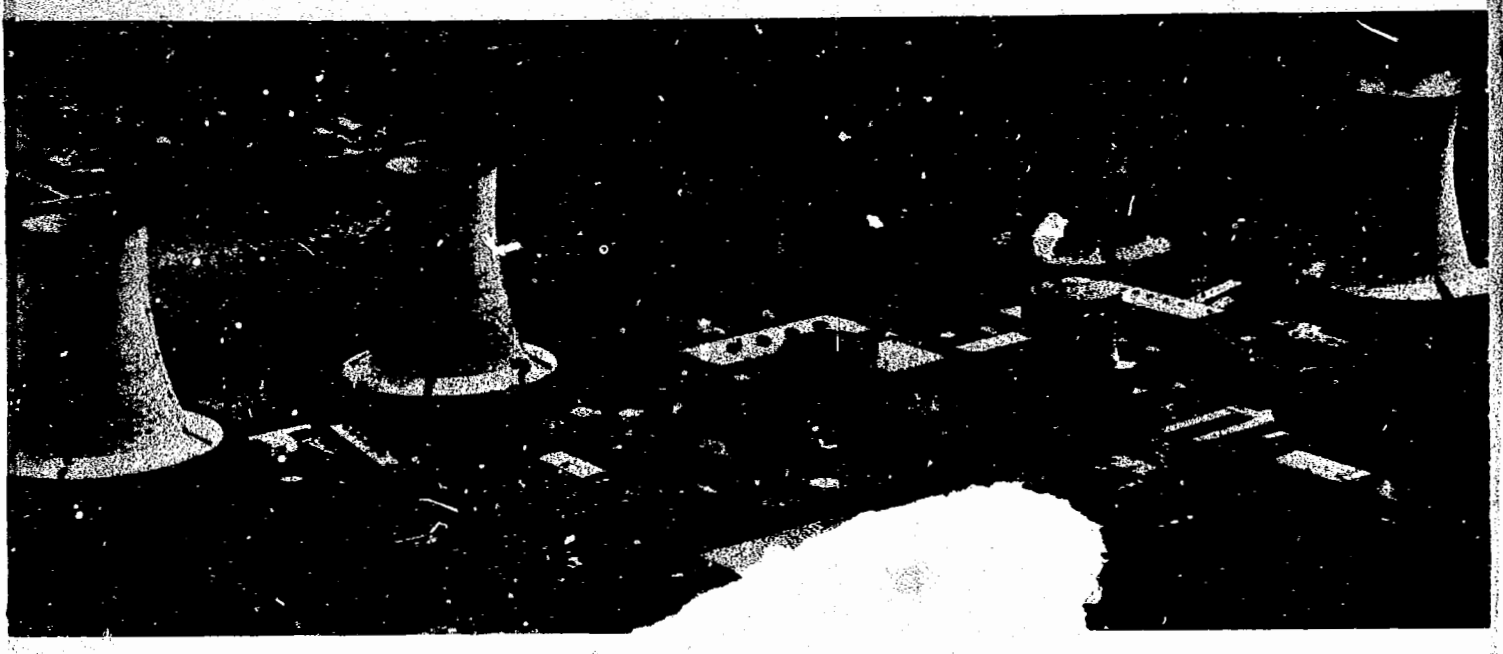

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MEASUREMENTS OF ^{129}I AND ... ...OACTIVE PARTICULATE CONCENTRATIONS IN THE TMI-2 CONTAINMENT ATMOSPHER... ...URING AND AFTER THE VENTING

J. E. Cline, et al.

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MEASUREMENTS OF ^{129}I AND RADIOACTIVE PARTICULATE CONCENTRATIONS IN THE TMI-2 CONTAINMENT ATMOSPHERE DURING AND AFTER THE VENTING

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SUMMARY

Measurements were made of the ^{129}I and ^{85}Kr concentrations in containment atmosphere during the purging of the Three Mile Island Unit 2 containment that occurred from June 28 until July 11, 1980. The measurements showed that although the airborne ^{129}I concentration was reduced by a factor of about 20 during the purge, within 15 days it had nearly returned to the prepurge value. Moreover, the initial fraction of organic iodine of 90% was reduced to about 40% during the purge and it returned to 90% in 15 days. It is believed that the equilibrium concentration and species distribution is the result of partitioning of iodine from the sump liquid and by deposition and resuspension of iodine from containment surfaces. The relative contribution to containment atmosphere from sump water and containment is not known. By comparison, the ^{85}Kr concentration decreased by a factor of 50 000 during the purge and recovered, most likely, by evolution from the sump water, to a level of 1/500 of its initial value after the main purge. Concentrations of ^{134}Cs, ^{137}Cs and ^{90}Sr were also measured in containment atmosphere.

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MEASUREMENTS OF ^{129}I AND RADIOACTIVE PARTICULATE CONCENTRATIONS IN THE TMI-2 CONTAINMENT ATMOSPHERE DURING AND AFTER THE VENTING

BACKGROUND

Previous studies of iodine at nuclear power plants suggested that, after escaping the pressure boundary, iodine was deposited and subsequently resuspended before appearing in ventilation exhaust air.[1,2,3] A mathematical model was derived to explain this behavior.[3] It was a two compartment model in which airborne iodine was allowed to deposit on building surfaces and resuspend later. The chemical form of iodine was assumed to remain unchanged. Laboratory tests were carried out to determine values for parameters in the model.[4] During these tests it was found that surface reactions with the elemental iodine formed organic and other forms of iodine. The model was modified to include the formation and resuspension of nonelemental forms of iodine and was tested with radioiodine measurements in containment atmospheres at operating PWRs.[5] One of the purposes for measuring ^{129}I prior to, during and after the purging of the TMI-2 containment atmosphere was to provide data for the model. Another purpose of the measurements was to document concentrations of other long-lived nuclides, e.g., ^{134}Cs, ^{137}Cs and ^{90}Sr.

A study[1] of the behavior of ^{131}I in the auxiliary and fuel-handling buildings at TMI, Unit 2 revealed that the most likely source of the activity in these buildings during the period April-September 1979 was evolution from the interior surfaces of the buildings. It is also most likely that the iodine deposited on the surfaces during or soon after the accident. Evaluations done[2] for the Nuclear Safety Analysis Center (NSAC) show that, relative to ^{137}Cs, about half of the ^{131}I present in the core at the shutdown cannot be accounted for in the reactor coolant, reactor building sump or atmosphere, or the auxiliary and fuel-handling buildings (liquids surfaces, filters, or releases). It is suggested in the study that the "missing" iodine was evolved either from the hot sump water or through flashing of the hot coolant as it was released into containment and that it deposited on surfaces. This continued until an equilibrium was established

between the sump, surfaces and atmosphere. The equilibrium appears to have been established rather quickly. Samples of containment atmosphere[2] taken on and following March 31, 1979 suggest that the iodine concentration decreased only with the decay half life of 8.04 days; this indicates that the activity level had reached equilibrium as early as three to six days after the accident (and subsequent isolation).

Radioiodine molecular species measurements made[6] June 26 and July 9, 1979 showed a relatively high concentration of organic ^{131}I, 86 and 79%, respectively, with the remainder equally split between the elemental and HOI forms. This iodine molecular composition is quite characteristic of iodine that has been "aged" through deposition and resuspension.

MEASUREMENTS

Samplers installed through a containment penetration sampled radioiodine and particulates from July 4 until August 12, 1980. Venting of containment was started June 28, 1980. It ended July 11 but was also done for very short times on August 1 and August 8.

Sampling System

The radioiodine and particulate sampler was installed in what is known as penetration R-626 in the Unit 2 containment building. This penetration is a 25-cm diameter pipe at the "358" level, about 3.1 m (10.5 ft) above the refueling floor. The physical arrangement is as shown in Figure 1.

The sampler pulled the containment atmosphere through a 1-cm diameter, 241-cm long stainless steel tubing that extended about 0.7 m beyond the containment wall. The tubing was connected in series to a flow measuring rotameter, the sampler and a carbon vane air pump. A glove box attached to the R-626 penetration housed the entire system. Sampling was done at a rate of 0.471 liters per second.

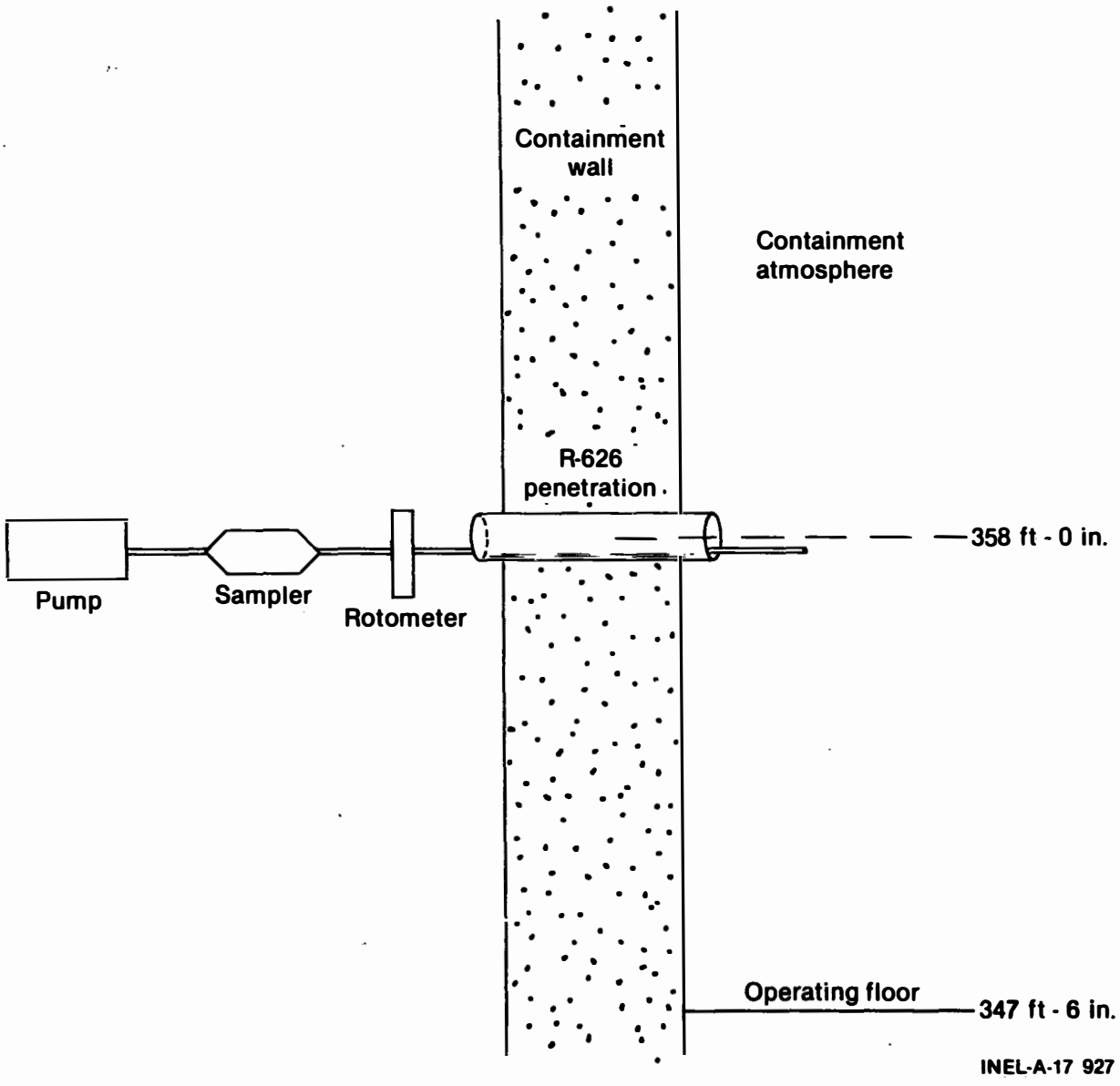

Figure 1. Location of sampler in R-626 containment penetration at TMI-2.

The iodine sampler was the SAI radioiodine molecular species sampler whose properties are described in Reference 3. This sampler has five components placed in a series: a particulate filter, a CdI_2 media, an IPH media, and two charcoal cartridges. The cartridges selectively absorb, respectively, particulate, elemental, HOI, and organic forms of iodine. The final charcoal cartridge is used to ensure no break through of iodine in the sampler.

Containment Venting

The main containment venting began June 28 and ended July 11, 1980. The initial venting used the hydrogen control system that vented from the "305'" level inside the steam generator "dee ring." The fast purge that began July 8 used the main venting lines that exhaust from the "347'" level, the refueling floor.

Venting was done cautiously with considerable starting and stopping. Table 1 gives the purge rates as a function of time for the venting. A histogram of these rates for the main venting is shown in Figure 2.

Although the main purge ended on July 11, there were additional short purges on August 1, August 8, August 14 and August 22 whose rates are shown in Table 1. A plot of the purging that includes these periods is shown in Figure 3.

Data Analysis

^{129}I Analysis

A thin window and thin crystal NaI(Tl) counted the radiation emitted in the decay of the ^{129}I activity. Each cartridge was counted nondestructively with the stream entry side toward the detector. Data were acquired using a pulse-height analyzer. In each case there was sufficient activity that this method of direct counting resulted in an analyzable spectrum of ^{129}I radiation.

TABLE 1. PURGE RATES OF TMI-2 CONTAINMENT FROM JUNE 28 TO AUGUST 23, 1980

Date	Time	Flow (cfm)	Date	Time	Flow (cfm)	Date	Time	Flow (cfm)
6/28	0800-0900	100		1700	150		0830	350
	1700-1800	15		1800	210		0845	240
	1800-2000	75		1900	210		0943	380
	2000-2100	96		2000	285		1219	565
	2100-2300	89		2100	80		1235	200
6/29	1400-1440	105		2200	150		1334	360
	1544	220		2300	220		1440-1445	0
	1600	130		2400	210		1445	215
	1700	90	7/1	0300	180		1649	460
	1800	100		0632	110		1933	180
	1900	94		0752	440		2133	470
	2000	102		0850	250		2231	230
	2100	108		0935	500		2332	430
	2200	110		1105	280	7/4	0030-0050	0
	2300	98		1132	190		0050	406
	2400	79		01-1312	0		0725	261
6/30	0100	80		1312	230		0827	485
	0200	90		1555	340		1645	400
				1725	520		1730	50
	0400	64		1925	300		2020	240
				2130	200		2120	142
	0600	100				7/4-5	2305-0319	0
	0700	130	7/2	0015-0532	0	7/5	0319	150
	0800	155		0532	258		0623	365
	0900	155		0928	490		0924	430
	1000	155		1200-1715	0		1042	570
	1100	150		1715	407		1920	250
	1200	155		1932	230		2120	332
	1300	165		2132	135		2220	165
	1400	160	7/3	0223	530		2330	340
	1500	190		0321	318	7/6	0031-0110	0
	1600	155		0			0110	150
7/6	0232	300		0909	2200			
	0555	540		0934	4000			
	0623	238		1055	4500			
	0723	540		1138	2900			
	2040	400		1250	4200			

TABLE 1. (Continued)

Date	Time	Flow (cfm)	Date	Time	Flow (cfm)	Date	Time	Flow (cfm)
7/7	0030-0105	0		1330	4350			
	0105	170		1442	2100			
				1755	1500			
	0125	89		1846	18500			
	0225	354	7/10	0148-1531	0			
	0425	445		1531	19000			
	0445	300		1634	18500			
	0525	153		2045	13000			
	0721	321		2145	16250			
	0810	540		2235	18500			
	2041	400	7/11	0100-0118	0			
	2133	260		0118	18500			
	2320	560		0933	0			
7/8	0037-0052	0	8/1	1300	0			
	0052	560		1400	7600			
	0622-1200	0		1520	0			
	1200-1300	1000	8/8	0900	4000			
	1600-1700	~1500		1000	8000			
	1800	1200		1100	9600			
	1850	500		1200	9800			
	1850	1750		1230	0			
	1900-2000	1000	8/14	1000	8000			
7/8-9	2000-0058	0						
	0058	560		1100-1500	9000			
	0430-0444	0		1600-2100	0			
	0444	1000		2100-2300	20000			
	0520	1800	8/22	1000	5000			
	0620-0700	0		1100	20000			
	0700	1000		1700	0			
	0730	1600						

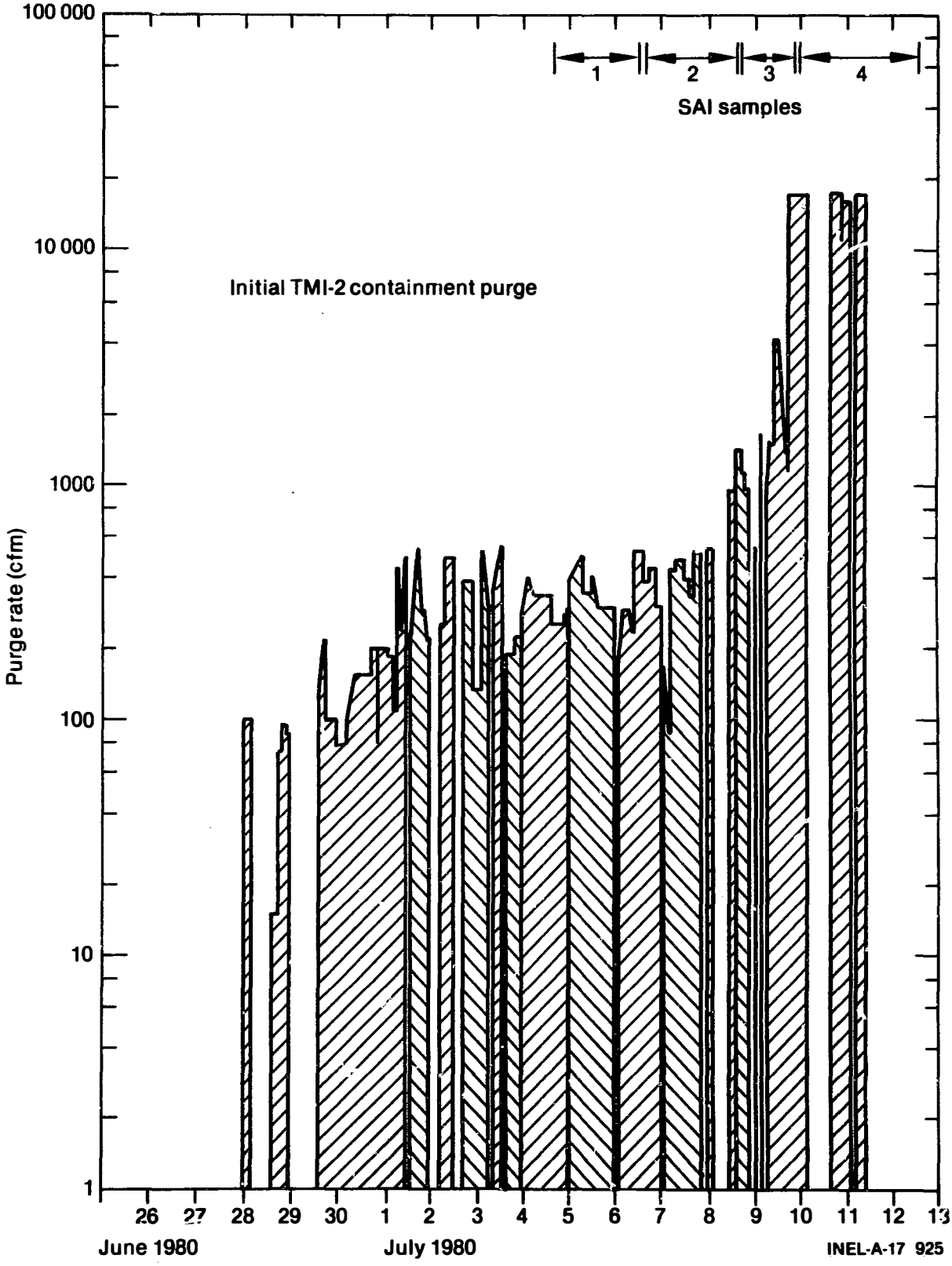

Figure 2. Histogram of TMI-2 containment purge rates during the period June 28 to July 12, 1980.

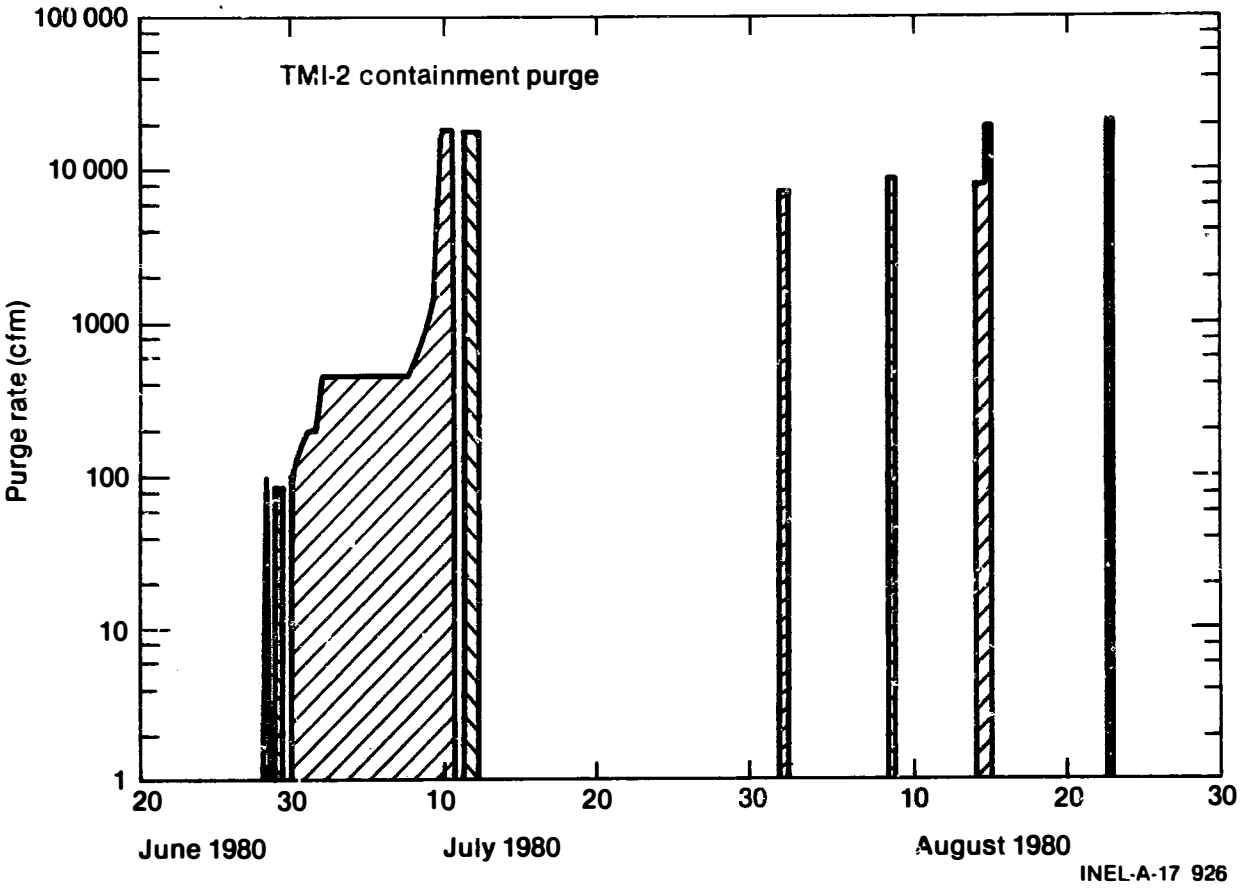

Figure 3. Histogram of TMI-2 containment purge rates during the period June 28 to August 23, 1980.

Efficiency calibration relied on the known intensities of the x-ray and 662-keV transitions from ^{137}Cs. The charcoal was removed from an unused cartridge, wetted with a solution containing about 1 μCi of ^{137}Cs, dried, mixed thoroughly, and loaded into a plastic cup. The filled cup was capped and sealed. This cup was analyzed for total Cs activity with a calibrated Ge(Li) detector (using the 662-keV gamma ray). We then calculated the emission rate of the K x-ray from the sample. Next, the sample in the cup was counted in a reproducible geometry using the thin window NaI(Tl) detector and the x-ray counting efficiency for this geometry was determined to be 1.90%. Three cartridges that had been counted nondestructively were taken apart and the charcoal mixed thoroughly and loaded in plastic cups identical to those for the ^{137}Cs secondary standard. These cups were then counted on the thin window NaI(Tl). The efficiency for the nondestructive geometry was thus determined and used for all of the analyses.

The principal assumption in the use of this calibration number was that the distribution of ^{129}I through the cartridges was the same for all cartridges. Roughly, the same volume of air was pulled through each cartridge in the sampling. The cartridges have shown reproducible activity distributions in past analyses. Hence, the assumption is considered as valid at least within the other sampling uncertainties.

Particulate Analysis

Analysis of the particulate filters for gamma-ray emitting nuclides used Ge(Li) spectrometers. The analysis revealed the presence of only the two cesium nuclides ^{134}Cs and ^{137}Cs. The filters were also analyzed nondestructively for ^{90}Sr activity by beta pulse-height spectrometry, and destructively for both ^{89}Sr and ^{90}Sr using the standard radiostrontium procedures at our laboratory.

Results

Table 2 gives the analysis results in terms of microcuries of ^{129}I per cubic centimeter of containment air for each sampling period. The table lists the values obtained for each component of the sampler as well as the

TABLE 2. ^{129}I CONCENTRATIONS FOR TMI-2 CONTAINMENT DURING VENTINGS. SAMPLES TAKEN FROM R-626 PENETRATION

Period (1980)	Concentrations ($\mu i/cm^3$)				Percentage of Total on Each Cartridge		
	Cadmium Iodide	Iodophenol	Charcoal	Total	CdI^2	IPH	Charcoal
7/4-7/6	7.69(-13)	5.40(-12)	2.14(-11)	2.76(-11)	2.1	19.1	78.8
7/6-7/8	5.69(-13)	2.99(-12)	1.10(-11)	1.46(-11)	3.9	20.6	75.5
7/8-7/9	3.39(-13)	1.60(-12)	2.85(-12)	4.80(-12)	7.1	33.3	59.5
7/10-7/12	2.89(-13)	7.84(-13)	7.43(-13)	1.82(-12)	15.9	43.2	40.9
7/12-7/15	4.55(-13)	2.56(-12)	4.18(-12)	7.21(-12)	6.3	35.7	58.0
7/15-7/18	5.69(-13)	3.19(-12)	1.20(-11)	1.58(-11)	3.6	20.2	76.2
7/21-7/22	1.18(-12)	3.09(-12)	2.30(-11)	2.72(-11)	4.3	11.4	84.3
7/23	1.27(-12)	2.94(-12)	2.25(-11)	2.67(-11)	4.7	11.0	84.3
7/25-7/26	1.24(-12)	2.56(-12)	2.53(-11)	2.91(-11)	4.2	8.8	87.0
7/30	1.12(-12)	3.25(-12)	3.99(-11)	4.44(-11)	2.5	7.3	90.2
8/2-8/3	9.28(-13)	2.06(-12)	2.82(-11)	3.12(-11)	3.0	6.6	90.4
8/5-8/6	7.91(-13)	3.01(-12)	2.95(-11)	3.33(-11)	2.4	9.0	88.6
8/7-8/8	9.87(-13)	2.58(-12)	3.51(-11)	3.88(-11)	2.6	6.7	90.8
8/9-8/10	7.47(-13)	2.62(-12)	2.10(-11)	2.44(-11)	3.1	10.8	86.2
8/12	5.10(-13)	2.53(-12)	2.72(-11)	3.03(-11)	1.7	8.3	90.0

total concentration. The table also contains columns that list the percentage of the total activity on each cartridge. Figure 4 shows a plot of the ^{129}I concentrations as a function of the sampling date.

Table 3 lists the measured concentrations of ^{134}Cs, ^{137}Cs and ^{90}Sr. Activities of ^{89}Sr were found to be below the minimum detectable limit. Figure 5 shows a graph of these concentrations.

For comparisons, Table 4 gives concentrations of ^{85}Kr in samples of containment air during the same time period as the ^{129}I sampling program. These data were obtained from the TMI staff The table gives the location where the sample was pulled. There were usually two samples taken. The results are given for both samples. Figure 6 shows a plot of the ^{85}Kr concentrations as a function of the sampling date.

DISCUSSION

The concentration of ^{129}I in the TMI-2 containment atmosphere before the purge was measured[7] to be about $7.5 \pm 2.0 \times 10^{-11}$ $\mu Ci/cm^3$. This value is comparable to the expected value of 6.7×10^{-11} $\mu Ci/cm^3$ that is based on earlier measurements of ^{131}I and relative isotope inventories of the iodine at the time of the accident. This agreement suggests that equilibrium of airborne iodine with the sump and building surfaces has been continuous since early in the accident.

Containment venting reduced the ^{85}Kr concentration by about a factor of 50 000. When the main purge was terminated, the activity rose nearly a factor of 100 to what appeared to be a new equilibrium level. The recovery was most probably the result of ^{85}Kr coming out of the sump water until a new equilibrium concentration was established. Ninety percent recovery was in about 17 days. Subsequent short purges continued to reduce the equilibrium concentrations of ^{85}Kr.

Iodine activity levels showed similar but significantly different behavior during and after the main venting. It is unfortunate that our measurements of ^{129}I did not begin at or before the venting began. The

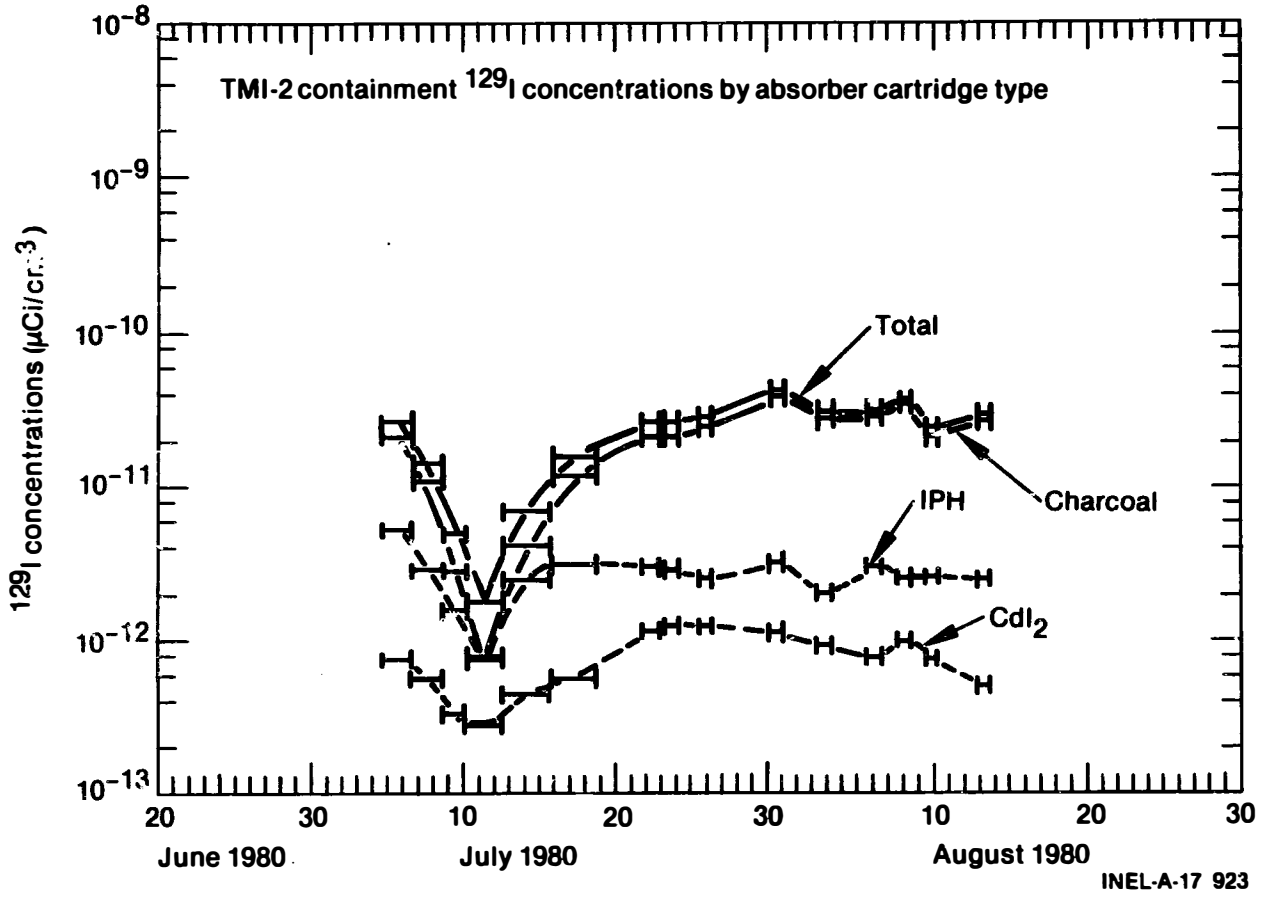

Figure 4. Plots of the ^{129}I concentrations as a function of time during the venting of the TMI-2 containment. The different curves show the concentrations of the different molecular forms of airborne iodine.

TABLE 3. PARTICULATE CONCENTRATIONS FOR TMI-2 CONTAINMENT DURING VENTINGS. SAMPLES TAKEN FROM R-626 PENETRATION
(Values in $\mu Ci/cm^3$)

Period (1980)	^{134}Cs	^{137}Cs	^{90}Sr	^{89}Sr
7/4-7/6	5.87(-12)	4.05(-11)	7.0(-12)	
7/6-7/8	2.84(-12)	1.80(-11)	6.4(-12)	
7/8-7/9	1.74(-11)	1.21(-10)	2.6(-11)	<1.1(-12)
7/10-7/12	1.95(-11)	1.40(-10)	4.67(-11)	<1.6(-12)
7/12-7/15	2.80(-11)	2.02(-10)	6.5(-11)	
7/15-7/18	2.46(-11)	1.78(-10)	7.94(-11)	
7/21-7/22	9.80(-11)	7.30(-10)	2.5(-10)	
7/23	2.76(-11)	1.96(-10)	7.4(-11)	
7/25-7/26	5.22(-11)	3.75(-10)	1.32(-10)	
7/30	1.096(-11)	7.62(-11)	2.6(-11)	
8/2-8/3	1.09(-11)	7.75(-11)	2.8(-11)	
8/5-8/6	8.40(-12)	5.89(-11)	2.12(-11)	
8/7-8/8	1.314(-11)	9.74(-11)	3.4(-11)	
8/9-8/10	1.507(-11)	1.123(-10)	3.8(-11)	
8/12	2.03(-11)	1.49(-10)	5.3(-11)	

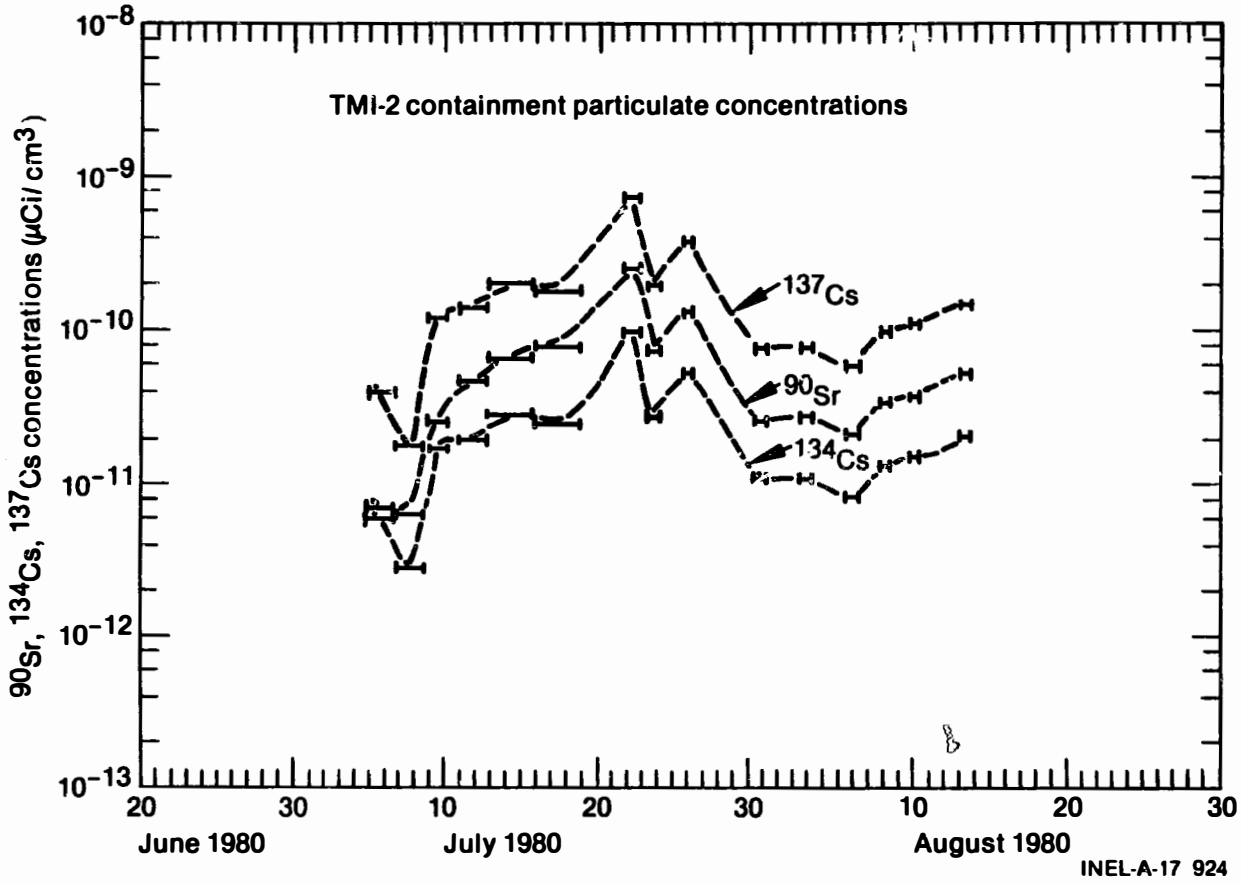

Figure 5. Plots of the particulate concentrations measured in TMI-2 containment atmosphere during the period July 4 to August 12, 1980.

TABLE 4. KRYPTON-85 CONCENTRATION FOR TMI-2 CONTAINMENT DURING VENTING
(Values of $\mu Ci/cm^3$)

Date 1980	HP-R-227 354' Sample 1	HP-R-227 354' Sample 2	HP-R-227 469' Sample 1	HP-R-227 469' Sample 2	HP-R-229X	HP-R-219	R-626
6/27	1.02		0.96				
6/29			0.95	0.88			
6/30	1.01	0.998	0.92				
7/1			0.88	0.89			
7/2			0.72	0.71			
7/3			0.61	0.61			
7/4			0.461	0.468			
7/5			0.358	0.372			
	0.342	0.328					
7/6			0.259	0.263		0.154	0.216
7/7	0.215	0.203					
					0.12		
					0.197		
7/8			0.134	0.135			
7/9	0.0072	0.0083	0.0094	0.0080			
7/10	1.7(-4)	1.7(-4)					
	1.5(-4)	1.9(-4)					
	2.0(-4)	1.8(-4)					
7/11			3.6(-5)	3.5(-5)			
			6.4(-5)	5.8(-5)			
7/15			6.4(-4)	6.2(-4)			
7/16	6.9(-4)[a]	6.7(-4)[a]	6.3(-4)	7.1(-4)			
7/17			7.5(-4)	7.7(-4)			
7/22			1.3(-3)	1.4(-3)			
7/23			1.4(-3)	1.4(-3)			
7/24			1.5(-3)	1.5(-3)			
7/25			1.6(-3)	1.6(-3)			
7/26			1.6(-3)				
7/27			1.7(-3)	1.7(-3)			
7/28			1.7(-3)	1.8(-3)			
7/29			1.9(-3)	1.7(-3)			
7/30			2.3(-3)	2.0(-3)			
7/31			3.4(-3)	3.3(-3)			
8/1			3.5(-3)	3.4(-3)			
			2.0(-3)	2.4(-3)			
8/4			2.5(-3)	2.3(-3)			
8/6			2.4(-3)	2.3(-3)			
8/8			2.4(-3)	2.4(-3)			
			1.4(-3)	1.4(-3)			

a. 315 ft.

TABLE 4. (continued)

Date 1980	HP-R-227 354' Sample 1	HP-R-227 354' Sample 2	HP-R-227 469' Sample 1	HP-R-227 469' Sample 2	HP-R-229X	HP-R-219	R-626
8/11			1.5(-3)	1.5(-3)			
8/13			1.6(-3)	1.6(-3)			
8/14			1.6(-3)	1.6(-3)			
			4.6(-4)	4.3(-4)			
8/15			<6.7(-6)	<7.0(-6)			
8/18			1.2(-4)	1.3(-4)			
8/20			5.6(-4)	6.0(-4)			
8/22			5.6(-4)	5.5(-4)			
			4.3(-5)	3.5(-5)			
8/25			1.0(-4)	1.2(-4)			
8/27			1.4(-4)	1.4(-4)			

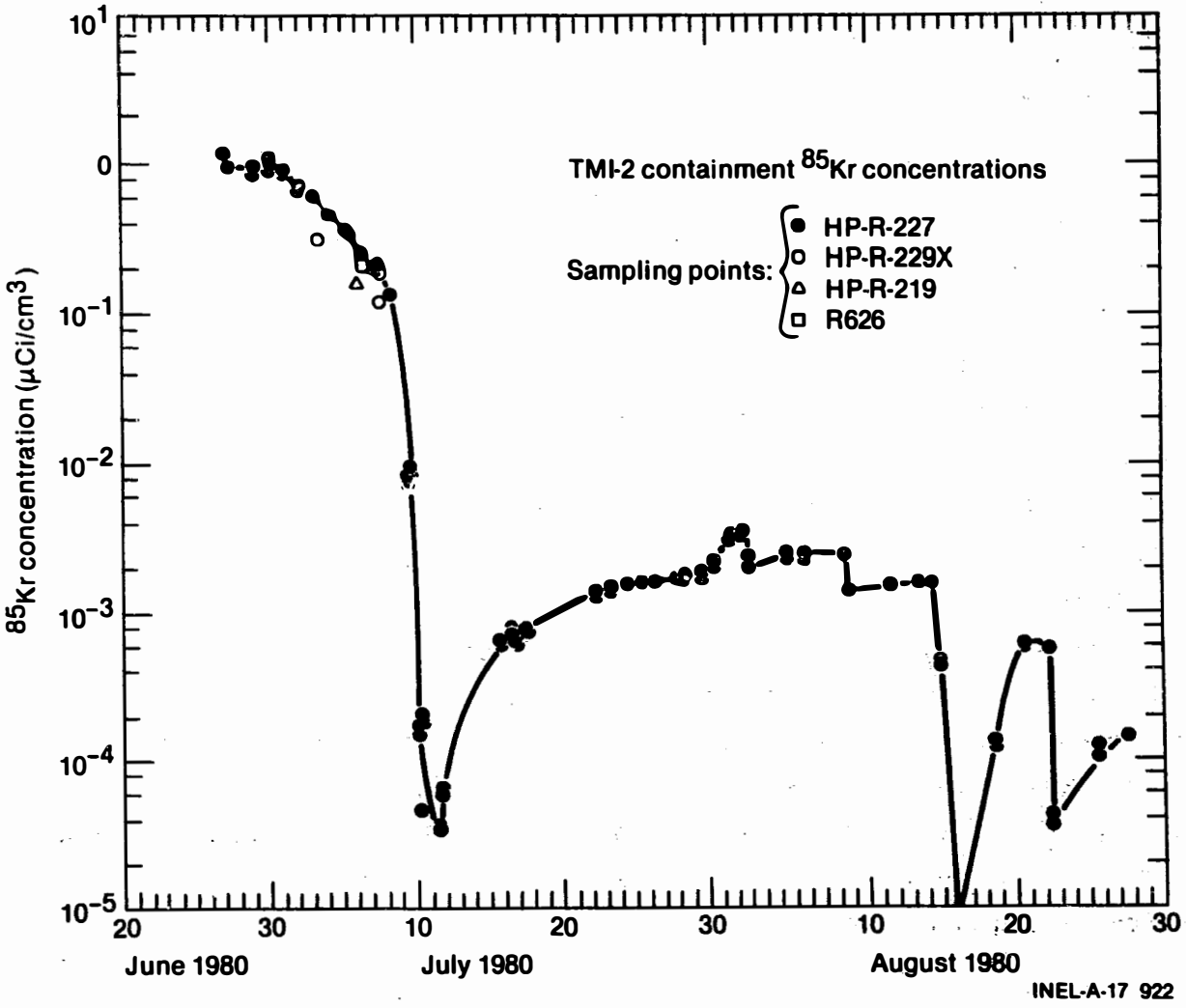

Figure 6. Plot of ^{85}Kr activity in TMI-2 containment during the period June 27 to August 28, 1980.

fact that they did not, results in some problems in comparing our measurements during and after the venting to those taken before the venting. Purging flow rates prior to the installation of the sampler was quite low (~200 cfm). By comparing the relative decrease of ^{85}Kr activity prior to the beginning of the sampling period, the equilibrium value on the scale of the present measurements is estimated to be about 4×10^{-11} $\mu Ci/cm^3$. It thus appears that the equilibrium concentration of total ^{129}I after the purge is about the same as that before the purge. Furthermore, each molecular specie of ^{129}I appears to recover to about the same concentration although the rate of recovery seems to be somewhat longer for I_2 and CH_3I than for HOI.

The makeup of the iodine species changed radically during the measurements with the relative concentrations of I_2 and HOI increasing to 16 and 43%, respectively, at the end of the purge and then returning to about 2.5 and 8% after the purging ended. The final equilibrium molecular species distribution is very similar to that of ^{131}I measured in June and July of 1979, mentioned in Section 1. The significant relative change of species can quantitatively be explained on the basis of partition coefficients between iodine and water[8] and deposition and resuspension coefficients for surfaces[4] for the different forms of iodine. Quantitative explanation would require computations involving the models developed[5] to describe the transport of iodine in systems. Changes in iodine species distribution were observed[9] during operations of "kidney filters" in containment at the Rancho Seco plant.

Concentrations of particulate activity was found to increase during the fast purge stage of the venting and to remain relatively high after the venting was terminated. A possible explanation is that the fast purge significantly disturbed the air currents in the vicinity of the sampler inlet, suspending considerable particulate activity into the air and that it remained suspended for a considerable time.

SUGGESTED ADDITIONAL WORK

Emission of iodine into the air from liquids and surfaces contributes largely to airborne iodine concentrations. This has significance in releases from nuclear reactors into confined areas from personnel safety or clean-up considerations.

Two additional tasks are desirable to fully understand the mechanism and implications of the iodine releases.

- Extend and apply the iodine model to the data to extract the needed resuspension, deposition, and partition coefficients.

- Measure the deposits of ^{129}I on the surfaces in TMI-2 containment to attempt to establish a better estimate of the relative contributions of the water and the surfaces.

The model in its current form does not include deposition of nonelemental forms of iodine. However, the fact that nonelemental forms of ^{129}I reached in equilibrium show that deposition does exist, and the model should be modified.

If the only source of ^{129}I were that deposited on interior surfaces early in the accident, the current model would predict that all the iodine would be in the air as organic and one purge would rid the atmosphere of it. It is possible that the sump water is also a source of ^{129}I. The model should be modified to include iodine partitioning after an accident as a source of iodine. Such a model would be capable of predicting iodine concentrations in containment after incidents such as those at Crystal River and Arkansas 1.

Using only airborne concentration measurements to verify and modify the model of iodine behavior provides only a portion of the evidence. Measurements of ^{129}I deposited on interior surfaces at TMI-2 offer along with air concentration measurements unambiguous evidence to verify or modify the model. Therefore, a program for measuring ^{129}I on surfaces at TMI-2 should be developed and carried out.

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