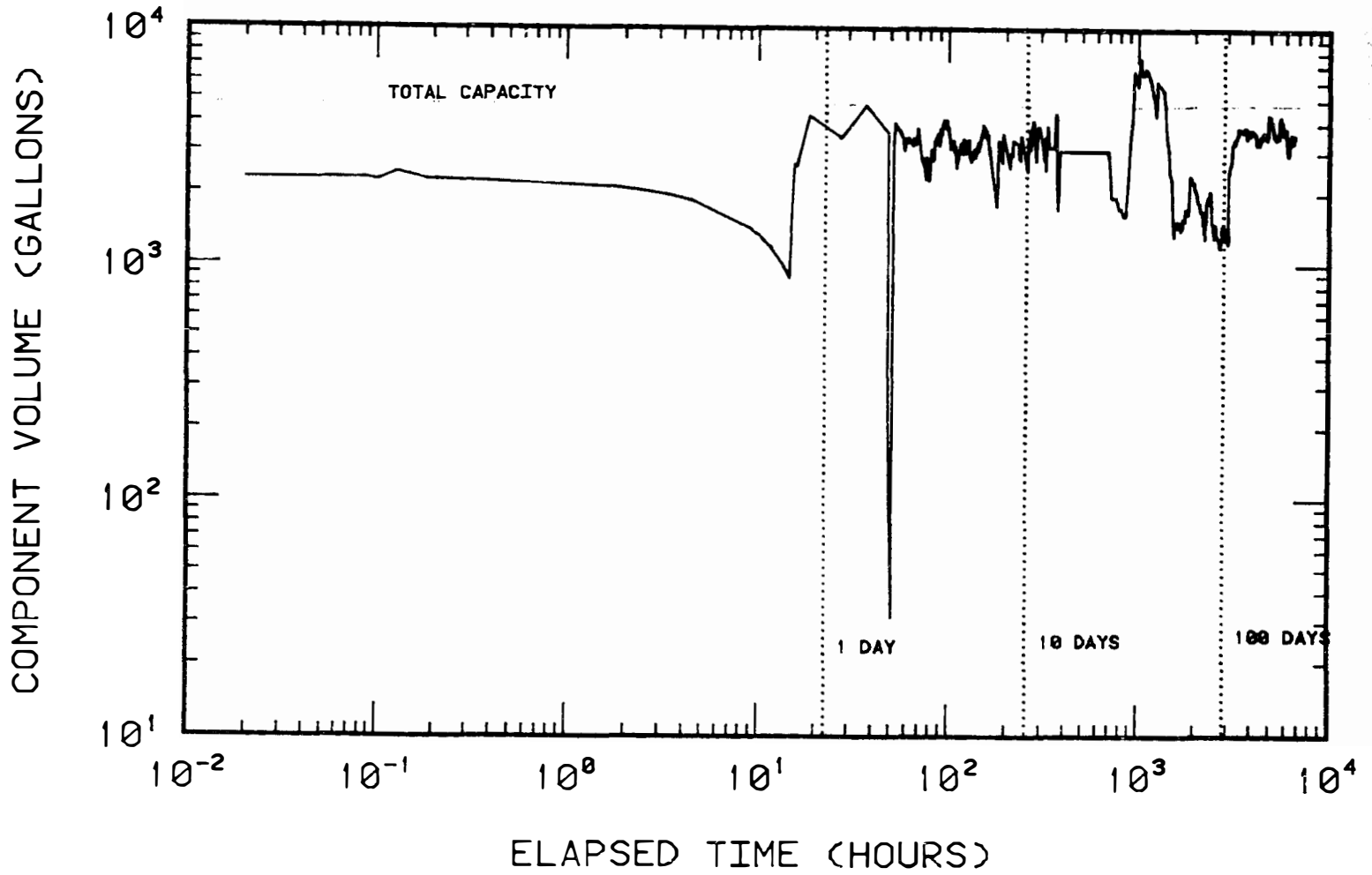


FIGURE 2-16

TMI-2 RCS H-3 INVENTORY

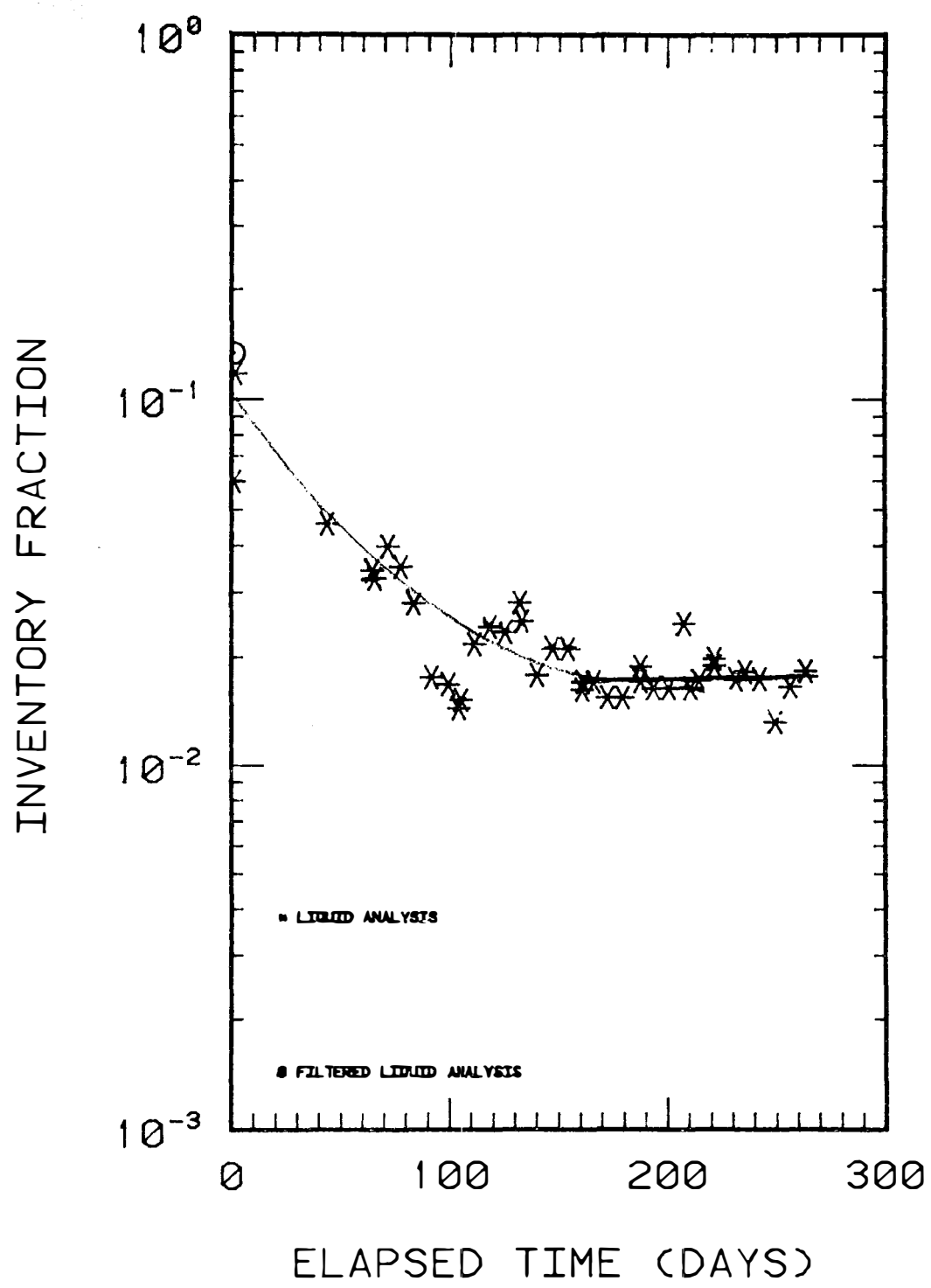


FIGURE 2-17

TMI-2 RCS SR-89 INVENTORY

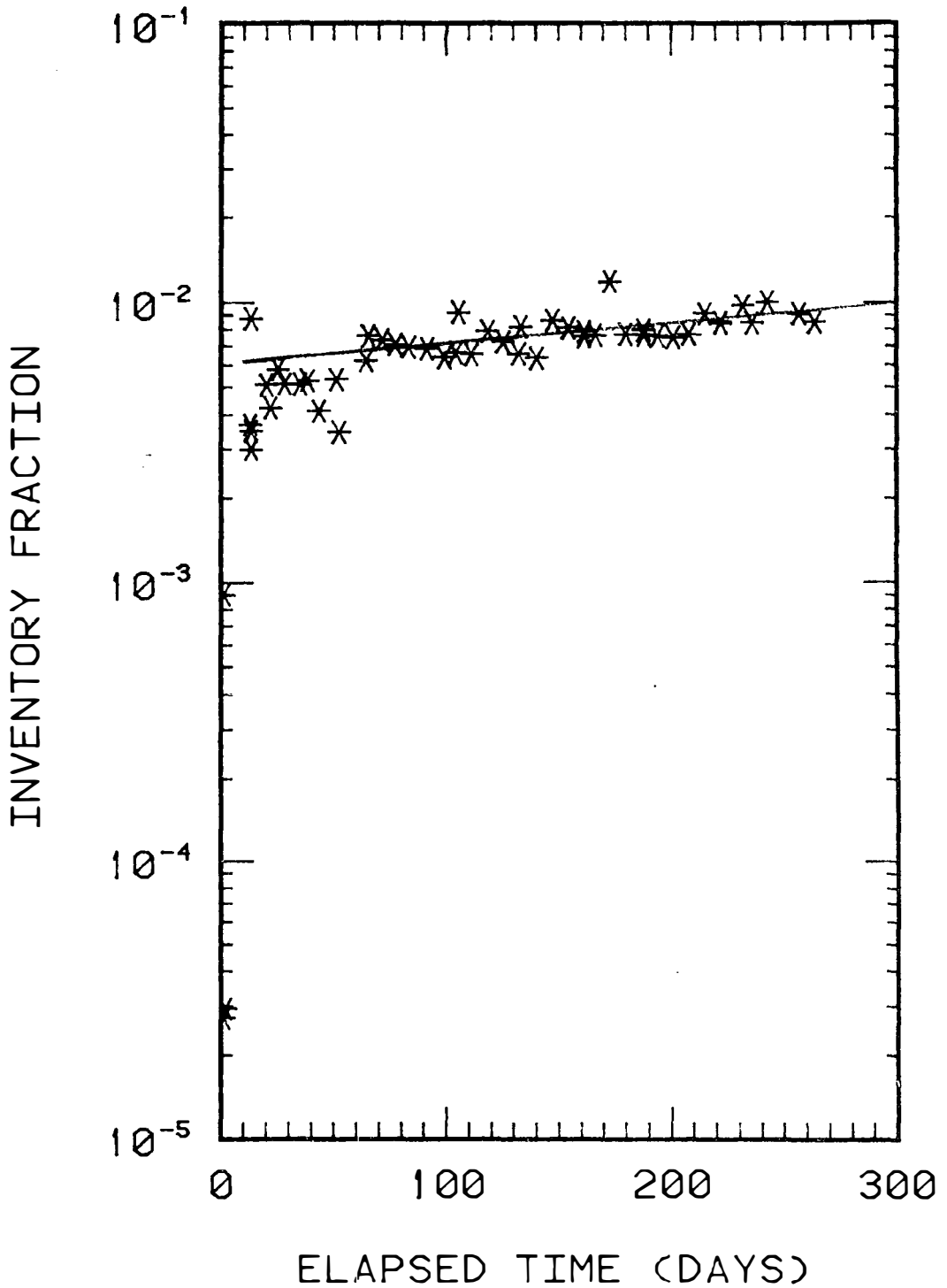


FIGURE 2-18

TMI-2 RCS SR-90 INVENTORY

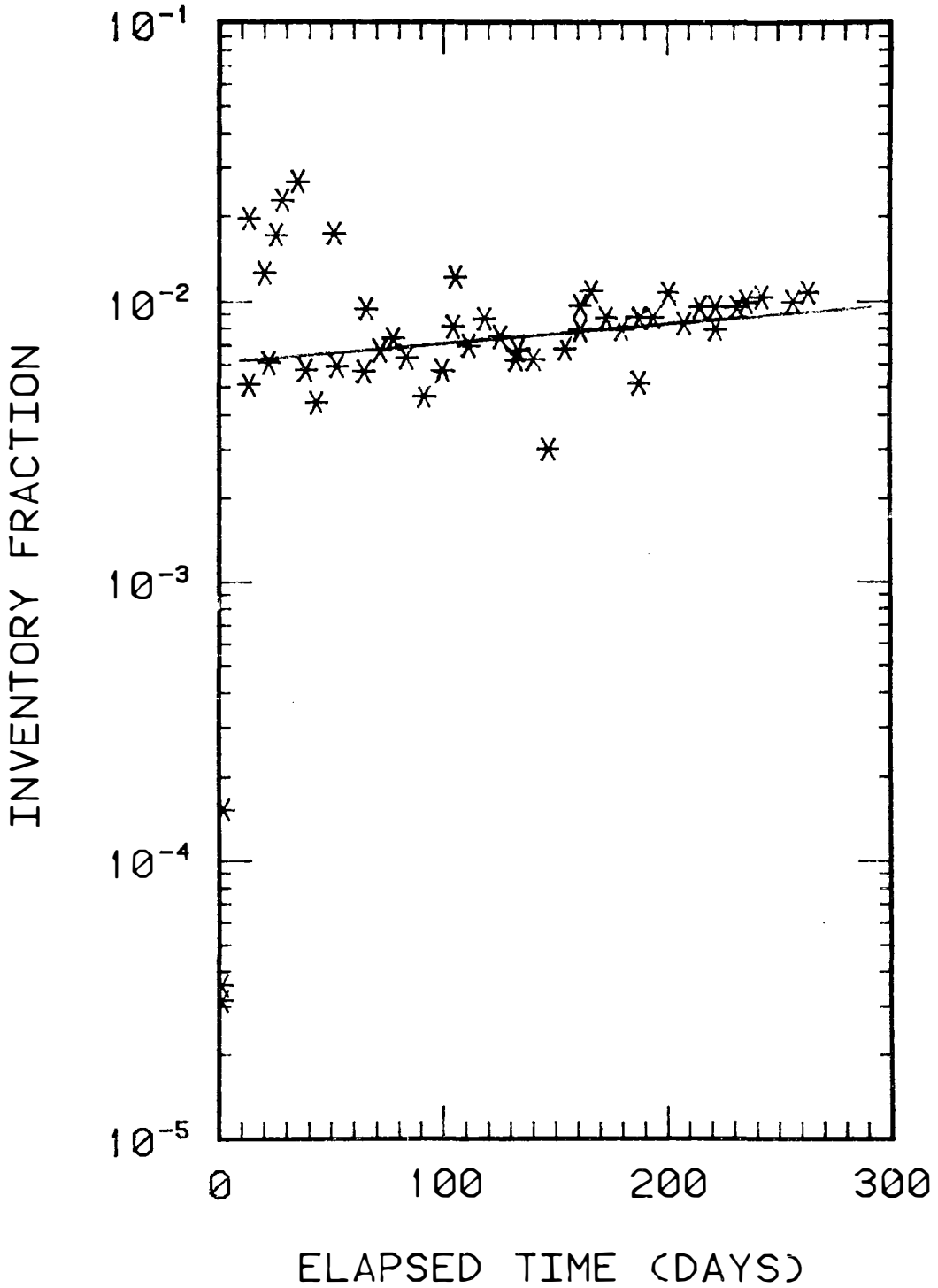


FIGURE 2-19

TMI-2 RCS I-129 INVENTORY

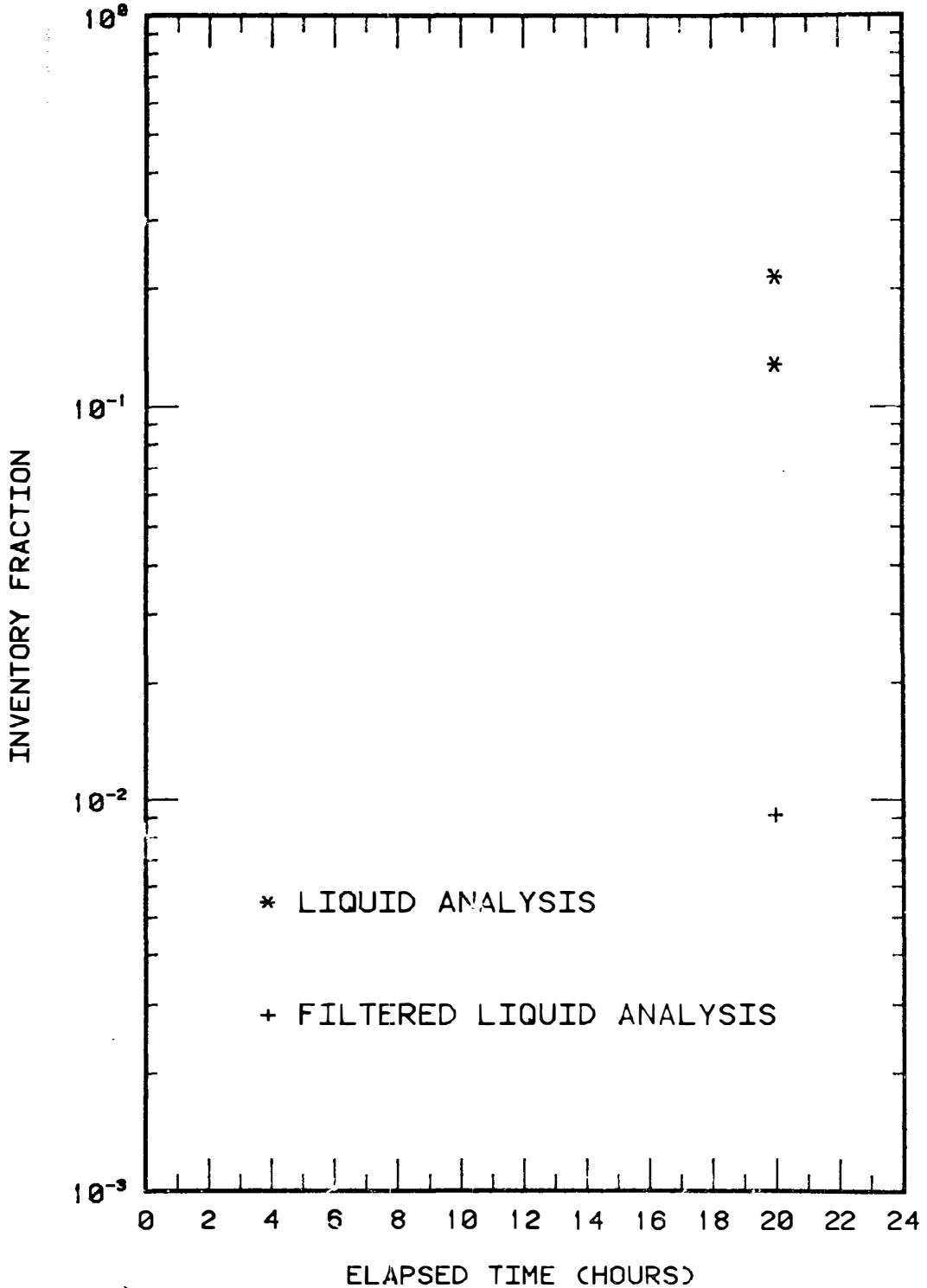


FIGURE 2-20

TMI-2 RCS I-131 INVENTORY

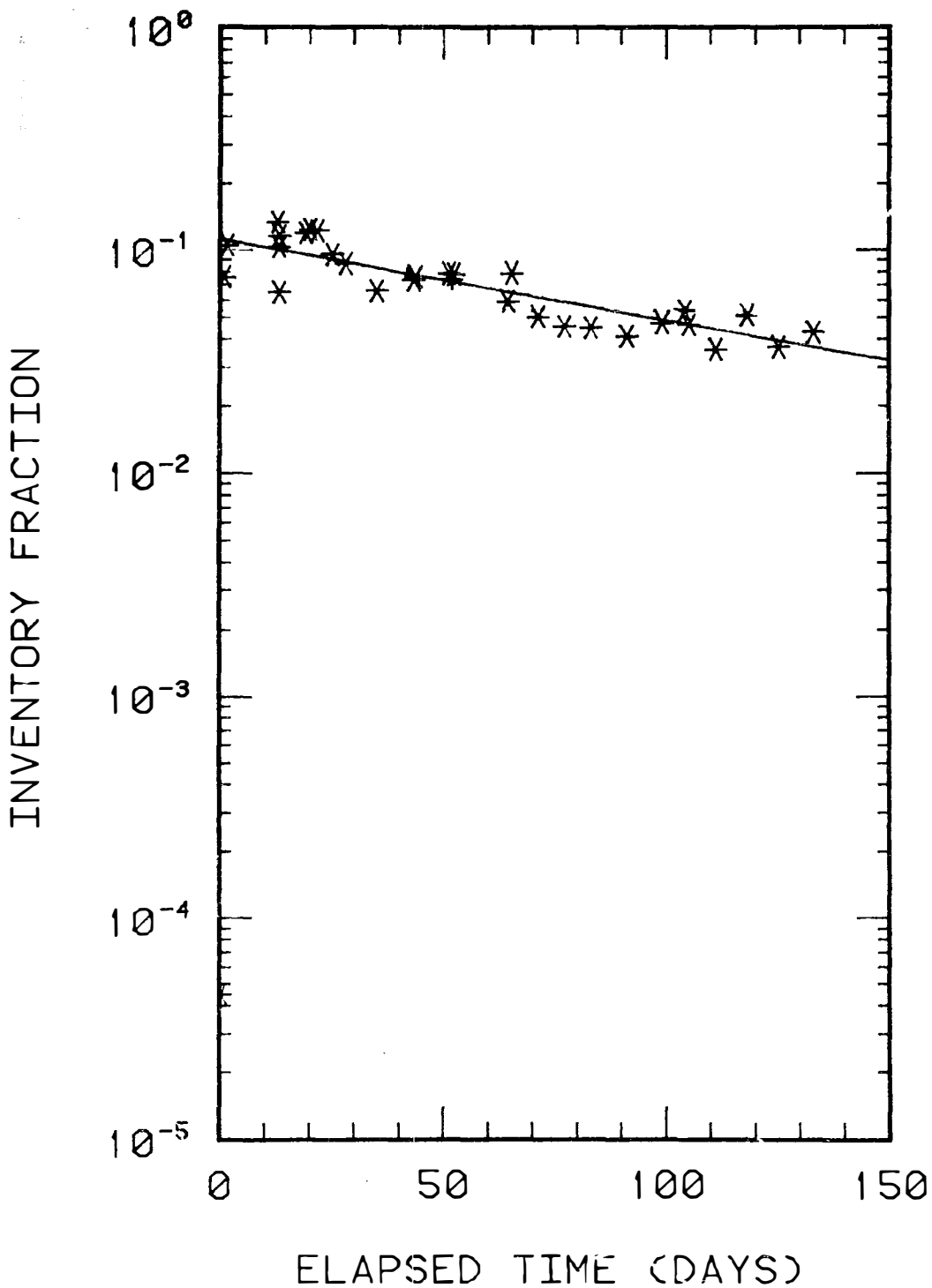


FIGURE 2-21

TMI-2 RCS CS-134 INVENTORY

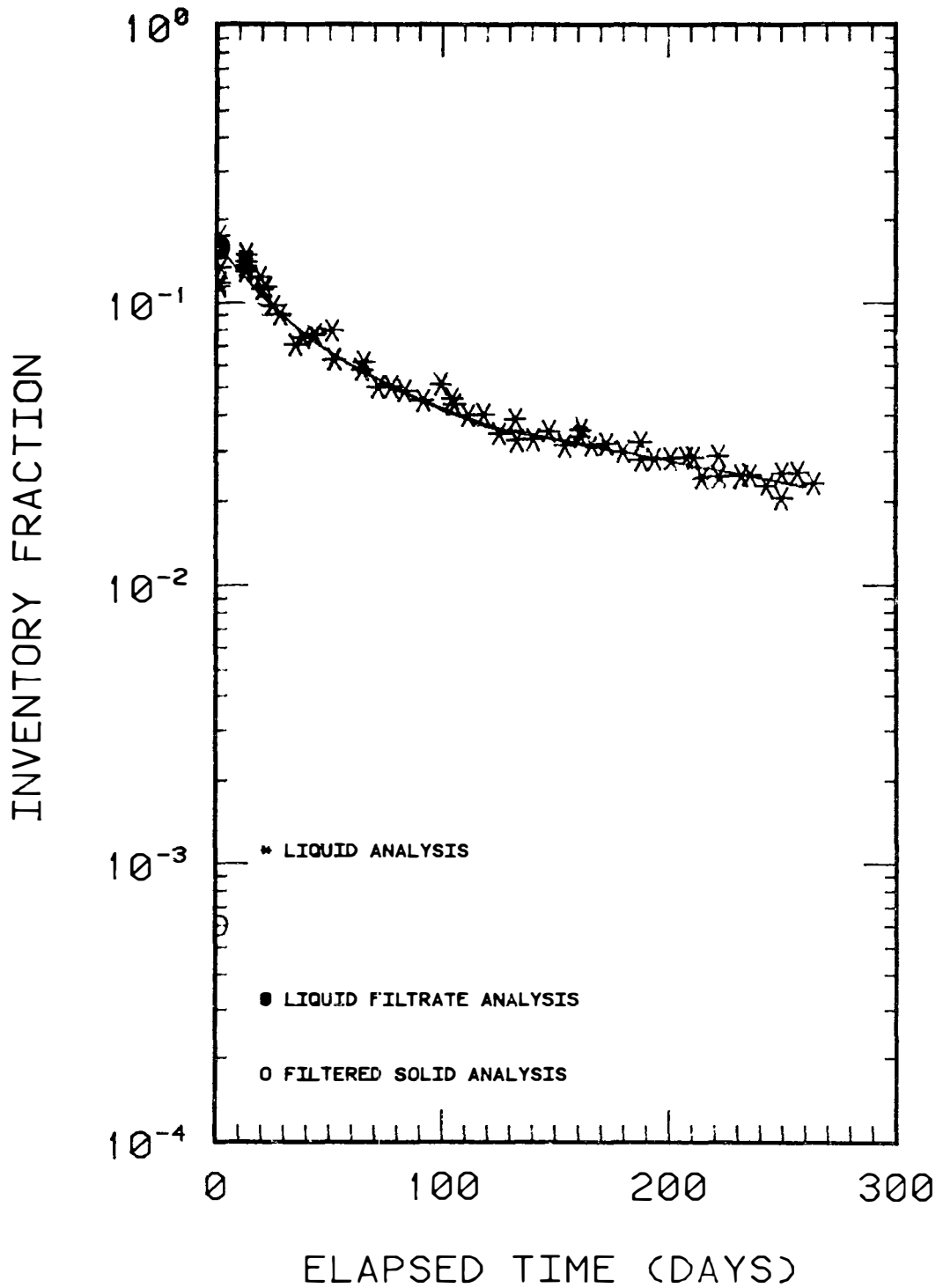


FIGURE 2-22  
TMI-2 RCS CS-137 INVENTORY

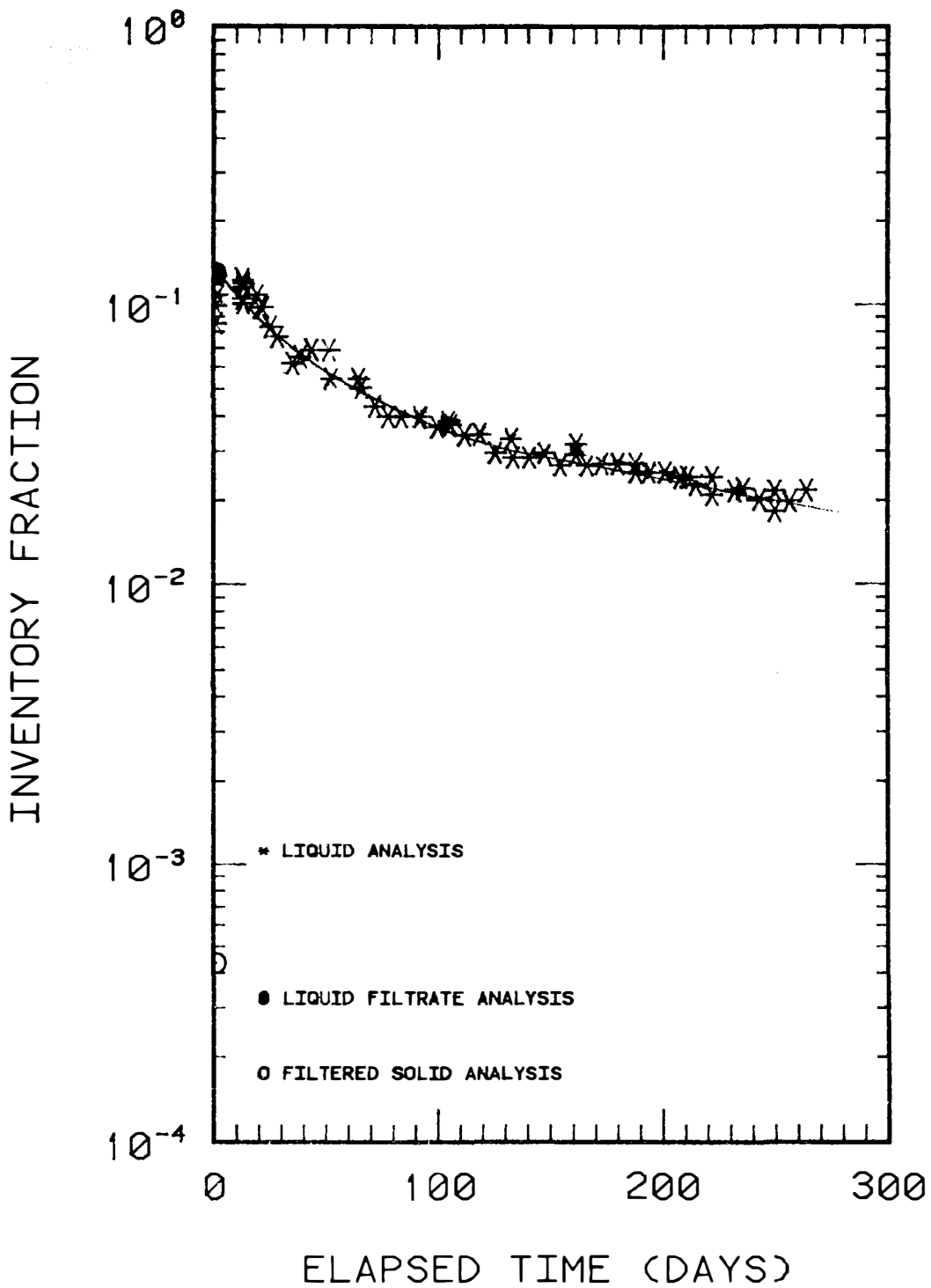
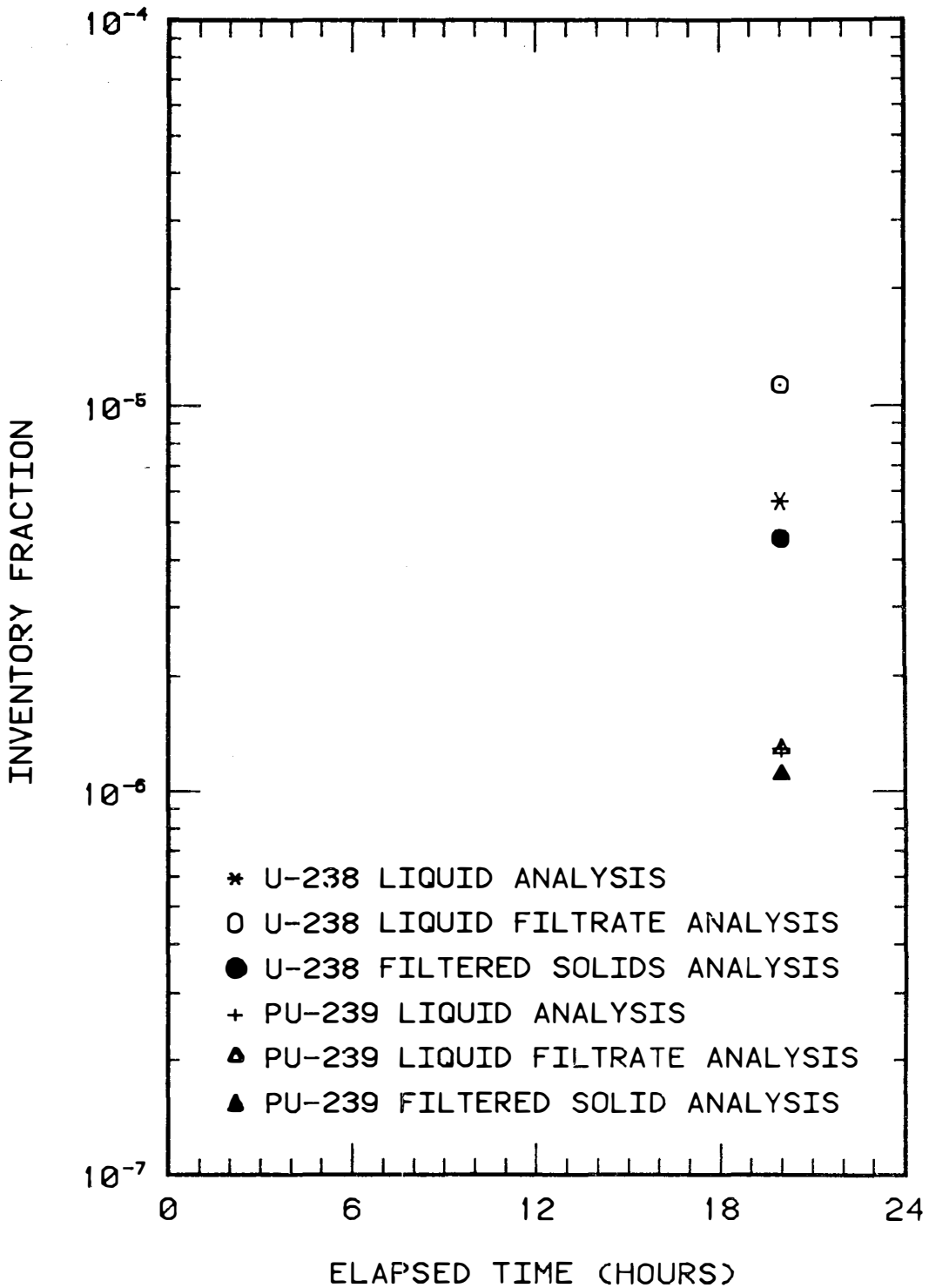




FIGURE 2-23

TMI-2 RCS U-238 AND PU-239 INVENTORY



which is very different from the postulated cesium behavior discussed later in this section.

Figures 2-19 and 2-20 provide the radioiodine inventory estimates for the RCS. The only I-129 inventory data found, are presented in Figure 2-19. The inventory fractions calculated fit well with the I-131 inventory fractions shown in Figure 2-20. The samples yield inventory fractions of 0.10 to 0.03 for the liquid in the RCS.

Figure 2-20 provides an insight into the time behavior of the RCS I-131 inventory. The results indicate that the iodine inventory fraction in the RCS was decreasing over the few months when data were available.

Figures 2-21 and 2-22 present the cesium inventory fraction data in the RCS for 1979. Although the Cs-134 data in Figure 2-21 are about 20 percent higher than those for Cs-137 in Figure 2-22, the curves are similar. The Cs-137 inventory fraction falls more rapidly during the first 2 months, then decreases at a slower exponential rate thereafter. The cesium inventory fraction in the RCS fell from about 0.15 to 0.02 during 1979. The depletion of cesium from the RCS is similar to I-131, since the slope of the tail is nearly equal to the slope of the curve of Figure 2-20.

Other data from the TMI-2 mass balance indicate that small inventories of radiogases were detected in the RCS liquid. These liquid analyses yield Kr-85, I-131, and Xe-133 inventory fractions in the ranges of  $3E-7$  to  $9E-6$ ,  $2E-8$  to  $3E-7$ , and  $6E-7$  to  $2E-5$ , respectively.

Figure 2-23 shows the small fuel material inventory fractions detected and analyzed in sample RCS-1. About  $10^{-5}$  of the U-238 and  $10^{-6}$  of the Pu-239 were detected in the RCS water. No analysis of trends was available with the data shown.

#### 2.3.1.3 Reactor Coolant Drain Tank

Steam and water were released from the RCS via the pressurizer to the reactor coolant drain tank during the accident and recovery periods. When the reactor coolant drain tank's rupture disk blew, reactor coolant was released to the reactor building basement. The basement water level increased steadily due to service water and RCS leakage, the latter partially via this tank. No radiochemical data have been reported to date for the reactor coolant drain tank. Thus for this study, the assumption has been made that the concentrations of this tank are similar to the RCS concentrations, since the RCS is the only source of activity. Figure 2-24 contains the calculated volume of the reactor coolant drain tank, which is 7500 gallons (full capacity) for times of interest. The estimated isotopic inventories would be proportional to the RCS inventories by liquid-volume ratio. This assumes continuous, good mixing from RCS leakage. Thus, the estimated isotopic inventory fractions would be 9 percent of the RCS inventories presented in the previous section.

#### 2.3.1.4 Reactor Coolant Bleed Tanks

The reactor coolant bleed tanks (RCBTs) are the largest storage tanks in the TMI-2 liquid waste system. These three tanks (RCBT-A, RCBT-B, and RCBT-C) normally provide supply or storage for water during startup or shutdown operations. The RCBTs were used during the TMI-2 accident for storage of excess diverted letdown and relief flows. The RCBTs also provided a source of batched water to the makeup tank to assure proper supply for RCS maintenance. Because of the large liquid volume, the reactor coolant bleed tanks are major sinks with significant inventory fractions.

The liquid volumes predicted for the RCBTs from the TMI-2 mass-balance data base are shown in Figures 2-25, 2-26, and 2-27 for RCBT-A, RCBT-B, and RCBT-C, respectively. The Figures show the liquid volume changes in the respective tanks for 1979 based on the data available to date. Due to frequently changing liquid volumes

FIGURE 2-24

TMI-2 REACTOR COOLANT DRAIN TANK LIQUID VOLUME

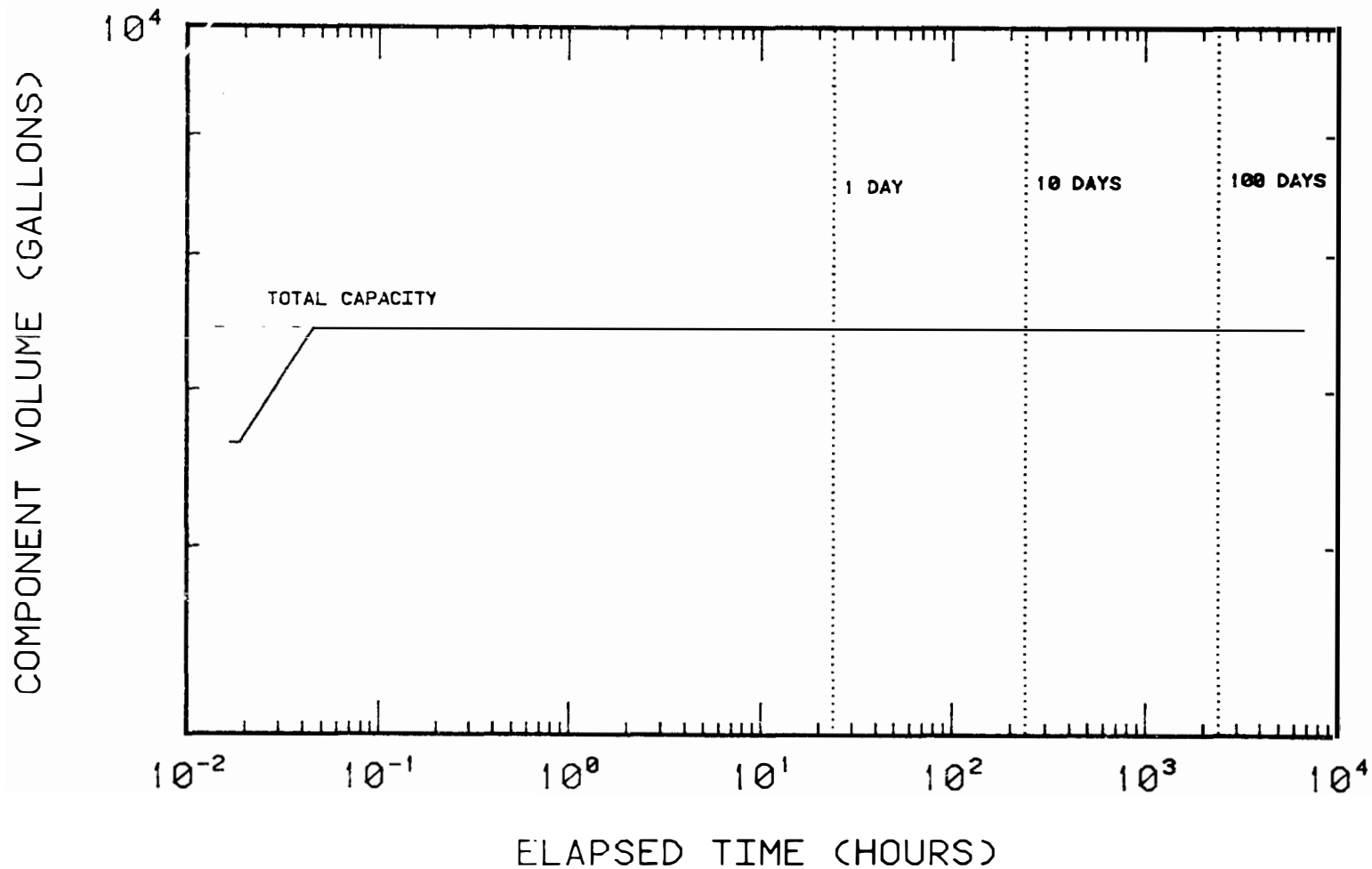


FIGURE 2-25

TMI-2 RCBT-A LIQUID VOLUME

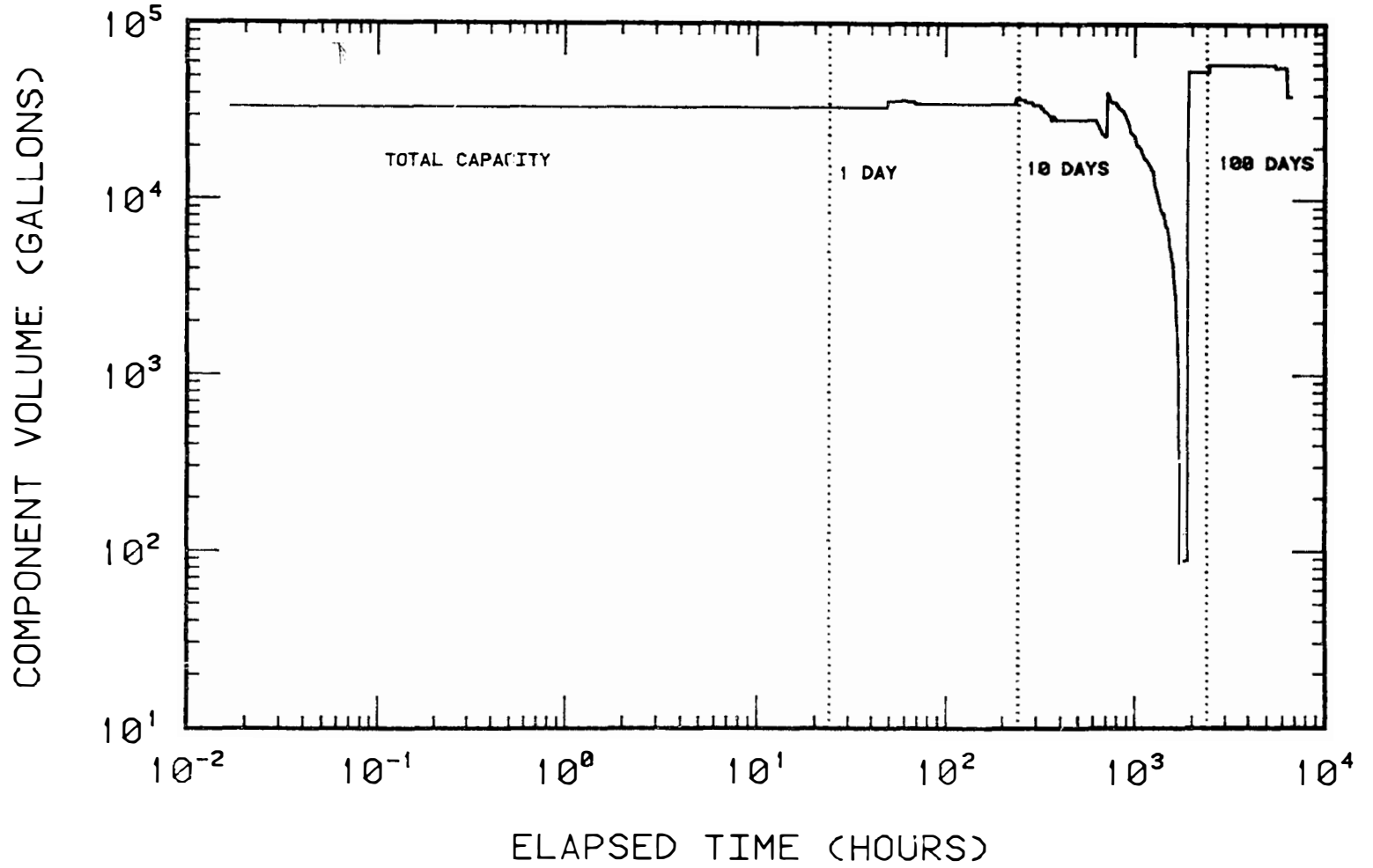
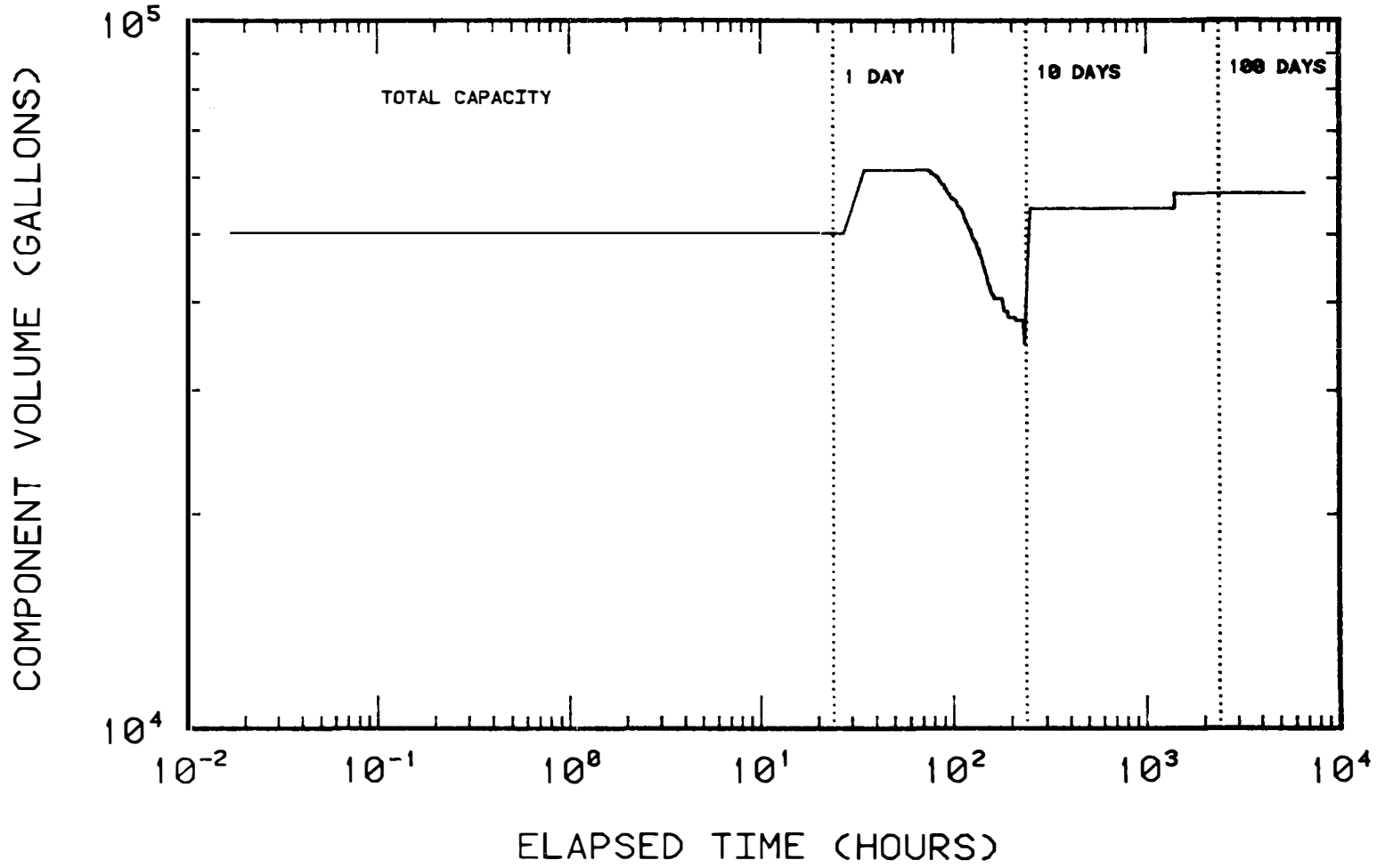


FIGURE 2-26

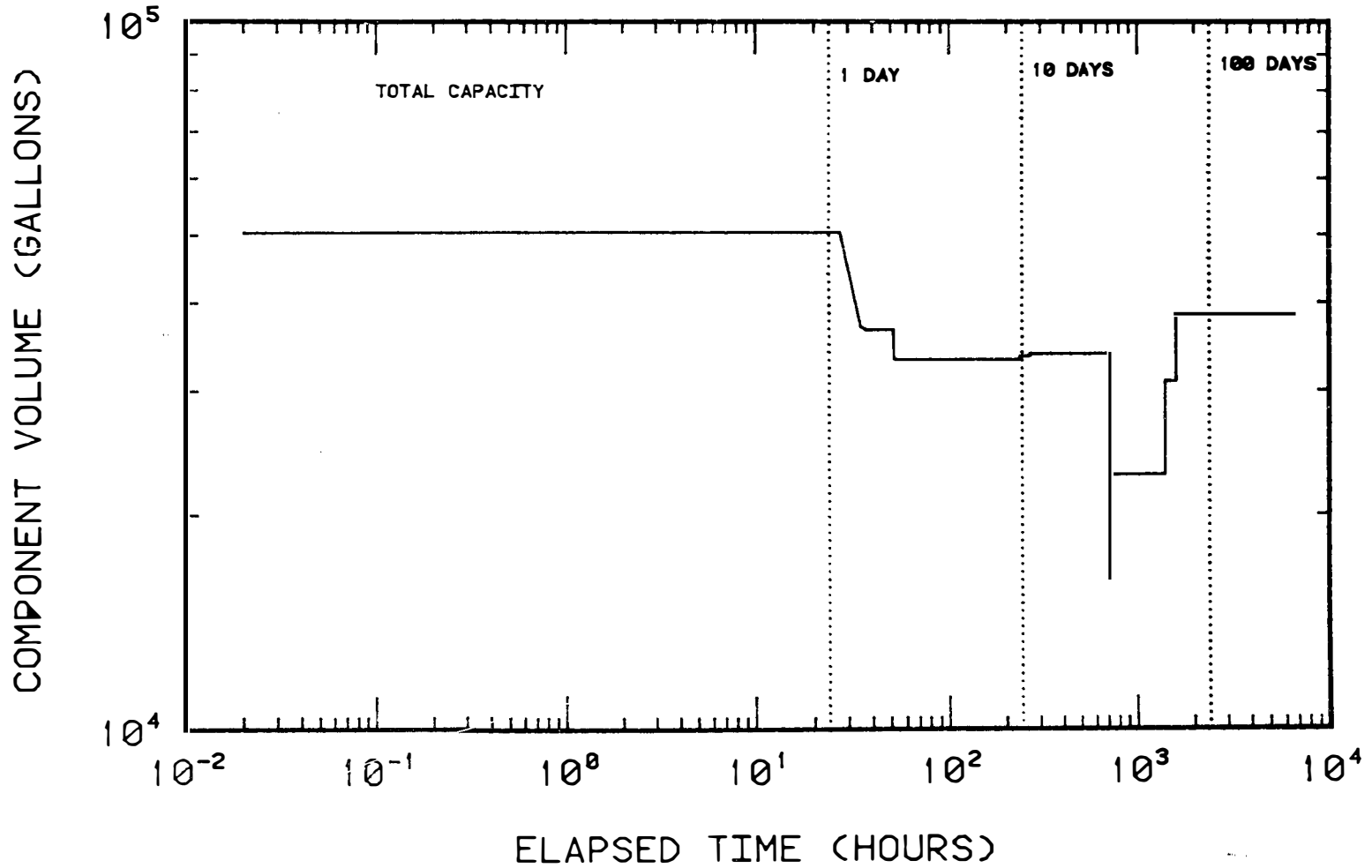
TMI-2 RCBT-B LIQUID VOLUME



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FIGURE 2-27

TMI-2 RCBT-C LIQUID VOLUME



and infrequent sampling, trend analyses for the RCBTs were not performed.

Figure 2-28 shows the calculated inventory fractions for tritium in the RCBTs. The data indicate that the tritium inventory in each of the RCBTs is on the order of 0.01.

Figures 2-29 to 2-33 provide the other isotopic inventory fractions for RCBT-A. The data in Figure 2-29 indicate that the strontium inventory fraction is approximately 0.0002. The radioiodine data and cesium data are shown in Figures 2-30 and 2-31. These inventory fractions range from 0.0001 to 0.01. Figure 2-32 presents the dissolved Xe-133 gas, which is small and insignificant, in the RCBT-A liquid. Finally, Figure 2-33 provides the fuel material inventory, which again is very small in the liquid--U-238 which is about  $4 \times 10^{-7}$  and Pu-239 which is about  $1 \times 10^{-7}$ .

Fewer radiochemical samples were taken during 1979 for RCBT-B and RCBT-C. Figures 2-34 and 2-35 provide the inventory fractions for RCBT-B. The I-131 and cesium inventories are about 0.0001 to 0.01 and 0.01, respectively. Similar data are presented in Figures 2-36 and 2-37 for RCBT-C. Again, the small amount of data available, with large volume changes, does not allow trends to become readily apparent.

### 2.3.2 Liquid Releases

Liquids from TMI-2 were released in a controlled manner during and following the accident. Liquid in the tankage of TMI-2 radwaste system prior to the accident was sent to TMI-1 to make storage available for additional water. Some of this water was sent through the IWTS (Industrial Waste Treatment System) and IWFS (Industrial Waste Filtration System) and subsequently released to the Susquehanna River along with water from TMI-1 refueling operations. Detailed information is available on this release path for April 1979.<sup>(8)</sup> The release of significance attributed to TMI-2



FIGURE 2-28  
TMI-2 RCBT H-3 INVENTORIES

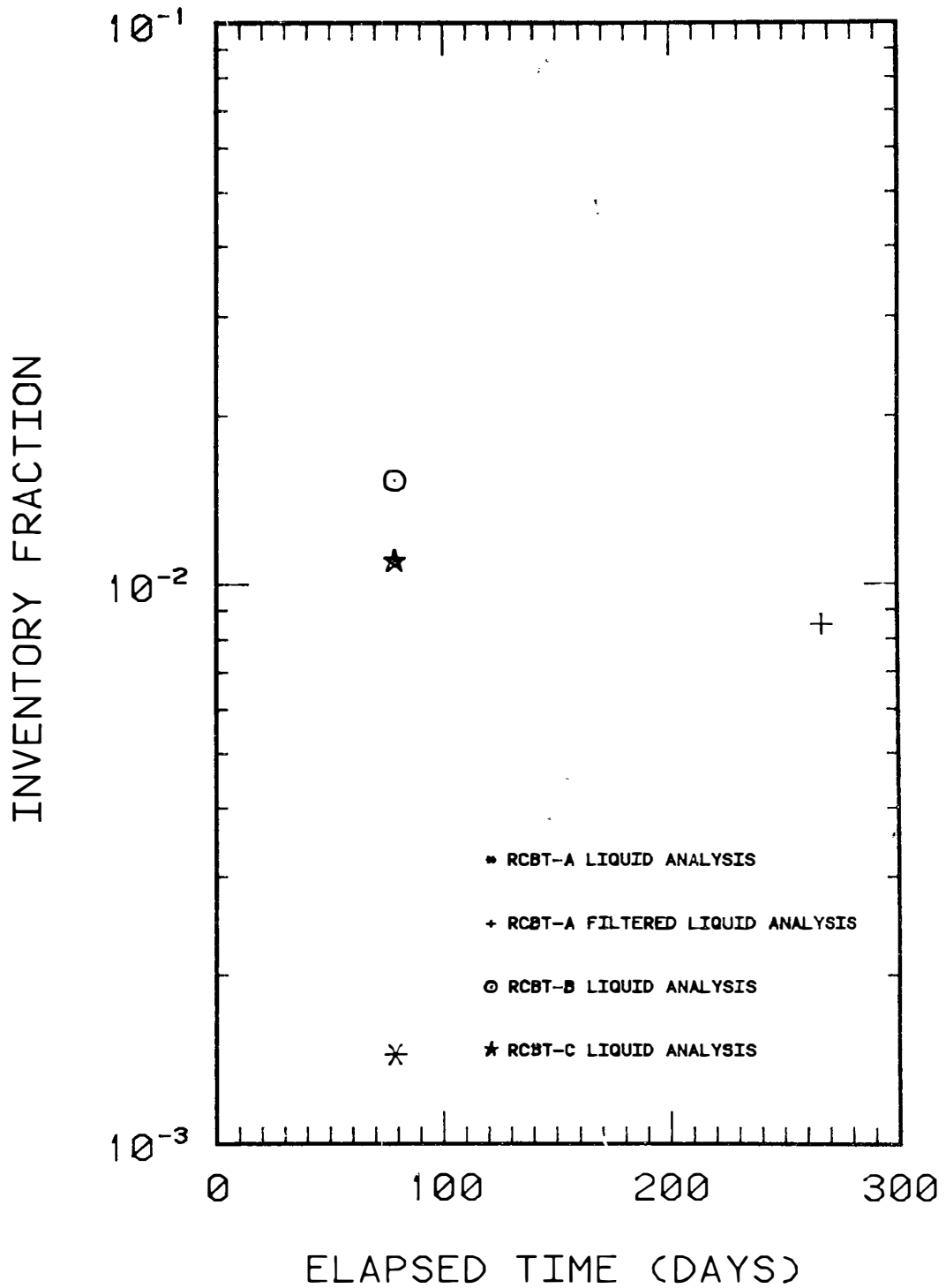


FIGURE 2-29  
TMI-2 RCBT-A SR-90 INVENTORY

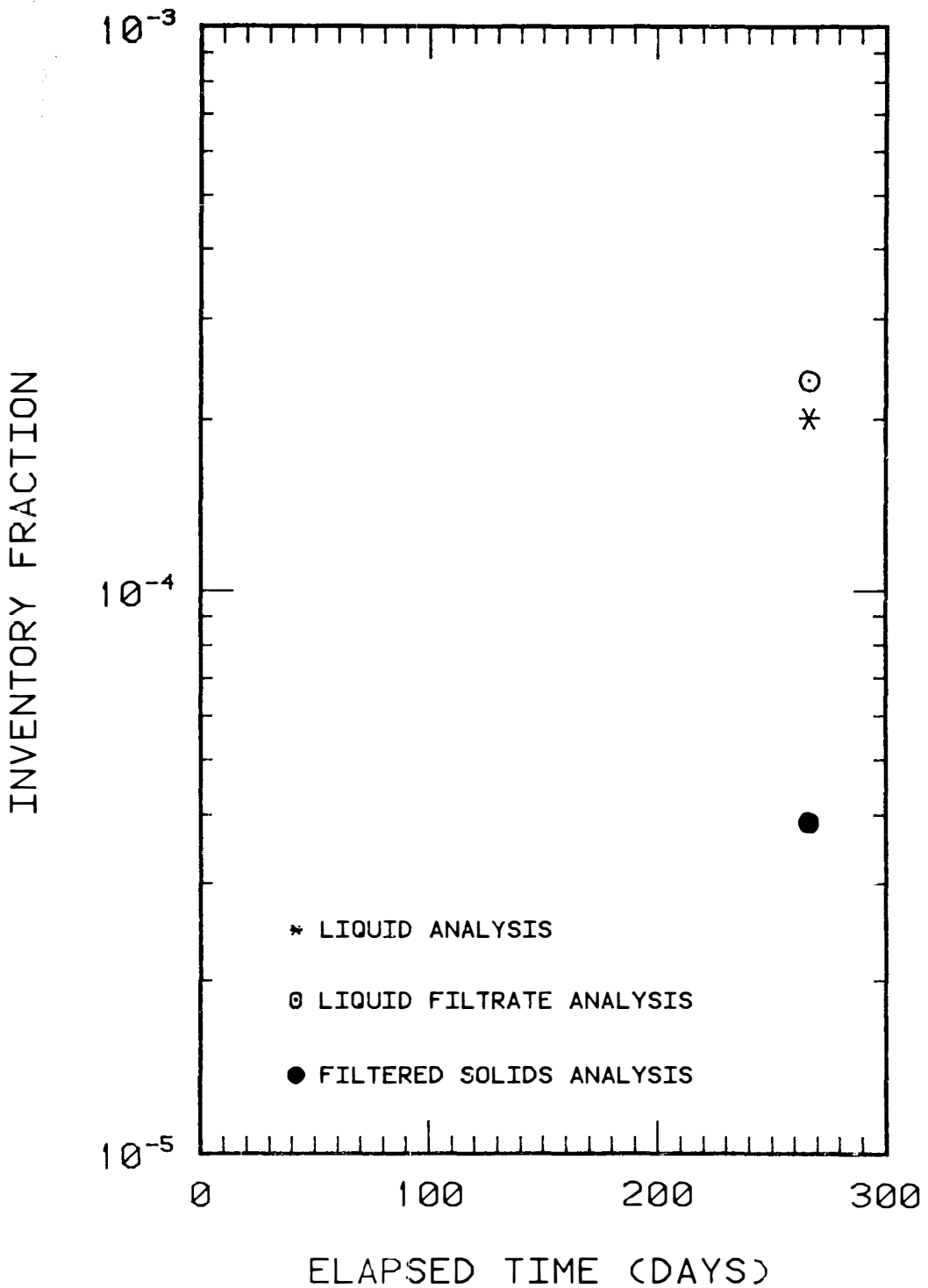


FIGURE 2-30

TMI-2 RCBT-A RADIOIODINE INVENTORY

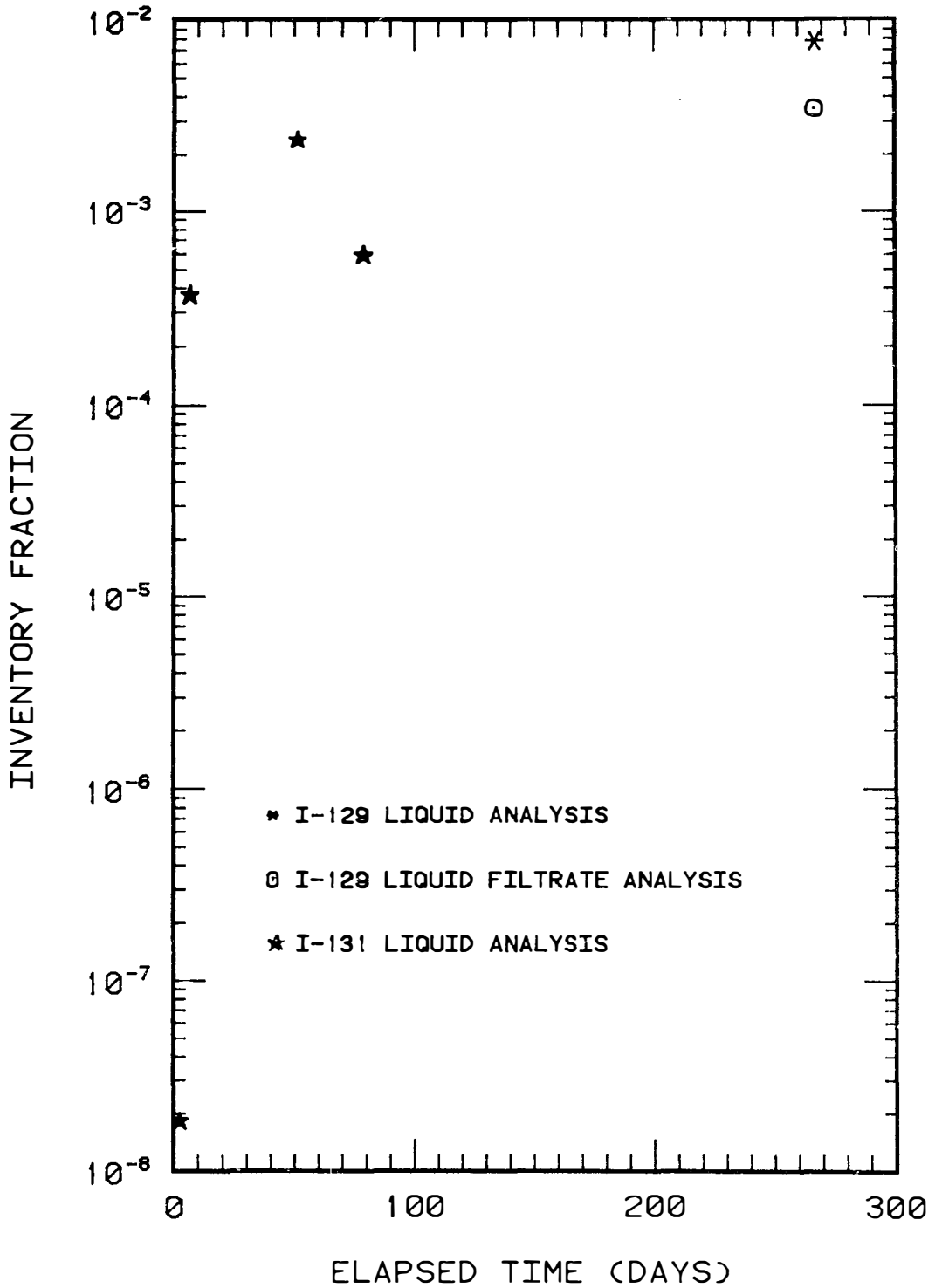


FIGURE 2-31

TMI-2 RCBT-A CESIUM INVENTORIES

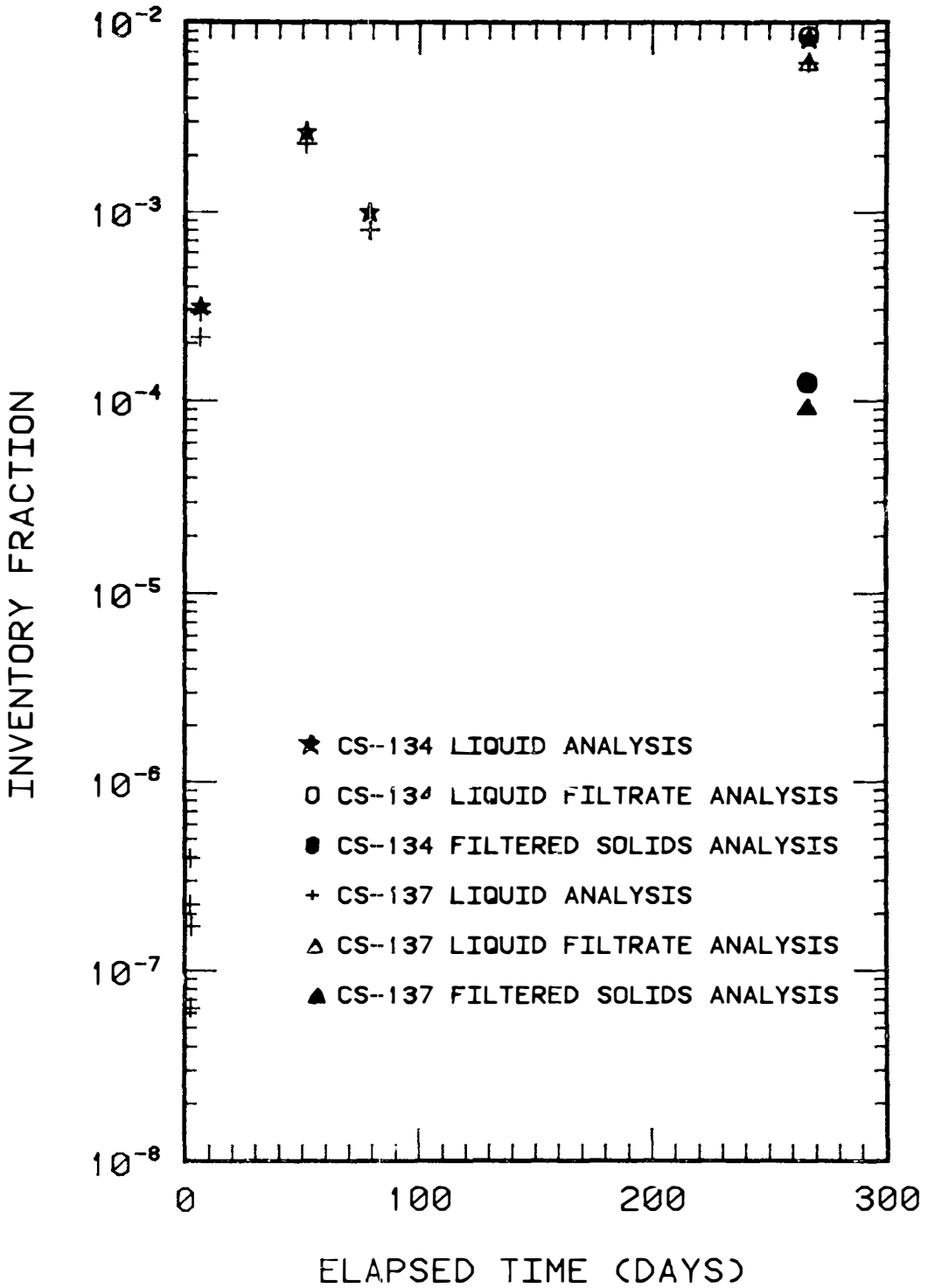


FIGURE 2-32

TMI-2 RCBT-A XE-133 INVENTORY

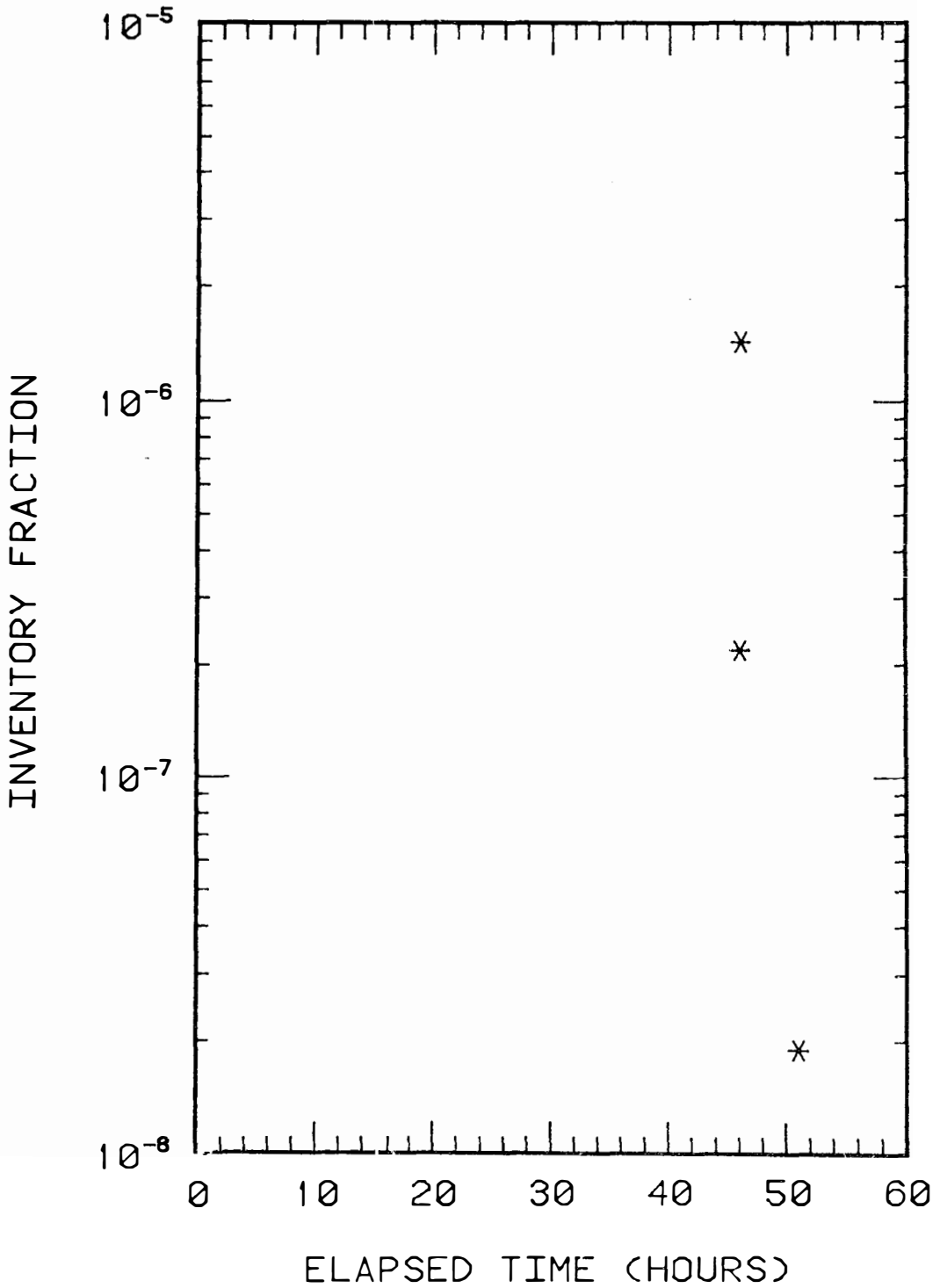


FIGURE 2-33

TMI-2 RCBT-A U-238 AND PU-239 INVENTORIES

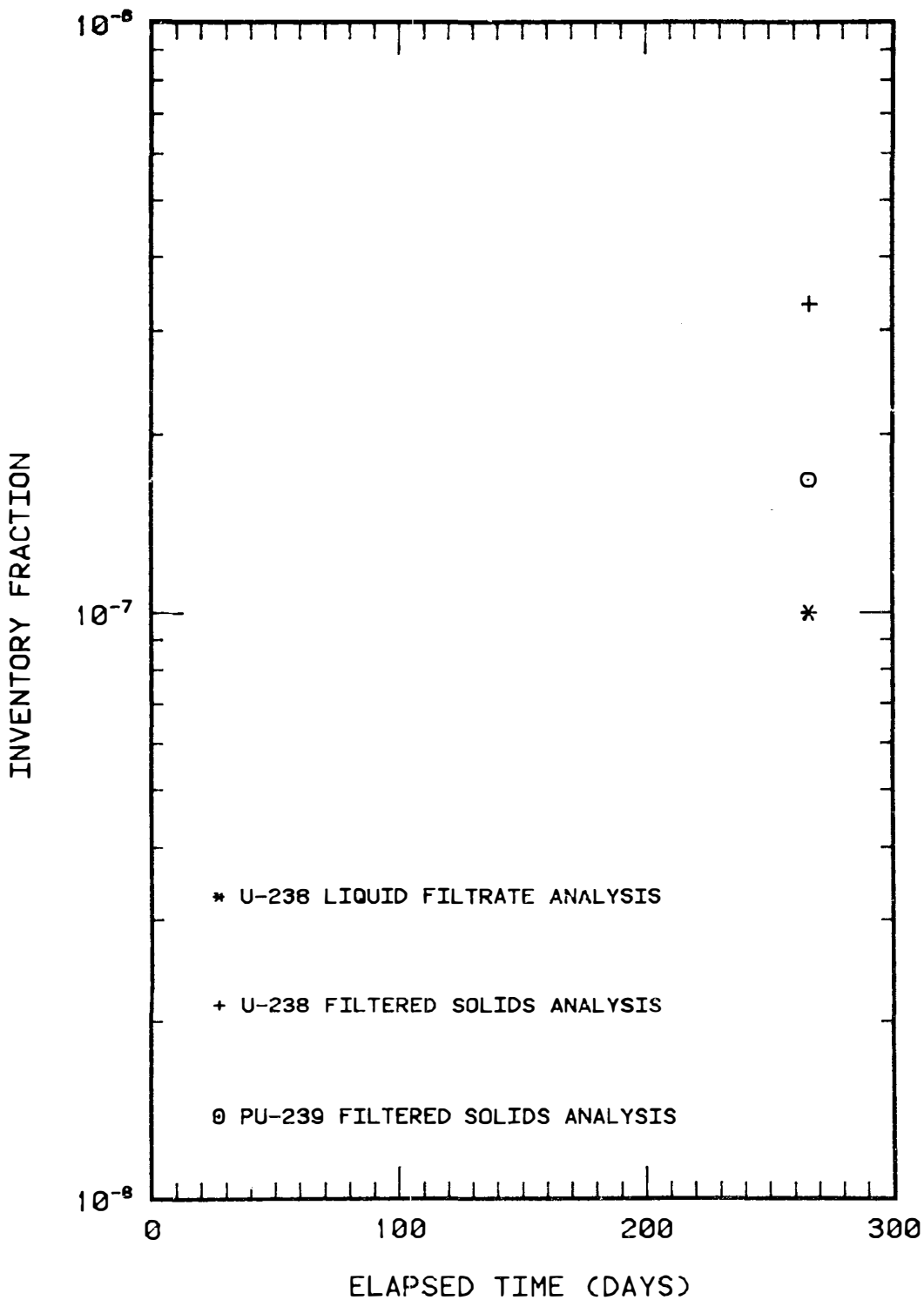


FIGURE 2-34

TMI-2 RCBT-B I-131 INVENTORY

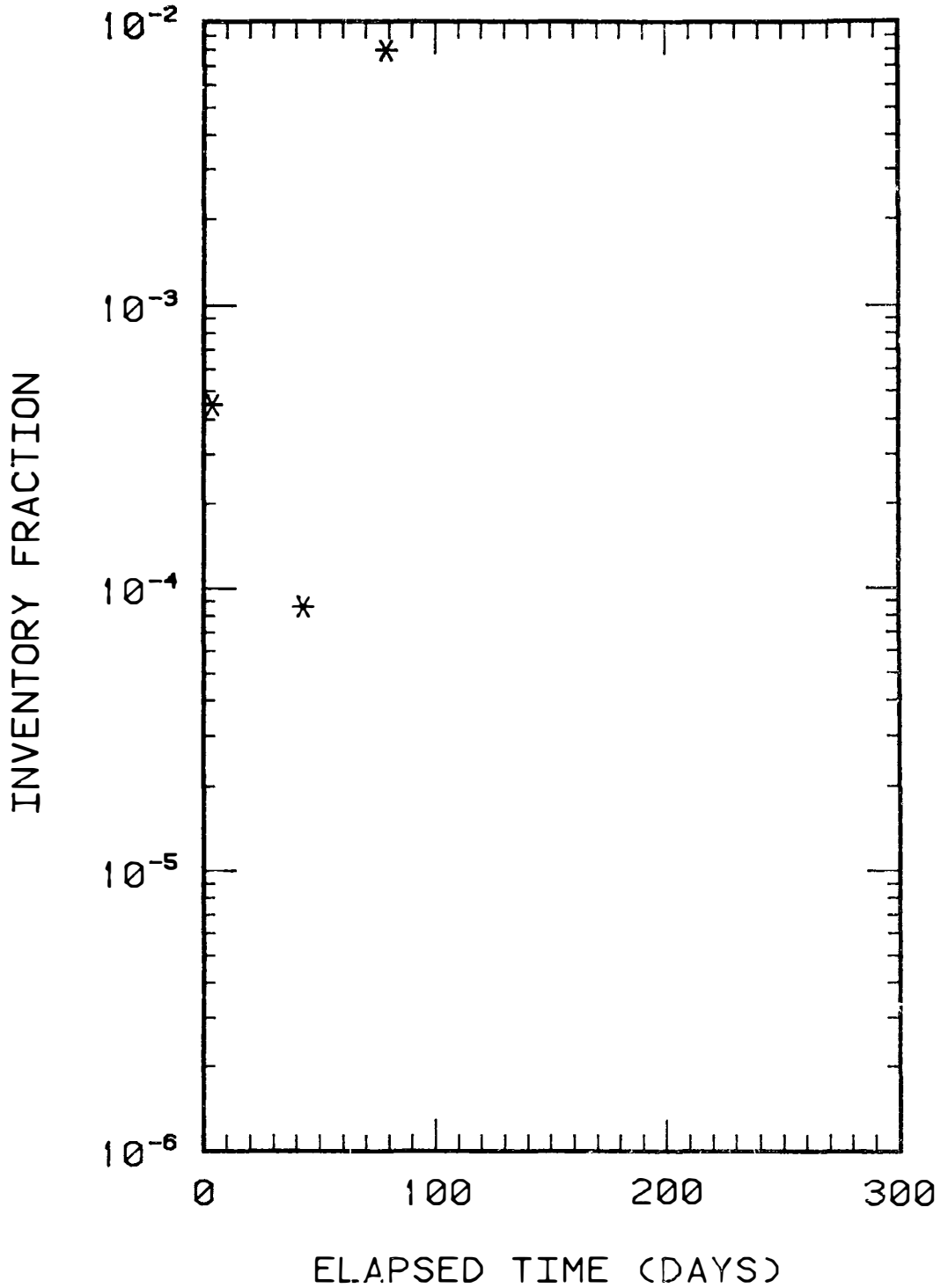


FIGURE 2-35

TMI-2 RCBT-B CESIUM INVENTORIES

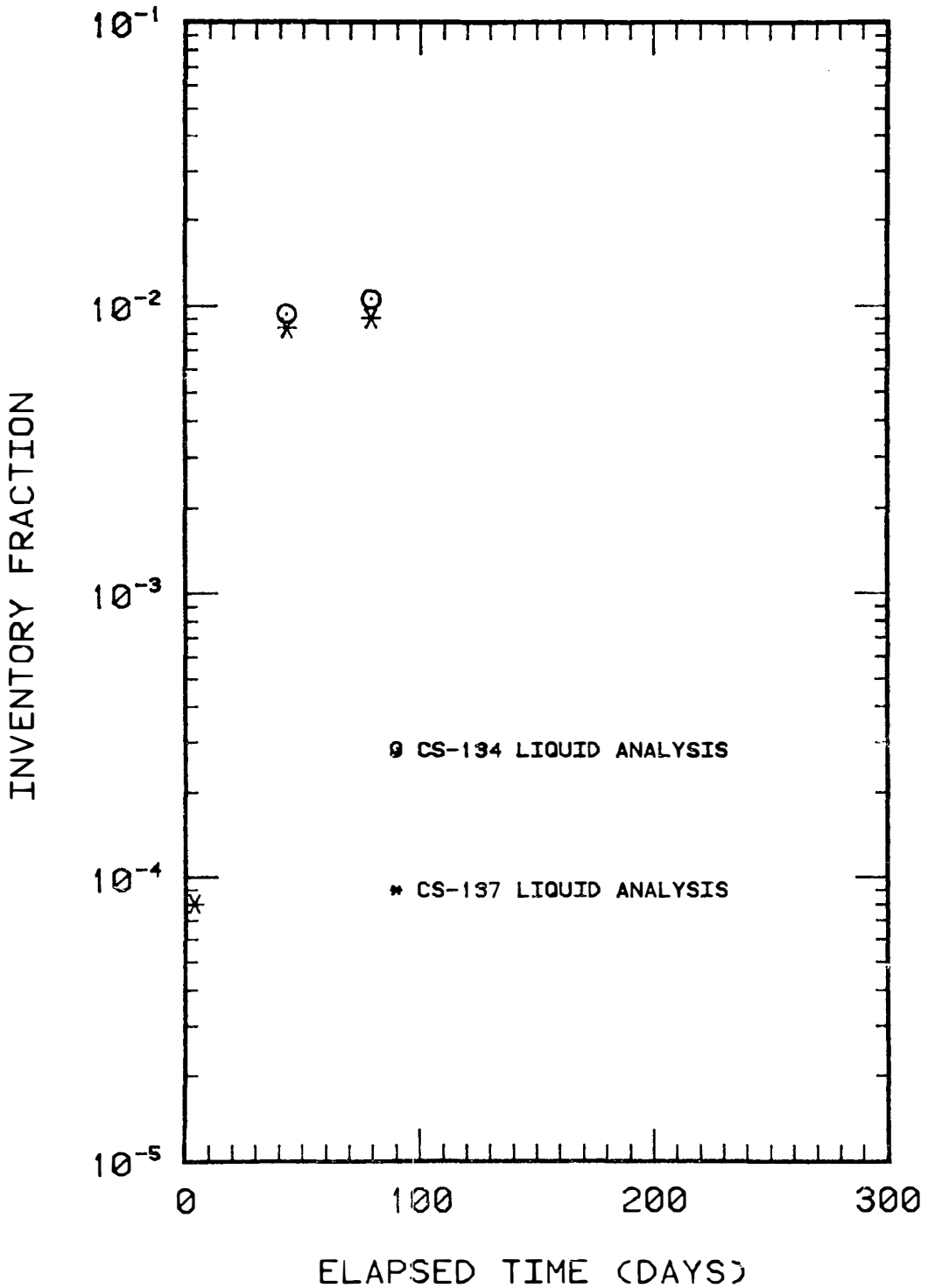




FIGURE 2-36

TMI-2 RCBT-C I-131 INVENTORY

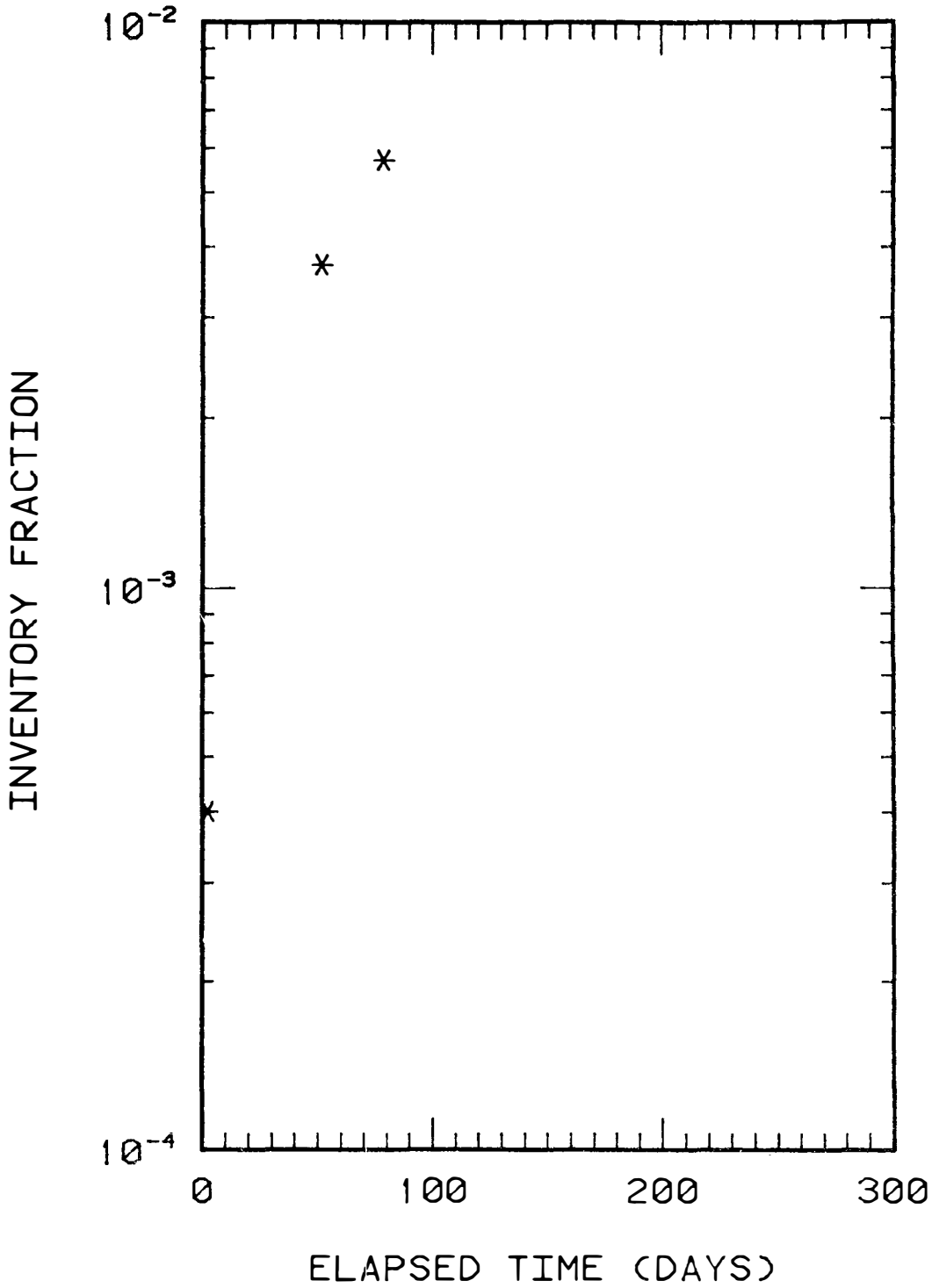
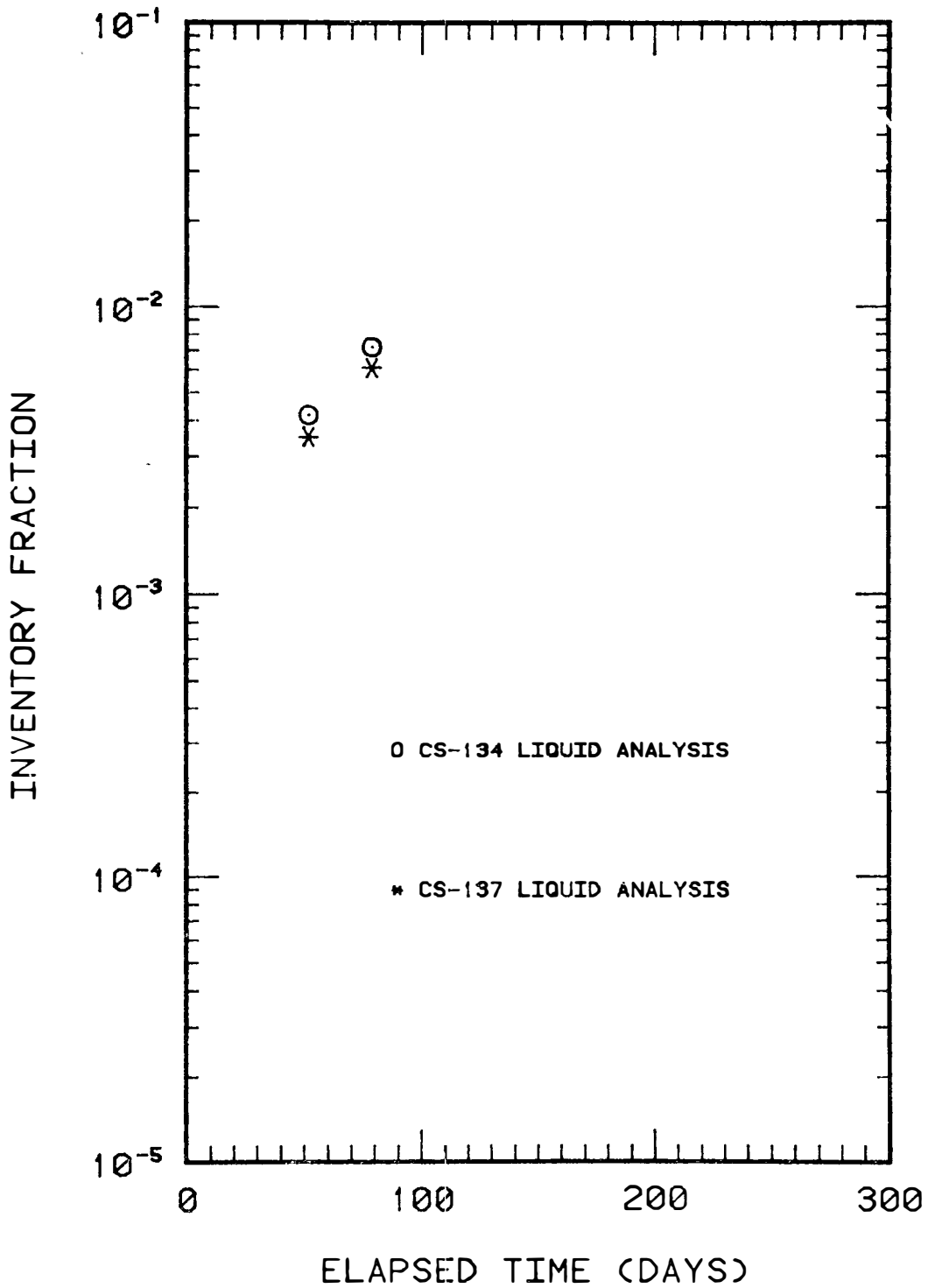


FIGURE 2-37

TMI-2 RCBT-C CESIUM INVENTORIES



was 0.235 Curies of I-131 for the period through April 1979. This represents an inventory fraction of about  $5 \times 10^{-7}$ .

## 2.4 INVENTORIES ON SURFACES AND IN SOLIDS

Following the accident at TMI-2, radionuclides were found on surfaces and in solid core debris transported to many components exposed to hot gases and liquids. In the following sections, a current assessment of such inventories is made.

### 2.4.1 Surface Inventories

Preliminary data indicate that significant inventory fractions of cesium and strontium have been found to date on the surfaces of the reactor containment building and plenum/reactor internals above the core.

Information on reactor building surface contamination has been collected<sup>(6)</sup> prior to surface decontamination. Based on mean surface concentrations and approximate surface areas, the reactor building surface isotope fractions were calculated and are presented in Table 2-4. It should be noted that data below the 305-foot level of the reactor building were not available and are not included in these estimates.

Other significant surface contamination was found on leadscrews removed from the reactor vessel for examination before head lift. In July 1982, three leadscrews were removed from their control rod guide tubes in the upper plenum assembly and reactor vessel head. The preliminary analytical results indicate that an estimated 4000 curies was deposited on stainless steel reactor vessel internals. This represents an estimated cesium inventory fraction of 0.05, as determined by the average surface activity measurement. The expected uncertainty in this number is large, based on the use of this single leadscrew data point extrapolated over the entire

Table 2-4  
 Reactor Building Surface  
 Inventory Fractions

Location & Elevation	Isotope Fraction			
	Sr-90	I-129	Cs-129	Cs-137
Floor 305 feet	$2.4 \times 10^{-6}$	$2.7 \times 10^{-5}$	$5.8 \times 10^{-5}$	$4.7 \times 10^{-5}$
Walls 305 to 345 feet 6 inches	$1.2 \times 10^{-7}$	$1.3 \times 10^{-4}$	$1.2 \times 10^{-5}$	$9.8 \times 10^{-6}$
Floor 347 feet	$4.4 \times 10^{-6}$	$5.4 \times 10^{-5}$	$4.4 \times 10^{-5}$	$3.7 \times 10^{-5}$
Walls 347 feet to Dome	$3.0 \times 10^{-7}$	$3.0 \times 10^{-7}$	$6.2 \times 10^{-6}$	$5.6 \times 10^{-6}$
Totals	<sup>a</sup> $7.2 \times 10^{-6}$	<sup>a</sup> $5.1 \times 10^{-4}$	$1.2 \times 10^{-4}$	$9.9 \times 10^{-5}$

<sup>a</sup>This value reflects incomplete data on the 347 foot level.

internals surface area; however, in the near future, with expected reactor vessel head removal, this cesium inventory will be better quantified.

#### 2.4.2 Solids Inventories

Radionuclides transferred with solid core debris by reactor coolant were deposited in the MUP system, the RCBTs, the reactor coolant drain tank, and the reactor building sump. The solids are characterized as suspensions or sludges. To date no data has been available for the reactor coolant drain tank solids, but solids in the other components have been characterized. The resulting inventory fractions are based on reported analysis and estimated quantities.<sup>(5)</sup>

The makeup and purification demineralizers cesium loadings were estimated by gamma-ray spectrometry.<sup>(5)</sup> The Cs-137 inventory fraction of demineralizer A was estimated to be 0.0044. It is estimated that the inventory fraction for both demineralizers is about 1 percent.<sup>(5)</sup> Inventory fractions have also been estimated for the MUP system filters.<sup>(5)</sup> These inventory fractions are considerably smaller, on the order of  $1 \times 10^{-6}$ . Table 2-1 shows these values since the MUP filters have been classified as minor sinks.

Table 2-5 presents the estimated solids inventory in the major sinks. The total cesium and strontium solids inventory fractions in the major sinks (besides the MUP demineralizers) are on the order of  $1 \times 10^{-3}$  and  $5 \times 10^{-4}$ , respectively.<sup>(5)</sup> The total solids inventory fractions identified to date are dominated by the MUP demineralizer inventory fractions. However, sampling and characterization of solids is subject to substantially greater uncertainty than is fluid sampling and these results must be viewed as very imprecise.

Table 2-5

Estimated Solids Inventory in Major Sinks

Component	State	Inventory Fraction		
		Sr-90	Cs-134	Cs-137
RCS	Suspended	$4 \times 10^{-6}$ to $1 \times 10^{-4}$	$2 \times 10^{-5}$ to $7 \times 10^{-4}$	$2 \times 10^{-5}$ to $4 \times 10^{-4}$
RCBTs	Suspended	$8 \times 10^{-5}$ to $1 \times 10^{-4}$	$5 \times 10^{-4}$ to $6 \times 10^{-4}$	$4 \times 10^{-4}$ to $5 \times 10^{-4}$
	Sludge	$4 \times 10^{-6}$ to $6 \times 10^{-4}$	$5 \times 10^{-6}$ to $8 \times 10^{-5}$	$5 \times 10^{-6}$ to $7 \times 10^{-5}$
RB basement	Suspended		$9 \times 10^{-8}$ to $6 \times 10^{-5}$	$9 \times 10^{-8}$ to $4 \times 10^{-5}$
subtotal		$9 \times 10^{-5}$ to $8 \times 10^{-4}$	$5 \times 10^{-4}$ to $1.4 \times 10^{-3}$	$4 \times 10^{-4}$ to $1.0 \times 10^{-3}$
MUP Demineralizers			$1.2 \times 10^{-3}$ to $2.3 \times 10^{-2}$	$1.2 \times 10^{-3}$ to $2.3 \times 10^{-2}$
Solids in major sinks		$9 \times 10^{-5}$ to $8 \times 10^{-4}$	$1.7 \times 10^{-3}$ to $3.4 \times 10^{-2}$	$1.6 \times 10^{-3}$ to $2.4 \times 10^{-2}$

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## 2.5 SUMMARY OF MASS BALANCE

A detailed accounting of the inventory fractions of major sinks and releases has been presented in the previous sections. These, combined with the inventory fractions of minor sinks presented in Table 2-1, provide the mass balance. An illustrative summary of the mass balance is presented in Table 2-6 for December 31, 1979.

Table 2-6 contains inventory data summarized for each element. The assumption is made that similar chemistry involving all isotopes of the same element is present. The total isotopic inventories are the sum of the major and minor sink inventory fractions.

The total isotopic inventories can be compared to influent inventories of the SDS and EPICOR-II liquid cleanup systems.<sup>(9)</sup> Table 2-7 contains a summary of the inventory fractions removed by SDS. Table 2-8 provides a comparison of SDS, EPICOR-II, and this study's accounted inventory fractions as presented in Table 2-6. Water processed by these cleanup systems found 66 percent of tritium, 47 to 50 percent of cesium and 2 percent of strontium. As an independent check, these cesium and strontium inventories agree well with the December 31, 1979, accounted total isotope fractions. Recognizing water recycle for cleanup and the inability to remove tritium in such cleanup, better agreement of tritium values will result from consideration of water recycle data not currently in the data base.

Uncertainties are discussed in the next chapter. The large inventory fractions of most radionuclides yet to be found indicate that future investigations into the core, the reactor vessel internals, the reactor building basement, the reactor coolant drain tank, and the RCS components are of utmost importance to closing the mass balance.

Table 2-6

Summary of Mass-Balance Inventory Fractions  
December 31, 1979

Component	Tritium	Krypton	Xenon	Strontium	Iodine	Cesium	Uranium	Plutonium
Reactor building basement	0.47			0.01	0.18	0.41	$<10^{-6}$	$<10^{-6}$
Reactor coolant system	0.018	$10^{-5}$	$<10^{-5}$	0.0098	0.014	0.018	$<10^{-4}$	$<10^{-5}$
Reactor coolant drain tank	0.002	$10^{-6}$	$<10^{-6}$	0.0009	0.001	0.002	$<10^{-5}$	$<10^{-6}$
Reactor coolant bleed tanks	0.030		$<10^{-5}$	$<10^{-3}$	0.02	0.02	$<10^{-6}$	$<10^{-6}$
Containment Air		0.473	0.28		$<10^{-3}$			
Releases to environment			0.071		$<10^{-6}$			
Makeup and Purification Demineralizers						0.012		
Reactor vessel internals/ plenum						0.05		
All major sinks	0.520	0.473	0.351	0.021	0.215	0.512	$<10^{-4}$	$<10^{-5}$
All minor sinks	0.010		$<10^{-3}$	0.001	0.006	0.002		
Total inventory fraction	0.530	0.473	0.351	0.022	0.221	0.514	$<10^{-4}$	$<10^{-5}$
Inventory fraction not found	0.470	0.527	0.649	0.978	0.779	0.486	$\sim 1.0$	$\sim 1.0$



Table 2-7

Summary of SDS Influent Inventory Fractions  
(Batch: L-45)

Component	H-3	Cs-134	Cs-137	Sr-90
RCBTs	0.0547 <sup>a</sup>	0.0046	0.0111	0.0017
RB sump	0.520 <sup>a</sup>	0.447	0.424	0.0020 <sup>a</sup>
RCS	0.0215	0.0112	0.0119	0.0191
Misc. flush	0.0212	0.0002	0.0002	8.60 x 10 <sup>6</sup>
Total	0.617 <sup>a</sup>	0.463	0.447	0.0223 <sup>a</sup>

<sup>a</sup>Data incomplete.

Table 2-8

## Summary and Comparison

Source	Inventory Fraction			
	H-3	Cs-134	Cs-137	Sr-90
EPICOR-II	0.042	0.039	0.027	0.001
SDS	0.617	0.463	0.447	0.023
EPICOR-II + SDS <sup>a</sup>	0.659	0.502	0.474	0.024
This Study <sup>b</sup> December 31, 1979	0.530	0.617	0.514	0.022

<sup>a</sup>Includes RCBTs, auxiliary sump, RCS, RB sump, and misc.

<sup>b</sup>Includes RCTSS, RCS, RB sump, RCBT, RCDT.

### 3. UNCERTAINTIES

Uncertainties in the mass balances arise from errors of two kinds: random errors associated with sampling and measurement activities, which may be expected to produce a more-or-less normal distribution about some expected value; and systematic errors, which may tend to bias measured or calculated results in a given direction, i.e., high or low. Random errors may be combined or propagated using standard statistical rules; systematic errors are not propagated in the same way.

In this study, uncertainties in the mass balances are estimated by two methods. First, the uncertainties are estimated by an analysis of random errors in the measured values that contribute to the mass balances. Second, comparisons of values determined from independent observations provide a basis for identifying possible systematic errors.

#### 3.1 ANALYSIS OF RANDOM ERRORS

The derivation of the relationships used to calculate the mass balance or total inventory fraction of an isotope at a chosen time is presented at the beginning of Chapter 2. Briefly, the mass balance is the sum over all components of the inventory fractions; the inventory fractions for each component are determined as the product of its component's volume and concentration divided by the total Three Mile Island Unit 2 (TMI-2) inventory of that component at the time of interest. The uncertainty in a mass balance due to random errors can be estimated from the uncertainty of the component inventories (as the root mean square value of the absolute uncertainties of the component inventories). The uncertainty in each component inventory can be estimated from the respective random errors in the volume, concentration, and total TMI-2 inventory (as the root mean square of the fractional or percentage uncertainties of the noted parameters). These do not, of course, consider systematic, or non-random errors.

### 3.1.1 Calculated TMI-2 System Inventories

The calculated total inventories in the data base are (as described in Appendix C, C.1.1.1) those from B&W's calculation<sup>(3)</sup> using their LOR2 code. Two other calculations are available for comparison; one is the ORIGEN computation<sup>(10)</sup> and the other is from CINDER-10.<sup>(11)</sup> Inventories of key isotopes for several decay times from ORIGEN and CINDER-10 are compared with LOR2 values in Table 3-1.

A critical comparison of these calculations has not been made; therefore, the results of these codes are not judged on the basis of systematic differences in the codes. Instead, the results are treated as independent estimates; the direct comparison of values from these codes provides at least a general idea of the variability of results from such calculations. It is noted that many of the comparable values are indeed of very similar magnitude. But there are differences. Table 3-2 presents a simple statistical treatment of the isotope inventories from the three codes (for zero time). The three values for each isotope were averaged, and the average deviations from the mean values were calculated. These average deviation values suggest that the calculations are reproducible within 10 percent for most isotopes.

Another measure of the accuracy of the calculated inventories is provided by ratios of isotopes of the same element in Table 3-3. The ratios should be unity (as the isotopes are transported independent of their atomic weight) if the calculated inventories are accurate. For the isotopes compared, that is, Cs-134:Cs-137 and Sr-89:Sr-90, identical transport of each isotope path is expected to occur so these ratios test the calculated values for the particular isotopes. The ratios suggest that the Sr-89:Sr-90 values are relatively accurate but the ratio for the cesium isotopes indicates that either the calculated inventory for Cs-137 is too high or the calculated inventory for Cs-134 is too low. Others<sup>(12,13)</sup> have suggested that the Cs-134 calculated inventory is too low.

Table 3-1  
Estimates of TMI-2 Fission Product Inventories

Source Reference <sup>a</sup> Time Isotope	CINDER	LOR-2	ORIGEN	CINDER	LOR-2	ORIGEN	CINDER	LOR-2	ORIGEN	CINDER	LOR-2	ORIGEN	CINDER	LOR-2	ORIGEN
	11 0	3 0	10 0	11 2h	3 2h	10 2h	11 50h	3 50h	10 48h	11 20.8d	3 20.1d	10 30d	11 1y	3 1y	10 1y
Inventory (Ci)															
Kr-85	b9.6(4)	9.6(4)	9.7(4)	9.6(4)	9.6(4)	9.7(4)	9.7(4)	9.7(4)	9.7(4)	9.6(4)	9.6(4)	9.7(4)	9.1(4)	9.1(4)	9.1(4)
Sr-89	6.3(7)	6.2(7)	7.3(7)	6.3(7)	6.2(7)	7.3(7)	6.1(7)	6.1(7)	6.9(7)	4.8(7)	4.8(7)	4.9(7)	4.8(5)	4.8(5)	4.9(5)
Sr-90	8.0(5)	7.8(5)	7.5(5)	8.0(5)	7.8(5)	7.5(5)	8.0(5)	7.7(5)	7.5(5)	8.0(5)	7.8(5)	7.5(5)	7.9(5)	7.6(5)	7.3(5)
Ru-106	2.9(6)	3.2(6)	3.3(6)	2.9(6)	3.2(6)	3.3(6)	2.9(6)	3.2(6)	3.3(6)	2.8(6)	3.1(6)	3.2(6)	1.5(6)	1.6(6)	1.7(6)
Te-129m	3.2(6)	3.3(6)	2.4(6)	3.2(6)	3.3(6)	2.4(6)	3.1(6)	3.2(6)	2.2(6)	2.1(6)	2.2(6)	1.4(6)	1.6(3)	1.7(3)	1.3(3)
I-129	1.7(-1)	1.7(-1)	2.1(-1)	1.7(-1)	1.8(-1)	2.1(-1)	1.7(-1)	1.8(-1)	2.1(-1)	1.8(-1)	1.8(-1)	2.1(-1)	1.9(-1)	1.9(-1)	2.2(-1)
I-131	6.4(7)	6.5(7)	7.0(7)	6.4(7)	6.5(7)	7.0(7)	5.4(7)	5.6(7)	5.1(7)	1.1(7)	1.2(7)	6.4(6)	1.4(-6)	1.5(-6)	1.5(-6)
Xe-133	1.5(8)	1.4(8)	1.5(8)	1.5(8)	1.5(8)	1.5(8)	1.3(8)	1.3(8)	1.1(8)	1.2(7)	1.4(7)	3.5(5)	3.1(-13)	3.3(-13)	2.0(-13)
Cs-134	1.3(5)	1.7(5)	1.6(5)	1.3(6)	1.7(5)	1.6(5)	1.3(5)	1.7(5)	1.6(5)	1.3(5)	1.7(5)	1.6(5)	9.6(4)	1.3(5)	1.1(5)
Cs-137	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.4(5)	8.2(5)	8.3(5)	8.2(5)
Ce-144	2.4(7)	2.3(7)	2.5(7)	2.4(7)	2.3(7)	2.5(7)	2.4(7)	2.3(7)	2.4(7)	2.3(7)	2.3(7)	2.3(7)	9.8(6)	9.6(6)	1.0(7)

<sup>a</sup>References:

11. T.R. England and W.B. Wilson, TMI-2 Fission Product Elemental and Isotope Inventories, LA-9622-MS, January 1983
3. TMI-2 Fuel History Isotopic Analysis, B&W, LOR-2, November 2, 1979.
10. R.G. Canada, NSAC ORIGEN Code Calculation of TMI-2 Fission Product Inventories, Technology for Energy Corporation, R-80-012, May 2, 1980.

b9.6(4) = 9.6 x 10<sup>4</sup>.

Table 3-2  
Variation Among Calculated Total Inventory Values

Isotope	Mean Value of Inventory at Zero Time (Ci)	Average Deviation <sup>a</sup> from Mean (%)
Kr-85	$9.6 \times 10^4$	0.4
Sr-89	$6.6 \times 10^7$	7.1
Sr-90	$7.8 \times 10^5$	2.3
Ru-106	$3.1 \times 10^6$	5.1
Te-129m	$3.0 \times 10^6$	12.8
I-129	$1.8 \times 10^{-1}$	9.8
I-131	$6.6 \times 10^7$	3.6
Xe-133	$1.5 \times 10^8$	2.7
Cs-134	$1.5 \times 10^5$	10.5
Cs-137	$8.4 \times 10^5$	0.0
Ce-144	$2.4 \times 10^7$	2.9

<sup>a</sup>Sum of the differences between each value and the mean of the three values, the sum divided by three.

Table 3-3  
Isotopic Concentration Ratios

Ratio <sup>a</sup>	Component	Number of Samples	RMS Value of Ratio <sup>b</sup>	Standard Deviation
(Cs-134/Cs-137) obs/(Cs-134/Cs-137) calc	RCS	150	1.20	0.12
"	RB Sump	14	1.19	0.39
"	WDL-T-1A <sup>c</sup>	5	1.26	0.15
"	WDL-T-1B <sup>c</sup>	3	1.71	0.62
"	WDL-T-1C <sup>c</sup>	3	1.23	0.06
(Sr-89/Sr-90) obs/(Sr-89/Sr-90) calc	RCS	87	0.95	0.40
"	RB Sump	3	1.42	0.08

<sup>a</sup>The ratio of measured concentrations of the identified isotopes divided by the ratio of the total inventories of these isotopes as calculated by the LOR-2 code for the same time as that at which the measurements were made.

<sup>b</sup>RMS refers to root mean square.

<sup>c</sup>Reactor coolant bleed tanks A, B, and C.

In summary, the cesium isotopic ratio information confirms that the calculated inventory of Cs-134 relative to that of Cs-137 is low by 20 percent. With other nuclides examined, the accuracy of the estimated inventories is indicated to be about 10 percent on the basis of comparability among code calculations.

### 3.1.2 Major Liquid Sinks

#### 3.1.2.1 Reactor Building Sump and Basement

Sampling procedures are presented in Appendix A. The procedures were well designed in the sense of assuring that sampling line volumes did not contaminate samples. Samples were taken at different elevations to detect concentration differences as a function of depth. It turned out that the differences in the water concentrations versus depth were small. Samples were not taken as a function of the other two dimensions of this large, essentially stagnant water body. Mixing would have tended to occur due to convection but would have been restricted by walls and structures. Also, only a few samples were taken over a long time span; the samples of the liquid volume were taken in August and November of 1979 and in May and September of 1981; removal of the water for decontamination through SDS began in September of 1981.

The analysis error was reported for many samples; the average errors and error ranges among those reported in the data base for reactor coolant, reactor building sump, and bleed tank samples are summarized in Table 3-4. These reported error values are used as an indication of the analysis errors generally for all the samples. As can be seen, they are generally a few percent for tritium and cesium isotopes and about 10 to 20 percent for strontium and iodine isotopes.

Table 3-5 presents information pertaining to the uncertainty in the volume of the water in the sump and basement. The data base values were extrapolated to key times at which other estimates<sup>(15,16)</sup> are



Table 3-4  
Reported Analysis Errors (Liquid Samples)

Isotope	Reactor Coolant		RB Sump		RCBTs	
	Avg. Error (%)	Error Range (%)	Avg. Error (%)	Error Range (%)	Avg. Error (%)	Error Range (%)
H-3	1.6	1.6	1.0	1		
Cs-134	1.9	0.5-8	0.6	0.5-0.7	14.3	8-20
Cs-137	2.7	0.2-17	0.9	0.6-2	22.9	7-100
Sr-89	18.1	14-20				
Sr-90	6.7	0.02-15	11.6	4-25		
I-131	12.0	12.0			9.1	4-25
I-129	5.9	4-8	21.8	13-50		

Table 3-5  
 Reactor Building Sump and Basement Water Volume

Date	Volume Added or Removed (gallons)	Total Volume (gallons)	Basis
DATA BASE			
Jan. 1, 1980	--	529,000	Data base (see Figure 2-9)
Sept. 1, 1981	114,000	643,000	Leakage at 0.13 gpm
Feb. 22, 1983	101,000	750,000	Leakage at 0.13 gpm
Feb. 22, 1983	368,000	1,122,000	Decon. water (15)
EG&G			
Sept. 1, 1981	--	641,000	(15)
ESTIMATES FROM SDS OPERATION			
Feb. 22, 1983	863,000	863,000	Removed to SDS <sup>(9)</sup>
Feb. 22, 1983	50,000	913,000	Estimated heel not pumped out

(15) T.E. Cox, et al., GEND-INF-011, Vol. III.

(9) T. Lookabill, EPICOR II and SDS Influent Sources and Concentrations, NUS-TM-349, April 1983.

available for comparison. The data base value for September 1, 1981, agrees very well with a value estimated by EG&G in cooperation with Bechtel, that is, 643,000 gallons (data base) versus 641,000 gallons (EG&G). The projection of the data base values to February 22, 1983, when most of the liquid had been processed through SDS for decontamination is based on the continuation of leakage at 0.13 gallons per minute plus a published estimate<sup>(15)</sup> of decontaminated flush water additions to the sump to yield a total for that date of 1,122,000 gallons. This value compares approximately with the total volume of the batches to SDS (of 863,000 gallons) plus a roughly estimated heel of water not removed (of 50,000 gallons) of 913,000 gallons. The heel volume is a rough estimate considering a volume of 6000 gallons per inch of depth plus the volume of the sump, incore cable chase, and elevator depression. On this basis, the projected volume from the data base is about 19 percent higher than the SDS removal volume plus heel. The latter comparison (for February 22, 1983) appears more significant than the former, which was a comparison of estimates made by two groups on the basis of essentially the same data. The SDS batch volumes and the estimated decontamination water volumes are based on measurements completely independent of the data base.

#### 3.1.2.2 Reactor Coolant

The sampling equipment and procedures are described in Appendix A. The sampling system appears to be well designed in terms of the procedural requirement to flush the sampling lines adequately before taking a sample. The system allows samples to be taken from the letdown line (which comes from the bottom of the cold leg before it feeds into coolant pump 1B) or from the bottom of the pressurizer. The reactor coolant was circulated by pumping until mid-April of 1979 and later by convection; therefore, the liquid volume was reasonably well mixed. The sampling error in this case would tend, therefore, to be smaller than the other components less well mixed.

A summary of the reported analysis errors is presented in Table 3-4. These errors are in the range of a few percent for tritium and cesium isotopes, and about 10 to 20 percent for strontium and iodine isotopes.

The volume of the reactor coolant system in the data base is 80,320 gallons. Since the maintenance of this system in a near-full condition was important to cooling requirements and safety, there is high confidence that the volume remained quite constant.

### 3.1.2.3 Reactor Coolant Bleed Tanks (RCBTs)

The sampling procedure for obtaining RCBT samples is described in Appendix A; the procedure provides adequately for flushing sample lines. These tanks were used; hence, the contained liquids would have been mixed to some extent due to pumping. The reported analysis errors are summarized in Table 3-4.

The volumes of the water in the bleed tanks varied due to pumping liquids in and out, as presented in Chapter 2. The volumes were therefore particularly uncertain due to the additive errors of many mass transfers.

### 3.1.3 Major Gaseous Sinks

Only the noble gases (represented by Kr-85 and Xe-133) were found primarily in gaseous sinks. The major sinks for these gases were the reactor building (in which the gases were held for decay and planned release in June to July 1981) and the early releases via the station vent during the period through April 1979. The measurements of the contents of these sinks are considered below.

#### 3.1.3.1 Reactor Building

The reactor building atmosphere samples were taken from a port about midway from top to bottom of the building (see Appendix A).

The gases would have tended to mix due to convection, but the maze of floors and walls would have inhibited the mixing process. Many measurements were made over a long period of time and the reproducibility of these data indicates the overall uncertainty in the values. The Kr-85 values show a narrower spread than those for Xe-133 and are therefore used to estimate the noble gas inventory. Also, the values do not appear to show any trend upward or downward with time (as would be expected since reactions with surfaces or significant dissolution into the water is not expected). The mean of the values of Kr-85 inventory in the reactor building is 0.47 and the standard deviation from this mean value is 0.18 or about 40 percent.

#### 3.1.3.2 Station Vent Releases

The station vent releases of noble gases during the early phase of the accident are very uncertain. The releases were not measured during the period of March 28 to March 31, 1979, but were estimated by scaling area monitor indications and by the use of TLD measurements coupled with dispersion calculations (see Appendix A). From March 31, grab sample and/or vent monitor data are available. The estimated release during the March 28 to March 31 period was over 80 percent of the total estimated release; hence, the uncertainty of the total release value is dominated by uncertainties in releases during the period without direct measurements. The gas release value should therefore be viewed as an order-of-magnitude estimate.

#### 3.1.3.3 Summation of Major Gaseous Sink Inventory Uncertainties

The uncertainty in the mass balance for noble gases can be indicated approximately by a skewed uncertainty limit. The lower limit is about 0.3 (i.e. as shown in Section 3.1.3.2, the reactor building value of 0.47 minus the standard deviation of about 40 percent) and the upper limit is unity.

## 3.2 DISCUSSION OF SYSTEMATIC ERRORS

The analysis of errors described above could be misleading due to undiscovered systematic errors. In order to disclose such systematic uncertainties, certain cases are discussed in which key results can be verified using independent data.

### 3.2.1 EPICOR II and SDS Data

The comparison of liquid inventories determined from component sampling and from EPICOR II and SDS data is presented in Table 3-6. The Sr-90 inventory from EPICOR II/SDS is probably low because concentrations for some of the batches were not available; hence the inventory in some batches is not included. The RCBT's estimate from EPICOR II/SDS is probably high; the total of batch volumes from the RCBTs exceeds the total capacities of the tanks, indicating that other miscellaneous liquids were also included. The volumes processed correspond to RCBT-A being about full, RCBT-B about twice full, and RCBT-C about full plus 10,000 gallons.

Another significant difference in the results is the Sr-90 inventories in the reactor coolant system (RCS): the EPICOR II/SDS value is about twice the component sample value. This could be a real difference (rather than an indication of error) due to leaching of strontium from the fuel during the batch-cleanup process in which portions of the reactor coolant were sequentially replaced by clean water.

The remaining results are the cesium values for the reactor building (RB) sump and RCS. The RB sump values agree within about 3 percent; the RCS values agree within 50 percent. Another comparison is the total values. The cesium values agree within 9 percent; the strontium values within 20 percent.

Table 3-6

Comparison of Inventories from Component Sampling and  
EPICOR II and SDS Data

Isotope	RB Sump	RCS	RCBTs	Total
EPICOR II AND SDS				
Cs-137	0.424 <sup>b</sup>	0.012	0.034 <sup>a</sup>	0.470
Sr-90	0.002 <sup>b</sup>	0.019	0.003 <sup>b</sup>	0.024
COMPONENT SAMPLES				
Cs-137	0.41	0.018	0.002	0.430
Sr-90	0.01	0.01	0.009	0.029

<sup>a</sup>Valuen may be high due to inclusion of miscellaneous liquids.

<sup>b</sup>Data incomplete (i.e., concentration not available for all batches).

In conclusion, the comparison of the EPICOR II/SDS results with component sample results disclose no gross unexplained discrepancies.

### 3.2.2 Ratios of Tritium to Other Nuclides

Tritium can be treated as a tracer for the reactor coolant as it existed soon after the accident; that is, tritium can be assumed to travel with the reactor coolant without loss in transit or storage. The summary of mass balance (Table 2-6) shows that the cesium-to-tritium ratios are reasonably constant; the values being 0.9 for the RB sump/basement, unity for the reactor coolant, and 0.7 for the RCBTs. This is a reasonable result since it would be expected that cesium would also tend to follow the reactor coolant with only limited losses (due to precipitation, plateout, or transfer to the gas phase). There appears, therefore, to be no readily apparent reason to suspect that the tritium or cesium inventories in major liquid sinks are grossly in error (e.g., due to transfer to other phases).

The iodine-to-tritium ratios are 0.4 for the sump/basement water, 0.8 for the reactor coolant, and 0.7 for the RCBT water. This could indicate that iodine precipitated or plated out on surfaces from the sump/basement water sufficiently to account for an additional 20 percent or so of the total iodine inventory. The possible sinks within the reactor building, in addition to the water, include (a) the solids and surfaces in contact with the water, (b) the gas phase, and (c) solids and surfaces in contact with the gas. Analysis of small amounts of solid material from liquid samples taken near the reactor building floor showed wide variations, considerable uncertainty, and the possibility of containing a few percent of the iodine. Surfaces below the high water mark have not been sampled. The gas phase data (presented in Chapter 2) indicate little iodine. The surfaces in contact with the gas phase have been sampled; the results are uncertain but allow the possibility of a significant amount of iodine (Section 2.4.1.). Further inves



tigations should be made to determine if such a quantity of iodine was transferred to the reactor building but not found there.

The strontium-to-tritium ratios are 0.02 for the sump/basement, 0.5 for the reactor coolant, and about 0.04 for the RCBTs. The amounts involved for strontium are smaller than for the other isotopes considered above so that poorer reproducibility of values may be expected. However, it seems clear that the ratio for the reactor coolant is much higher than the others. The time dependence of strontium in the reactor coolant shows a very definite increase with time; also the SDS data analysis indicates that strontium was continuing to appear in solution at the time of the SDS cleanup campaigns. The present conclusion is therefore that the data are all interconsistent and probably indicate a process of leaching from the fuel.

### 3.2.3 Early Samples

Retrospective extrapolation of the mass balance data to the time frame of hours and the transfer of reactor coolant through the PORV to the reactor building appears to be a reasonable and potentially useful exercise (see Chapter 4). Such a study will tend to make the first reactor coolant samples and their analyses particularly important. However, these early samples may have been taken under difficult circumstances; hence, details of the sampling procedures may have been compromised. An effort to determine any peculiarities of these earliest samples, for example by means of discussion with the those who took the samples, should be made.

## 3.3 SUMMARY OF UNCERTAINTIES

The analysis of ratios of tritium to other nuclides suggests that there may have been a large unidentified sink for iodine in the reactor building; this is a postulate that should be investigated further. This sink may have been surfaces and/or solids in the basement or surfaces at the higher elevations in the building.

Substantially more strontium is found in the reactor coolant than in other major liquid sinks; this is consistent with other data and with continual leaching of strontium from surfaces and/or solids in the primary system.

The data from the earliest reactor coolant samples are particularly important since it is of interest and importance to retrospectively extrapolate these data in studies of mass transfer, including transfer through the PORV to the reactor building basement. Therefore, it is recommended that the effort be made to estimate the reliability of these data, e.g., to determine if some aspects of the sampling procedures were compromised due to high radiation fields or other special problems.

## 4.0 CONCLUSIONS AND RECOMMENDATIONS

The results of the FY1983 work on the Three-Mile Island Unit 2 mass-balance are summarized below to provide a perspective of their significance, which is discussed to provide a background for speculation and the development of postulates. Finally, recommendations for future work are made on the basis of the results, their significance, and speculations about them.

### 4.1 SUMMARY OF RESULTS

The results are, of course, based on and limited by the data available. In general, the present data base includes information for all potentially significant sinks for radioactive material outside the reactor coolant system. The data also characterize the reactor coolant and (in a preliminary way) the surface contamination in the plenum region of the primary system.

The results are expressed in terms of inventory fractions; these values are the calculated inventories in particular phases/components (e.g., liquid in the reactor coolant system) divided by the initial core inventories. The inventories in each phase/component are calculated as the product of a concentration and volume (or mass or area); the initial core inventories are the values calculated by B&W's LOR2 code.

Inventory fractions are presented for the major and minor sinks (Table 4-1). For the minor sinks these values correspond to the times at which samples were taken and together represent less than 1 percent of the inventories of the isotopes considered (i.e., tritium cesium, iodine, strontium, and noble gases). These minor sinks include solids in the makeup and purification (MUP) system filters, and water in liquid waste (WDL) system tankage (such as the neutralizer tanks, auxiliary building sump tanks, auxiliary building sump, miscellaneous waste holdup tank, contaminated drain tanks, and piping). Reactor building surfaces (at 305 feet and

above) are included as a minor sink. Releases to the river are considered very minor.

Inventory fractions are also presented for major sinks (which are arbitrarily defined as those containing more than 1 percent of one or more of the key isotopes). Insofar as the data allowed, these inventory fractions are presented as a function of time (over the days/weeks/months time-frame for which the data pertain). The major sinks are the reactor building sump and basement water, the reactor coolant, the reactor coolant drain tank water (assuming its concentrations are similar to those in the reactor coolant), the RCBTs, reactor building atmosphere and gaseous releases to the environment (major for noble gases only), the reactor vessel plenum surfaces (for cesium based on preliminary data) and the makeup and purification (MUP) demineralizers (for cesium). These inventory fractions (for a time of about December 31, 1979 or for the times of measurement in the cases of gaseous releases, demineralizers and surfaces) are shown on Table 4-1. The inventory fractions are presented as a function of time in Chapter 2 for each of these sinks except the reactor coolant drain tanks (because that tank has not been sampled), the gaseous releases to the environment, the MUP demineralizers, and the plenum surfaces contamination. Only the reactor coolant data, which consist of many samples, show clearly defined trends. These trends are shown in Figure 4-1; the tritium, cesium and iodine inventory fractions continually decline at approximately the same rate. The strontium values increase with time.

#### 4.2 SIGNIFICANCE AND IMPLICATIONS OF RESULTS

Table 4-1 indicates that about 50 percent of the noble gases, 53 percent of the tritium, 51 percent of the cesium (i.e., Cs-137), 22 percent of the iodine, 2 percent of the strontium, and less than 0.01 percent of the uranium/plutonium have been found.

Table 4-1  
Summary of Mass Balance Results

Component	Inventory Fractions					
	Tritium	Noble Gases	Strontium	Iodine	Cesium <sup>C</sup>	U & Pu
RB Sump & Basement	0.47		0.01	0.18	0.41	$<10^{-6}$
Reactor Coolant	0.018	a $<10^{-5}$	0.01	0.014	0.018	$<10^{-4}$
RCDT	0.002	a $<10^{-6}$	0.001	0.001	0.002	$<10^{-5}$
RCBT's	0.030	b $<10^{-5}$	$<10^{-3}$	0.02	0.02	$<10^{-6}$
Containment Air		a 0.47		$<10^{-3}$		
Gaseous Releases		b 0.07		$<10^{-6}$		
Plenum Surfaces					0.05	
MUP Demineralizers					0.012	
Minor Sinks	0.010		0.001	0.006	0.002	
Total	0.53	0.5	0.02	0.22	0.51	$10^{-4}$

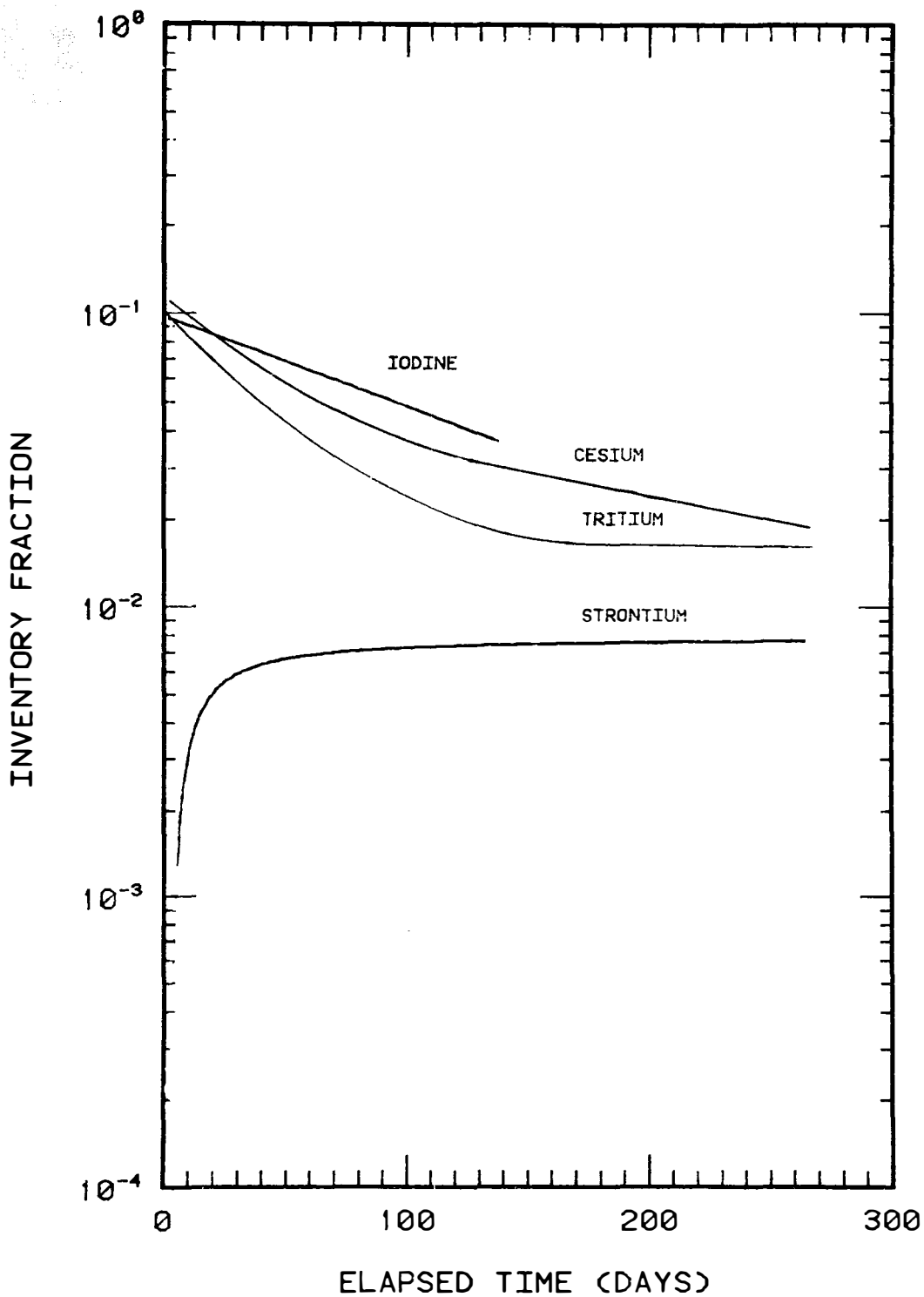
<sup>a</sup>Krypton.

<sup>b</sup>Xenon.

<sup>c</sup>Cesium-137.

FIGURE 4-1

TRENDS IN REACTOR COOLANT INVENTORIES



In principle, the noble gases mass-balance should indicate the fraction of failed fuel. However, the noble gas mass-balance is very uncertain and can only indicate a range of 30 to 100 percent fuel failure.

Tritium is potentially useful as a tracer of the reactor coolant as it existed soon after the accident. The tritium data used in this way, along with liquid mass-transfer computations, can help disclose buildup or losses of other nuclides in particular components. However, since only half of the tritium has been found, it appears that the other half remained in the reactor coolant system solids (i.e., fuel, hydrided zirconium). In order to confidently use the tritium as a tracer, it will be necessary to demonstrate that the tritium in the reactor coolant system solids/surfaces has stayed there without gradual leaching into the reactor coolant. The trend in the tritium inventory in the reactor coolant (Figure 4-1) is approximately explained by the dilution of the reactor coolant due to makeup for leakage with clean water; the correlation of the observed tritium inventories with the dilution should be quantified as well as possible to support the use of tritium as a tracer.

The iodine mass-balance (Table 4-1) appears to be low; that is, it would be surprising if indeed the 78 percent so far unaccounted for had remained in solids and/or on surfaces in the reactor coolant system.

The iodine inventories in the reactor coolant and the RCBTs are similar to those of cesium and tritium but relatively less iodine was found in the reactor building sump/basement. One hypothesis that would explain the relatively low iodine recovery is that a large amount (about half) of the iodine lost to the sump/basement may have transferred to solids or surfaces not yet sampled.

The strontium in the reactor coolant increased throughout the 300 days considered. This implies that strontium was transferring from solids or surfaces in the primary system to the liquid.

#### 4.3 RECOMMENDATIONS

##### 4.3.1 Mass Transfer Model Applications

The present data base contains nearly all of the data available to characterize the radionuclide inventories in TMI-2 except for the solids and surfaces in the reactor coolant system.

For tritium, the available data account for about half the total and the other half may remain in the reactor coolant system surfaces as hydrides and solids (relatively intact fuel pellets). It appears that these nuclides were partially released into the reactor coolant during the accident and since that time the released fraction has remained in the water and the unreleased fraction remained in solids or surfaces. This hypothesis can be substantiated by a mass transfer analysis of the reactor coolant to determine if the reactor coolant inventory versus time behavior can be explained, as suspected, by dilution with clean water used to make up for leakage.

For strontium, only a small fraction (about 2 percent) of the total is accounted for, and it is a reasonable speculation that the remainder will be found in primary surfaces and solids. The strontium inventory in the reactor coolant shows a buildup with time indicating transfer of strontium from the primary solids/surfaces to the liquid. The kinetics of this process can be clarified by an incremental analysis (similar to that described above for tritium) to distinguish the effects of leaching rate from those of dilution. Strontium leaching kinetics may also be determined by studying the behavior of strontium in reactor coolant during the batch replacement of coolant with water during the submerged demineralizer system processing campaigns. Understand



ing the strontium behavior (and possibly other slowly leached species) may aid in the core-removal strategy and post removal handling and transport.

In the case of the iodine, 22 percent of the inventory has been found but it seems reasonable to suspect that more iodine had been transferred to the reactor building basement water than was subsequently found in the basement water. Between the time of the accident and late August when the basement water was first sampled, it seems possible that some iodine may have transferred to surfaces or solids. The iodine inventory transferred to the basement can be estimated using the mass-balance model and consideration of the volumes and concentrations of reactor coolant that went to the basement.

The existing data base chronology of liquid transfers contains some anomalies that are apparent because the chronology indicates certain components to be filled beyond capacity for brief periods. These anomalies should be worked out so that components are never overfilled and the water balance among components is retained. This improvement in the data base will allow the mass transfer analyses described earlier to be done more accurately.

#### 4.3.2 Data Base and Model Improvements

The data base should be expanded to include additional isotopes (i.e., Sb-125, Te-125m, Te-127m, Ru-106, Ce-144, Ag-110m, Cd-113m, and In-114m). The antimony and tellurium isotopes are of interest because they are precursors of iodine isotopes. Ruthenium is of concern in terms of health impact; the cerium isotope is important because many of the analyses (especially for solid samples) include values for cerium. The silver, cadmium, and indium are of interest as activation products from control rod materials and are also potential getters for iodine.

There are not many data available for the isotopes noted above, as well as for some already in the data base (e.g., I-129, Te-129m, and Ru-106). A systematic study of the data available for the major liquid sinks, possible use of isotopic ratio analyses, possible stable isotope analyses, and available archived samples should be made with the objective of developing a program to disclose more about iodine and its precursors.

The General Public Utilities (GPU) data (which, for example, contains pH data) should be made accessible to the mass-balance data base by means of a tape archive of the GPU data and the software to transfer the GPU data to the mass-balance data base format.

The data base should be extended to new data from samples of solids and surfaces in the reactor coolant system. The extension should include the provision for encoding the location of samples in a way that facilitates the calculation of inventories over surface areas and volumes/masses of solids.

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The Radiochemical Concentration Module links the sample and analysis tables of the data base to the user-specified set of model components and identifies samples that could potentially represent the isotopic contents of the components at the mass-balance time. This model also affords the user with a variety of numerical tools and options for selection and calculation of representative concentration data should concentrations be unavailable at the mass-balance time. Numerical analysis options include an exponential or linear functional fit of component-specific sample concentrations to predict the concentrations of the key isotopes at the mass-balance time. The code employs regression techniques which also allow for the elimination of spurious data. This module provides additional capability independent of mass-balance calculations--the ability to determine isotopic ratios and to calculate root mean square values and standard errors within any set of user-specified data.

The final module, the Isotope Fraction Calculation Module, calculates the component inventories and the inventory fractions, that is, the fractions of the available core inventory present in the components at the mass-balance time. The module also totals the inventories and inventory fractions for the isotopes over all of the considered components.

APPENDIX A

TMI-2 MASS BALANCE: SAMPLING OF MAJOR SINKS

## A.1 REACTOR COOLANT SYSTEM AND REACTOR COOLANT BLEED TANK SAMPLING

The reactor coolant system (RCS) and reactor coolant bleed tanks (RCBTs) (as well as other tanks) were sampled by General Public Utilities (GPU) using the Nuclear Sampling System, which employed a sampling station common to and located in Three Mile Island Unit 1 (TMI-1). This system is described in the final safety analysis report (FSAR)<sup>(1)</sup> and was used until June 17, 1980. Procedure Z-95, "RCS Sampling and Pressurizer Sampling for Total Gas" was used with this system during April through September.<sup>(2)</sup> Similar procedures, R-2-79-66<sup>(3)</sup> and/or R-2-79-112<sup>(4)</sup> with the same title were applicable during the period September 1979 through June 17, 1980. On June 17, 1980, the Temporary Nuclear Sampling System became operational; this system employs the same sampling lines as the original system but uses a temporary sampling station located in Three Mile Island Unit 2 (TMI-2); this allowed TMI-2 sampling without further involvement of TMI-1. The applicable procedure for this system is Procedure 2104-4.62, "Temporary Nuclear Sampling System";<sup>(5)</sup> this procedure incorporates into one document, sampling procedures for several components, including points in the RCS, the RCBTs, and the miscellaneous waste holdup tank (MWHT). These sampling systems are shown on available drawings.<sup>(6)</sup>

The procedures and drawings were used to determine the sampling line volumes and the minimum flush volumes. Inquiries were made to determine the sample container material. The line volumes and flush volumes are shown on Table A-1; stainless steel (304L) sample bombs were used for RCS sampling prior to November 1981; after that time for RCS and for all times for the RCBTs and MWHT, 125-milliliter polyethylene or polypropylene bottles were used.

Table A-1

TMI-2 Sampling System Line Volumes and Flush Volumes

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Sample Point	Line Size (in.)	Length of Line (ft)	Line Volume (gal)	Flush Volume (gal)
Original Nuclear Sampling System (to June 17, 1980)				
RCS	3/8	881.5	2.75	30
RCBTs	1/2	223.5	1.47	30
MWHT	1/2	406.5	2.68	30
Temporary Nuclear Sampling System (after June 17, 1980)				
RCS	3/8	665.5	2.08	15
RCBTs	1/2	307.5	2.02	30
MWHT	1/2	459.5	3.03	30

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## A.2 REACTOR BUILDING BASEMENT WATER SAMPLING

### A.2.1 SAMPLES TAKEN ON AUGUST 28 AND NOVEMBER 15, 1979

The first samples were taken from the reactor building basement water by means of a tygon tube inserted into penetration 401.<sup>(7)</sup> The end of the flexible tubing was weighted and a rotary flex-type pump was used to pull liquid up the tubing, through a sample bomb, and to a large container of demineralized water. The procedure indicates that the tubing was dry on installation and was backflushed into the containment with air between samples; sampling water was drawn upward until liquid was observed in the section of tygon tubing above the sample bomb.

The procedure does not indicate how the position of the end of the tygon tubing above the floor of the reactor building basement was determined.

Results were reported by the Oak Ridge National Laboratory (ORNL)<sup>(8)</sup> for samples taken on August 28, 1979, from the "top," "middle," and "bottom" of the water volume. SAI<sup>(9)</sup> reported that the "bottom" sample was taken 3 inches from the floor and the "middle" sample was taken 60 inches from the floor. The results of the November 15, 1979, samples have also been reported.<sup>(10)</sup>

### A.2.2 SAMPLES TAKEN ON MAY 14, AND SEPTEMBER 24, 1981

These samples were taken with a specially designed thief sampler that enabled opening of previously evacuated 125-milliliter glass bottles at determined elevations under the water. The sampler is described in detail.<sup>(11)</sup> This sampler was used on May 14, 1981, by lowering it through the covered hatch on the 305-foot elevation. Samples were taken at 0, 5-3/8, 47-3/4, and 84-3/4 inches above the floor.

A similar sampler was used on September 24, 1981, through the open stairwell on the 305-foot level; only a bottom sample was taken at that time.

#### A.2.3 SAMPLES TAKEN AFTER SEPTEMBER 24, 1981

The draining of the reactor building basement water (and its decontamination via submerged demineralizer system and EPICOR II) started in September 1981; hence, later samples were taken with the water level down from the maximum. These samplings are described elsewhere.<sup>(10)</sup>

On June 23, 1982, only a thin layer of water was left on the floor; a sample was obtained using a metal scoop. Again on January 11, 1983, a specially designed suction sampler was used to obtain a sample of the material near the floor.

#### A.3 REACTOR BUILDING AIR SAMPLING

Reactor building air was sampled through the R-626 penetration located at elevation 358'6". This elevation compares with the basement floor level of 280'6" and the elevation at the top of the building of 469'10".<sup>(12)</sup> The procedure<sup>(13)</sup> required purging the lines for 5 minutes at a rate of 10 liters per minute prior to mounting a particulate filter sampler and/or a multiple cartridge sampler for iodine forms (containing layers of cadmium iodide, iodophenol, silver zeolite, and charcoal) and the withdrawal of 1 cubic centimeter samples with a hypodermic syringe or 25 cubic centimeters into an evacuated bomb sampler.

#### A.4 SAMPLING OF GASEOUS RELEASES

Prior to the accident, xenon was monitored on the station vent flow at TMI-2 with a continuous monitor that went off the scale soon after the accident started. A grab sampling program was initiated on March 31, 1979, and continued through the month

of April. The station vent monitor was again operating on April 18 and thereafter. (14)

During the early period from the time the continuous monitor went off the scale until the grab sampling program was initiated, releases of xenon were estimated by scaling of area monitor readings (which fluctuated over a factor of 3-4 from mean values) and from TLD measurements coupled with dispersion calculations. (14)

## REFERENCES FOR APPENDIX A

1. TMI-2 Final Safety Analysis Report, Section 9.3.2, "Process Sampling Systems."
2. TMI Nuclear Station Special Operating Procedure, Z-95, "RCS Sampling and Pressurizer Sampling for Total Gas," May 4, 1979.
3. TMI Nuclear Station Special Operating Procedure, R-2-79-66, "RCS Sampling and Pressurizer Sampling for Total Gas," September 14, 1979.
4. TMI Nuclear Station Special Operating Procedure, R-2-79-112, "RCS Sampling and Pressurizer Sampling for Total Gas," December 20, 1979.
5. TMI-2 Operating Procedure, 2104-4-62, Temporary Nuclear Sampling System, June 1980.
6. Drawings pertaining to the GPU Sampling Systems:
  - Burns and Roe TMI-2, Plan Elevation Drawing 2415
  - Burns and Roe TMI-2, Plan Elevation Drawing 2505
  - Burns and Roe TMI-2, Plan Elevation Drawing 2506
  - Burns and Roe TMI-2, Plan Elevation Drawing 2507
  - Burns and Roe TMI-2, Plan Elevation Drawing 2167
  - Burns and Roe TMI-2, Plan Elevation Drawing 2168
  - Burns and Roe TMI-2, Plan Elevation Drawing 2065
  - Burns and Roe TMI-2, Plan Elevation Drawing 2066
  - Gilbert Associates, TMI-1 Plan Elevation Drawing E-304-684
7. TMI Nuclear Station, Unit 2 Recovery Operating Procedure, Reactor Building Water Sampling 2104-4.17 (Rev. 1 dated October 12, 1979).

8. R.E. Brookshank and W.J. Armento, Post-Accident Cleanup of Radioactivity at the TMI Nuclear Power Station, ORNL-TM-7081, February 1980.
9. C.A. Pelleter, et al., Preliminary Radioiodine Source Term and Inventory Assessment for TMI-2, GEND-028, March 1983.
10. T.E. Cox, J.T. Horan, and G. Worku, Reactor Building Basement Radionuclide and Source Distribution Studies, GEND-INF-011, Volume III, June 1983.
11. T.E. Cox, J.T. Horan, C.V. McIsaac, Reactor Building Basement Radionuclide Distribution Studies, GEND-INF-011, Volume II, October 1982.
12. TMI-2 FSAR, Figure 6.2-33.
13. TMI Nuclear Station Unit 2 Operating Procedure 2104-4.19, Containment Air Sampling Procedure, October 12, 1979.
14. GPU-TDR-059, Assessment of Off-Site Radiation Doses from the Three Mile Island Unit 2 Accident, Keith Woodard (PL&G), July 31, 1979.

APPENDIX B

TMI-2 MASS-BALANCE: MASS TRANSFER MODEL

## B.1 DESCRIPTION OF MODEL

The concept of radionuclide mass balance is essentially one of accounting arithmetic in which the liquid or gaseous contents (volume) of system components are multiplied by their corresponding known concentrations for each of the key isotopes to obtain component inventories at chosen mass-balance times. These resultant inventories are normalized to the LOR2 total core inventory predicted for each of the key isotopes forming the inventory fractions present in system components at the mass balance time; inventory fractions are then summed over all system components as a measure of balance closure.

Based on the simplified methodology described above, a series of five interactive computer modules outlined in Figure B-1 has been developed to access the data in the Three-Mile Island Unit 2 (TMI-2) mass-balance data base and facilitate mass-balance calculations for any number of system components at any selected mass-balance time. The overall system accommodates a variety of analytic techniques including regression analysis or user-supplied input to estimate or represent concentrations/system data in the event of insufficient radiochemical concentrations or system component mass-transfer data. In addition, the modules provide special analytic capability (i.e., isotopic ratio analyses and volumetric mass balances). These capabilities are discussed in more detail below.

The five interactive modules, which are executed sequentially, are identified as the Introductory Text Module, Problem Initialization Module, Volume Computational Module, Radiochemical Concentration Module and Isotope Fraction Calculation Module.

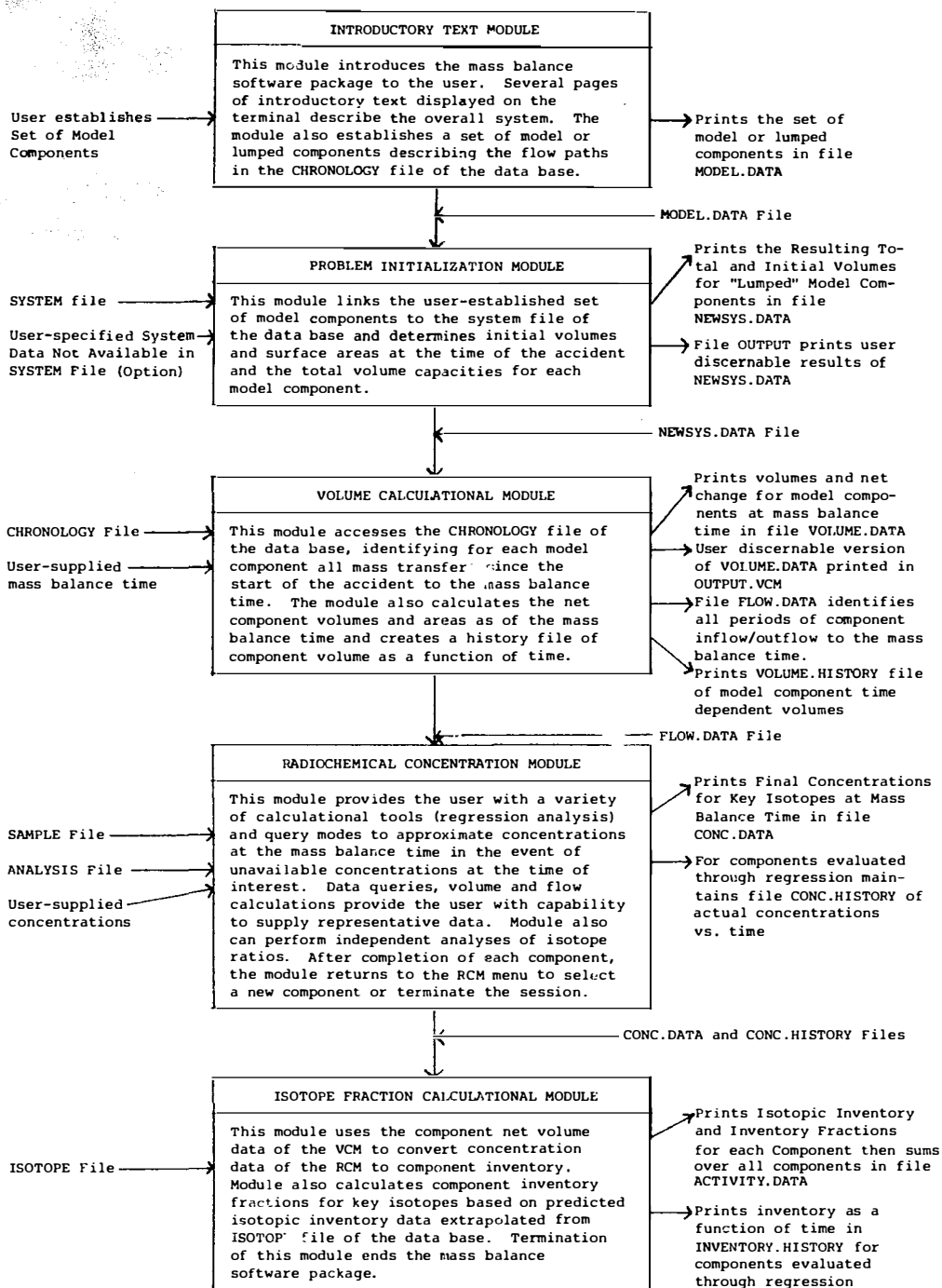


Figure B-1

TMJ-2 RADIOCHEMICAL MASS BALANCE  
COMPUTER CODE SYSTEM



The source coding was written in FORTRAN-77 and installed on the NUS Prime Computer. Verification of the modules was accomplished by comparing results of hand calculations to code calculated results for a variety of representative sample problems. A description of the modules and numerical methods employed are presented in following sections.

## B.2 INTRODUCTORY TEXT MODULE

The Introductory Text Module (ITM) is the first of five modules that support the radiochemical mass-balance calculations. Its main functions are to introduce the user to the overall system and the options it affords and to establish a set of state-specific (i.e., LIQ, GAS, etc.) model components to be evaluated along with their associated constituent components.

The liquid flowpath model presented in Chapter 4 (Figure 2-8) forms the basis for mass-balance calculations, from which a set of model components is developed. The model component can function as the overall system designator as in the reactor coolant system (RCS) with all of its constituent components (i.e., RC-P-1A) or as a lumping of like components further subdivided into constituent or specific components. This relationship between constituent and model component is demonstrated by lumping of A, B, and C reactor coolant bleed tanks (RCBTs) under model component WDL-T-1.

After the introduction to the system, the module prompts the user for all of the required input and creates an external file, MODEL.DATA, comprised of model and constituent components. Table B-1 shows the resultant set of model and constituent components for the flowpath model noted above.

Table B-1

User-Supplied Model Component Data

24326  
 17RCS                    LIQ  
 RC-T-J  
 RC-T-2  
 RC-H-1A  
 RC-H-1B  
 RCP-C-1  
 RCP-C-1A1  
 RCP-C-1A2  
 RCP-C-1B1  
 RCP-C-1B2  
 RCP-P-1  
 RCP-H-1A  
 RCP-H-1B  
 RC-P-1&2  
 RC-P-1A  
 RC-P-1B  
 RC-P-2A  
 RC-P-2B  
   3MU-C-1                LIQ  
 MU-C-J  
 MU-C-1A  
 MU-C-1B  
   1MU-J-FE              LIQ  
 MU-J-FE  
   3MU-F-5                LIQ  
 MU-F-5  
 MU-F-5A  
 MU-F-5B  
   3MU-K-1                LIQ  
 MU-K-J  
 MU-K-1A  
 MU-K-1B  
   3MU-F-2                LIQ  
 MU-F-2  
 MU-F-2A  
 MU-F-2B  
   1MU-T-1                LIQ  
 MU-T-1  
   4MU-P-1                LIQ  
 MU-P-1  
 MU-P-1A  
 MU-P-1B  
 MU-P-1C  
   1DH-T-1                LIQ  
 DH-T-1  
   1WDL-T-3                LIQ  
 WDL-T-3  
   1RB SUMP                LIQ  
 RB SUMP  
   1WDL-T-2                LIQ  
 WDL-T-2  
   1WDL-T-1A              LIQ  
 WDL-T-1A  
   1WDL-T-1B              LIQ

(Continued from Column 1)

WDL-T-1B  
   1WDL-T-1C              LIQ  
 WDL-T-1C  
   1AUX SUMP              LIQ  
 AUX SUMP  
   1WDL-T-5                LIQ  
 WDL-T-5  
   3WDL-T-8                LIQ  
 WDL-T-8  
 WDL-T-8A  
 WDL-T-8B  
   3WDL-T-11              LIQ  
 WDL-T-11  
 WDL-T-11A  
 WDL-T-11B  
   1UNIT 1                 LIQ  
 UNIT 1  
   1CON2                    GAS  
 CON2  
   1STATION VENT          GAS  
 STATION VENT  
   1ATMOSPHERE            GAS  
 ATMOSPHERE  
   5NR-P-1                 LIQ  
 NR-P-1  
 NR-P-1A  
 NR-P-1B  
 NR-P-1C  
 NR-P-1D

### B.3 PROBLEM INITIALIZATION MODULE

The second module, the Problem Initialization Module (PIM), assists the user in initializing the problem by linking the system table of the data base to the flow path model established in the previous module. For each model component, the module identifies and sums initial and design volumes and areas over all of the constituent components. Using the RCBT model component example presented in the previous section, WDL-T-1, the initial and design volumes of constituent components WDL-T-1A, WDL-T-1B, and WDL-T-1C are totaled and written to a file identified as new system data (NEWSYS.DATA) for use by subsequent modules. The Problem Initialization Module provides a list of model (lumped) components, total component volumes and areas, and the component source volumes at the time of the accident.

The module, which is executed interactively, provides the user with a list of system components for which volume or surface area data does not exist in the data base and provides the option to input such data. In the example case for the overall mass flow model, this option was declined and the list of components for which system data are lacking is provided in Table B-2. Lack of volume data for the components identified in Table B-2 is acceptable for components like RCP-C-1 which is represented as a lumped component in the chronology table by the constituent component in the system table (RCP-C-1A1, 1A2, 1B1, 1B2). Similar explanations for WDL-T-1, WDL-T-8, and WDL-T-11 apply. For the component sinks identified in Table B-2, (i.e., Unit 1, atmosphere, auxiliary floor) component volumes cannot be quantified. For those components such as NR-P-1 (river water pumps) that lack the necessary information, a mass-balance calculation cannot be performed without entry of representative data. Table B-3 presents a summary of the data assimilated by the Problem Initialization Module.

Table B-2

Component Designations Within The Model  
For Which System Data Are Lacking

THIS MODEL CONTAINS 24 COMPONENTS.  
THE SYSTEM FILE HAS 219 CARDS.

THERE IS NO MATCH IN SYSTEM DATA FOR RCP-C-1	OF RGS	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR RC-P-162	OF RGS	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-C-1	OF MU-C-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-C-1A	OF MU-C-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-C-1B	OF MU-C-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-1-FE	OF MU-1-FE	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-5	OF MU-F-5	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-5A	OF MU-F-5	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-5B	OF MU-F-5	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-K-1	OF MU-K-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-K-1A	OF MU-K-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-K-1B	OF MU-K-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-2	OF MU-F-2	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-2A	OF MU-F-2	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-F-2B	OF MU-F-2	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-P-1	OF MU-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-P-1A	OF MU-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-P-1B	OF MU-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR MU-P-1C	OF MU-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR WDL-T-8	OF WDL-T-8	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR WDL-T-11	OF WDL-T-11	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR UNIT 1	OF UNIT 1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR CON2	OF CON2	WITH STATE GAS
THERE IS NO MATCH IN SYSTEM DATA FOR STATION VENT	OF STATION VENT	WITH STATE GAS
THERE IS NO MATCH IN SYSTEM DATA FOR ATMOSPHERE	OF ATMOSPHERE	WITH STATE GAS
THERE IS NO MATCH IN SYSTEM DATA FOR NR-P-1	OF NR-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR NR-P-1A	OF NR-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR NR-P-1B	OF NR-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR NR-P-1C	OF NR-P-1	WITH STATE LIQ
THERE IS NO MATCH IN SYSTEM DATA FOR NR-P-1D	OF NR-P-1	WITH STATE LIQ

Table B-3

Total and Initial Volumes and  
Areas of Model Components

\*\*\*\*\*  
THIS IS THE NEW SYSTEM DATA FOR 24 MODEL COMPONENTS  
\*\*\*\*\*

COMPONENT	STATE	TOTAL VOLUME	TOTAL AREA	INITIAL VOLUME	INITIAL AREA
RCS	LIQ	8.86E+04	0.00E-01	8.24E+04	0.00E-01
MU-C-1	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
MU-1-FE	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
MU-F-5	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
MU-K-1	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
MU-F-2	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
MU-T-1	LIQ	4.49E+03	0.00E-01	2.28E+03	0.00E-01
MU-P-1	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
DN-T-1	LIQ	4.56E+05	0.00E-01	4.49E+05	0.00E-01
WDL-T-3	LIQ	4.86E+03	0.00E-01	6.64E+03	0.00E-01
KB SUMP	LIQ	2.09E+03	0.00E-01	8.74E+02	0.00E-01
WDL-T-2	LIQ	1.90E+04	0.00E-01	1.39E+04	0.00E-01
WDL-T-1A	LIQ	7.55E+04	0.00E-01	3.32E+04	0.00E-01
WDL-T-1B	LIQ	7.55E+04	0.00E-01	5.02E+04	0.00E-01
WDL-T-1C	LIQ	7.55E+04	0.00E-01	5.02E+04	0.00E-01
AUX SUMP	LIQ	6.37E+03	0.00E-01	4.84E+03	0.00E-01
WDL-T-5	LIQ	2.89E+03	0.00E-01	2.50E+03	0.00E-01
WDL-T-8	LIQ	1.51E+04	0.00E-01	1.68E+04	0.00E-01
WDL-T-11	LIQ	4.64E+03	0.00E-01	2.53E+03	0.00E-01
URIT 1	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01
CUN2	GAS	0.00E-01	0.00E-01	0.00E-01	0.00E-01
STATION VENT	GAS	0.00E-01	0.00E-01	0.00E-01	0.00E-01
ATMOSPHERE	GAS	0.00E-01	0.00E-01	0.00E-01	0.00E-01
NK-P-1	LIQ	0.00E-01	0.00E-01	0.00E-01	0.00E-01

#### B.4 VOLUME CALCULATIONAL MODULE

The third module, the Volume Calculation Module (VCM), supports subsequent modules of the system by linking the chronology table of the data base to the new system file of model components and identifying mass transfers from the start of the accident to the mass-balance time. The module prompts for one user-specified input parameter during execution of this module, the mass-balance time. It bounds the consideration of mass transfers on a model component by model component basis to the period of time between start of the accident and mass-balance time.

The module serves two specific functions in the determination of radionuclide mass balances. It determines the component volume balances and their net increase or decrease, for all of the model components at the mass-balance time. The results of the component volume balances are maintained in a file identified as VOLUME.DATA and are used by the last module to convert component-specific isotopic concentrations to inventories. These results are also presented in user-discernible output identified as OUTPUT.VCM (Table B-4). The negative component volumes at the MBT shown in Table B-4 indicate that further chronology development may be warranted or that these components, as in the river water, functioned as the source of inflow for other components. Also, to support a special concentration case in which component inflow occurred near the mass-balance time and concentrations are known at times other than the mass-balance time, the code identifies for each model component, all additions and withdrawals as well as their associated flow rates and the other components involved in the transfer. These data reported in file FLOW.DATA are accessed by the radiochemical module.

Table B-4

Volumes and Accumulated Volume Changes

at an Example MBT

VOLUMES ARE AT:

YEAR: 79  
 MONTH: 6  
 DAY: 28  
 HOUR: 4  
 MINUTE: 0  
 SECOND: 0

\*\*\*\*\*  
 \* TABLE OF COMPONENT VOLUMES \*  
 \*\*\*\*\*

COMPONENT	STATE	VOLUME	CHANGE IN VOLUME
RCS	LIQ	8.032E+04	-2.080E+03
MU-C-1	LIQ	6.140E+01	0.000E-01
MU-1-FE	LIQ	2.840E-02	0.000E-01
MU-F-5	LIQ	8.980E+00	0.000E-01
MU-K-1	LIQ	5.800E+02	0.000E-01
MU-F-2	LIQ	8.980E+00	0.000E-01
MU-T-1	LIQ	1.668E+03	-6.121E+02
MU-P-1	LIQ	1.460E+01	0.000E-01
DH-T-1	LIQ	1.612E+05	-2.878E+05
WDL-T-3	LIQ	7.437E+03	7.970E+02
RB SUMP	LIQ	4.943E+05	4.935E+05
WDL-T-2	LIQ	-2.334E+04	-3.724E+04
WDL-T-1A	LIQ	5.388E+04	2.068E+04
WDL-T-1B	LIQ	5.723E+04	7.028E+03
WDL-T-1C	LIQ	3.838E+04	-1.182E+04
AUX SUMP	LIQ	2.082E+04	1.598E+04
WDL-T-5	LIQ	-6.050E+04	-6.300E+04
WDL-T-8	LIQ	1.014E+04	-6.659E+03
WDL-T-11	LIQ	-2.241E+04	-2.494E+04
UNIT J	LIQ	1.008E+05	1.008E+05
CON2	GAS	2.120E+06	0.000E-01
STATION VENT	GAS	-4.057E+09	-4.057E+09
ATMOSPHERE	GAS	4.057E+09	4.057E+09
NR-P-1	LIQ	-1.822E+05	-1.822E+05

VOLUME UNITS ARE GALLONS FOR LIQUIDS AND CUBIC FEET FOR GASES

The Volume Calculational Module serves one other function independent of the radionuclide mass-balance calculations: it maintains a history file of model component change in source volume from the start of the accident to the mass-balance time.

#### B.5 RADIOCHEMICAL CONCENTRATION MODULE

The Radiochemical Concentration Module links the SAMPLE and ANALYSIS tables of the data base to the user specified set of model components and identifies samples which could potentially represent the isotopic contents of the components at the mass-balance time. This module is developed as a tool to be used in determining component-specific concentrations at the mass-balance time based on available sample and analysis data. Input to the Radiochemical Concentration Module consists of the external flow data and new system files developed by previous modules; output consists of a file list of model components and their isotopic concentrations at the mass-balance time.

This module, executed interactively, also affords the user with a variety of numerical tools and options for selection and calculation of representative concentration data should concentrations be unavailable at the mass balance time. At various times during execution, the user is presented with an option to reject representative concentrations or data and return to the menu of model components. The Radiochemical Concentration Module evaluates only one model component at a time and accommodates only one mass-balance time during an execution session.

Numerical analysis options made available to the user include an exponential or linear functional fit of component-specific



sample concentrations to predict the concentrations of the key isotopes at the mass-balance time. The code employs regression techniques, which also allow for the elimination of data points that are significantly outside of the regression line (i.e.,  $2\sigma$ ) and then the refitting of the data. Under this option, a file of the actual concentrations as a function of time is maintained by the system and allows time-dependent calculation of isotopic inventories. These methods are applicable to components like the RCS which experiences relatively little fluctuation in component volume.

In addition to the methods described above, the Radiochemical Concentration Module permits review and selection of a representative sample. Based on a code search of the flow data generated by the Volume Calculation Module, the Radiochemical Concentration Module identifies periods of component inflow, between the sample time and mass-balance time, related sources of inflow and their concentrations. If concentrations in the related components are not known, the user may enter representative concentrations. The resultant concentrations at the mass-balance time are determined according to conservation of mass and volume.

This module provides additional capability independent of mass-balance calculations, to study the relationship between key isotopes through ratio analysis. For a user-defined component and set of key isotopes (e.g., Cs-134 and Cs-137) and time period, the code determines the ratios of the sample concentrations, ratios of the predicted core inventory at corresponding concentration analysis times, and then the resultant "ratio of ratios" or isotope ratio. This is a statistical tool that permits analysis of deviation of the resultant ratio from a "true value" (i.e., unity) as a measure of data uncertainty.

## B.6 ISOTOPE FRACTION CALCULATION MODULE

The final module, the Isotope Fraction Calculation Module, calculates the component inventories and the inventory fractions, that is, the fractions of the available core inventory present in the components at the mass balance time. The module also totals the inventories and inventory fractions for all of the key isotopes over all of the considered components.

As discussed in Section B.5, for those components and isotopes that have been evaluated according to regression techniques, the module allows the user to determine the component inventories as a function of time after the accident.

The input for this module consists of the component volume balance data and concentration data as of the mass-balance time. Table B-5 presents an example output from this module.

## B.7 RADIOCHEMICAL CONCENTRATION MODULE NUMERICAL METHODS

### B.7.1. REGRESSION ANALYSIS

The Radiochemical Concentration Module program provides regression analysis capability to predict component-specific isotopic concentrations at the mass-balance time on the basis of either a linear or exponential functional fit of the data. This method, which utilizes the method of least squares to minimize the deviation in the concentration ( $y$ -direction or ordered pairs  $(x,y)$ ) for the most probable curve (linear or exponential), is particularly useful in determining concentrations at times in which there are no sample data. It is the responsibility of the user to determine the applicability of the two functional fits to existing data.

Table B-5

TMI Mass Balance Sample Results

1 TIME ACTIVITY CALCULATIONS FOLLOW FOR 24 COMPONENTS

THE CHOSEN MASS BALANCE IS AT TIME: 79/ 6/28 AT 4 0 0 HOURS.

1 ACTIVITY IN RCS

COMPONENT	STATE	ISOTOPE	COMPONENT ACTIVITY (CURIES AT MBT)	ISOTOPE ACTIVITY (CURIES AT MBT)	ISOTOPE FRACTION (AT MBT)
RCS	LIO	137-CS	3.489E+04	8.483E+05	4.057E-02
RCS	LIO	134-CS	7.468E+03	1.544E+05	4.837E-02
RCS	LIO	3-H	8.959E+01	3.806E+03	2.354E-02
RCS	LIO	89-SR	1.265E+05	1.832E+07	6.902E-03
RCS	LIO	90-SR	5.504E+03	7.723E+05	7.127E-02
RCS	LIO	131-I	1.215E+03	2.468E+04	4.940E-02

1 ACTIVITY IN RA SUMP

COMPONENT	STATE	ISOTOPE	COMPONENT ACTIVITY (CURIES AT MBT)	ISOTOPE ACTIVITY (CURIES AT MBT)	ISOTOPE FRACTION (AT MBT)
RA SUMP	LIO	137-CS	3.296E+05	8.483E+05	3.922E-01
RA SJNP	LIO	134-CS	7.995E+04	1.544E+05	5.178E-01
RA SJNP	LIO	90-SR	4.682E+03	7.723E+05	5.959E-03
RA SJNP	LIO	89-SR	1.627E+05	1.832E+07	8.880E-03
RA SJNP	LIO	131-I	4.458E+05	2.468E+04	1.809E-01
RA SJNP	LIO	3-H	1.948E+03	3.806E+03	5.119E-01
RA SJNP	LIO	131-I	4.866E+03	2.468E+04	1.978E-01

1 ACTIVITY IN JDL-T-11

COMPONENT	STATE	ISOTOPE	COMPONENT ACTIVITY (CURIES AT MBT)	ISOTOPE ACTIVITY (CURIES AT MBT)	ISOTOPE FRACTION (AT MBT)
JDL-T-11	LIO	133-I	-1.849E-03	2.468E+04	-7.515E-08
JDL-T-11	LIO	134-CS	-1.026E-02	1.544E+05	-6.647E-08
JDL-T-11	LIO	137-CS	-3.809E-02	8.483E+05	-5.961E-08

\*\*\*\*\*  
TOTAL ISOTOPE FRACTIONS  
\*\*\*\*\*

ISOTOPE      TOTAL FRACTION

3-H	5.355E-01
85-KR	8.000E-01
89-SR	1.578E-02
90-SR	1.389E-02
129-I	8.000E-01
131-I	4.281E-01
133-HE	8.000E-01
134-CS	5.662E-01
137-CS	4.328E-01
238-U	8.000E-01
239-PU	8.000E-01
106-TU	8.000E-01
144-CE	8.000E-01
129MTE	8.000E-01

### B.7.1.1 Linear Fit of Data

The necessary equations for determining a linear fit of the data,  $y = a_0 + a_1 x_1$  are as follows:

$$a = \frac{\sum y \sum (x^2) - \sum x \sum xy}{n \sum (x^2) - (\sum x)^2}$$

$$a = \frac{n \sum xy - \sum x \sum y}{n \sum (x^2) - (\sum x)^2}$$

$n$  = no. of ordered pairs  $(x, y)$ ,  $x$  = time,  $y$  = concentration

The standard deviation of the actual concentrations about the regression line can be defined as follows. This calculation is important to determine the applicability of the function to the data.

Where  $R = y_i - (a_0 + a_1 x)$ ,  $y_i$  = actual concentration.

### B.7.1.2 Exponential Fit of Data

An exponential fit of concentration data is determined through regression analysis by linearizing the exponential and calculating the above coefficients,  $a_0$ ,  $a_1$ :

$$\begin{aligned} y &= Ae^{Bx} \\ \text{where } x &= \text{time} \\ y &= \text{concentration} \end{aligned}$$

By taking the natural log (Ln) of both sides the exponential is transformed to linear form:

$$\begin{aligned} \ln y &= \ln A + Bx, \text{ or } y = a_0 + a_1 x \\ \text{where } y &= \ln Y \\ a_0 &= \ln A \\ a_1 &= 1B \end{aligned}$$

This equation has linear form and therefore all of the equations of the method of least squares apply to determine  $a_0$  and  $a_1$ . Then by substitution and taking the antilog of both sides, the following exponential fit results:

$$y = Ae^{Bx}$$

$$\ln y = \ln a_0 + e^{a_1 x}$$

$$y = e^{\ln A + Bx} = e^{a_0} e^{a_1 x} = e^{a_0 + a_1 x}$$

The standard deviation of the concentration data about the predicted exponential function is determined by:

$$\sigma = \left[ \frac{\sum (R^2)}{n - 1} \right]^{1/2}$$

$n$  = number of ordered pairs  
 $R = y_i - e^{a_0} e^{a_1 x}$   
 $y_i$  = actual concentration

### B.7.2 CONSERVATION OF MASS AND VOLUME CALCULATIONS

The Radiochemical Concentration Module option "No Special Analysis" allows the user to select a representative sample analysis from the data base, from which the isotopic contents of a component can be determined at the mass-balance time. In the event that there is inflow to the component between the mass-balance time and sample time, resultant concentrations are calculated according to conservation of volume and mass; otherwise, sample analysis concentrations need only be decay corrected to the mass balance time. The equations used to determine resultant concentrations at the mass-balance time if

inflow occurs between the sample time ( $t_s$ ) and the mass-balance time (XMBT) are:

$$C(XMBT) = \frac{C(t_s) * V(t_s) + ISIGN * \sum_{i=1}^n C_n V_n}{V(t_s) + ISIGN \sum_{i=1}^n V_n}$$

Where  $V(t_s)$  = component volume at sample time

$V_n$  = volume of inflow  $n$  between  $t_s$  and XMBT  
 $= F_n \Delta t'$

where  $F_n$  = flow rate of inflow  $n$  and

$\Delta t'$  = actual flow period

$C_n$  = Concentration of the inflow source

ISIGN = +1 if XMBT >  $t_s$   
 -1 if  $t_s$  > XMBT

As mentioned above, this method is used for all cases of component inflow for the specific case where there is both inflow and outflow the same methodology is employed by assuming a quasi-steady-state outflow.

### B.7.3. ISOTOPE RATIO ANALYSIS

Isotope ratio analysis capability was developed to assess the standard error associated with isotopic concentrations on a component by component basis. For a user-specified component, set of isotopes (e.g., C-134 and C-137), and time period after the accident, the code determines the following ratios:

1. Ratios of concentrations (samples).
2. Ratios of predicted core inventory of the set of isotopes corresponding to the analysis times at which item 1 concentrations were reported.

3. Ratio of the items 1 and 2 ratios, which if there were no errors associated with concentrations, would be equal to unity.

The code then uses statistical methods to determine the standard deviation of the resultant ratios (item 3) based on the mean value determined by the square root of the sum of the squares or on a user-supplied number, such as 1.0.

The ratio of the ratios is defined as follows:

$$R=M/C$$

where M = ratio of measured isotope concentrations

C = ratio of total amounts of the same isotopes from LOR2.

Therefore

$$M = c_1/c_2$$

$$C = a_1/a_2$$

The predicted ratio, C, is evaluated as follows:

for time <40 days, values are interpolated from the ISOTOPE table of the data base as follows:

$$C = \frac{a_{t,1} e^{-\lambda_1(t_x - t_t)}}{a_{t,2} e^{-\lambda_2(t_x - t_t)}}$$

where  $t_x$  = time of interest

$t_t$  = time corresponding to the set of values in the isotope table closest to the time of interest

$a_{t,1}$  = value in ISOTOPE table for  $t_x$  for isotope 1

$a_{t,2}$  = value in ISOTOPE table for  $t_t$  for isotope 2

$\lambda_1$  = decay constant for isotope 1

$\lambda_2$  = decay constant for isotope 2

for time > 40 days, values are extrapolated from the 40-day values in the ISOTOPE Table as follows:

$$C = \frac{a_{40,1} e^{-\lambda_1(t_x - t_{40})}}{a_{40,2} e^{-\lambda_2(t_x - t_{40})}}$$

where  $t_x$  = time of interest,  $t_{40}$  = time at 40 days

$a_{40,1}$  = value in ISOTOPE table at  $t_{40}$

$a_{40,2}$  = value in ISOTOPE table at  $t_{40}$  for isotope 2

$\lambda_1$  = decay constant for isotope 1

$\lambda_2$  = decay constant for isotope 2

The deviation from the true value of ratios is then calculated according to the following two options:

1.  $T$  = value input by user (e.g. value of unity or a value calculated from radionuclide parent-daughter and mass transfer chronologies)

2. 
$$T = \left[ \frac{1}{n} \sum (R_i - T)^2 \right]^{1/2}$$

(e.g., calculate "true value" as the root-mean-square value of a set of comparable R values)

Based on these results, data uncertainty is addressed as follows:

For a set of comparable R values



$$r_i = T - R_i \quad \text{and}$$

$$\sigma_R = \left[ \frac{1}{n-1} \sum r_i^2 \right]^{1/2}$$

$\sigma_m$  (ratio of measured concentration):

$$R = M/C$$

assuming no error in C,

$$\frac{\sigma_R}{R} = \frac{\sigma_M}{M}$$

$$\sigma_M = \frac{M}{R} \sigma_R$$

Sigma for concentration values:

$$M = \frac{C_1}{C_2}$$

$$\frac{\sigma_M}{M} = \left[ \left( \frac{\sigma_{C_1}}{C_2} \right)^2 + \left( \frac{\sigma_{C_2}}{C_2} \right)^2 \right]^{1/2}$$

assume  $\frac{\sigma_{C_1}}{C_1} = \frac{\sigma_{C_2}}{C_2} = \frac{\sigma_C}{C}$

$$\frac{\sigma_M}{M} = \sqrt{2} \frac{\sigma_C}{C}$$

$$\sigma_C = \frac{C}{\sqrt{2} M} \sigma_M = \frac{C}{\sqrt{2} R} \sigma_R$$