Session 20

AIR CLEANING TECHNOLOGY AT THREE MILE ISLAND

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CHAIRMAN: John Collins
Nuclear Regulatory Commission

INVESTIGATION INTO THE AIR CLEANING ASPECTS OF THE THREE MILE ISLAND ACCIDENT
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OPENING REMARKS OF SESSION CHAIRMAN:

Welcome to the session entitled, Air Cleaning Technology at Three Mile Island. I have been asked by several people to comment a little further concerning a remark I made at lunch yesterday as to why we cancelled the first entry into TMI containment. Cancellation was due to the fact that we were advised by our staff in Washington the night before that the breathing equipment that Metropolitan Edison had proposed to use, a unit manufactured by Bio-Marine, had been modified by Bio-Marine and the modification voided the certification of the equipment. The NRC regulations require that the licensees use approved equipment. The modifications in no way hampered breathing air; in fact, it actually increased the amount of air that the user would have received. But the fact that it was modified without recertification by NIOSH, meant that I could not permit them to make that entry. I hope that clears up the comment I made.

We have four papers to be presented in this session, dealing with various aspects of the air cleaning systems that were in place at the time of the accident at TMI. As I mentioned in my keynote speech, we have learned a lot about the accident and also that we do not know as much about air cleaning systems as we thought we did prior to the accident. We have a lot of work yet to be accomplished in this area.

Our first paper this morning is entitled, "Investigation into the Air Cleaning Aspects of The Three Mile Island Accident," and is authored by Dr. Bellamy. He has worked with various groups within NRC, e.g., the Effluent Treatment Systems Branch, the Accident Analysis Branch, and several months ago he was assigned to my staff at TMI as a Section Chief of our Technical Support Section. In this capacity, he is responsible for the review of all the technical activities associated with the cleanup of TMI-2.

Our second paper for this morning is entitled, "Studies of Airborne Iodine at TMI-2, Sources and Filtration." It will be presented by J.E. Cline, who with C.A. Pelletier arrived at TMI shortly after the accident. They were very instrumental in helping to quantify and characterize the releases of radioiodine in the plant.

The third paper this morning is entitled, "Radioiodine Adsorption Geometry Effects." As I mentioned yesterday, when the decision was made to change out the charcoal adsorbers in the Auxiliary and the Fuel Handling Building filtration systems, I contacted the authors and solicited their assistance in trying to analyze samples of the charcoal in an effort to determine the cause of charcoal degradation. Their paper will present some of these findings.

The last paper this morning is entitled, "Evaluation of Carbons Exposed to the Three Mile Island Accident." At the same time that I contacted Dr. Kovach about analyzing charcoal, I also asked Dr. Deitz to do similar tests on that same charcoal and he will share his findings with us.

INVESTIGATIONS INTO THE AIR CLEANING ASPECTS OF THE THREE MILE ISLAND ACCIDENT

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ABSTRACT

It would be difficult to underestimate the impact of the Three Mile Island accident on the nuclear industry. In the aftermath of the March 28, 1979 stuck-open pressurizer relief valve, an enormous and uncalculable amount of time, energy, and expense have been consumed attempting to analyze the causes of the accident and the impact, and to ensure that a similar accident does not occur again. Numerous investigative groups were formed to answer the questions of who, what, when, where, why, and how, and the published analyses have been read and studied with great interest. The industry has seen immediate hardware and operational changes at existing facilities, increased inspection by regulatory agencies, consideration of long-term changes in design requirements, and a licensing pause.

Of the many real and perceived dangers resulting from the accident, the release of radioactive materials in gaseous effluents cannot be ignored. The millions of curies of noble gases released were largely untreated, whereas the tens of curies of iodine passed through filtration systems--some of these systems were part of the facility on March 28, others were added post-accident specifically to reduce the magnitude of the releases. The investigative groups have analyzed in great detail the pre-accident design of the filtration and ventilation systems, their condition at the time of the accident, their usefulness in mitigating gaseous radioactive releases to the environment, and the post-accident mitigating air-cleaning actions taken by the utility. Numerous recommendations for enhanced filtration system performance resulted from the efforts of these investigative groups.

Investigative groups that have placed heavy emphasis on analysis of the air cleaning systems have included the President's Commission on the Accident at Three Mile Island (chaired by J. Kemeny), the Office of Inspection and Enforcement of the U. S. Nuclear Regulatory Commission (two studies done independently, one by an Investigative Team, the other by a Special Review Group), internal USNRC Task Forces formed specifically for this purpose (termed "Lessons Learned"), and an independent USNRC Special Inquiry Group staffed primarily by USNRC but under the outside supervision of attorney M. Rogovin (the Rogovin Report). Their interim and final reports, attached appendices, and supplementary and supporting documents are numerous. This paper will summarize the efforts of these groups in the air cleaning area. It will serve as a conpendium of the many available reports, and also be one source for the results of all the investigative groups. This paper will briefly discuss the mechanisms whereby gaseous radioactive materials were released to the environment, but the emphasis will be to summarize and discuss, in

one source, the efforts of the various investigative groups for the Three Mile Island accident pertaining to air cleaning technology, the recommendations these groups have made, and the potential impact on the nuclear industry of the implementation of these recommendations.

I. INTRODUCTION

The United States experienced the worst accident in the history of commercial nuclear power generation on March 28, 1979, at the Three Mile Island Station Unit No. 2. The resultant release of approximately 2.5 million curies of noble gases and 15 curies of iodine-131 led to sincere concern for the public health and safety, but in fact, the maximum probable offsite dose to an actual individual was on the order of 37 mrem (1) (compared to a background level of 116 mrem/yr). The accident and subsequent actions attracted significant interest from every sector of the world-wide population, technical and non-technical, and resulted in numerous investigative groups. One of the areas of concern for these investigative groups has been the air cleaning aspects of the accident, to include the status of the gaseous radwaste systems at the time of the accident, the development of a source term estimating the releases, the use of the air filtration systems in operation at the time of the accident to mitigate the releases, and the supplementary systems added during and after the accident to further mitigate the releases.

II. THREE MILE ISLAND GASEOUS RADWASTE SYSTEM

The gaseous radwaste system at Three Mile Island was designed and built in accordance with regulations and industry criteria in order to satisfy the requirements of Appendix I to 10 CFR Part 50 for maintaining releases "as low as reasonably achievable" (ALARA). The process gas system is designed to collect and store the radioactive gases stripped from the primary coolant in the letdown line and also the gases from the reactor building vent header and vent gases from equipment. The low pressure vent header collects these gases and pipes them to waste gas compressors for compression and storage in the waste gas decay tanks. After holdup for radioactive decay, the gases are released to the environment through HEPA filters and charcoal adsorbers. Ventilation exhaust systems are provided for the main condenser vacuum pump discharge (added after the accident), the turbine gland seal condenser discharge (primarily for clean steam),the auxiliary building exhaust, the fuel handling building exhaust, the reactor building purge and the hydrogen purge unit. Of primary interest are the auxiliary and fuel handling building exhaust systems, which played a significant role in reducing the release of gaseous radioactive iodine resulting from the March 28 accident, and the reactor building and hydrogen purge unit, which played a significant role in later cleanup operations.

The auxiliary building ventilation system is composed of two 30,000 cfm trains, each consisting of a prefilter, HEPA filter, 2 inch deep carbon adsorber, second HEPA filter, and fan. All ventilation air from the auxiliary building is designed to be processed by these cleanup components at all times-there is no bypass line. The entire ventilation system is designed for continual use during normal operation of the reactor. There were no technical specifications for balancing of ventilation flows or inplace testing of the exhaust air filtration components until the recovery effort began in the summer of 1979.

The fuel handling building ventilation system is composed of two 18,000 cfm trains, each consisting of a prefilter, HEPA filter, 2 inch deep carbon adsorber, a second HEPA filter, and a fan. Although a bypass is installed around these components to prevent their degradation and to preserve them for postaccident situations, the filter systems had been manually valved into service prior to March 28. Since the completion of acceptance testing in February 1978, all ventilation flow has been continuously routed through all the cleanup components. The fuel handling building ventilation exhaust system is an engineered safety feature system designed to operate in a postaccident environment. TMI-2 technical specifications require periodic inplace testing of the exhaust units to verify that the systems are ready to perform after an accident. However, exemptions to pertinent sections of these technical specifications were granted until the first refueling outage for TMI-2, which has not yet occurred.

The technical specification exemptions pertained to the carbon that was approved for installation in the fuel handling building ventilation filters. The carbon did not meet the specifications of, and its testing did not meet the recommendations of, NRC Regulatory Guide 1.52 (Revision 1, July 1976). The carbon did, however, satisfy the utility specification to remove 99.95% elemental iodine and 85% methyl iodide when the new carbon was tested at a relative humidity of 90% and at a residence time of 0.25 seconds. The exempted technical specifications would have required 99.9% for elemental iodine and 99% for methyl iodide. The exemption allowed carbon with a methyl iodide removal efficiency of 96.97% to be installed, but more importantly, waived the requirement for a representative sample of carbon to be removed every 720 hours of filter system operation and tested for degradation.

The waiver of the technical specification assumes added importance when it is relized that the basis for the waiver includes the assumption that the ventilation systems for Unit 1 and 2 are independent. However, the spent fuel areas are in direct communication for the two units, and any gaseous radioactive material present in either spent fuel area will be exhausted via both fuel handling building ventilation systems. More emphasis needs to be placed on interties between reactor units, buildings, and systems in future air cleaning work.

February 1978 was the installation date for the filters and cleanup components in the auxiliary and fuel handling buildings. They were inspected and satisfactorily tested inplace at that time, but they were not tested or inspected thereafter. Final painting and cleanup of these buildings between February and December 1978 generated significant amounts of fumes and aerosols that degraded the cleanup components far beyond any degradation that would have been permitted by technical specifications and periodic inplace testing. It is clear that although ANSI N510-1975 recommends the installation of prefilters as the only cleanup component during the final stages of construction at a nuclear power station and to dispose of these prefilters prior to station startup, and install all new components after completion of construction, this practice was not followed at Three Mile Island Unit 2. The degradation of these components led to higher radioiodine releases, by approximately a factor of five, than would have been experienced if no degradation had occurred.(2)

The reactor building air purge unit is composed of two 25,000 cfm trains. Cleanup components include a prefilter, HEPA, 2 inch deep carbon adsorber, second HEPA, and a fan. This system was not used in response to the March 28 accident. It was used during the purging of the krypton-85 from the containment structure in July 1980, when conditions allowed a high purge flow rate. The carbon adsorbers were removed prior to the purge since the iodine-131 had decayed to less than detectable concentrations in the 15 month period after the accident.

The hydrogen purge unit was designed to process 150 cfm of air from the reactor building through a prefilter, HEPA, 2 inch deep carbon adsorber, and a second HEPA. This system was modified in early 1980 prior to the containment purge to allow purging at flow rates less than 1000 cfm.

Both the reactor building and hydrogen purge units operated satisfactorily in order to allow the reactor building to be purged of approximately 43,000 curies Kr-85. Complications to these systems arose during testing prior to purging. For the hydrogen purge, an access door upstream of the first bank of HEPAs needed to remain open during DOP testing and fitted with a metal inlet port to allow DOP injection. Thus, after satisfactory DOP testing, the access door would be open until physically closed, in effect requiring another DOP test. The lack of planning during design for inplace test is obvious, and resulted in operational problems. Modifications to the system included a larger capacity fan, which required a right-angle exit and in actuality reduced the flow rate to a maximum of 560 cfm.

For the large capacity purge of containment, ductwork downstream of the fan was subjected to a positive pressure. Since krypton-85 would be present in this ductwork during the purge, all connections, seams and doors were sealed with duct sealant on the inside and outside, then tested. Nonetheless, significant concentrations of krypton-85 leaked into the auxiliary building at the onset of the fast purge, requiring termination. Subsequent analysis and inspection, including physically climbing into the ductwork revealed no leak paths. However, a concrete penthouse at the foot of the discharge stack was found to have an open door, resulting in significant leakage. Closing and sealing of the door eliminated the leakage and permitted the purge to continue. This incident emphasizes the importance of an overall system approach to air cleaning. The best filters will not provide satisfactory air cleaning if the filters are not integrated into acceptable ductwork and an overall system.

III. THE ACCIDENT

At 4:00 a.m. on March 28, 1979, TMI-2 experienced a turbine trip due to the stopping of condensate pumps. These pumps stopped automatically when their inlet and outlet valves closed. The reactor coolant pressure increased, and to protect the reactor coolant system, a pilot-operated relief valve (PORV) on the top of the pressurizer opened. The reactor automatically shut down eight seconds later, and should have resulted in the PORV closing. This valve however, stuck open, and water was continually removed from the reactor coolant system to the reactor coolant drain tank. A rupture disc failed on this tank at 4:15 a.m., allowing primary coolant to enter the reactor building sump. This sump automatically started, transferring over 8000 gallons into the 700 gallon auxiliary building sump tank. Liquid overflowed to the auxiliary building sump, which caused water to back up through the floor drains in both the auxiliary and fuel handling buildings. This water was not highly contaminated. Other non-contaminated water entered the buildings from four river water pumps, which exhibited leaks estimated to be on the order of gallons per minute.

After core damage, the primary coolant became extremely radioactive (15,000 uCi/cc). Radioactive material was transported out of the reactor by the letdown line of the makeup and purification system. It was necessary to maintain some letdown flow to the makeup and purification system to ensure safe cooldown of the reactor. Leaks in the makeup and purification system (located in the auxiliary building), which release small amounts of radioactive material in normal operation, released large amounts of radioactive materials during the accident, even though the letdown flow was reduced from its normal volumetric flow of 45 gallons per minute to about 20 gallons per minute. The letdown flow was, in fact, the major path for transferring radioactive material out of the reactor. This source of liquid radioactivity was released to the auxiliary building, and subsequently spread throughout the auxiliary and fuel handling buildings due to the presence of non-contaminated water.

The release of radioactive gases into the auxiliary and fuel handling building occurred by direct gas leakage and leakage of radioactive liquid from which radioactive gases evolved. Direct leaks of radioactive noble gases were the major source of releases to the environment. Leaks in the vent header system and the waste gas decay system were the primary mechanisms for the direct release of radioactive materials to the environment. The high pressure in the reactor coolant drain tank prior to rupture pressurized the vent header, and caused at least six leaks in the vent header system.(3) The high pressure also forced liquid through gas vent lines to the vent header that damaged component check valves. These check valves are designed to permit flow only from the component to the vent header and not in the opposite direction. With their failure, radioactive gases traveled from the vent header, where they are designed to collect, to the individual tanks. These tanks have relief valves set at a lower pressure (for example, the reactor coolant evaporator is set at 10 psig), which relieve directly to the stack. Thus, a pathway exists for transfer of radioactive gases from the primary coolant out the stack (see Figure 1).

Two other sources of direct gaseous leakage were present. The first was from the waste gas system compressor which exhibited significant leaks when operated.(4) The second was via venting of the makeup tank. Degassing of the letdown line in the makeup tank (see Figure 1) resulted in increased pressures in this tank. This high pressure opened a liquid relief, depleting the supply of borated water, and also increased the probability that relief valves on the reactor coolant bleed holdup tanks would open, permitting uncontrolled releases to the environment. In order to decrease makeup tank pressure, the vent was opened on this tank. Unfortunately, a momentary reading of 1200 mr/hr above the stack on Friday morning, March 30, triggered the unnecessary evacuation recommendations.

IV. MITIGATION OF GASEOUS RELEASES

The buildings and equipment at the Three Mile Island Station provided substantial mitigation of the release of radioactive materials to the environment. To prevent gaseous releases, the containment structure, simply by its leak-tight presence, prevented large quantities of radioactive material from being released (the building has remained negative in pressure differential since the hydrogen burn of the afternoon of March 28). The piping and tanks in the auxiliary building, and to some extent, the building itself, also retained quantities of radioactive material to provide holdup for decay, or containment of the radionuclides for later treatment.

The existing auxiliary and fuel handling building air filtration systems prevented essentially all of the particulate material, and the bulk of the

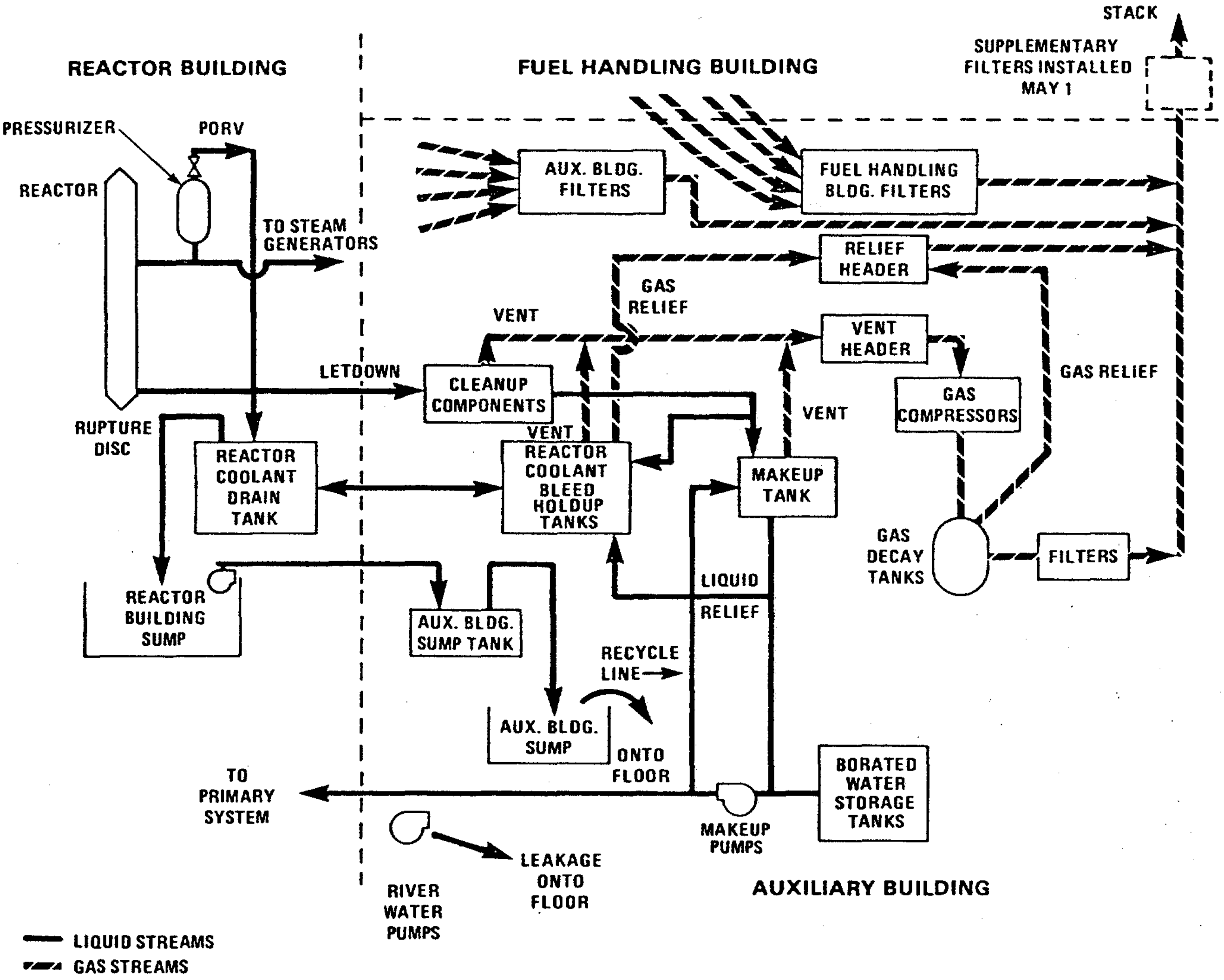

FIGURE 1. RELEASE PATHWAYS FOR THE THREE MILE ISLAND ACCIDENT.

radioiodines, from being released to the environment. Two of the investigative reports for the accident, "A Report to the Commissioners and the Public", directed by attorney Mitchell Rogovin (known as the Rogovin Report) (5), and the "Report of the President's Commission on the Accident at Three Mile Island", chaired by John G. Kemeny (known as the Kemeny Report) (6), highlighted the use of these air filtration systems. The "Technical Staff Analysis Report on Iodine Filter Performance" to the Kemeny Report (7) concluded that the carbon adsorbers in these two filter systems exhibited a decreased removal efficiency during the accident due to pre-accident weathering. Laboratory data indicate tht the carbon in the four trains during the accident was capable of removing between 49 and 75% methyl iodide at 95% relative humidity, depending on the individual train. The spread is considered an indication of unbalanced ventilation flows.

The Rogovin Report analyzed the available data in greater detail. The carbon removed from the four filter trains was layered, and analyzed for adsorbed activity and the pH of water extract. Although removal efficiencies ranged from 49 to 75% for methyl iodide at 95% relative humidity (as stated above), elemental iodine efficiencies were as high as 99.9%. Summation of the total amount of iodine captured yielded 112 curies, versus 13 curies released through the filters up to the time of the filter changeout. This translates into a decontamination factor of 9.5. It is concluded that the filter systems installed at the time of the accident operated at an efficiency of 89.5% (equivalent to a DF of 9.5) for all species of iodine.

The apparent discrepancy in the two methods used to predict carbon performance (testing of carbon for retention efficiency versus analysis of the carbon for adsorbed iodine compared to measured releases downstream) identifies the lack of an appropriate procedure for analyzing carbon that has been exposed to a large iodine source term. ASTM techniques would require loading the beds with a small amount of test iodine compared to the accident generated adsorbed iodine, which will be masked in the counting analyses. Species analysis of air samples taken upstream and downstream of the filter trains (8) indicated a concentration of 7.3×10^{-8} uCi/cc for particulates, which as stated above, was essentially all removed by the redundant banks of HEPAs. This analysis also showed concentrations of 6.7×10^{-8} uCi/cc for elemental iodine, 7.9×10^{-8} uCi/cc for methyl iodide, and 4.8×10^{-8} uCi/cc for hypoiodous acid. This species data is summarized in Table I.

TABLE I

IODINE SPECIES PRESENT DURING THE ACCIDENT

SPECIES	CONCENTRATION uCi/cc	PERCENT OF NON-PARTICULATE FORM
Elemental Iodine-I_2	6.7×10^{-8}	35%
Methyl Iodide-CH_3I	7.9×10^{-8}	40%
Hypoiodous Acid-HOI	4.8×10^{-8}	25%
Particulate Iodine	7.3×10^{-8}	--

In order to further protect the health and safety of the public, Metropolitan Edison (the licensee for Three Mile Island) decided to obtain a supplementary air filtration system to further mitigate radioiodine releases. Four 30,000 cfm filter units were installed on the roof of the auxiliary building. Each unit consisted of a heater, prefilter, HEPA, 2 inch deep carbon adsorber (KI_3-impregnated carbon) and a second HEPA. These units were airlifted from Richland, Washington, where they were scheduled for installation at the WPPSS Nuclear Units 1 and 4. These filter units were installed in early April 1979, in series with the existing auxiliary and fuel handling building filters, and therefore, all ventilation air was filtered twice prior to release to the environment since May 1, when system operation began. Since these supplementary auxiliary building filtration units were put in service, iodine releases have been negligible.

Replacement carbon for these ventilation filter systems was primarily a carbon co-impregnated with amine and potassium iodide. This carbon performed extremely well, although analysis results were hampered by the inability to obtain representative adsorbent samples from the installed banks. New carbon was certified to remove at least 95% methyl iodide, and 99.9% elemental iodine, at 95% relative humidity. After five months of service the carbon was still extremely effective in removing radioiodines (84.3 to 98.1% methyl iodide removal at 95% relative humidity, and 99.4 to 99.9% methyl iodide removal at 30% relative humidity). All of the available data is tabulated in Appendix II.2 "Carbon Performance with Time", to Volume II of Part 2 of the Rogovin Report. The efficient operation of carbon with co-impregnants has led to serious consideration of co-impregnated carbons for future nuclear use.

It is also interesting to note that replacement carbon for the existing auxiliary and fuel handling buildings was a co-impregnated carbon. Degradation of this carbon was followed very systematically as a function of time for a five month period after the accident. Testing was performed for methyl iodide removal efficiency, and also physical properties of the carbon. Methyl iodide removal efficiencies decreased from over 99% to 83% at 95% relative humidity, but remained at 99.9% at 30% relative humidity. Physical properties analyzed included moisture content, and the pH of water extract. Moisture content data varied significantly due to environmental conditions, and there are no apparent trends in the data. The pH data showed some decrease with time of operation, but not significant. All of these data are available in Appendix II.2 to the Rogovin Report.

Further data and analysis are discussed elsewhere in these proceedings by Dietz et al, "Evaluation of Carbons Exposed to the Three Mile Island Accident".

Significant sampling problems arose. Although test procedures conformed to the recommendations of Regulatory Guide 1.52 (Revision 1), the data lack consistency. Two types of impregnated carbon were obtained from two sources, and carbon sampling methods did not conform to industry standards. The carbon samples removed for analysis were shipped in plastic bags, with incomplete data on cells sampled, location in bank, date obtained, and type of charcoal. There is also no means to ascertain whether the sample was properly mixed to assure homogeneity prior to shipment. In addition, different cells with different operating histories have been removed for sampling. This removal resulted in some of the carbon being tested that had been used to refill a test cell at the previous sampling, and not testing other carbon that had been in service since the changeout. These sampling problems resulted in nonrepresentative samples with results that may be neither reproducible nor valid.

Nonetheless, based on the negligible iodine releases after the accident, it is concluded that the co-impregnated carbon performed satisfactorily in reducing radioiodine releases to the environment.

Noble gas releases to the environment were not mitigated by the ventilation systems, but some delay time was provided by the installed tanks and components. Based on the pathways discussed earlier, $2.37 \times 10^{+6}$ curies of total noble gases have been predicted to be released as a result of the accident. Direct measurement of this release, however, is not possible since the radiation monitor installed on the stack for this purpose was designed for normal operation only, and went off-scale early in the accident. An indirect method was used to predict the noble gas releases, based on a corelation between an area gamma monitor in the auxiliary building and the stack monitor. The area gamma monitor remained on scale during the accident, and was sensitive to changes in the radioactive release rates since it is located close to the exhaust ductwork.

The buildup of noble gases due to offgassing of the primary coolant caused the waste gas decay tanks to approach their capacity, causing concern for uncontrolled releases. Therefore, vent lines from these tanks were installed back to containment for transfer of these radioactive gases. This operation was performed satisfactorily, and has led to a recommendation that other reactors consider the installation of such lines for use after accidents only. In fact, Three Mile Island Unit I has committed to the installation of such lines prior to restart.

One further filter system that warrants discussion is the ventilation system used for the building housing the EPICOR II liquid radwaste cleanup system. EPICOR II is a system of demineralizers designed to assist in the recovery operation by treating the 500,000 plus gallons of liquid in auxiliary building tanks generated by the accident, and additional water generated by decontamination efforts. The building housing the EPICOR II system is ventilated via a filtration system prior to release to the environment. The filtration system consists of a prefilter, a HEPA filter, 2 inch deep carbon adsorber, and a second HEPA filter. The carbon adsorber section is of the gasketless design. During the summer of 1979, prior to filter system startup in October, the water sprays for the carbon were somehow activated, resulting in the carbon being wet, and leaving approximately eight to ten inches of water on the housing floor. Within days, the wet carbon was removed. From October 1979, until the present, the filtration system was operated continuously, except for a second changeout of carbon due to a second inadvertent activation of the water sprays (time frame unknown). Inspection of the screens in September of 1980 showed no visible corrosion to any part of the metal. Thus, it would appear that the much-publicized potential corrosions of screens may be eliminated, and at least reduced by quick removal of the wet carbon.

A minor problem noted with the gasketless design of the EPICOR II filtration system is the bowing of the screen where it is attached to the frame. This leads to an increase in the bed depth which is a positive aspect, but may lead to gross failure. No indication of any carbon was observed on the floor of the housing upon inspection in September 1980, and the system appeared quite sturdy.

V. INVESTIGATIVE GROUPS

In addition to the President's Commission and the Rogovin Report, two internal groups of the U. S. Nuclear Regulatory Commission placed heavy emphasis on the air cleaning aspects of the Three Mile Island accident. Their efforts resulted in the Office of Inspection and Enforcement's Investigative Report, and the Office of Nuclear Reactor Regulation's Lessons Learned Reports.

The Inspection and Enforcement (I&E) Investigative Report established the basic facts concerning the accident, and evaluated the performance of the licensee, Metropolitan Edison, as a basis for corrective and enforcement action. This report did not evaluate any other group, or their input into the accident. A comprehensive study of Met Ed's conformance to regulations, regulatory guides, procedures and safety analysis report commitments was performed. Their major findings with importance to air cleaning are summarized below:

1. Preoperational testing of atmospheric monitors was incomplete. Chart recorder readings were not calibrated, hindering postaccident evaluation.
2. Tests showed that ventilation exhaust flow rates in the auxiliary and fuel handling buildings were different than designed. This would lead to the difference in carbon performance indications as already discussed. This imbalance still exists at TMI today.
3. A number of maintenance problems with the waste gas system were outstanding at the time of the accident, specifically with improper operation of the waste gas compressor, and leaks in the makeup tank vent valve. Correction of these work items would have reduced the release of noble gases.
4. Leaking ventilation bypass dampers were welded shut, requiring all ventilation air to pass through the carbon. This substantially degraded the carbon, resulting in increased radioiodine releases during the accident. Aerosols generated during final painting with the cleanup components installed further degraded the carbon. And, as previously discussed, inplace or laboratory testing of the carbon in accordance with technical specifications had been waived. (New dampers were to be installed at the first refueling outage, and the technical specification waiver was to be null and void at that time. However, the first refueling outage for Unit 2 has yet to occur).

The NRC staff investigation of the accident resulted in a task force being formed to identify the lessons learned from the accident. Their recommendations resulted in short term requirements, long term requirements, and a task action plan for implementation purposes. This investigation received review at all levels of the NRC, including the Commissioners. The applicable changes proposed for licensees in the area of air cleaning are itemized below.

1. Insufficient instrumentation was available to adequately follow the course of the accident. The noble gas stack monitor went offscale early in the accident, and inadequacies in obtaining radioiodine and particulate effluent samples have already been discussed. The recommendation includes providing high range radiation monitors for noble gases in plant effluent

lines (and a high range monitor inside containment). Also, instrumentation should be provided for monitoring effluent release lines to measure and identify radioiodine and particulate radioactive effluents under accident conditions.

In the area of control room habitability, instrumentation should be provided for accurately determining in-plant airborne radioiodine concentrations to minimize the need for unnecessary use of respiratory protection equipment. Twice during the accident masks were donned by all control room personnel when intake monitors for control room air alarmed high. However, the masks were designed for particulate protection only, not the noble gas or particulate fractions, and therefore provided minimal protection for the operators. The use of the masks increased the "confusion factor" in the control room, and severely hindered communications.

2. Establish an onsite technical support center (TSC) separate from, but in close proximity to, the control room for engineering and management support of reactor operations in the event of an accident. Among the design criteria for the TSC, is the requirement that the TSC be habitable to the same degree as the control room for postulated accidents. This includes permanent ventilation systems with particulate and charcoal filters. The design and testing guidance of Regulatory Guide 1.52 should be followed, except the systems need not be qualified as engineered safety feature. Thus, the systems need not be redundant, seismic, instrumented in the control room, automatically activated or have HEPA filters tested at a quality assurance station or satisfying the quality assurance of Appendix B to 10 CFR Part 50.

The NRC Action Plan for actions judged necessary to correct or improve the regulations or operation of nuclear facilities based on TMI-2 experience has been published as NUREG-0660, "NRC Action Plan Developed as a Result of the TMI-2 Accident," May 1980. The Plan identifies priorities, lead NRC offices, implementation schedules, and fiscal resources. Those actions pertaining to air cleaning are summarized below.

1. Requirements for post-accident sampling of containment atmosphere for noble gases, iodines and nonvolatiles include obtaining the sample within one hour and analyzing within two hours.
2. The need for additional accident monitoring instrumentation as concluded by the Lessons Learned Task Force is implemented as a high priority item.
3. NRC will evaluate potential sources of airborne radioactivity due to primary coolant that may be in systems outside containment. Leaks in the letdown line and the makeup and purification system, and the vent gas system have been identified as resulting in the major release path for noble gases.

   Evaluations will include leakage detection and control, overpressurization design, pressure relief mechanisms, flow restriction, permanently installed paths to vent gaseous systems to containment and discharge points for vent-gas systems.

4. Research has been requested to ascertain viable alternatives to eventual discharge of long-lived noble gases to the environment.
5. Ventilation systems should be reviewed in order to determine if adequate control of airborne radioactive material can be achieved. This will include upgrading of filtration systems and implementation of surveillance testing for all engineered safety feature and non-engineered safety feature filtration systems (Regulatory Guide 1.52 and 1.140 respectively). Research will be initiated to evaluate carbon and other radioiodine collection media performance under accident conditions. Factors to be evaluated will include degradation due to normal operating conditions, poisoning, bed depth, types of carbon impregnants, radiation degradation effects, influence of high noble-gas and radioiodine concentrations, and bleeding of radioiodine after collection. Feasibility will also be investigated for (a) requiring inplace online testing of ventilation systems (such as continuous upstream/downstream sampling) to ascertain overall filter system performance, (b) requiring the development of procedures to evaluate spent carbons exposed to accident conditions, and (c) requiring committed filtration systems for accidents only. Based on the results of this research, the appropriate regulatory guides may be revised.
6. Potential control room operator doses will be evaluated to verify that the workers are adequately protected from radiation and radioactive materials, and can continue to operate the plant in a safe manner following an accident. Airborne radioactive material ingress and radiation penetration from sources internal and external to plant structures will be considered.

The fullest possible understanding of the accident, both from a technical point of view and of a regulatory process, was undertaken by a special inquiry of the NRC to review and report on the accident. An outside law firm was contracted to conduct the inquiry, in recognition of the potential conflict of interest problem if the NRC staff directed and undertook the inquiry. Although volunteer NRC staff comprised the bulk of the Special Inquiry Group, technical consultants and investigatory lawyers supplemented the effort. The product of the Special Inquiry Group, "Three Mile Island, A Report to the Commissioners and the Public," also is known as the Rogovin Report, for the director of the inquiry, Mitchell Rogovin. This report placed heavy emphasis on the air cleaning aspects of the accident.

Those aspects different from the conclusion of other investigatory reports are indicated below, along with those that warrant repetition.

1. A major finding of the Rogovin Report is that the radwaste systems as operating at the time of the accident provided substantial mitigation of the release of radioactive materials to the environment, even though the design bases of the systems were exceeded. For all the lessons learned, and

faults uncovered during all the investigations, it is important to realize, and to emphasize that the health and safety of the public was protected at all times.

2. The design and review of the Unit 2 ventilation systems did not consider the interties with Unit 1. This is especially evident in the fuel handling buildings, where there is direct communication between the spent fuel pools. This was not considered in the technical specification exemptions allowed for the Unit 2 fuel handling building ventilation filters.
3. Leakage of radwaste system components in the makeup and purification system, and the waste gas system (especially the compressor), led to the most significant releases of radioactive materials.
4. The gaseous radwaste system components were provided with manual vents to reduce pressure in these components when necessary. The vents were piped to the waste gas system and therefore the gases were treated prior to release to the environment. However, the system design also includes automatic relief valves which provide relief directly to the atmosphere as unfiltered gas. These relief valves did operate during the accident.
5. Modifications to the ventilation and gaseous radwaste systems initiated after the accident (supplementary auxiliary buiding filter systems, vent lines from gas decay tanks to containment) helped mitigate releases to the environment.
6. For the auxiliary and fuel handling building filter systems, continual use of these systems placed the carbon in a degraded condition on March 28, 1979. This contributed to increased radioiodine releases over that expected with unused carbon. If carbon had been in place at the time of the accident that satisfied the technical specifications, radioiodine releases would have been lower by a factor of 5 (based upon postaccident determinations of carbon retention efficiencies). Even in a degraded state, the carbon installed at the time of the accident provided a decontamination factor of 9.5 (89.5% efficiency) for all species of radioiodine. However, during the first week of the accident, neither inplace testing nor laboratory testing of carbon samples was adequate to analyze the effectiveness of the ventilation exhaust filters.
7. The auxiliary and fuel handling building air filtration systems were operable during the accident and provided substantial mitigation of radioiodine prior to release to the environment, and also collected essentially all of the radioactive particulate material. Each filter system had identical safety-grade cleanup components, although the auxiliary building system was not designed safety-grade. The safety grade versus nonsafety grade designations of these systems were meaningless, and a nonsafety grade system provided significant mitigation of radioactive releases.

VI. CONCLUSION

Although the Three Mile Island accident of March 28, 1979, released 2.4 million curies of noble gases and 15 curies of iodine-131, the gaseous radwaste and ventilation systems prevented these releases from being considerably higher. The systems operated as designed, but in their operation a number of problems arose that have been discussed. Many of these problems and concerns have led to recommendations for hardware changes to future systems, upgraded surveillance requirements for existing systems, and, in some cases, completely new ventilation and air filtration systems. The implementation of these systems is expected to further ensure the health and safety of the public from gaseous radioactive material releases after an accident at a nuclear power station.

REFERENCES

1. "Population Dose and Health Impact of the Accident at the Three Mile Island Nuclear Station," U. S. Nuclear Regulatory Commission NUREG-0558, May 10, 1979.
2. "Three Mile Island A Report to the Commissioners and to the Public," M. Rogovin, Director, Nuclear Regulatory Commission Special Inquiry Group, Volume II, Part 2, p. 347.
3. Id. at p. 352.
4. Deposition of J. T. Collins, U. S. Nuclear Regulatory Commission, taken by the NRC Special Inquiry Group, September 1979, p. 85.
5. "Three Mile Island A Report to the Commissioners and to the Public," M. Rogovin, Director, Nuclear Regulatory Commission Special Inquiry Group, Volumes I and II.
6. "Report of the President's Commission on the Accident at Three Mile Island, The Need for Change: The Legacy of TMI," J. Kemeny, Chairman, October 1979.
7. "Technical Staff Analysis Report on Iodine Filter Performance," to the President's Commission on the Accident at Three Mile Island, William M. Bland, Technical Assessment Task Force.
8. "Three Mile Island A Report to the Commissioners and to the Public," M. Rogovin, Director, Nuclear Regulatory Commission Special Inquiry Group, Volume II, Part 2, p. 359.

DISCUSSION

FIRST: Throughout your discussion you commented that the NRC was going to do this, or was considering that, or might do something else in terms of mitigating a series of events such as you described. Now, it has been seventeen months since this occurrence. Is this what you would term "deliberate speed ahead" for the NRC?

BELLAMY: As Chairman of the Session, Mr. Collins, would you like to answer?

COLLINS: As I mentioned in the keynote speech, I think that in the development of many of the short-term lessons, we looked at all of the causes of the accident and implemented what were the essential things that had to be corrected on a short-term basis. They were implemented, and had to be implemented, by January 1, 1981. We are, of course, continuing to investigate other systems, such as the filtration systems. I am sure that within the next short period of time, we will see further implementation.

STUDIES OF AIRBORNE IODINE AT TMI-2, SOURCES AND FILTRATION*

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Summary

In April 1979, SAI began an evaluation of the behavior of airborne radioiodine at Unit 2 of the Three-Mile-Island nuclear plant. The experimental portion of the radioiodine sampling program continued from May until September 1979. Two specific tasks were addressed in the study to be reported here. These were:

1. Identify specific sources of radioiodine within the auxiliary and fuel-handling buildings.
2. Measure the retention efficiencies of the auxiliary and fuel-handling building charcoal filters and the supplementary filter banks on the auxiliary building roof for ^{131}I.

Some details of the measurements of radioiodine within the auxiliary and fuel-handling buildings are summarized in the following table. Within the 281 foot

Area	Fraction of Total Ventilation Flow (percent)		Fraction of Total Radioiodine Release (percent)	
Auxiliary Building Total	65		76	
281-foot level		10		25
305-foot level		25		2
328-foot level		30		49
Fuel Handling Building	35		24	

---

* Work supported in part by the Electric Power Research Institute, General Public Utilities Corporation and the Department of Energy.

level, the principal contributor was the let-down pipe chase and valve room (23%). The molecular form of the iodine in both buildings was predominantly inorganic. During the period May to September, the inorganic fraction increased from approximately 70 to 90 percent. The average normalized release rate for radioiodine for this plant (release rate divided by reactor coolant concentration) was comparable to that from other PWR plants. This suggests that the releases were from paths similar to those in other plants, typical paths such as leaking valves, pump seals, etc. Arguments are presented that suggest that the majority of radioiodine released from mid May on was from surfaces rather than current leaks, and that the major deposition of iodine on these surfaces occurred early in the accident.

Decontamination factors (dF) for the internal auxiliary building charcoal filters showed a slight decrease with time from late May until late August 1979. The factor for inorganic iodine decreased from well over 100 to about 25 at the end of June where it remained until the termination of sampling. The factor for organic iodine remained constant at about 17 during the entire period.

Decontamination factors for the internal fuel-handling building charcoal filters for organic iodine showed a decline from about 15 in early June to about 8 in late August. The factor for inorganic iodine was higher, ranging from about 16 to 53.

The filters on the auxiliary building roof showed considerably higher dFs, averaging about 230 for total iodine. Inorganic iodine dFs were slightly higher than those for organic. The roof filters had less weathering of the charcoal than the inside filters which probably accounts for the higher decontamination factors.

Measurements were made of the ^{129}I and particulate concentrations during the purging of the Unit 2 containment atmosphere that occurred from June 28 until July 11, 1980. Iodine modecular species samplers were used in the measurements. 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 has 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. The equilibrium concentration in both total iodine and in species distribution was being maintained by partition of iodine from the liquid and by deposition and resuspension of iodine from surfaces. The dominant source of iodine, either the sump water or the containment surfaces, is not known at this time. By comparison, the

Section 1

PROGRAM TO IDENTIFY SOURCES OF AIRBORNE RADIOACTIVITY WITHIN THE AUXILIARY AND FUEL-HANDLING BUILDINGS

Well after the accident, the auxiliary and fuel-handling buildings at Three-Mile-Island Unit 2 continued to exhibit high airborne concentrations of radionuclides. A program was begun early in May to identify major injection points of ^{131}I into these buildings. The objective was to eliminate the sources, substantially reducing the activity leaks in the building. SAI installed twenty-four samplers in ventilation exhaust ducts and specific room air points. Sampling continued at various locations from May 11, 1979 until September 4, 1979 when iodine activities had dropped to levels that were difficult to measure. A report (1) on this work has been published.

1.1 SUSPECTED PRINCIPAL CONTRIBUTORS TO RELEASES FROM AUXILIARY AND FUEL-HANDLING BUILDINGS

The primary coolant letdown and makeup systems and the waste gas system were suspected to be the principal contributors to the release of radioactivity into the auxiliary and fuel-handling buildings. Hence, much of the activity was directed toward a study of these components.

1.1.1 Location of Plant Components

The three reactor coolant makeup and purification pumps are located on elevation 281 in the auxiliary building. The reactor coolant bleed hold-up tanks are also located on this level. In addition, some letdown system valves and penetrations are located on this floor in the area of the reactor building annulus next to makeup pump room "C". The waste gas decay tanks and compressors are located on the 305-foot level of the auxiliary building as are the makeup tanks and demineralizers. The makeup and purification valve aisle is located in the fuel-handling building, also on the 305-foot level. The radiation level in this area remained quite high. The 328-foot level of the auxiliary building houses the filtration system for both the auxiliary and fuel-handling buildings and the core flood makeup tank around which there was some high activity spillage in May. The corridor ^{85}Kr concentration was reduced by a factor of 50000 during the purge and recovered by evolution from the sump water to a level of 1/500 of its initial value after the main purge.

on the 328-foot level in the fuel-handling building is a ventilation flow path from the reactor building annulus that contains letdown system penetrations and valving.

1.1.2 Building Ventilation System and Sampler Locations

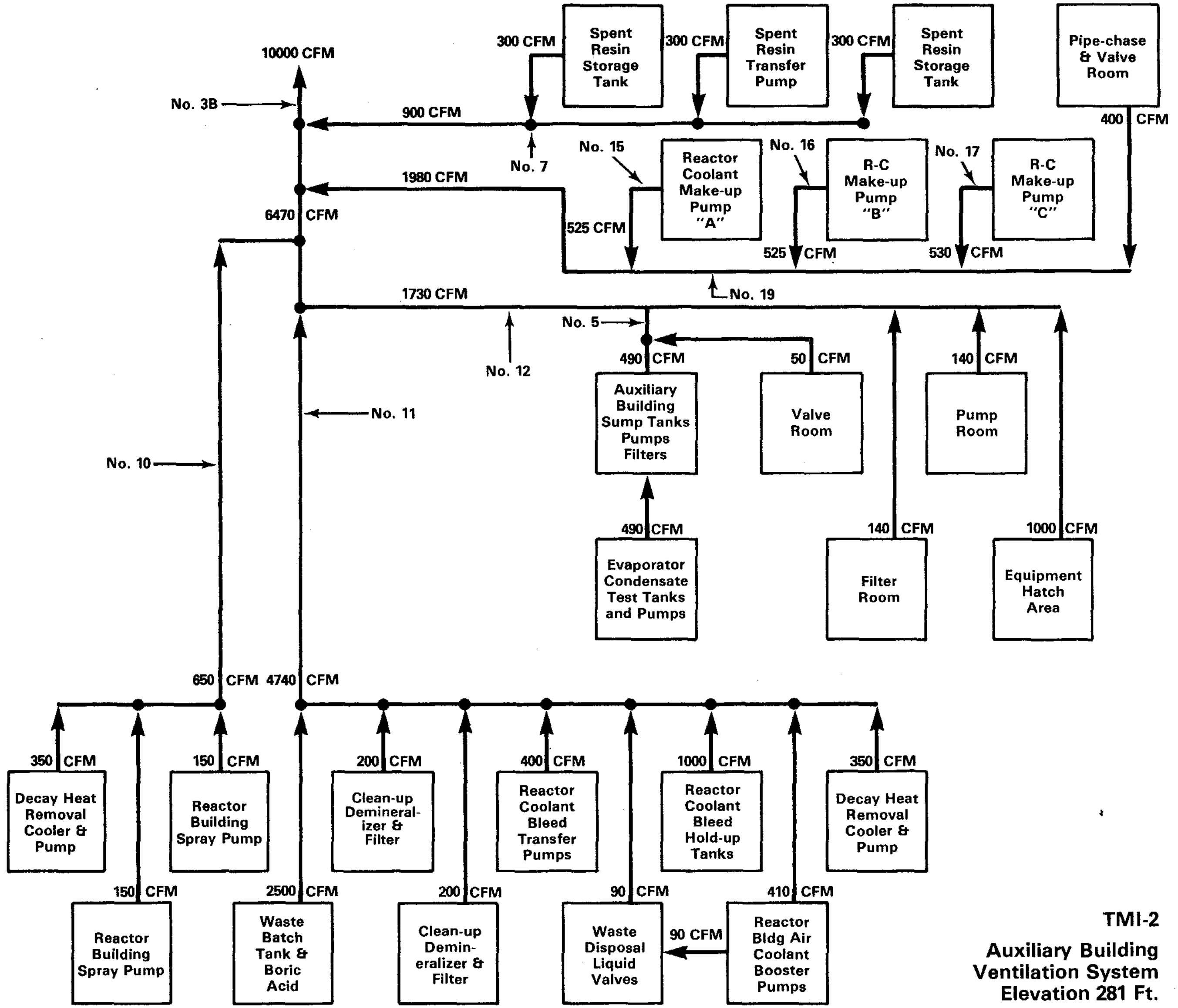
The ventilation systems of the TMI-2 auxiliary and fuel-handling buildings were constantly being changed during the course of the measurement program. Supply and exhaust fans were being turned on and off; dampers were being closed and opened; filters, installed on the building roof, were being tested and balanced. As a consequence, at any time it was impossible to know just what the ventilation flow rates were or that they would remain constant during a sampling period. Therefore, in order to calculate absolute or relative release rates for the various sample points it is necessary to assume that the exhaust flow rates approximated the design flow rates. All of the following discussions assume design flows. The accuracy of the release rates is therefore unknown but is probably better than a factor of two. Relative release rates are probably much better than this.

A flow diagram of the design ventilation system is given in Figure 1.1 for the auxiliary building 281-foot elevation. A number of samplers were installed to sample the ventilation air from this level because of the large number of potential release contributors in the area. The locations of these samplers are indicated on the figure with a sampler number and an arrow showing the location in the system. As can be seen in the figure, many of these samplers were located to monitor the reactor coolant high-pressure injection system and the auxiliary building sump system.

Figure 1.2 shows the design flows for the ventilation system for the 305-foot elevation of the auxiliary building. Two samples were installed to sample the air from this level; one in the makeup tank room and the second in the total exhaust duct.

The floor plan of the 328-foot elevation consists principally of open areas. Sampling was limited to those general areas with relatively high flow rates.

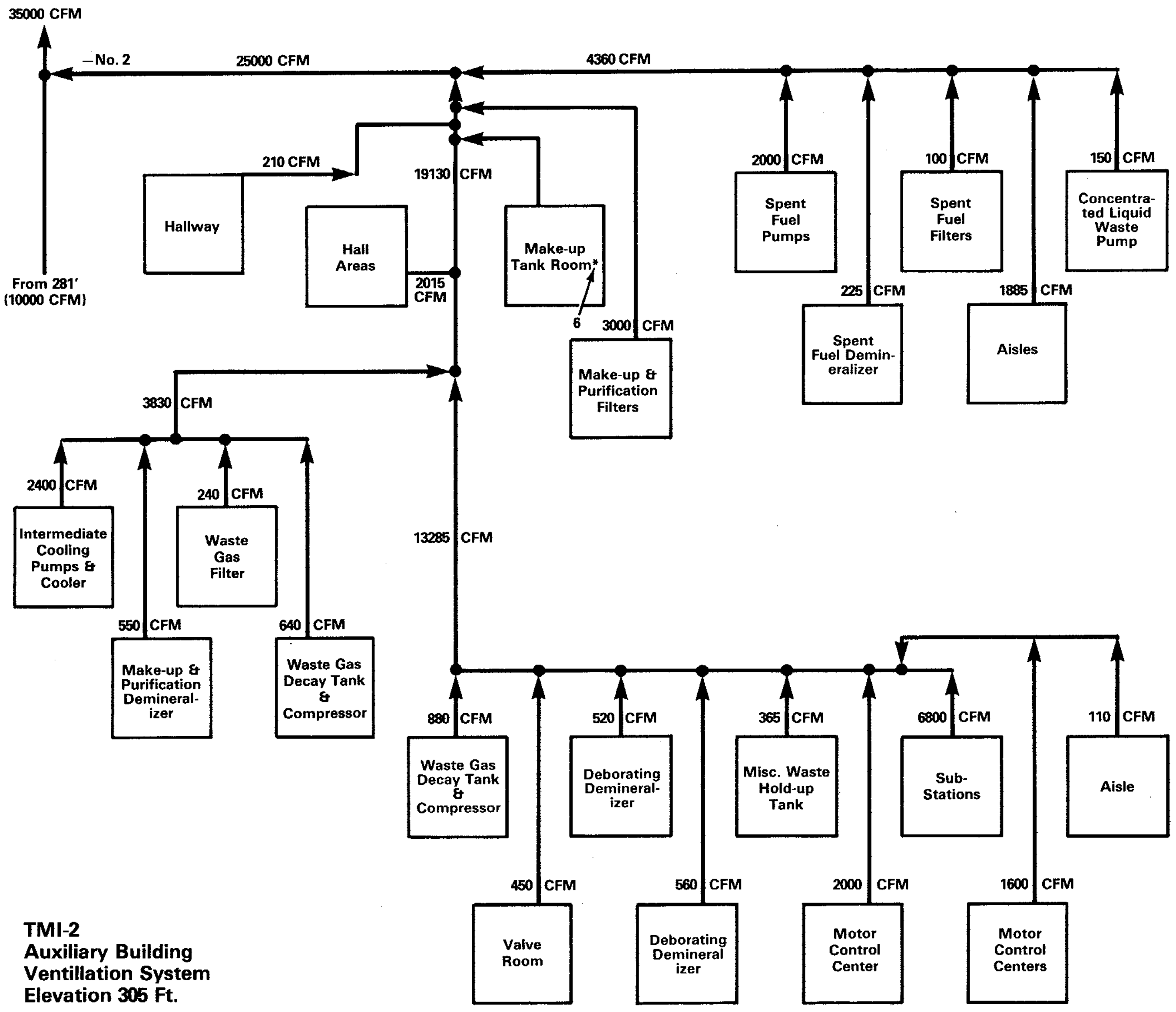
The fuel-handling building also has many open areas. An area known as the "reactor chase" is a portion of the reactor building annulus that connects to the fuel-handling building. The flow is from the 281-foot elevation upward with the exhaust at the 347-foot level through the pipe chase. It was suspected to be a large source of activity.



TMI-2

Auxiliary Building Ventilation System Elevation 281 Ft.

Figure 1.1



TMI-2
Auxiliary Building
Ventillation System
Elevation 305 Ft.

Figure 1.2

The auxiliary and fuel-handling building charcoal filter system is shown in Figure 1.3. This system is located on elevation 328-foot of the auxiliary building. The location of the SAI samplers are indicated as AE-1 through AE-8. The plant monitoring systems are shown as the HP-R locations.

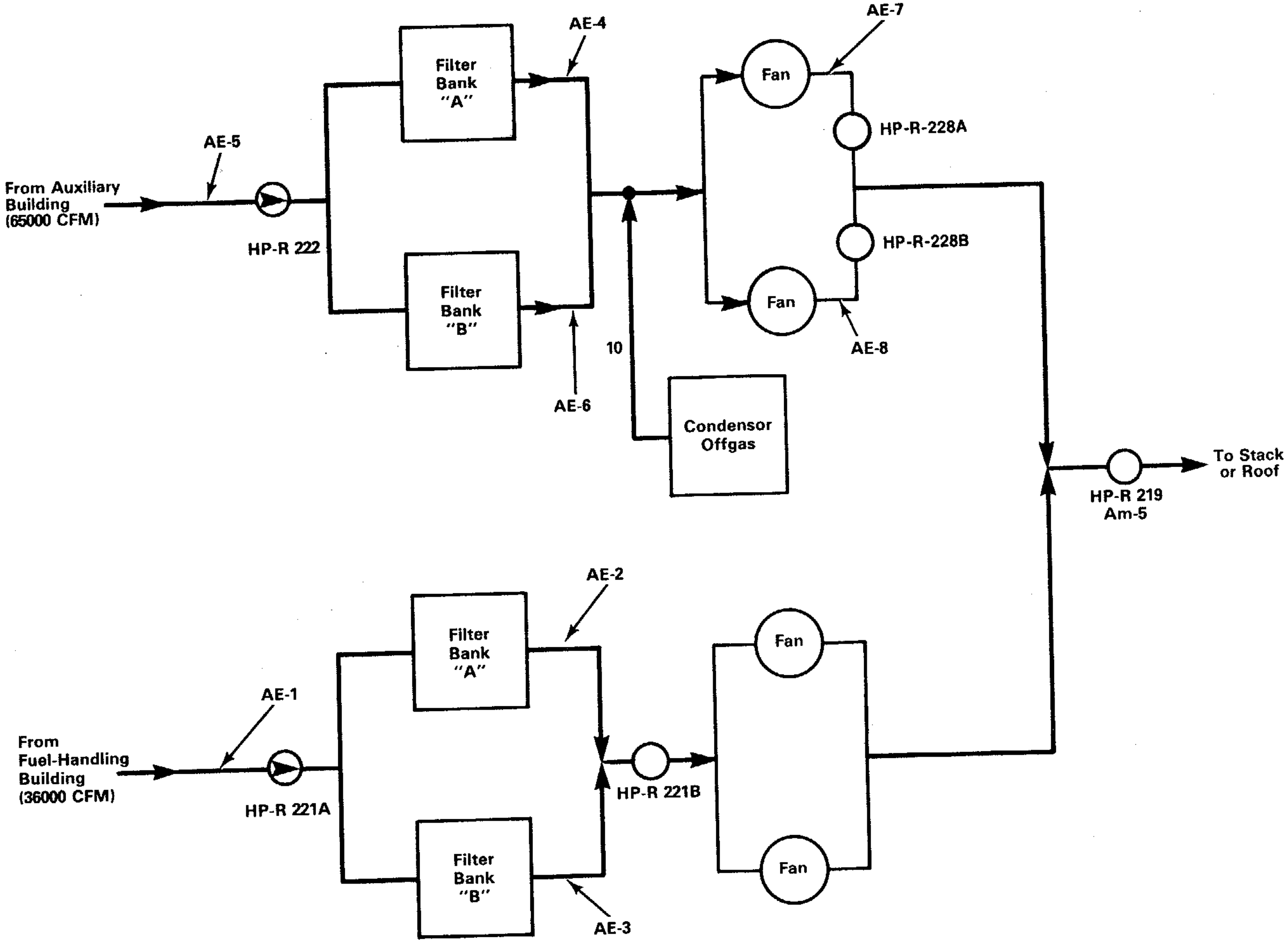
1.1.3 Operational Activities Since the Accident That Might Contribute to Activity Releases in Auxiliary and Fuel-Handling Buildings

The early period of the accident was characterized by the movement of large volumes of water throughout the auxiliary building by means of the letdown and makeup systems. During the first few days, a feed and bleed operation was used to degas the primary system. In this operation, reactor coolant was let down to the coolant bleed tanks and to the makeup tanks so that the non-condensable vapors could be removed. The gas was released through the gas vent header to the compressors and into the waste gas decay tanks. The decay tanks, after March 30, were released back into containment through the hydrogen recombiner system. Leakage from any of these components could contribute substantially to radioactivity levels in the auxiliary building since the systems were processing the highly radioactive primary coolant. The vent header was known to leak and there was a gaseous release from the plant each time it was used.

Similarly, the makeup system has been in continual usage since the accident using letdown coolant that has been degassed and diluted with a small amount of fresh boric acid solution. Any leakage in the high-pressure makeup pumps will release reactor coolant water into the air at pressures and temperatures that will vaporize the liquid after release. Except for short periods during March 28 and during the period May 9 to May 13, makeup pump "B" was running continuously. Pump "C" ran only for a short period during March 28 and pump "A" ran during March 28 and during the period May 9 to May 13.

The letdown system also ran continuously after the accident. The reactor containment penetrations and the valving and piping for the system are in the reactor building annulus, both in the auxiliary building and in the fuel-handling building portions. Some of these valves have leaked and there was high activity liquid in the pit of the annulus in the fuel-handling building.

Early in the accident the auxiliary building sump was known to have overflowed, pushing water up through the floor drains on the 281-foot elevation of the auxiliary building. Much of the water that covered the floor was relatively low



TMI-2 — Auxiliary and Fuel Handling Buildings Ventilation and Monitoring Systems

Figure 1.3

activity but some was not. The sump, however, continued to be a suspected source of airborne activity.

In May (17-19) some highly contaminated water was spilled on the floor in the auxiliary building on the 328-foot elevation near the core flooding makeup tanks. This water is suspected to have been primary coolant water that backed up through the core flood addition line. The area was a possible contributor to high airborne concentrations.

1.2 SAMPLING AND DATA ANALYSIS

1.2.1 Sampling and Analysis Techniques

Both molecular species and total iodine sampling was done in these measurements. Analysis of the collected samples was by gamma-ray spectrometry using Ge(Li) detectors. The equipment was located in the SAI trailer laboratory at the Three-Mile-Island site.

1.2.2 Release Rates for ^{131}I

Graphs of the ^{131}I release rates as a function of time for the fuel-handling and auxiliary buildings are shown in Figures 1.4 and 1.5, respectively. These release rates were calculated by multiplying the measured concentrations by the design flow rate. Although the trend of each of these curves demonstrates a decay curve having about the eight-day half life of ^{131}I, there are substantial fluctuations seen in the data. Some of these fluctuations resulted from difficulties in obtaining the samples. However, many of the observed fluctuations most probably reflected real changes in the iodine concentrations at the sample locations.

1.2.3 Normalized Release Rates

A technique for expressing the release rate of radioactivity from a plant is to express it in terms of the concentration of that nuclide in the reactor coolant at the time of the sampling. This concept of normalized release rate was developed (2,3,4) in previous studies of releases from light-water reactors. The use of normalized release rates has two principal advantages in the discussion of data from Three-Mile-Island. Firstly, it allows a better view of the release rates as a function of time because it eliminates the reduction in levels that are due to radioactive decay. Secondly, it removes the dependence of the releases on the condition of the reactor fuel so that plants can be compared one with another. It

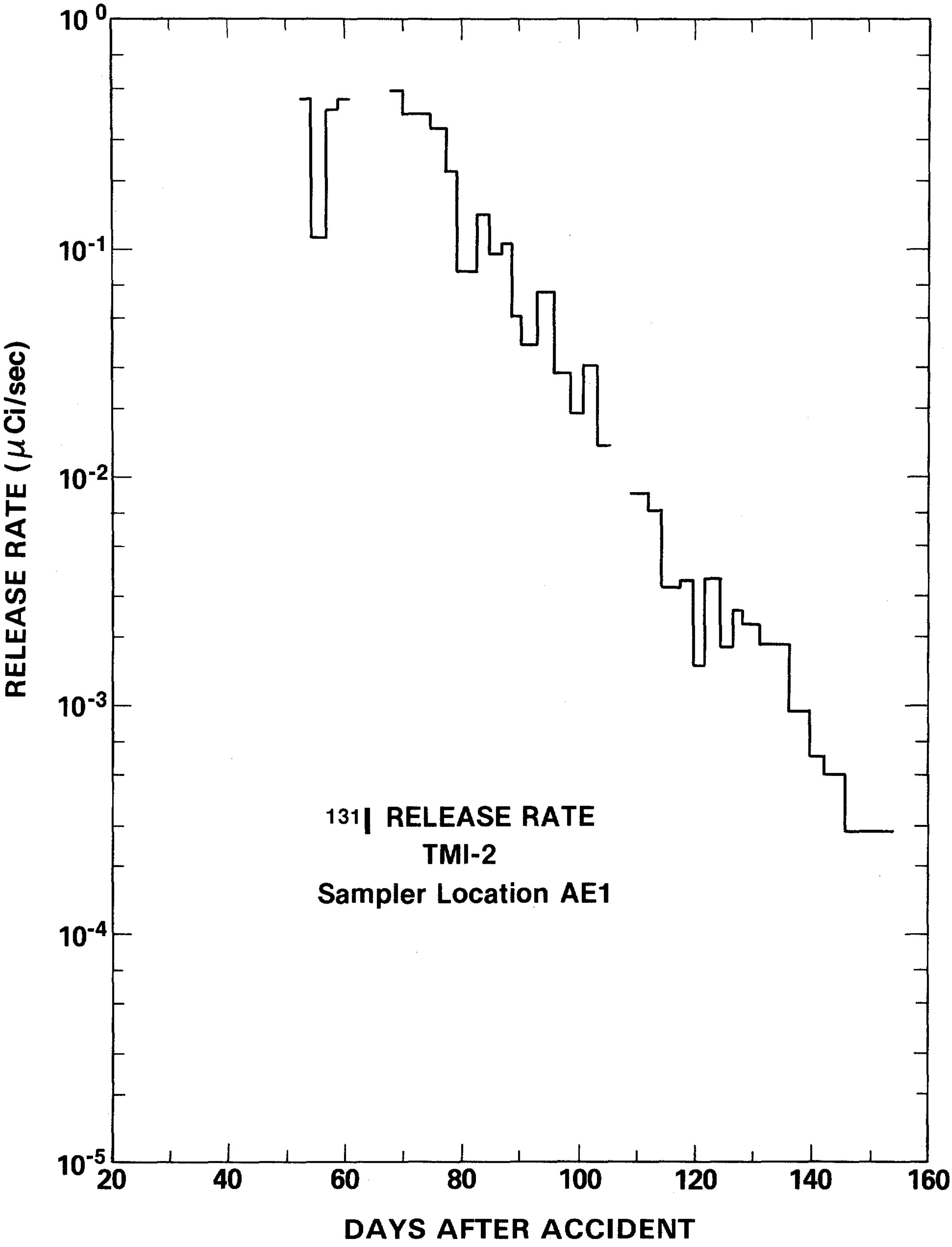

10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
RELEASE RATE (μCi/sec)
20
40
60
80
100
120
140
160
DAYS AFTER ACCIDENT
^{131}I RELEASE RATE
TMI-2
Sampler Location AE1

Figure 1.4

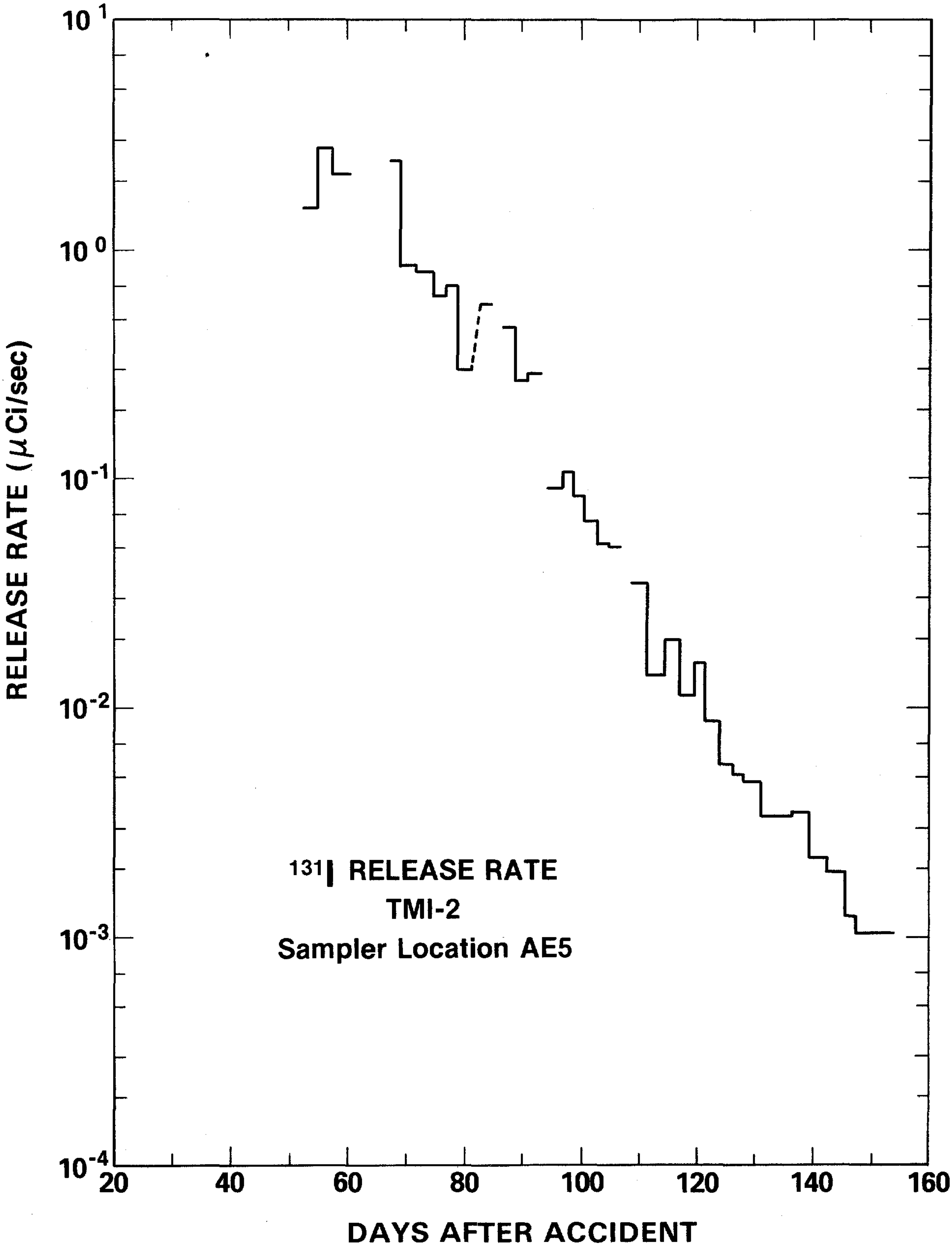

10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
RELEASE RATE (μCi/sec)
20
40
60
80
100
120
140
160
DAYS AFTER ACCIDENT
^{131}I RELEASE RATE
TMI-2
Sampler Location AE5

Figure 1.5

has in the past (2,4) allowed a more reliable identification of sources of airborne radioactive material.

Normalized release rates have been determined for all of the samplers. Two nuclides were treated in the analyses, ^{131}I and the particulate, ^{137}Cs. Figure 1.6 shows the concentration of these two nuclides in the reactor coolant during the sampling period. The ^{137}Cs activity in the coolant decreases with an apparent 78.7-day half life (determined by least-squares fit to the data). This can be explained by the dilution of the coolant by clean makeup water and leakage of mixed coolant into the reactor building sump. The ^{131}I activity decreases with an apparent 7.2-day half life. When the dilution factor is included, the remaining decay term is the proper 8.04-day half life characteristic of ^{131}I.

Figures 1.7 and 1.8, respectively, show the normalized release rates for ^{131}I and ^{137}Cs as a function of time for samples in the input to the fuel-handling and the auxiliary building charcoal filters. These data are presented as typical of all of the samplers. Many of the normalized release rates for ^{137}Cs, although showing considerable fluctuations, show a downward trend whereas many of those for ^{131}I indicate an increase with time. If the ^{131}I data were normalized with a curve having an 8-day decay half life rather than the 7.2-day decay of the coolant, the apparent increase in the ^{131}I normalized release rate would not be there.

1.2.4 Molecular Distributions of Iodine

The SAI iodine molecular species sampler was used continuously at the samplers located at the same two filter inlets and for some samples at other locations.

Organic Iodine Fraction

Figure 1.9 shows the fraction of total airborne iodine that was organic at the inlet to the fuel-handling building filters as a function of time. Figure 1.10 shows a similar distribution as at the inlet to the auxiliary building filters. There is a similarity between these two sets of data; the fraction of total iodine that was organic decreased in both buildings to about 10% by the middle of August. The organic fraction measured in other sampler locations showed similar values, 49 ± 2%.

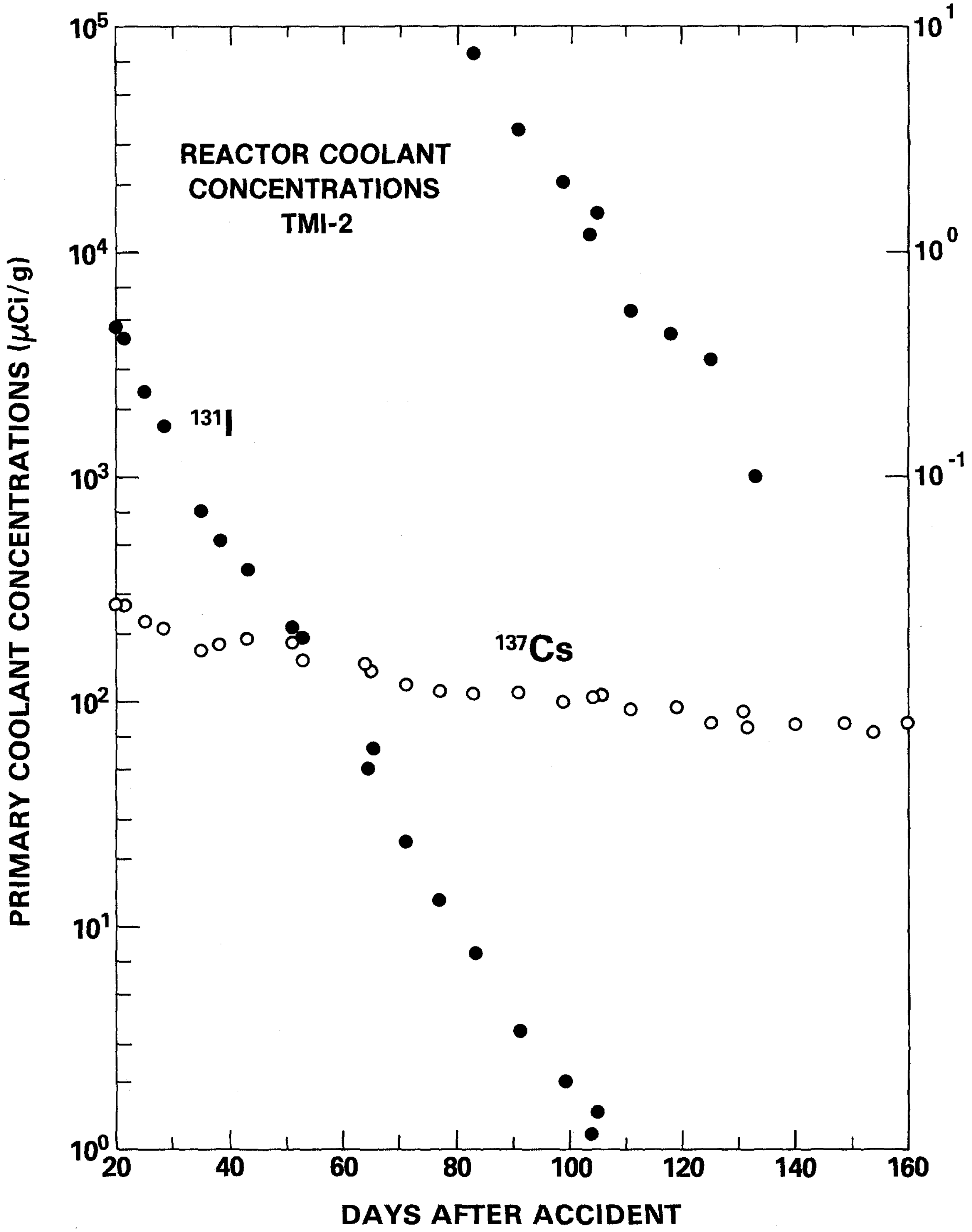

REACTOR COOLANT
CONCENTRATIONS
TMI-2
131I
137Cs
PRIMARY COOLANT CONCENTRATIONS (μCi/g)
DAYS AFTER ACCIDENT
10^5
10^4
10^3
10^2
10^1
10^0
10^1
10^0
10^-1
20
40
60
80
100
120
140
160

Figure 1.6

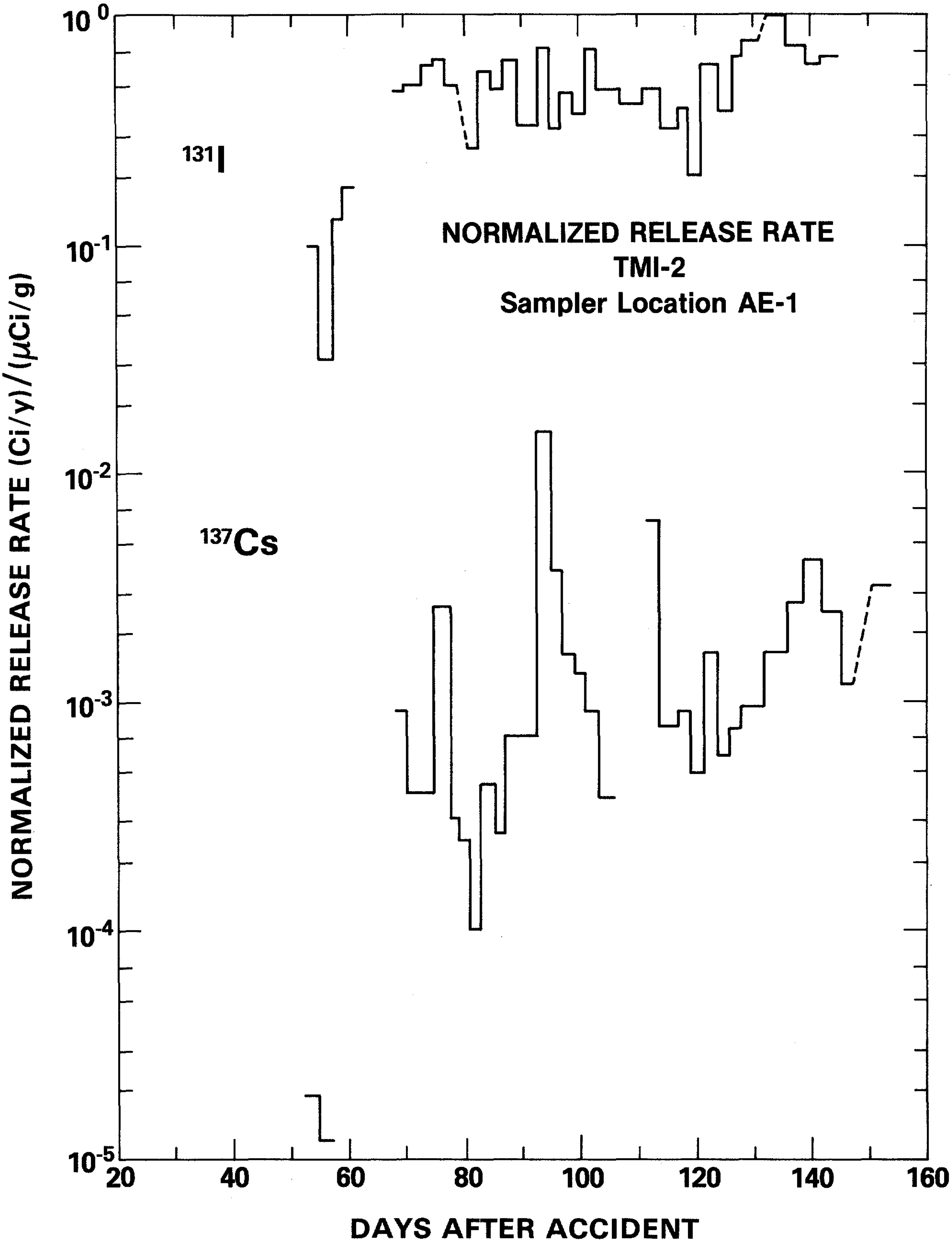

NORMALIZED RELEASE RATE
TMI-2
Sampler Location AE-1
^{131}I
^{137}Cs
NORMALIZED RELEASE RATE (Ci/y)/(μCi/g)
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
20
40
60
80
100
120
140
160
DAYS AFTER ACCIDENT

Figure 1.7

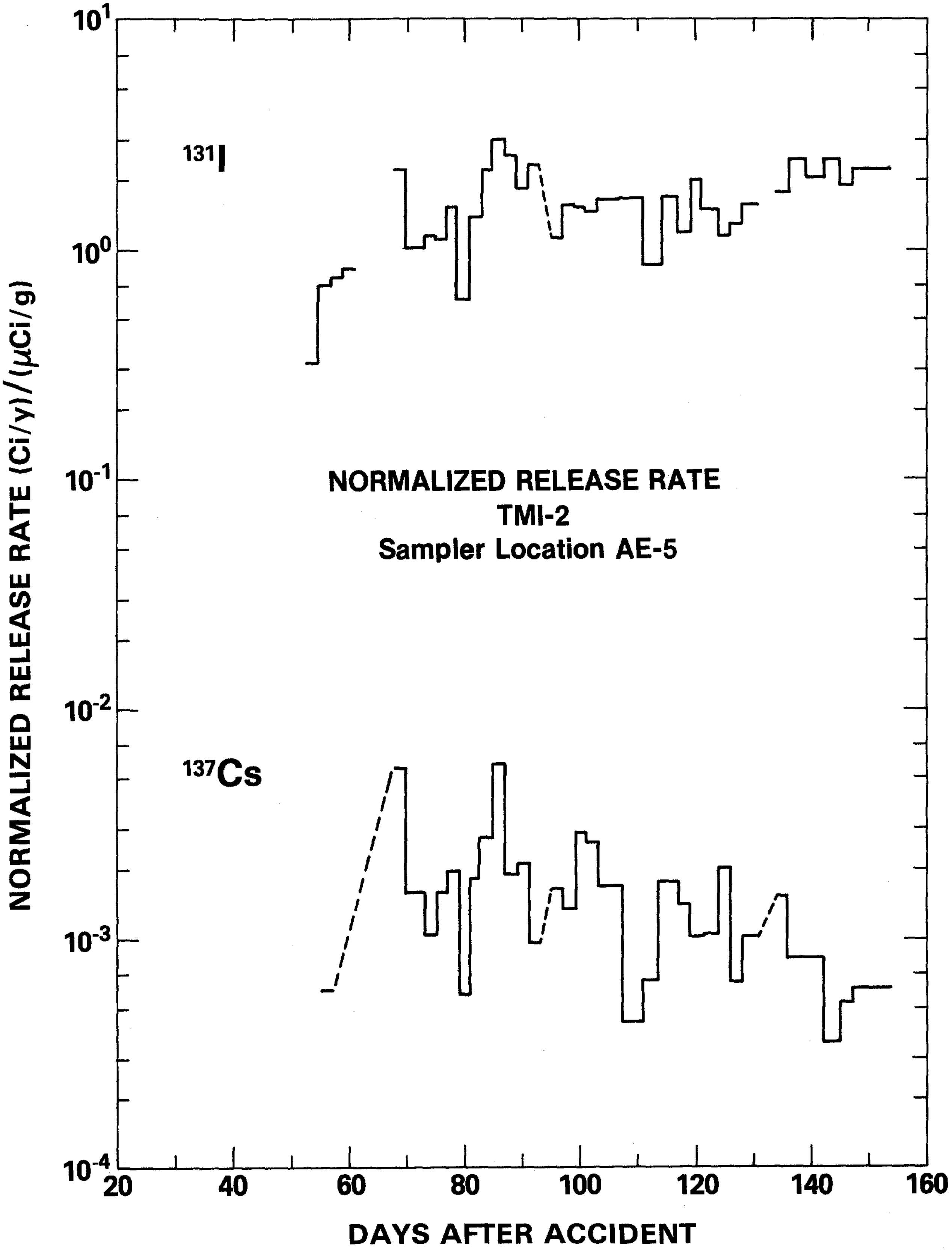

10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
NORMALIZED RELEASE RATE (Ci/y)/(μCi/g)
^{131}I
^{137}Cs
NORMALIZED RELEASE RATE
TMI-2
Sampler Location AE-5
20
40
60
80
100
120
140
160
DAYS AFTER ACCIDENT

Figure 1.8

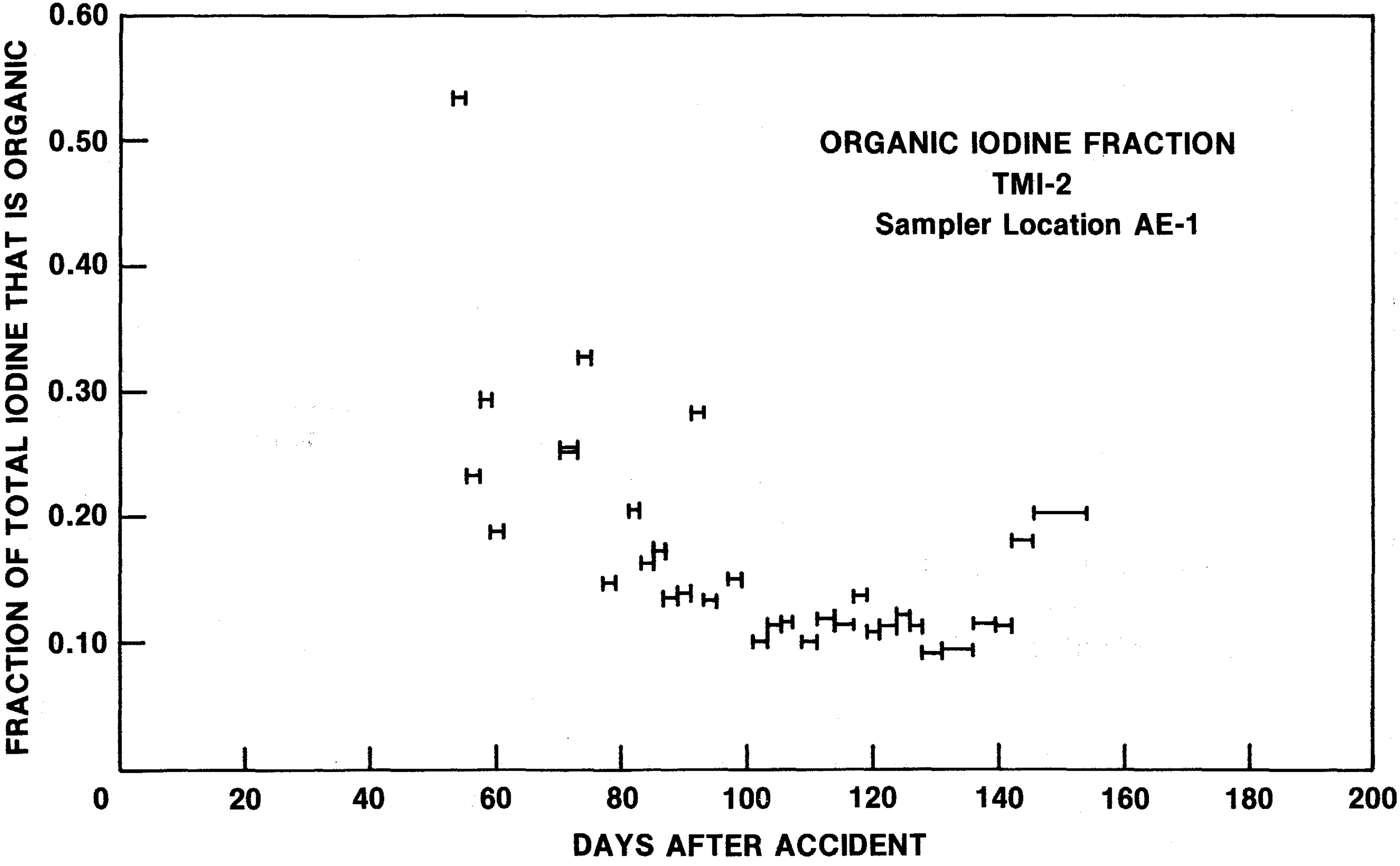

ORGANIC IODINE FRACTION
TMI-2
Sampler Location AE-1
FRACTION OF TOTAL IODINE THAT IS ORGANIC
0.60
0.50
0.40
0.30
0.20
0.10
0
20
40
60
80
100
120
140
160
180
200
DAYS AFTER ACCIDENT

Figure 1.9

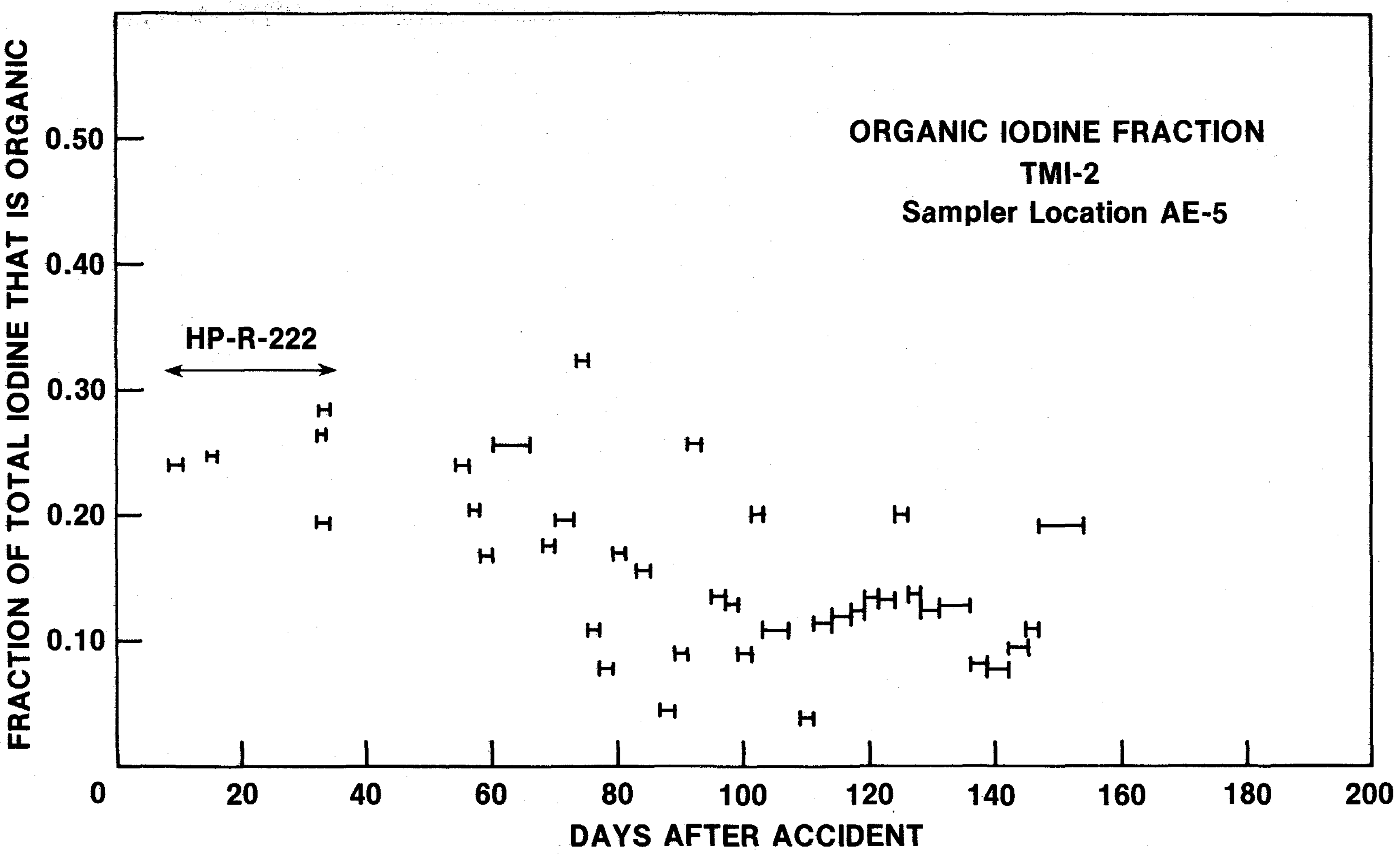

ORGANIC IODINE FRACTION
TMI-2
Sampler Location AE-5
HP-R-222
FRACTION OF TOTAL IODINE THAT IS ORGANIC
0.50
0.40
0.30
0.20
0.10
0
20
40
60
80
100
120
140
160
180
200
DAYS AFTER ACCIDENT

Figure I.10

Inorganic Fraction of Iodine

The inorganic fraction was absorbed as in the past (2,3) by using the cadmium iodide and iodophenol adsorbants. Although the fraction of total inorganic iodine measured was quite consistent between measurements, the relative fraction on the two components varied considerably. (The media used had been previously tested in the laboratory and had been found to perform properly). We have seen considerable evidence in some past studies (5,6) that the species of inorganic iodine generally thought to exist as airborne activity from nuclear plants, elemental and hypoiodous acid, are accompanied by at least one additional form. It is well beyond the scope of the present study to discuss this evidence other than to mention that the observed fluctuations in the TMI data of the split in the inorganic fraction are consistent with the previous evidence and could be explained by the existence of other forms of inorganic iodine.

1.3 SUMMARY

1.3.1 Principal Contributors to Releases

Table 1.1 gives a summary of results from all of the sampler stations used in the attempt to identify sources of airborne radioiodine in the auxiliary and fuel-handling buildings. The table which includes sample number, location, sampling dates, duct flows and average normalized release rates, is organized according to area within the two buildings. The average normalized release rates were calculated as the average rate over the sampling period. Since the average normalized release rates were relatively constant (within a factor of 2) during the sampling periods, the calculation of an average normalized release rate for each sample location is felt to be useful for purposes of comparison of the sample locations. Also included in the table is the percentage of total ventilation flow that each duct represents and the percentage of total release that each duct represents.

Several features are apparent in these comparisons. The auxiliary building as a whole contributes a slightly higher fraction of the total iodine release than of the total ventilation flow. Within the building, the principal contributors were the 328-foot elevation (50% of the total release) and the 281-foot elevation (24% of total release). The 305-foot elevation contributed only 2% of the total release in 25% of the total flow. From the 281-foot elevation, sampler position 19 contributes most of the release. Within the total area being measured by this sample, makeup pump rooms "B" and "C" contribute only about 2.5% which leaves

Table 1.1

SUMMARY OF NORMALIZED RELEASE RATES FROM SAI SAMPLERS AT TMI-2.
INCLUDED ARE THE PERCENTAGES OF TOTAL VENTILATION FLOW AND TOTAL ^{131}I RELEASE FOR COMPARISON

Sample #	Location	Sampling Dates	Duct Flow (CFM) (Design Flow)	Percent Of Total Flow	^{131}I Average Normalized Release Rates (Ci/y/μCi/g)	Percent Of Total Release
	Auxiliary Building					
AE-5	Total Aux. Bldg. Input to Filters	5/19-9/4/79	65000	65.0	1.57	76.1
	328' Total Exhaust (No Sampler-Results by difference)	--	30000	30.0	(1.03)	49.9
13	Duct from large open area of the 328' level	6/2-9/4/79	13800	13.8	9.68(-2)	4.7
	305' Total (No Sampler-Results by difference	--	25000	25.0	(4.6(-2))	2.2
2	(305' + 281') Total Exhaust	5/11-9/4/79	35000	35.0	0.544	26.4
6	Make-up tank room	5/16-6/25/79	1000	1.0	7.61(-3)	0.4
3B	281' Total Exhaust	5/26-9/4/79	10000	10.0	0.498	24.1
10	Decay Heat Vault, Reactor Bldg. Spray Pumps	6/3-6/13/79	650	0.7	3.81(-3)	0.2
11	Reactor Coolant Bleed Tank System, Clean-up demineralizers	6/3-9/4/79	4740	4.7	9.04(-2)	4.4
12	Aux. Bldg. Sumps, Valve rooms, pump room	6/3-6/13/79	1730	1.7	2.76(-3)	0.1
5	Aux. Bldg. Sump tanks	5/19-5/31/79	540	0.5	3.68(-3)	0.2

TABLE 1.1
(continued)

19	Reactor Coolant Make-up pumps "B", "C", Let-down system pipe chase and valve room	7/13-9/4/79	1455	1.5	0.515	25.0
17	Reactor Coolant Make-up pump room "C" room air	6/17-9/4/79	530	0.5	9.32(-3)	0.5
16	Reactor Coolant Make-up pump room "B" room air	6/17-9/4/79	525	0.5	4.39(-2)	2.1
7 15	Reactor Coolant Make-up pump room "A" exhaust air	5/19-5/29/79 6/17-9/4/79	525	0.5	1.71(-2)	0.8
	<u>Fuel-Handling Building</u>					
AE-1	Total Fuel Handling Building Input to filters	5/19-9/4/79	35000	35.0	0.493	23.9
14	Reactor annulus - pipe chase at 305'	6/2-9/4/79	900	0.9	1.77(-2)	0.9
18	Reactor annulus - pipe chase at 281'	6/23-9/4/79	900	0.9	7.22(-3)	0.3
4	Make-up and purification valve room	5/12-7/9/79	130	0.1	4.49(-3)	0.2
3A	281' Total Exhaust	5/17-5/26/79 7/9-9/4/79	3045	3.0	6.34(-2)	3.1

about 22.5% of the total released activity as coming from the letdown system pipe chase and valve room. Makeup pump "B" has been running nearly continuously since the accident and has a known small leak in one of the seals. Hence, the relatively large contribution from this pump room (2.1% in 0.5% of the flow) was expected. The auxiliary building sump system was not a significant contributor to the release of activity.

The main conclusion is that although there are areas of the auxiliary building that were contributing substantially to the release of radioiodine (such as the letdown pipe chase and valve room), the radioiodine was liberally distributed throughout the building, probably being evolved from the surfaces of walls, floors, ceilings and machinery.

1.3.2 Comparison of Normalized Release Rates from TMI with those of Previous Studies

The combined average normalized release rate of ^{131}I from the auxiliary and fuel-handling buildings to the input to the charcoal filters is 2.1 (Ci/year)/(μCi/g). This value can be compared with a value of 16.4 (Ci/year)/(μCi/g) obtained in previous studies (4) at TMI, Unit 1, or 15.5 (Ci/year)/(μCi/g) obtained at Calvert Cliffs. These latter two figures represent yearly averages and include periods of operation as well as shutdown modes. The mode of operation considered most comparable to the abnormal conditions at TMI, Unit 2 is what was defined in reference 3 as "mode 3", a period that includes periods of cold and hot shutdowns but does not include any refueling operations. For this mode, the average normalized release rate at TMI, Unit 1 (4) for auxiliary building inlet to the filters was 4.2 (Ci/y)/(μCi/g), a value quite comparable to the value of 2.1 for Unit 2.

Table 1.2 provides a summary comparison of normalized release rates from TMI-2 to those from three other PWR's that were studied previously. The comparison is for ^{137}Cs as well as for ^{131}I. Two conclusions are apparent from the table: (1) the releases of ^{131}I in the auxiliary and fuel-handling buildings of Three-Mile-Island, Unit 2 during the period following the accident when expressed in terms of normalized release rates are very comparable to those from other PWR's during normal initial shutdown conditions and (2) the level of ^{137}Cs releases at TMI-2 were substantially lower during this same period when expressed in the same manner.

Table 1.2

SUMMARY OF AVERAGE NORMALIZED RELEASE RATES FROM TMI-2 DURING THE PERIOD MAY TO SEPTEMBER, 1979. AVERAGE RATES FROM THREE OTHER PWR'S ARE INCLUDED FOR COMPARISON

AUXILIARY BUILDING FILTER INLET

Nuclide	TMI Unit 2	TMI Unit 1	Calvert Cliffs 2)	Ginna 3)
^{131}I	1.57	2.02	1.78	1.47
^{137}Cs	0.0016	0.103	0.0088	0.13

FUEL-HANDLING BUILDING FILTER INLET

Nuclide	TMI Unit 2	TMI 1) Unit 1	Calvert Cliffs 4)	Ginna 4)
^{131}I	0.49	2.19	--	--
^{137}Cs	0.0014	0.018	--	--

Notes

1. Mode 3 from reference 4.
2. Mode 2 (operational plus hot shutdown), no mode 3 data on Auxiliary Building total at this plant.
3. Sum of mode 3 sampler areas 1, 4 and 5 reported in reference 4.
4. No distinct building as such at these plants.

1.4 DISCUSSION

Interpretation of the data was neither simple nor straightforward. None-the-less, although not unique the following provides, in our judgment, the most likely scenario of iodine release that is consistent with the results.

Thesis

Iodine was most probably being released into the ventilation system of the auxiliary and fuel-handling buildings by resuspension from the surfaces of walls, floors, ceilings and machinery. Furthermore, there are indications that the bulk of this iodine was deposited during the early part of the accident, possibly during the first day.

Facts in Support of the Thesis

1. The normalized release rates of iodine increase slightly during the measurements. This indicates that the release rates do not correlate with the reactor coolant that is decaying with an apparent seven-day half life. Airborne iodine levels decayed with the eight-day half life characteristic of ^{131}I. Hence, the emission rate of iodine was not associated with the current concentration in the coolant.

2. Normalized release rates of iodine and cesium are not correlated to each other nor is the cesium release rate correlated well to the coolant concentration. Cesium, being a particulate material rather than a volatile substance like iodine, would not be expected to have the same resuspension mechanism as iodine. The cesium may be resuspended due to the cleaning operations in the buildings.

3. The normalized release rate for cesium is much lower than that found in other operating PWR's and the release rate decreases with time. If the reservior of activity available for resuspension was deposited during the early portion of the accident, the cesium activity on surfaces may have been depleted by previous resuspension and by the cleanup operation in the building. Deposition of iodine, however, has been found in previous studies (7) to be very extensive and resuspension would not be expected to deplete the reservior in any short time.

The main release of iodine into the building from the reactor system could have occurred during March 28 during the time when the reactor primary coolant was a two phase system. In this case, the carry over of cesium into the buildings through the letdown system would be proportionally less than in normal operation since the transfer into the steam generator and the letdown system would have been via the steam phase.

4. The ventilation systems for the auxiliary and fuel-handling buildings were turned off during the 28th of March. The fuel-handling building was stagnant for 220 minutes during the morning and afternoon. Stagnant air would increase greatly (7) the deposition of iodine. This would also cause the iodine to deposit more uniformly throughout the building.

5. The molecular species distribution of ^{132}I and ^{131}I in the auxiliary building air on April 7 and again on April 13 were substantially different. Whereas ^{131}I showed about 30% organic component, the ^{132}I was about 90% organic. Thus ^{132}I and ^{131}I had different sources. By the time of the measurement, the 2.3-hour ^{132}I activity had its origin as the daughter of 78-hour ^{132}Te. It is assumed that little or no Te activity has escaped the reactor vessel even in the coolant and that the sources of any ^{132}I activity was the reactor coolant water (having been injected in the water by the decay of ^{132}Te). Hence, the ^{132}I was probably from fresh coolant leaks and the ^{131}I was from an older inventory.

Facts Possibly in Conflict with the Thesis

1. Measured molecular species of ^{131}I at TMI were predominantly inorganic. In past studies (2,3) the presence of inorganic iodine has indicated "fresh" sources of iodine rather than sources resuspended from surfaces. It is possible, however, that the observed species distribution was caused by the caustics and thiosulfates used throughout the buildings in attempts to stabilize the emission of iodine or by the chemicals used in the cleaning operations. Iodine concentration and pH have been shown (5) to have substantial effects on both the rate and molecular form of iodine emanation.

2. There were relatively large fluctuations in the measured iodine concentrations between measurements. Resuspended iodine should be expected to show a more regular release rate. As was stated in Section 1.0, some of these fluctuations resulted from difficulties in the sampling. The remainder of the fluctuations are of unknown origin. Possible explanations that are still consistent with the thesis being proposed are: (1) fluctuations in the ventilation flow rates or air currents and (2) effects caused by local temperatures, relative humidities or chemical fumes from decontamination efforts.

Section 2

PERFORMANCE OF CHARCOAL ADSORBERS

2.1 AUXILIARY BUILDING VENTILATION EXHAUST

2.1.1 Description of Adsorbers

There are two banks of charcoal in service at TMI-2. Each bank contains 90 trays of charcoal with an average weight of 34 Kg. The effective bed depth of the system is two inches with a total area of 1200 square feet (both banks combined). Originally the system was loaded with charcoal impregnated with KI_3. After April 20, 1979, bank "A" was loaded with a charcoal having an impregnant containing a mixture of KI and TEDA.* On April 24 bank "B" was put into service with 79 of the trays containing KI_3 impregnated charcoal and 11 trays of KI, TEDA impregnated charcoal.

2.1.2 Sampling Program

Independent samplers were installed before and after the filters. AE-5 sampled a plenum into which all air was exhausted prior to its entering the filters. Samplers AE-4 and -6 were installed to sample air down stream of the filters. It should be re-emphasized that throughout the measurement period working conditions were less than ideal. In a study of this nature carried out under more normal circumstances, the samplers would have been verified for representativeness prior to the start of sampling. This was not feasible at TMI and representativeness checks were not started until August 1979 and not completed until December. These checks are made by releasing helium at a known rate into the ventilation exhaust system ahead of the particular sampling point and measuring the concentration at the sampler location.

All sampling points under-responded to releases from the 281' level and over-responded to releases from the 305' level. While responses to releases from various levels are interesting, and show that the air in the ducts was unmixed, they are

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* Provided by Nuclear Consulting Services, Inc., Columbus, Ohio.

not of direct concern in measuring filter performance. What is important is the relative response of samplers before and after the filters. If AE-5 is taken as 1 (inlet) then the response of the average of AE-4 and -6 (outlet) was 1.3, 1.4 and 0.92 for releases from the 281-foot, 305-foot and 328-foot levels. The response of the outlet monitor relative to the inlet monitor was 1.05, 0.33 and 4.0 for releases on the 281-foot, 305-foot and 328-foot levels.

The AE samplers were installed late in May and it was not until May 24 that a complete set of samples was collected. Initially, the sampling period was two days. As iodine concentrations decreased, the sampling period was increased. Most samples measured the chemical form of iodine. All samples were counted by SAI using Ge(Li) detectors.

2.1.3 Adsorber Performance and Discussion

Decontamination factors (DF) from the AE samplers, defined as the ratio of the inlet to the outlet iodine concentrations, were computed for each period. Figure 2.1 shows the DF for organic and inorganic iodine plotted as a function of time. The DF for inorganic iodine appeared to decrease with time until the end of June when it leveled off at approximately twenty five. Except for the high DF measured from June 6 to 13 the DF for organic iodine remained relatively constant. The average DF not including the high value was seventeen. It should be kept in mind that because of the non mixing problems described above the outlet concentrations may be biased high in relation to the inlet concentration. Depending on the source of the iodine the real DFs could be as much as 1.4 times higher.

The DFs as measured by the HPR-222 and 228 monitors were much lower than those measured by the AE samplers. This was due mainly to the fact that the concentrations measured by HPR-222 were much lower than those measured by AE-5. This finding supports other evidence given below that the majority of the iodine was released from the 281' and 328' levels.

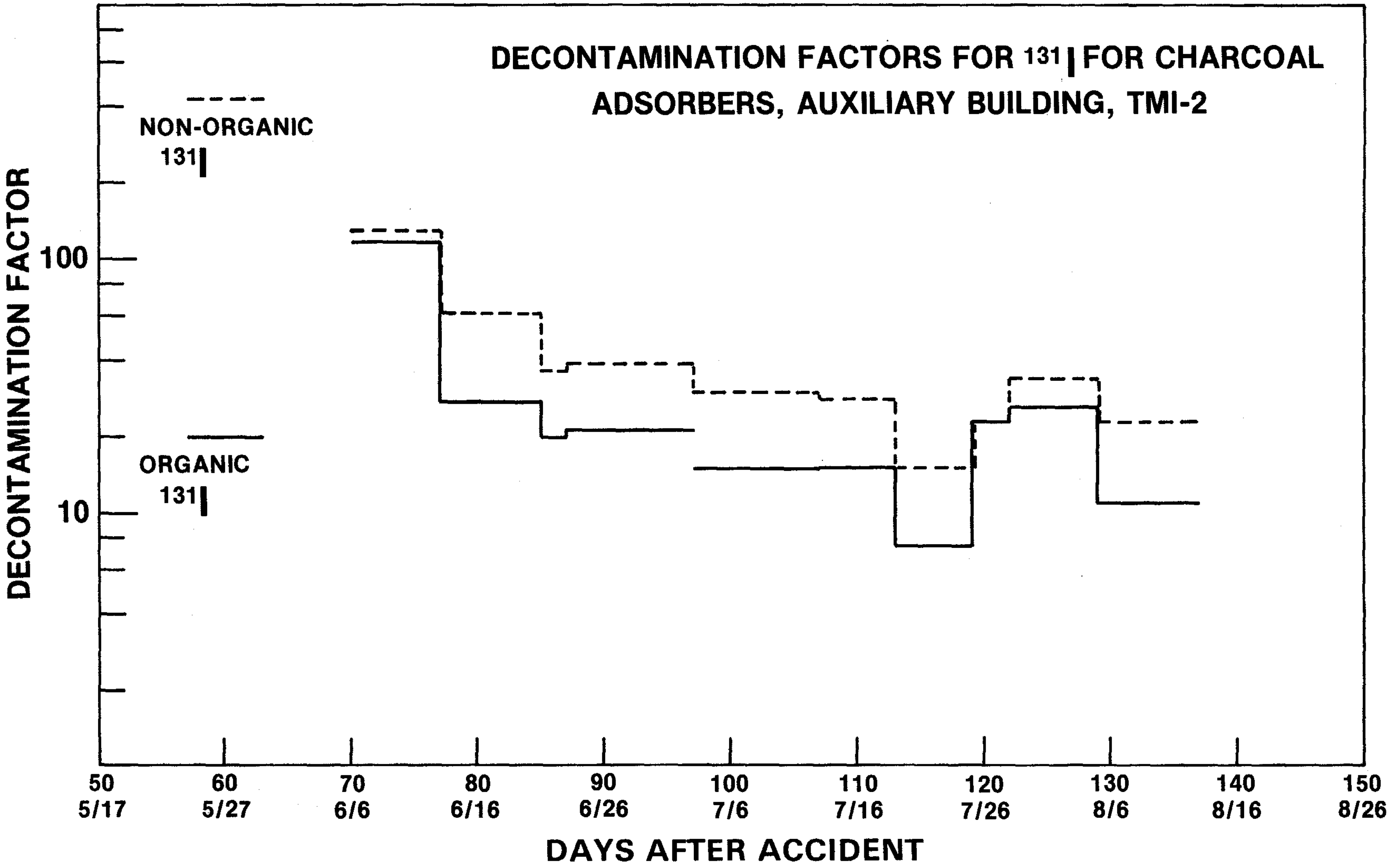
DECONTAMINATION FACTORS FOR 131I FOR CHARCOAL ADSORBERS, AUXILIARY BUILDING, TMI-2
NON-ORGANIC 131I
ORGANIC 131I
DECONTAMINATION FACTOR
100
10
50 5/17
60 5/27
70 6/6
80 6/16
90 6/26
100 7/6
110 7/16
120 7/26
130 8/6
140 8/16
150 8/26
DAYS AFTER ACCIDENT

Figure 2.1

2.2 FUEL-HANDLING BUILDING VENTILATION EXHAUST

2.2.1 Description of Adsorbers

As in the case of the auxiliary building, there are two banks of adsorbers for the fuel-handling exhaust. Bank "A" was changed out with KI, TEDA loaded charcoal on April 24, 1979. Bank "B" was changed out with KI, TEDA loaded charcoal on May 23, 1979. Each bank was composed of 60 trays each with an average weight of charcoal of 34 Kg.

2.2.2 Sampling Program

Plant monitors up and down stream were designated HPR-221A and 221B. In addition, independent samplers were installed. Sampler AE-1 measured ^{131}I concentrations upstream and samplers AE-2 and -3 measured ^{131}I concentrations downstream. For the same reasons as for monitors HPR-222 and 228, ^{131}I concentrations measured by HPR-221A and 221B are biased somewhat low. However, this inaccuracy was small in relation to the sampling biases measured.

The fuel-handling building ducts also were not mixed. The downstream sampler HPR-221B over responded relative to the upstream sampler HPR-221A for releases at all three levels. The over responses were 1.4, 3.2 and 2.7 for releases from 328', 305' and 281' levels, respectively. There were similar problems with the AE samplers; the ratio of the average outlet response (average of AE-2 and -3) and the inlet response was 0.46 and 1.5 for releases from the 328' and 281' level.

2.2.3 Adsorber Performance and Discussion

Figure 2.2 shows the DFs for the fuel-handling building adsorber computed from the concentrations measured by the AE samplers. The initial value was quite low and may reflect the recent change out of the charcoal in bank "B". Daily measurements at the HPR-221A and 221B locations showed that the DF did not start to increase substantially until two days after the May 23 change out date. And then it increased steadily until June 1. Starting with the measurement on June 6, the measured DF for organic iodine was approximately fifteen. Subsequent measurements were lower and there may have been a slow downward trend. Excluding the initial measurement, the average DF for organic iodine was eight which is approximately 1/3 the average DF for organic iodine for the auxiliary building filters. The DF for inorganic iodine was higher than that for organic. Toward the end of the measure-

DECONTAMINATION FACTORS FOR ^{131}I FOR CHARCOAL ADSORBERS, FUEL HANDLING BUILDING EXHAUST AND TOTAL VENTILATION EXHAUST, TMI-2

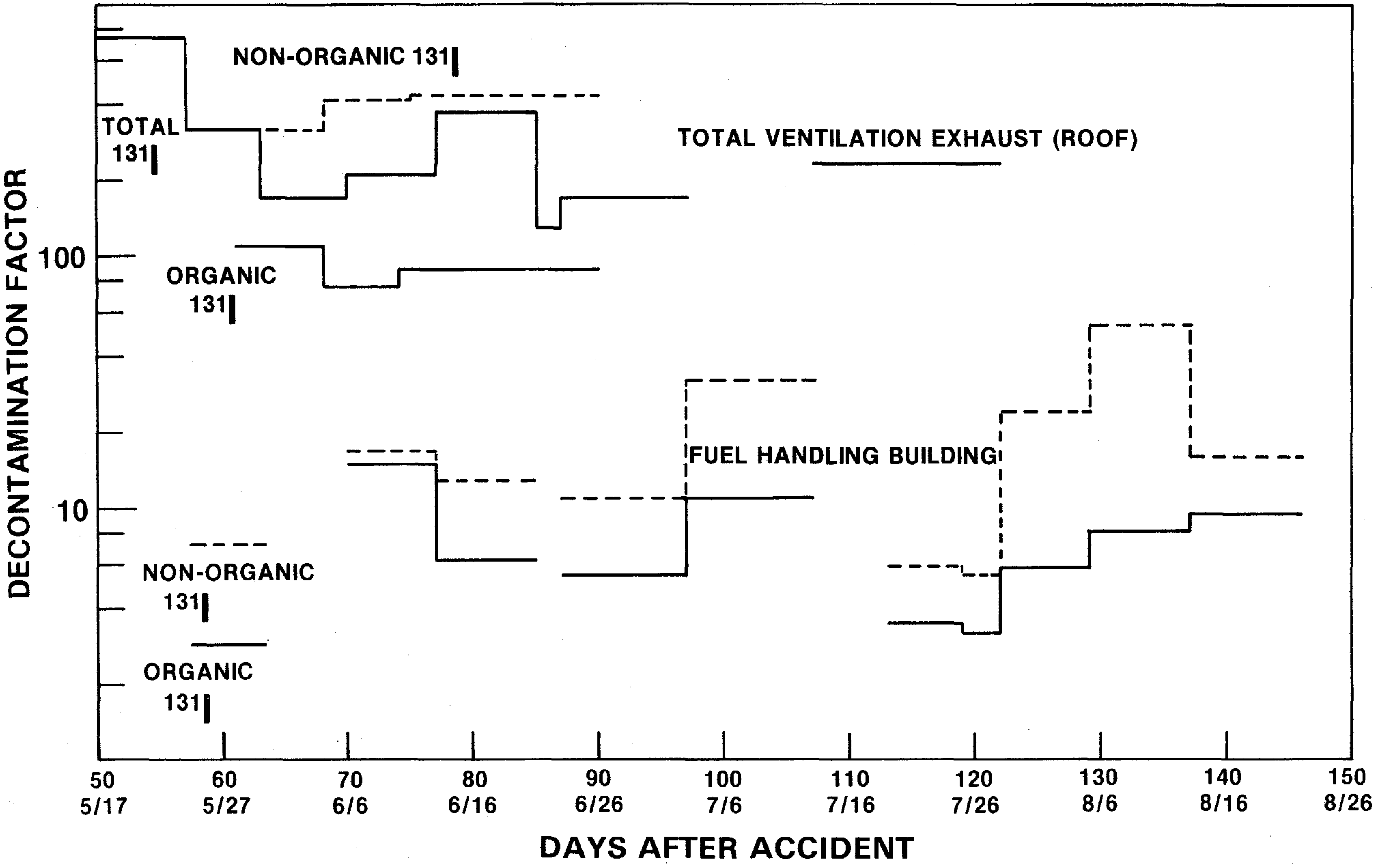

Figure 2.2

ment period it ranged from sixteen to fifty-three, which is in the same range as that for the auxiliary building.

2.3 AUXILIARY BUILDING ROOF ADSORBERS

2.3.1 Description of Adsorbers

On May 17 the unit vent was capped and the ventilation exhaust from the auxiliary and fuel-handling building, coupled with the ventilation exhaust air from the control and service building, was passed through charcoal adsorbers which had been installed on the roof. The supplemental adsorbers had an effective bed depth of 2" and the charcoal was impregnated with KI_3. There were four sampling program trains installed and normally only three were used. The four trains had a common inlet which was monitored. The monitor had the designation AM-5. The effluent from each train was monitored. The sampling line for each monitor was provided with a "T" which was used to collect species samples. However, during the early days of measurement, when construction activities were still in progress, we found it impossible to collect our own samplers consistently. Therefore, the concentrations used in this report are those measured on charcoal filters used in the plant monitors. The flow rate of each monitor was approximately three CFM. Sampling times ranged from one day in the beginning when concentrations were relatively high to five days or longer toward the end of the measurement period when the concentrations were low.

2.3.2 Adsorber Performance and Discussion

Figure 2.2 shows the DF for total ^{131}I as a function of time. Also included are the DFs for organic and inorganic iodine measured from May 28 through June 25. After May 24 there appears to have been little decrease, the average DF being 230. Over the period May 28 to June 25 when the DF for total ^{131}I was 210, the average DF for organic and inorganic ^{131}I was 88 and 400, respectively. These are substantially higher than the comparable DFs for the auxiliary and fuel-handling building filters.

We can only speculate as to why the roof adsorbers performed so much better than the inside adsorbers. It may be that KI_3 charcoal has a greater efficiency for removing iodine than KI, TEDA at least in the early life of the charcoal. Another possible explanation concerns differential weathering of the charcoal. It is well established (8) that contaminants in air can, over a period of time, degrade the iodine adsorption ability of charcoal. It may be that contaminants are being adsorbed on the auxiliary and fuel-handling building charcoal and thereby degrading its performance. However, except for the 7,000 cfm from the control and service buildings, the roof adsorbers are exposed to air which has had most of the contaminants removed and therefore have not weathered as fast.

Section 3

MEASUREMENTS OF ^{129}I IN TMI CONTAINMENT DURING AND AFTER THE VENTING

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 (1,3,10). A mathematical model (10) was derived to explain this behavior. 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 (7). 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 non-elemental forms of iodine and tested with radioiodine measurements in containment atmospheres at operating PWRs. 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.

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 (10) 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 (10) taken on and following 31 March

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 (11) made 26 June and 9 July 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 (2) of iodine that has been "aged" through deposition and resuspension.

3.2 MEASUREMENTS

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

3.2.1 Sampling System

The radioiodine 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 meters (10.5 feet) above the refueling floor.

The sampler pulled the containment atmosphere through a 1-cm diameter, 241-cm long stainless steel tubing that extended about 0.7-meter 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.

The iodine sampler was the SAI radioiodine molecular species sampler whose properties are described in Reference 2. This sampler has five components placed in 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 assure no break through of iodine in the sampler.

3.2.2 Containment Venting

The main containment venting began 28 June and ended 11 July 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 8 July used the main venting lines that exhaust from the "347'" level, the refueling floor. Venting was done cautiously with considerable starting and stopping. Although the main purge ended on 11 July, there were additional short purges on 1 August, 8 August, 14 August and 22 August. A plot of the purging that includes these periods is shown in Figure 3.1.

3.2.3 Data 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 non-destructively 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.

3.2.4 Results

Table 3.1 gives the analysis results in terms of microcuries of ^{129}I per cubic centimeter of containment air for each sampling period. Values were obtained for each component of the sampler as well as the total concentration. The table also contains columns that list the percentage of the total activity on each cartridge. Figure 3.2 shows a plot of the ^{129}I concentration as a function of the sampling date. For comparison, Figure 3.3 shows a plot of the ^{85}Kr concentrations during the same period.

3.3 DISCUSSION

The concentration of ^{129}I in the TMI-2 containment atmosphere before the purge was measured (14) to be about 7.5×10^{-11} μCi/cc. This value compares favorably with the expected value of 6.7×10^{-11} μCi/cc derived using earlier measured values of ^{131}I and the relative isotopic concentrations of iodine in the core 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}K concentration by about a factor of 50000. 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.

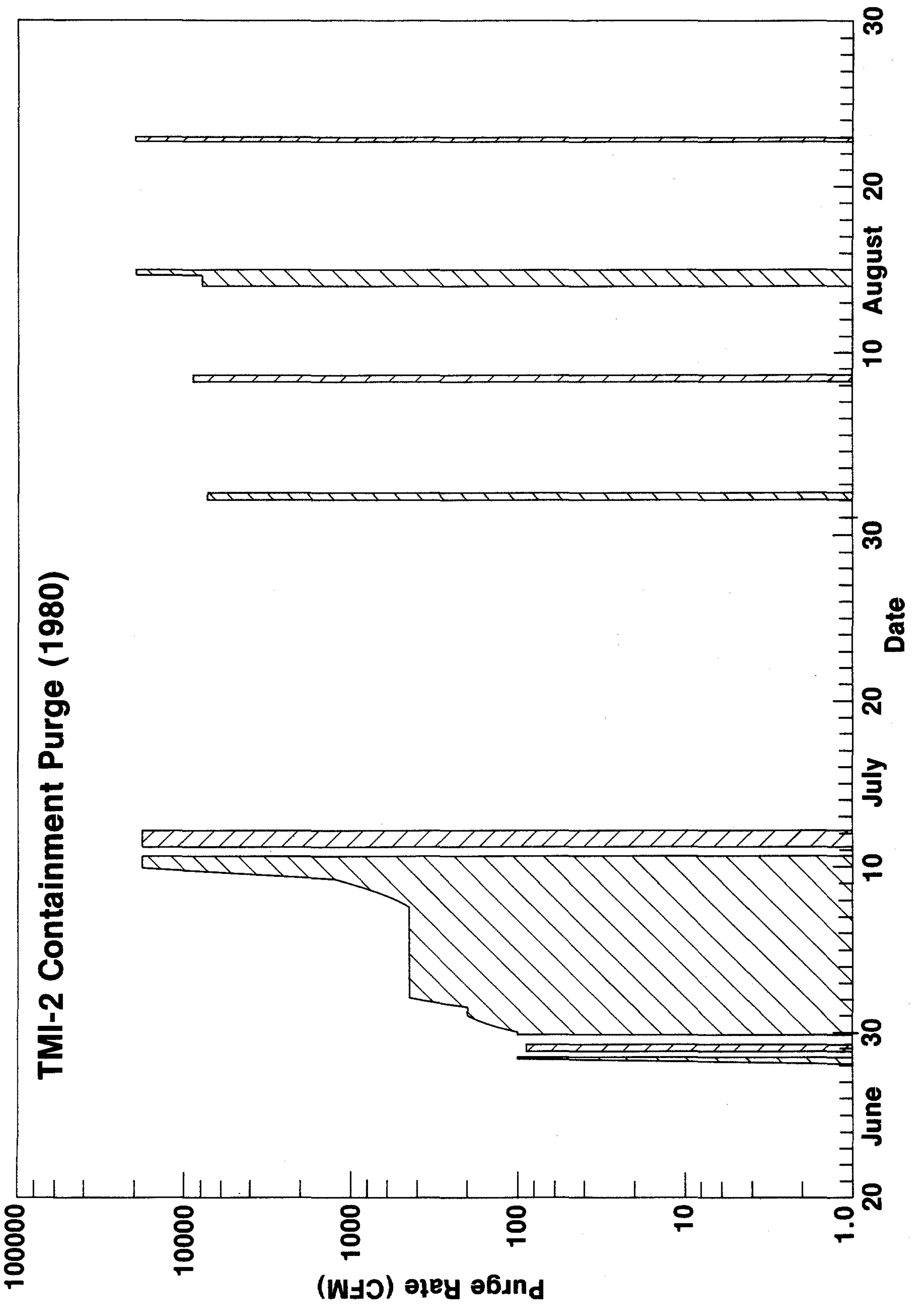
TMI-2 Containment Purge (1980)
Purge Rate (CFM)
100000
10000
1000
100
10
1.0
Date
20
June
30
10
July
20
30
10
August
20
30

Figure 3.1

Table 3.1

IODINE 129 CONCENTRATIONS FOR TMI-2 CONTAINMENT DURING VENTINGS
SAMPLES TAKEN FROM R-626 PENETRATION

	Concentrations (μCi/cc)				Percentage of Total on Each Cartridge		
Period (1980)	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

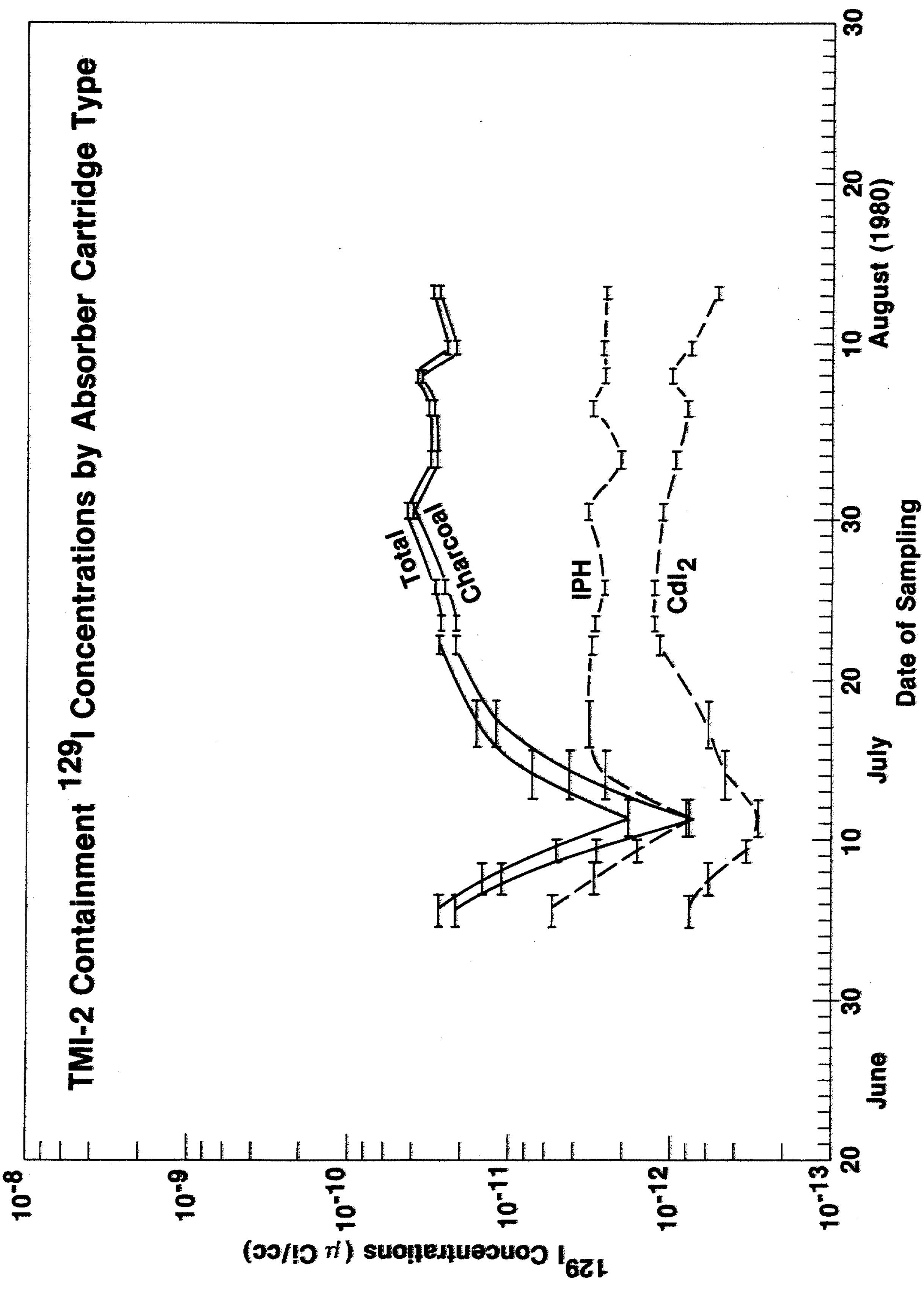
TMI-2 Containment 129I Concentrations by Absorber Cartridge Type
Total
Charcoal
IPH
CdI2
129I Concentrations (μ Ci/cc)
10-8
10-9
10-10
10-11
10-12
10-13
20
30
10
20
30
10
20
30
June
July
August (1980)
Date of Sampling

Figure 3.2

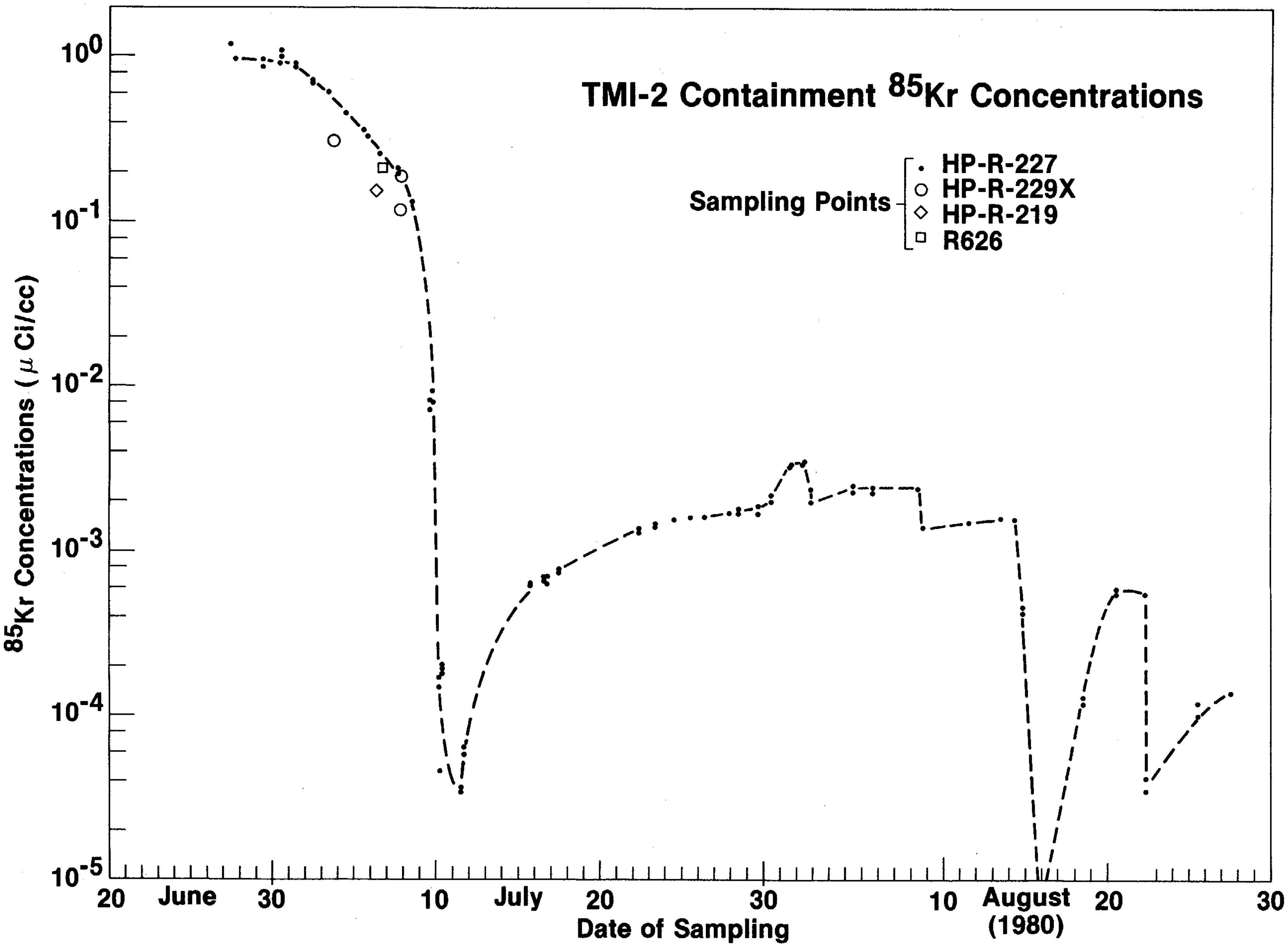
TMI-2 Containment ^{85}Kr Concentrations
Sampling Points
HP-R-227
HP-R-229X
HP-R-219
R626
^{85}Kr Concentrations (μCi/cc)
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
20
June
30
10
July
20
30
10
August
(1980)
20
30
Date of Sampling

Figure 3.3

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 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). Hence, the equilibrium value on the scale of the present measurements is estimated to be about 4×10^{-11} μCi/cc. 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 (9) and deposition and resuspension coefficients for surfaces (7) for the different forms of iodine. Quantitative explanations are being attempted and will be reported in future reports.

REFERENCES

1. ^{131}I Studies at TMI-Unit 2, J. E. Cline, et al., EPRI NP-1389, April 1980.

2. Sources of Radioiodine at Boiling Water Reactors, EPRI NP-495, February 1978, C. A. Pelletier, E. D. Barefoot, J. E. Cline, R. T. Hemphill, W. A. Emel, and P. G. Voilleque´.

3. Sources of Radioiodine at Pressurized Water Reactors, EPRI NP-939, November 1978, C. A. Pelletier, J. E. Cline, E. D. Barefoot, R. T. Hemphill, P. G. Voilleque´ and W. E. Emel.

4. Airborne Particulate Releases from Light-Water Reactors, EPRI NP-1013, March 1979, J. E. Cline, C. A. Pelletier, J. P. Frederickson, P. G. Voilleque´ and E. D. Barefoot.

5. Nuclear Power Plant Related Iodine Partition Coefficients, Final Report to EPRI Project RP 962-1, C. A. Pelletier and R. T. Hemphill, October 1979.

6. Radioiodine Emissions from LWR's - Assessment of Exposure Problems, private communication from J. E. Cline from proposal submitted to U. S. Department of Energy.

7. Surface Effects in the Transport of Airborne Radioiodine at Light Water Nuclear Power Plants, EPRI NP-876, September 1978, R. T. Hemphill and C. A. Pelletier.

8. "Effects of Weathering on Impregnated Charcoal Performance," Victor R. Deitz, NUREG/CR-0771, May 10, 1979.

9. Nuclear Power Plant Related Iodine Partition Coefficients, R. Hemphill, et al, EPRI NP-1271, December 1979.

10. Private Communication, SAI and NSAC, C. A. Pelletier and A. Miller.

11. Reactor Containment Species Samples, private communication, P. G. Voilleque´ to C. Montgomery, 11 July 1979.

12. NSAC EPRI ORIGEN CODE Calculation of the TMI-2 Fission Product Inventory, R. G. Canada, Technology for Energy Corporation Report R-80-012, May 1980.

13. Containment Sump Sample Analyses, private communication from R. E. Brooksbank (ORNL) to B. Rusche (Met. Ed.), 12 September 1979.

14. This value was reported by EG&G, Idaho and Exxon Nuclear, Idaho from their measurements, J. Mandler, personal communication, July 1980.

DISCUSSION

KOVACH: I have two questions. The first is, do you know if the filters which were reinstalled in the Fuel Handling and Auxiliary Building were tested in place after the reinstallation? And I have the same question for the group filters: were they in-place tested after installation to evaluate for by-passers?

COLLINS: The filters were not in-place tested after installation. Since there were high radiation readings in and around the filter enclosures, I made the decision not to require the licensee to in-place test the filters. You must also recognize that the effluent from these systems was further cleaned up by the supplemental system which was installed shortly after the accident and which was in-place tested prior to operation.

KOVACH: I would like to make clear that while they were in operation, the two systems were always in series.

CLINE: That is correct, and is most likely the reason that the second filter bank had the higher efficiency.

COLLINS: It looks to me like the evidence is that a pre-filter helps. That will make Mr. Wilhelm very happy.

RADIOIODINE ADSORPTION GEOMETRY EFFECTS

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Abstract

Some effects of, and information obtainable from, geometric considerations of the internal geometry of an adsorption bed are the subject of this paper. Two major areas are covered. First, the capture/release data obtainable with proper analysis of a heavily loaded (with I131) bed. Second, the effects of sampling adsorbents for the standard ASTM D3803-79 efficiency tests relative to preserving the internal geometry of the sample vs a homogeneously mixed sample.

Analysis of highly contaminated carbon from the TMI Unit II accident and subsequent laboratory simulation tests confirm that useful data on iodine captured and released can be obtained under certain conditions.

Experimental test data per ASTM D3803-79 indicates that a homogeneously mixed sample of a poisoned adsorbent is always a conservative result when compared to a sample tested with its internal geometry preserved. That is, the test efficiency from a homogeneously mixed sample is always slightly lower than the same sample tested with the internal geometry preserved. This result confirms the essentially universal sampling and testing practices in use in the United States today will err only in the direction of conservatism (i.e., maximum safety).

Part I

While the efficiency testing of adsorbents has been rather thoroughly covered in the last 25 years and has been the subject of various test standards starting with RDT M16-1T (various revisions) and currently ASTM D3803-79 there was no standard method to handle the highly loaded Type II trays from TMI Unit II in the spring of 1979. The trays initially sent for analysis were chosen as the highest activity tray from each system. These trays had as much as one curie of I131 on them so the standard efficiency tests were obviously not applicable. The data that was of most critical importance at that time was an analysis of the isotopes on the carbon. Specifically we analyzed for cladding or transuranium isotopes which would indicate fuel melting (there were none) and any estimate of I131 release. The approach used to estimate I131 release was to core the two inch deep bed into four equal segments of one-half inch each and plot the I131 activity on each segment.

At the time of the accident four filter trains were postulated to be in operation, these are Trains A & B of the Auxiliary Bldg. (AHF 14A & 14B) and Trains A&B of the Fuel Handling Bldg. (AHF 10A & 10B).

Although evaluation of the adsorbent from these trains indicates that not all trains were operating and some of the trains may have been turned on sometime after the presence of I131 in the areas of the buildings where the train intakes are.

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*Currently with Ohio EPA.

On 12 April 1979 and subsequent to this date adsorbers were removed from the trains and transported to NUCON's Columbus laboratory for evaluation.

While initially only the methyl iodide removal efficiency of the adsorbent was to be evaluated a more thorough evaluation permitted the generation of useful results in relation to the adsorbent behavior and the evaluation of the 131 Iodine release.

Samples were obtained by sliding a 9" X 8" X 1/32" steel plate along the bottom (outlet) screen of an opened portion of the adsorber. The plate provided a solid substrate which could be lifted from the adsorber after insertion of four (4) 2" diameter glass tubes into the adsorbent until flush contact was made with the previously inserted plate. The open end of the glass cylinders were fitted with a "plunger" consisting of a 2" cardboard disc secured with wadded tissue paper. The disc and tissue served to hold the sample core securely in its insitu orientation, i.e., no mixing of the granules occurred when the plate and cylinders were inverted.

The plate with the cylinders held flush was inverted. Excess carbon on the plate was therefore removed. The open end of the glass cylinders was exposed in this maneuver without spillage or mixing of the core samples.

Each core was divided into 4, 1/2" samples by pushing the disc and tissue plunger while holding the cylinder in an upright position.

The plunger was pushed in 1/2" increments and the cylinder top leveled with a straightedge.

This was performed over a tissue lined tray (which prevented scattering of the particles) and the tissue changed after each 1/2" increment was obtained.

In this way, four (4), 1/2" samples per core were obtained.

Each sample was thoroughly mixed and precisely 0.5000 g of each sample fixed in a small separate cylinder to be placed in the Germanium well crystal for multichannel analysis for I131 content.

Table 1 details the results of the segmental analysis. Table 2 shows the results of the moisture and pH determinations.

Table 1

	Auxiliary Bldg.		Fuel Handling Bldg.	
	AHF 14A	AHF 14B	AHF 10A	AHF 10B
Adsorber Serial No.	2652	2525	2759	2603
MSA Carbon Lot No.	165	159	157	155
	Activity in µC/g carbon on 28 March 1979			
First 0.5 inch	10.1	15.4	34.5	76.1
Second 0.5 inch	4.4	4.9	6.4	26.5
Third 0.5 inch	2.5	3.1	0.9	18.5
Fourth 0.5 inch	1.7	1.5	0.1	15.1
Shipping Carts No.	USAF	3	2	1

Direction of flow from first toward fourth.

AHF 14A & 14B contained 90 adsorbers each with an average carbon weight of 34.0 kg.

AHF 10A & 10B contained 60 adsorbers each with an average carbon weight of 34.0 kg.

Table 2

	Auxiliary Bldg.				Fuel Handling Bldg.			
	AHF 14A		AHF 14B		AHF 10A		AHF 10B	
Adsorber Serial No.	2652		2525		2759		2603	
MSA Carbon Lot No.	165		159		157		155	
	Activity in μC/g carbon on 28 March 1979							
	H_2O	pH	H_2O	pH	H_2O	pH	H_2O	pH
First 0.5 inch	2.53	4.3		3.4		4.1		3.2
Second 0.5 inch	3.41	4.5		4.0		4.7		3.4
Third 0.5 inch	2.28	5.9		4.3		4.5		3.9
Fourth 0.5 inch	1.05	5.6		4.6		4.5		3.9

Data from Table 1 is plotted on semi-log paper on Figures 1 & 2. It is seen that the last three points show an exponential drop-off with distance through the carbon bed. Therefore, they can be represented by:

$$C = C_o e^{-\alpha x}$$

where C_o is the maximum surface concentration of I-131 on the entry face, and the drop-off rate going from sample-to-sample and x is the sample position; viz, x = 0 is the entry face and x - 4.0 is the exit face. The amount of I-131 captured by the carbon beds (on 28 March 1979) can be calculated as:

$$C'_c = \int_0^4 c dx = \frac{C_o}{\alpha} (1 - e^{-4\alpha})$$

This analysis will show less iodine captured than actually is on some of the beds. (Particularly for the Fuel Bldg AHF10B System.) However, it is expected that the retained form of iodine in the inlet face of the bed would be elemental iodine, while the penetrating (and integrating) form is organic iodine.

Similarly the amount of I-131 released (circa 28 March 1979 assuming that all laden airflow occurred then) can be modeled as the integral spatial concentration, C, beyond the last sample ($x \geq 4$);

$$C'_R = \int_4^\infty c dx = \frac{C_o}{\alpha} (e^{-4\alpha}) \text{ mixrocuries}$$

the ratio of I-131 released to that which was captured is

$$\frac{\text{RELEASE}}{\text{CAPTURE}} = \frac{C'_R}{C'_c} = \frac{e^{-4\alpha}}{1 - e^{-4\alpha}} = (e^{4\alpha} - 1)^{-1}$$

The summary captured release data is shown in Table 3.

FIGURE 1 TMI UNIT II Auxiliary Bldg.

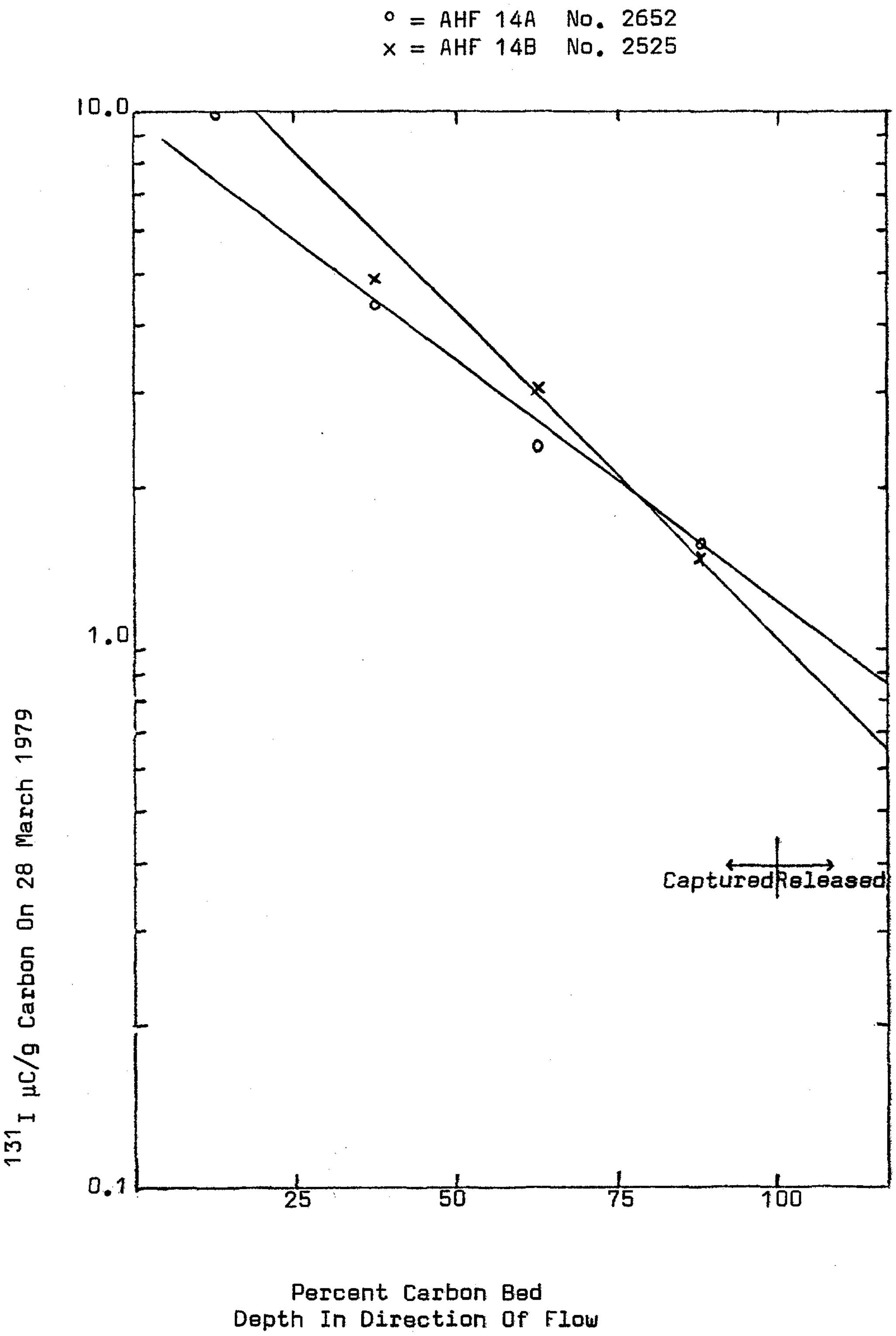


FIGURE 2 TMI UNIT II
Fuel Handling Bldg.
o = AHF 10A No.2759
x = AHF 10B No.2603

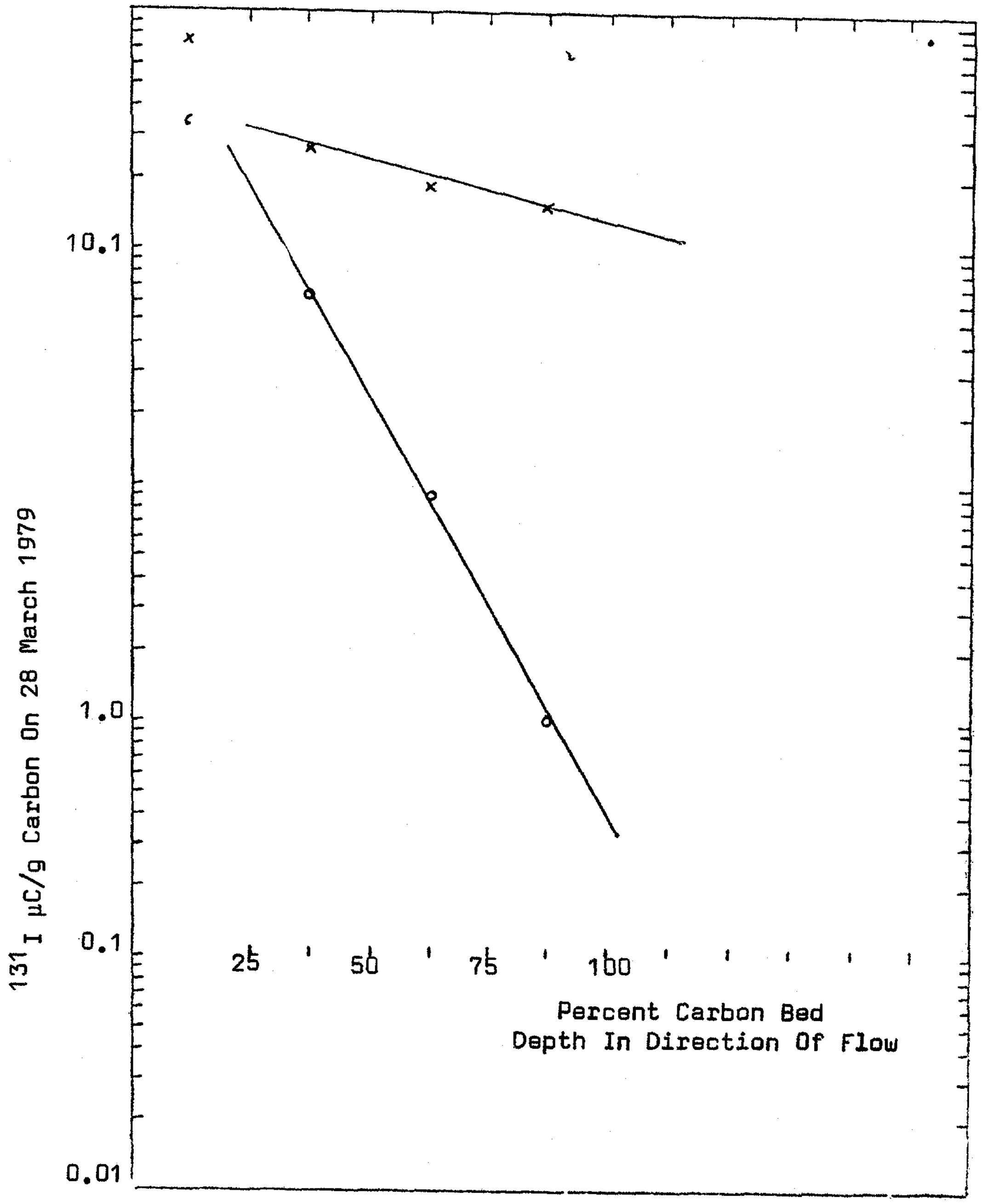


Table 3 Summary of Captured & Released 131-Iodine

	Curies of 131 Iodine Captured	Released	% Efficiency
Auxiliary Bldg Trains			
AHF 14A	11.94+	2.09	85.1+
AHF 14B	14.57+	1.51	90.6+
Fuel Handling Bldg Trains			
AHF 10A	36.64+	$9.03\mathrm{X}10^{-3}$	99.9+
AHF 10B	48.25+	23.23	67.5+
Total in four systems	111.4	26.84	80.6+

Notice must be taken that these figures are based on analysis of the most active trays and are, therefor, the worst case situation. The actual system or train activity was undoubtedly less on the average.

The results indicate that only one of the adsorber trains AHF 14A behaved as expected and presumably designed based on regulatory criteria.

General comments can be made concerning the impregnated carbon type. It is a KI-I_2 impregnated carbon containing free iodine. This was observed during the moisture content determination using the ASTM xylene method where the xylene turned purple. The pH values obtained were again very low. Evans evaluated pH and iodine/potassium ratio dependence of iodine penetration (1) (2).

It is also indicated that the design air flow of 36,000 CFM is not equally distributed between 10A and 10B systems but 10B is handling a larger portion of the air flow. This was confirmed during later inspections by NUCON Field Test Engineers and discussions with plant personnel.

The total curies of I131 released as calculated by this method was 26.84. Given the fact it is based on the most active trays it is in good agreement with other independent estimates. Even with this good agreement formal conformation of the approach was undertaken. A series of tests were performed using elemental iodine only, methyl iodide only and mixed elemental/methyl iodide as a challange gas through a homogeneously mixed bed of poisoned carbons. The carbon was obtained from trays that had been in actual service in various power plants. No attempt was made to identify the poisons. The carbons used were all impregnated with KI_3 and had an efficiency range of about 50% to 80% at 30°C, 95% RH per ASTM D3803-79. All tests were run per ASTM D3803-79 at 30°C, 95% RH, 16 hour pre-equilibration, 2 hour loading, 2 hour post sweep, 2 inch deep X 2 inch diameter test bed (segmented into 4 one-half inch segments with stainless steel screens), challange concentration of 1.75 mg/m^3 and a 4 inch guard bed of known high efficiency ($>$ 99.9%) carbon. The guard bed would capture all the challange released by the test bed and be the experimental basis for the calculated release comparison.

The data for activity on each Test Bed and Guard Bed segment as well as the measured capture and release (i.e., the total activity on each full bed) is given in Tables 4, 5 and 6. Sample nomenclature is: E1-E4 for elemental iodine (I_2) tests, 01-04 for organic iodide (CH_3I) tests and M1-M4 for mixed species tests. The mixed species test were conducted with approximately equal amounts of elemental and organic components by both chemical and activity measurements.

Table 4 Elemental Iodine Challange

Segment Counted	Sample	Measuted Activity of I^{131} on Sample in Microcuries E-1	E-2	E-3	E-4
TB 1		9.40×10^{-2}	3.05×10^{-2}	8.21×10^{-2}	1.20×10^{-1}
TB 2		1.78×10^{-3}	1.68×10^{-3}	4.74×10^{-3}	1.05×10^{-2}
TB 3		1.22×10^{-3}	1.58×10^{-3}	1.90×10^{-3}	2.22×10^{-3}
TB 4		1.11×10^{-3}	1.27×10^{-3}	2.17×10^{-3}	2.64×10^{-3}
GB 1		1.06×10^{-3}	3.39×10^{-4}	8.63×10^{-4}	9.85×10^{-4}
GB 2		1.66×10^{-3}	2.61×10^{-4}	5.96×10^{-4}	8.72×10^{-4}
Captured		9.82×10^{-2}	3.50×10^{-2}	9.09×10^{-2}	1.35×10^{-1}
Released		2.72×10^{-3}	6.00×10^{-4}	1.46×10^{-3}	1.86×10^{-3}
Efficiency (per D3803)		97.30%	98.31%	98.90%	98.64%

Table 5 Methyl Iodide Challange

Segment Counted	Sample	Measured Activity of I^{131} on Sample In Microcuries O-1	O-2	O-3	O-4
TB 1		1.04×10^{-1}	1.25	8.44×10^{-1}	1.93
TB 2		9.47×10^{-2}	8.22×10^{-1}	4.61×10^{-1}	1.49
TB 3		9.57×10^{-2}	8.27×10^{-1}	4.84×10^{-1}	9.54×10^{-1}
TB 4		9.05×10^{-2}	5.09×10^{-1}	3.13×10^{-1}	7.69×10^{-1}
GB 1		2.78×10^{-1}	2.24	1.78	1.35
GB 2		9.08×10^{-4}	3.11×10^{-3}	5.20×10^{-3}	7.75×10^{-3}
Captured		3.85×10^{-1}	3.40	2.10	5.14
Released		2.79×10^{-1}	2.24	1.79	1.36
Efficiency (per D3803)		58.01%	60.25%	53.68%	79.17%

Table 6 Mixed I_2 + CH_3I Challange

Segment Counted	Sample	Measured Activity of I^{131} On Sample In Microcuries			
		M-1	M-2	M-3	M-4
TB 1		$4.81X10^{-1}$	$5.59X10^{-1}$	$3.90X10^{-1}$	2.40
TB 2		$1.78X10^{-2}$	$1.38X10^{-1}$	$3.26X10^{-2}$	$1.16X10^{-1}$
TB 3		$1.28X10^{-2}$	$9.49X10^{-2}$	$2.87X10^{-2}$	$6.83X10^{-2}$
TB 4		$1.08X10^{-2}$	$8.70X10^{-2}$	$2.69X10^{-2}$	$6.99X10^{-2}$
GB 1		$5.27X10^{-2}$	$2.59X10^{-1}$	$4.58X10^{-2}$	$1.20X10^{-1}$
GB 2		$7.37X10^{-4}$	$6.20X10^{-4}$	$5.64X10^{-4}$	$1.04X10^{-3}$
Captured		$5.22X10^{-1}$	$8.79X10^{-1}$	$4.80X10^{-1}$	3.90
Released		$5.35X10^{-2}$	$2.60X10^{-1}$	$4.63X10^{2}$	$1.21X10^{-1}$
Efficiency (per D3803)		89.76%	77.20%	91.20%	95.63%

This data is plotted in Figures 3, 4 and 5. It is plotted as activity vs bed position to show the shape of the activity distribution. A sharp knee is noticed in the elemental and mixed challange plots. This is due to the differences in trapping efficiency for elemental and organic forms of iodine. The organic forms for the elemental challange come from reactions between the iodine and the organic poisons on the carbon. The plots for the elemental and mixed challanges should be considered as the sum of two plots. This allows straight line (on semilog paper) equations to be used and is necessary for the release calculations. The methyl iodide plots are essentially straight lines and may be so treated.

The slope of the elemental iodine capture is quite steep and will not under any credible circumstances become zero (i.e., paralle to the X axis) for LWR systems. Methyl iodide and/or other organic forms may indeed reach an equilibrium condition and the slope will then be zero. Under varing parameters encountered in the real world where temperature, RH, flow and inlet concentrations may all vary considerably the slope may even go positive for short periods. This phenomenon has been tentatively identified by inference at TMI Unit II post accident conditions. A detailed treatment of long term use of a carbon system under equilibrium conditions (i.e., zero slope) is given in "Gas Clean-Up System For Vented Containment."(3)

It must be stressed that the subject method for release estimation is valid only under specific conditions. The carbon must have sufficient effective efficiency for the plotted activity to have a reasonable negative slope. Obviously if it is zero or near zero the integration will give an infinite release. This is correct to the extent that it is telling us that the carbon has no efficiency so that all the organic iodine that goes in will come out minus only the small quantity delayed by the bed.

Flow must be constant for any inference to be made. If the flow varies then any quantitative calculation of release is invalid. You may however infer some data as to the efficiency of the carbon from the shape of the plot, given the flow rate the time just before the tray was removed.

Recognition that this method is intended for specific analysis by highly qualified personnel who are knowledgeable not only with the analysis technique but

FIGURE 3

Elemental Challenge
ASTM D3803
30°C
95% RH

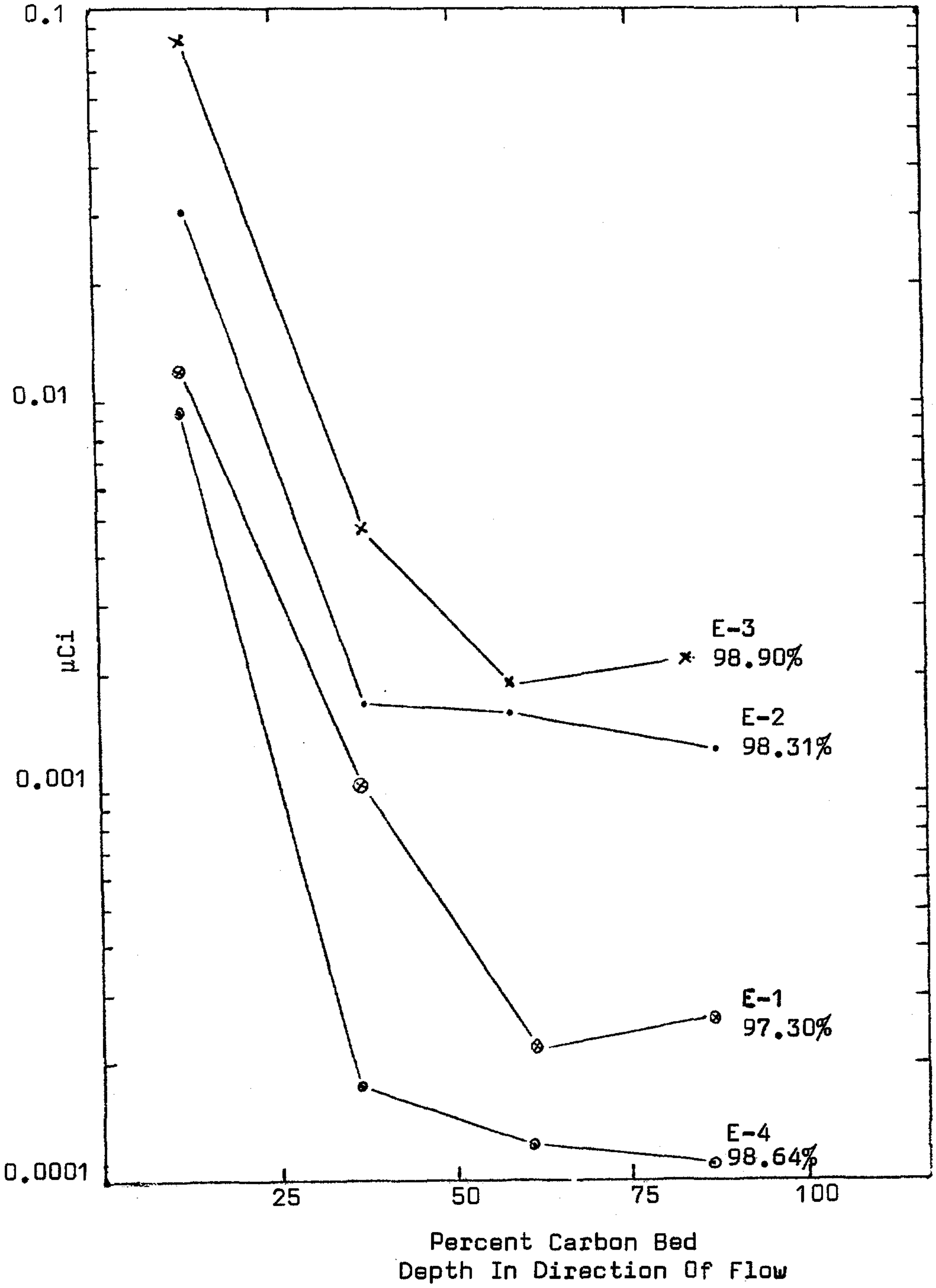


FIGURE 4

Organic Challenge
ASTM D3803
30°C
95% RH

0-1 = o —— o 58.01% Efficiency
0-2 = x —— x 60.25% Efficiency
0-3 = • —— • 53.68% Efficiency
0-4 = △ —— △ 79.17% Efficiency

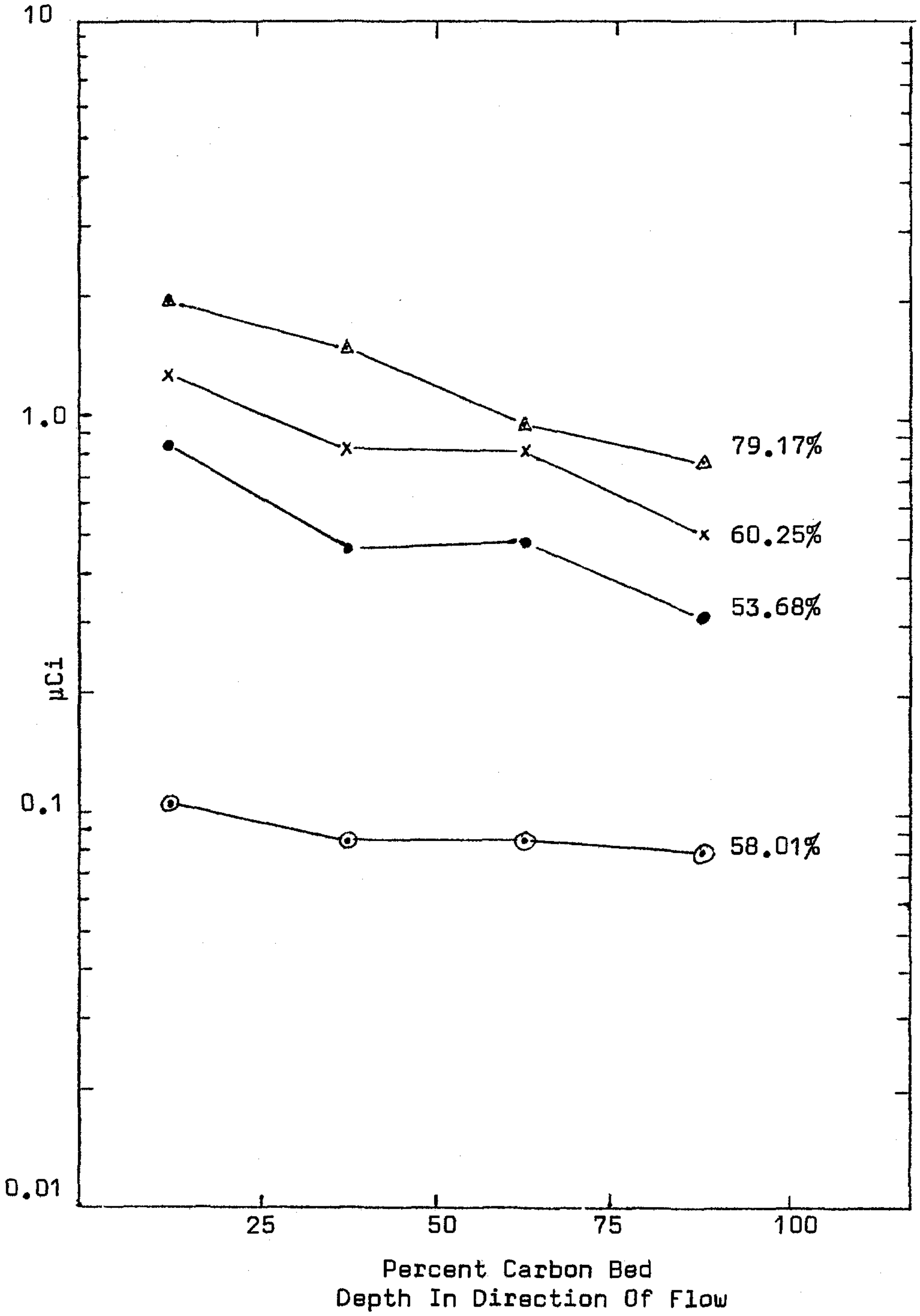


FIGURE 5A

Mixed Challenge
ASTM D3803
30°C
95% RH

M-1 = •——• 89.76% Efficiency
M-2 = x——x 77.20% Efficiency
M-3 = o——o 91.20% Efficiency

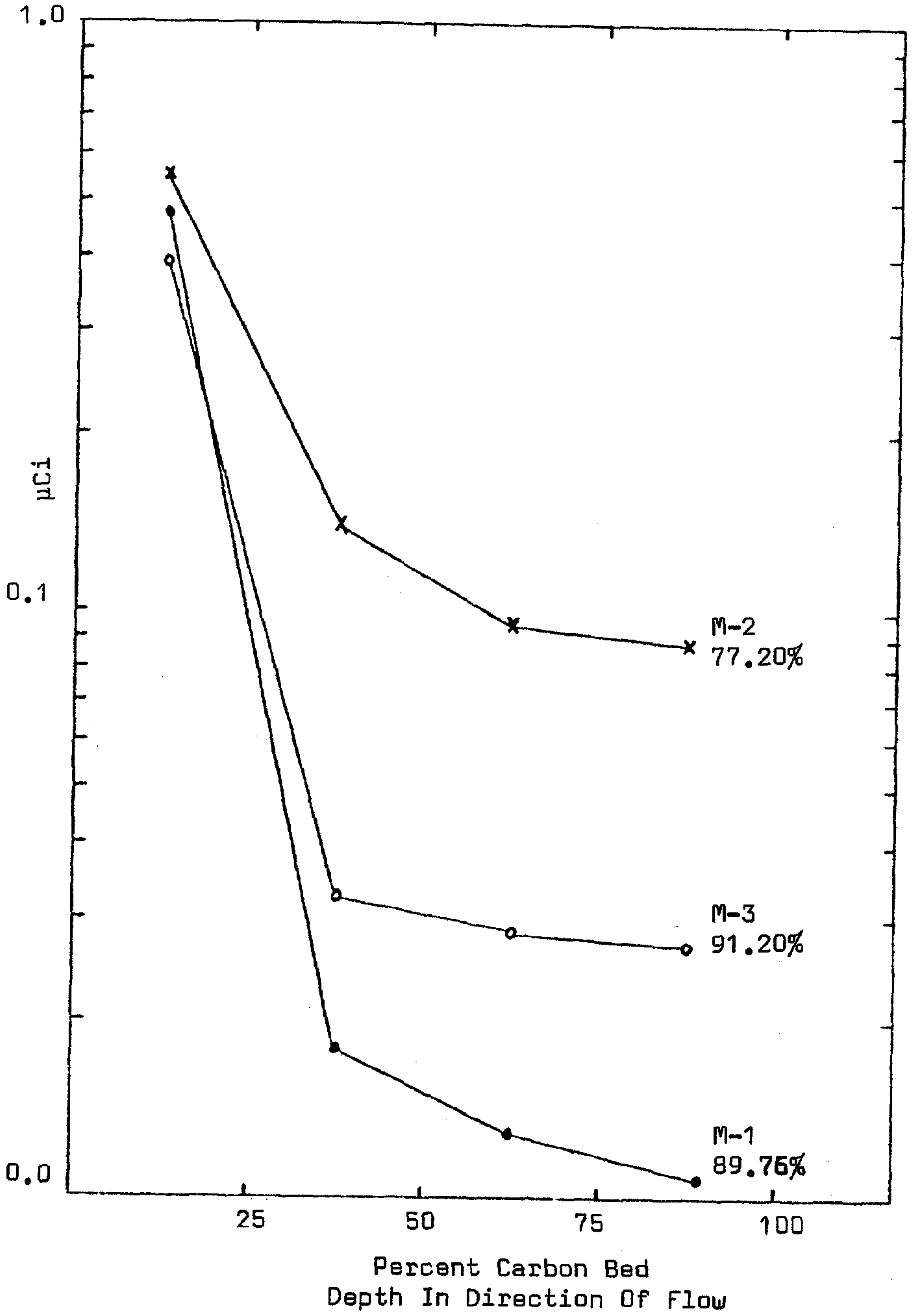
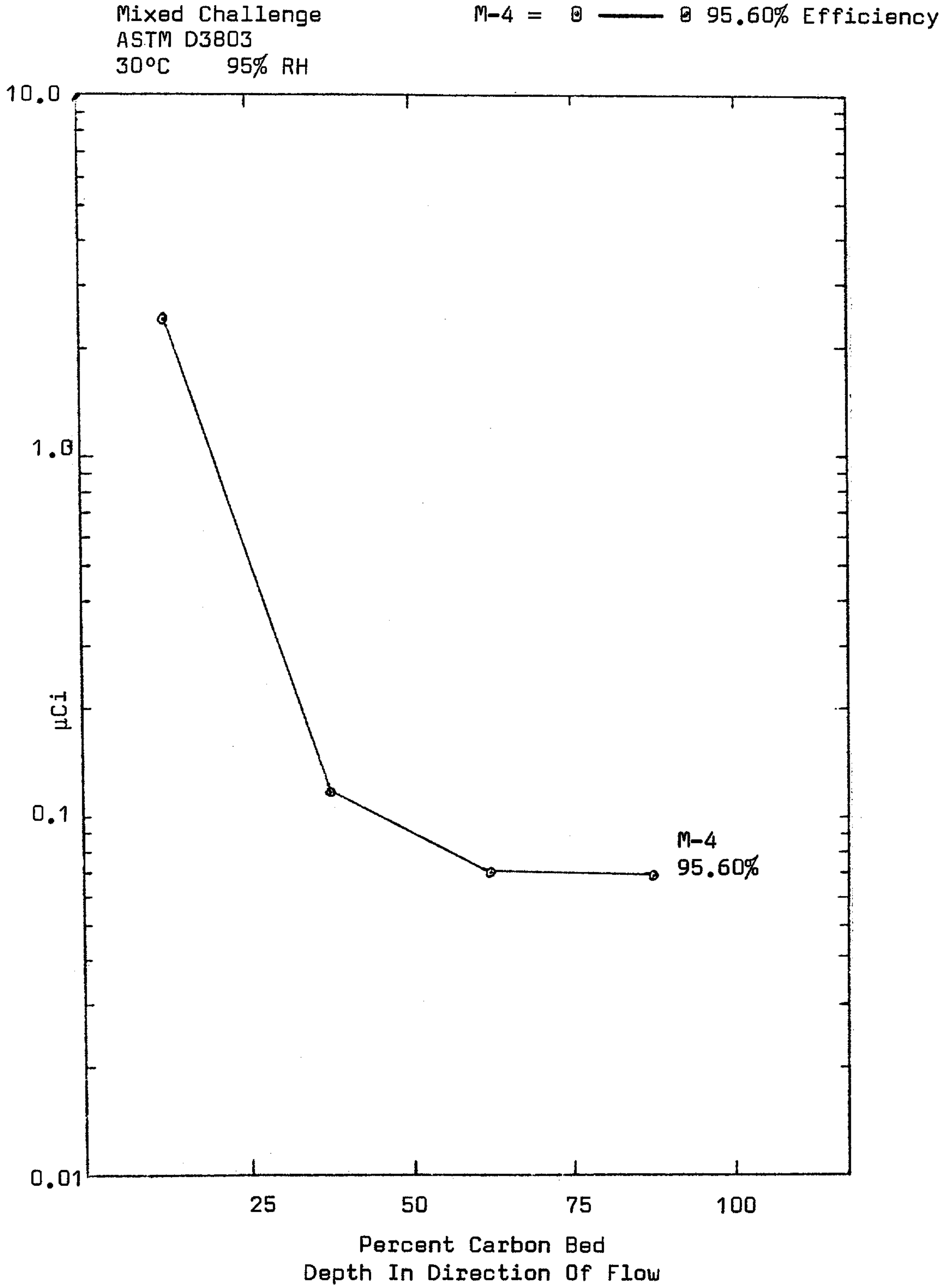


FIGURE 5B


Mixed Challenge
ASTM D3803
30°C 95% RH
M-4 = ⊘ —— ⊘ 95.60% Efficiency
10.0
1.0
0.1
0.01
µCi
25
50
75
100
Percent Carbon Bed
Depth In Direction Of Flow
M-4
95.60%

the systems involved and their operating history is mandatory.

Actual experimental confirmation of the capture/release analysis technique consists of using the plotted Test Bed data to calculate the release and comparing it with actual release from the Guard Bed data. The analytical technique and a sample calcuation follows with the results tabulated in Tables 7, 8 and 9.

The radioiodine release is calculated from the capture data as plotted in Figures 4, 5 and 6. Equation $C = C_o e^{\alpha x}$ is used for a straight line extention on semilogrimithic paper. It is fitted to the applicable section of the plot. In the organic iodide challange where there is no knee in the plot the entire plot is used. For the elemental and mixed challange, or any empirical plot where a knee is present, only the section of the plot to the right of the knee is applicable. This section represents the penetrating portion of the activity. Review of these plots shows that considerable care is required in proper choice of the data points used, and in fitting the straight line equation to them.

The actual parameters used are C_o and α, where C_o is the inlet face concentration and α is the slope.

C_o is the concentration at $X = 0$ of the straight line extention of the chosen section of the plot. α is the drop off rate of concentration or the slope.

Intergation of $C = C_o e^{\alpha x}$ from the outlet face ($X=4$) of the carbon bed to infinity gives the calculated activity released.

$$C_{Release} = \int_4^{\infty} C dx = \int_4^{\infty} C_o e^{\alpha x} dx = \frac{C_o}{\alpha} e^{-4\alpha}$$

Note this applies only when α is negative which is the only appliable case.

Reveiw of the plots shows that there can be considerable latitude in the possible choice of the straight line extention. The low loadings of the experimental data vs the high loadings of the TMI Unit II data undoubtedly contribute to this spread. Many real parameters such as poison movement during the test, or incident, unequal inlet concentrations over time, varing temperature, varing RH, etc., in a real incident will all add to the possible spread of the data points. For this reason graphic choice of the straight line extention to set C_o and α is recommended. It follows that a knowledgeable and experienced technical person must perform this critical task.

Table 7 Elemental Iodine Challange

	Release In Microcuries	
Sample	Calculated	Actual
E-1	3.87×10^{-3}	2.72×10^{-3}
E-2	3.69×10^{-3}	6.00×10^{-4}
E-3	5.28×10^{-3}	1.46×10^{-3}
E-4	1.39×10^{-4}	1.86×10^{-3}

Table 8 Organic Iodide Challenge

Sample	Release In Microcuries Calculated	Actual
O-1	0.143	0.279
O-2	5.67	2.24
O-3	2.12	1.19
O-4	1.98	1.36

Table 9 Mixed Species Challenge

Sample	Release In Microcuries Calculated	Actual
M-1	4.71×10^{-2}	5.22×10^{-2}
M-2	9.61×10^{-1}	2.60×10^{-1}
M-3	4.13×10^{-2}	4.63×10^{-2}
M-4	1.32×10^{-1}	1.21×10^{-1}

Attention is again called to the fact that for these experiments the activities used were many orders of magnitude lower than seen in the trays from TMI Unit II. It is believed most actual accident circumstances that would require such analysis would have much higher activity. The low activity complicates the analysis to the extent that very small errors from whatever source, in the number of counts will drastically change the form of the plot. Further it is hoped that the efficiency of the actual carbon inplace in operating plants will meet the regulatory and design requirements which are usually over 90% or 95% at 30°C 95% RH for organic iodide.

Part II

The carbon (or other adsorbent) in a nuclear power plant is required by license to be subjected to a laboratory test to confirm its efficiency for I131. Since this is a laboratory test the sample of adsorbent must be removed from the system and transferred to the testing laboratory. This is usually accomplished by either sending a small sample canister containing the adsorbent or by obtaining, by any of a number of methods, a bulk sample. In the first case the internal geometry of the adsorbent is usually preserved and in the second it is never preserved.

The reason that an adsorbent which at one point was able to meet a set of efficiency criteria and later fails the same test is chemical poisoning of the adsorbent. There are an almost unlimited number of possible compounds that act as poisons for the impregnated carbon used in current plants. This poisoning will always start at the inlet face and progress through the bed in an identifiable mass transfer zone (MTZ). The MTZ will vary greatly in shape and rate of movement depending on the airflow velocity, temperature, chemical poison, RH, type of carbon, etc. At some point in time the bed will have a well defined poisoned section, partially poisoned section and a "clean" section. Under continued adverse conditions the MTZ will progress through the bed and it will become fully poisoned. The requirement for periodic testing is specifically setup to prevent this from occuring.

Given the current situation that some samples are taken in bulk and some in canisters the effect on test results of preserved vs non-preserved geometry is of great interest. A set of methyl iodide efficiency experiments was designed to provide data to investigate this question.

Selecting a poison turned out to be the first problem. To eliminate a number of variables resulting from adding an actual chemical poison to a fresh carbon it was decided to simulate totally poisoned carbon with gravel. A gravel was chosen that had essentially the same density as the carbon and could be screened to the same (8X16 U.S.) mesh size as the carbon. The carbon chosen was NUCON NUSORB KITEG-II* which meets all the requirements of US NRC Regulatory Guide 1.52. Initial tests showed that the gravel would mix homogeneously with the carbon and exhibited no tendency to separate. Further a very sharp transition between the carbon and gravel could be obtained and varified due to the difference in color (light gray/pink vs black). The use of a zero efficiency gravel to simulate poisoned carbon also provides the absolute limiting case and any real world situation could be no worse.

The actual experiments consisted of two (2) sets of tests, one with pre-equilibration and one without. All tests were in conformance with the procedures of ASTM D3803-79 at the parameters indicated. Each series consisted of pairs of tests at the same level of simulated poisoning with one sample a mixed test bed and one sample with geometry preserved. That is a gravel layer of appropriate thickness at the inlet section of the test bed over a carbon layer with a sharp flat interface. The results were then plotted for each of the two (2) series of experiments.

One series was performed at 25°C, 95% RH and no pre-equilibration, the other was at 30°C, 95% RH and 16 hours pre-equilibration. A 2 hour loading, and 1.75 mg/m^3 was standard.

The results are tabulated and plotted in Tables 10 and 11 and Figures 6 and 7.

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*Trademark of Nuclear Consulting Services, Inc.

Table 10

Gravel (Inches)	Carbon (Inches)	% Bed Poisoned	Efficiency (%)	
			Mixed	Layered
0	2	0	NA	99.9+
1/4	1 3/4	12.5	99.75	99.86
1/2	1 1/2	25.0	98.42	99.84
3/4	1 1/4	37.5	97.66	99.81
1 1/4	3/4	62.5	87.99	96.15
1 3/4	1/4	87.5	70.99	78.91
2	0	100	0.67	0.67

Test Conditions:
Temperature 25°C
R.H. 95%
Inlet Concentration 1.75 mg/m^3 CH_3I/CH_3I^{131}
Face Velocity 40 FPM
Prehumidification None
Post Sweep None
Loading Period 2 Hours

Procedure and test apparatus per ASTM D3803-79.

Table 11

Gravel (Inches)	Carbon (Inches)	% Bed Poisoned
0	2	0.0
1/4	1 3/4	12.5
1/2	1 1/2	25.0
3/4	1 1/4	37.5
1	1	50.0
1 1/4	3/4	62.5
1 1/2	1/2	75.0
1 3/4	1/4	87.5
2	0	100.0

Test Conditions:
Temperature 30°C
R.H. 95%
Inlet Concentration 1.75 mg/m^3 CH_3I/CH_3I^{131}
Face Velocity 40 FPM
Prehumidification 16 Hours
Post Sweep 2 Hours
Loading Period 2 Hours

Procedure and apparatus per ASTM D3803-79.

FIGURE 6

Efficiency per ASTM D3803-79
25°C 95% RH
No pre-equilibration, No post sweep

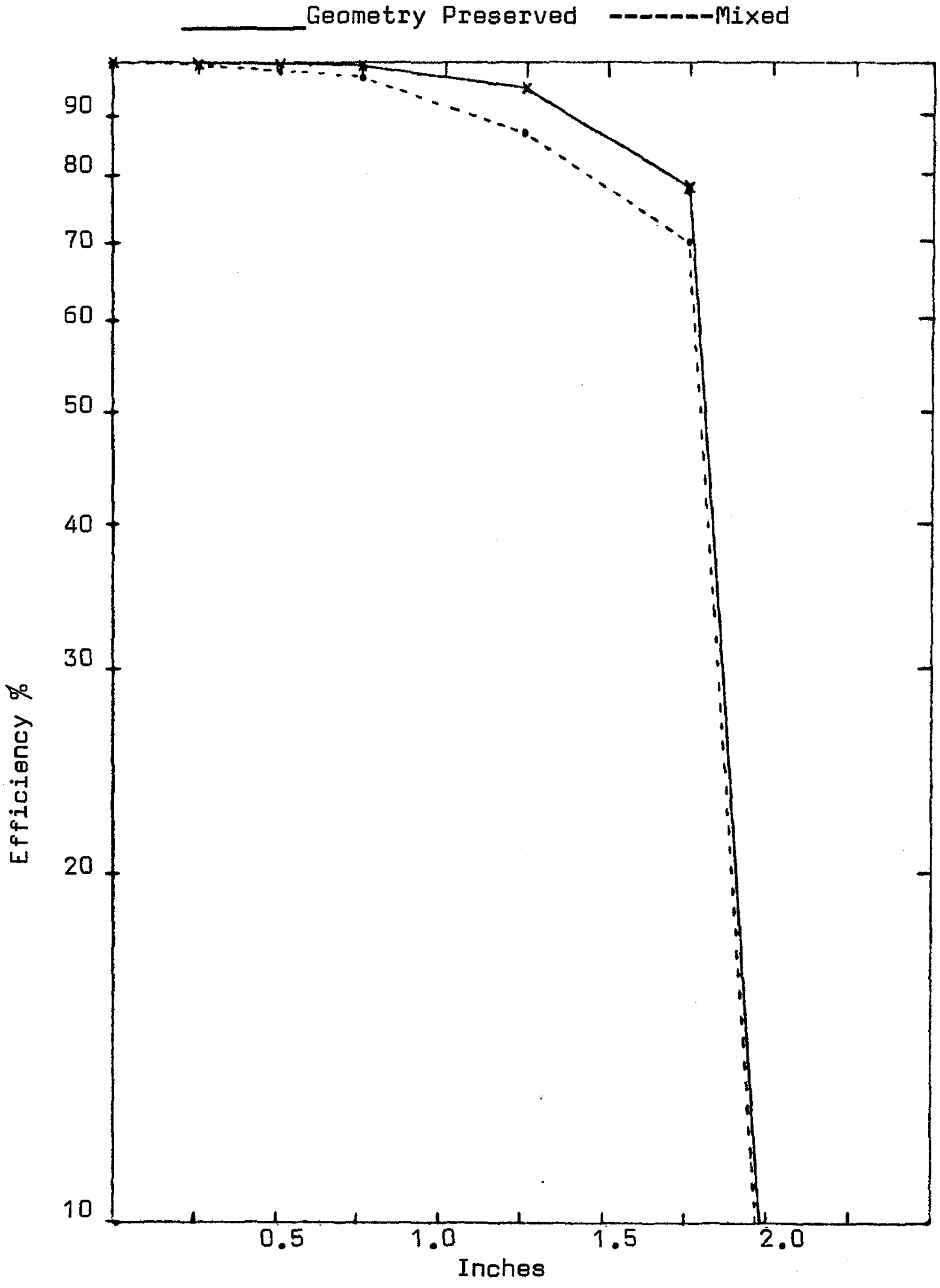
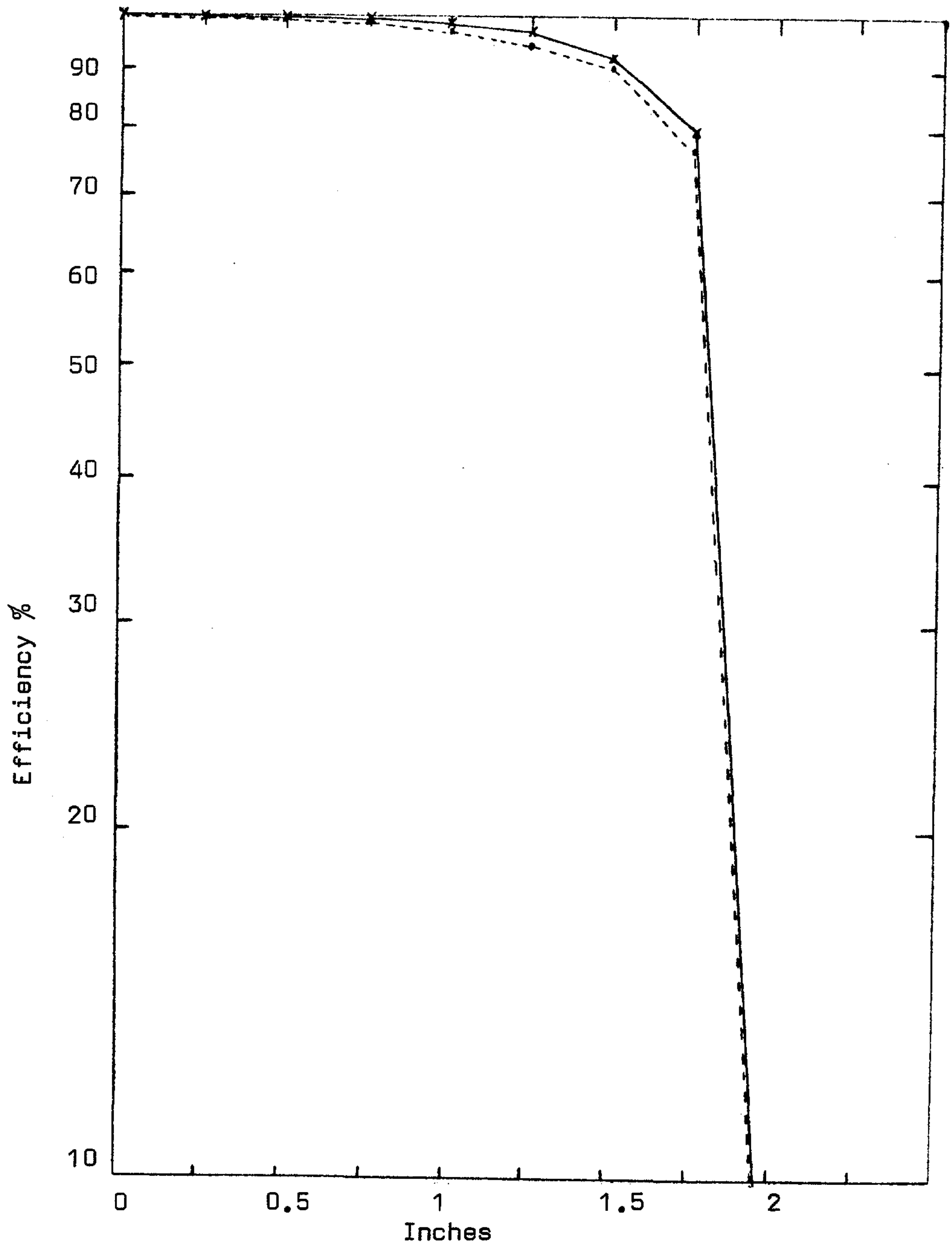


FIGURE 7

Efficiency per ASTM D3803-79
30°C 95% RH
16 Hour Pre-equilibration, 2 Hour Post Sweep

——————— Geometry Preserved

- - - - - - - - - Mixed



The results clearly and consistantly show that a mixed bed will give a slightly lower efficiency than a bed with geometry preserved. This means that the laboratory test results, as currently obtained, are always conservative. Since we have stressed that the simulated "zero" efficiency gravel is the worst case actual tests will have less difference in efficiency results. This set of experiment is therefor the limiting case.

Given that actual results will always show less difference than found here and the very high cost of trying to provide provisions for testing the geometry preserved we suggest that these experiments confirm the current approach as technically and economically sound.

References

1. Confinement Of Airborne Radioactivity Progress Report: July-December 1973 DP-1355 E.I. du Pont de Nemours and Co., A. G. Evans, L. R. Jones.

2. Confinement Of Airborne Radioactivity Progress Report: January-December 1976 DP-1463 E. I. du Pont de Nemours and Co., A. G. Evans.

3. Gas Clean-Up System For Vented Containment: 14th ERDA Air Cleaning Conference, CONF-760822, Volume II Pages 749-760, J. L. Kovach.

EVALUATION OF CARBONS EXPOSED TO THE THREE MILE ISLAND ACCIDENT

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Abstract

One of the lines of defense that served to mitigate the radiological effects of the accident at Three Mile Island was the activated carbon installed in ventilation air flows. Filters in the Auxiliary and Fuel Handling Buildings of Unit 2 adsorbed tens to hundreds of curies of iodine-131, preventing the release to the environment. The carbon exposed to the accident has been replaced and the spent carbon has been analyzed in the laboratory. Independent analyses were performed for the two filter trains in both the Auxiliary and Fuel Handling Buildings, replaced at various times after the accident. The results of these analyses are compared to new (unexposed) carbons.

In addition to measurements of the penetration of methyl iodide-131 according to the procedures in RDT M16-1, the penetration of methyl iodide-127 (non-radioactive) was determined using chromatography with an electron capture detector. The "bleeding" of methyl iodide-127, which had been adsorbed by the carbon during the dose period, was observed in the subsequent air purge conducted at the same flow, relative humidity, and temperature. An analogy is drawn with the release of I-131 during the Three Mile Island accident.

The simultaneous emission of iodine, carbon monoxide, and carbon dioxide during programmed heating (3^{o} per minute) in excess air was determined up to the spontaneous ignition temperature. Although elementary iodine was the main iodine species, the TMI-exposed carbons gave an appreciable organic iodide emission which became measurable at $80^{o}C$. The emission far exceeded that observed with new KI_x impregnated carbons, indicating the presence of less stable iodine-carbon complexes in the service-aged carbons.

I. Introduction

One of the lines of defense that served to mitigate the radiological effects of the accident at Three Mile Island was the activated carbon installed in ventilation air flows. Filters in the Auxiliary and Fuel Handling Buildings of Unit 2 adsorbed tens to hundreds of curies of iodine-131, preventing the release to the environment. The carbon exposed to the accident has been replaced and the spent carbon has been analyzed in the laboratory.

The retention of iodine by nuclear carbons after adsorption is a very important property. Elemental iodine retained by physical adsorption alone would be desorbed to the air flow after a small temperature increase. Iodine, however, held by chemical combination with the carbon network, would be more stable and require a higher temperature to decompose the carbon-iodine covalent bond. Chemical modification of the carbon network and of the impregnants by weathering will also influence the retention since the surface composition is modified by the accumulated contaminants.

Independent analyses were performed on carbon samples withdrawn from the two filter trains in both the Auxiliary and Fuel Handling Buildings, replaced at various times after the accident. The results of these analyses are compared to new (unexposed) carbons. The "bleeding" of methyl iodide-127, which had been adsorbed by the carbon during the dose period, was observed in the subsequent air purge conducted at the same flow, relative humidity, and temperature that were used in the dosing period. There is a striking resemblance between the ^{131}I "bleeding" or penetration at Three Mile Island and that observed in the laboratory with $^{127}ICH_3$ which was followed for five days.

II. Nuclear Carbon Samples

The samples were received via the Nuclear Consulting Services, Inc., and are listed in Table I. The locations of sampling and the points in time of withdrawal are given by Collins, Travers and Bellamy.(1) The carbons from Unit 2 had been subjected to the incident of 28 March, but had decayed to a safe level for investigation at NRL. The radioactivity of the carbons from Unit 1 were not above background activity.

Table I. Carbon samples from Three Mile Island .

Reactor	Label Information	Date Received	NRL Number
Unit 1	Replacement Cell 217, MSA Carbon Lot C-201, No service since April, 1974	26 April	5144
Unit 1	Met Ed Cell 76-217; Same as MSA Lot C-201	11 May	5147
Unit 1	Met Ed Cell #250	11 May	5149

(continued)

Table I (continued)

Reactor	Lable Information	Date Received	NRL Number
Unit 2	Met Ed Cell #2652 A train, Aux. Bldg.	11 May	5148
Unit 2	Met Ed Cell #2525 B train, Aux. Bldg.	18 June	5152
Unit 2	Met Ed Cell #2759 A train, Fuel Handling	18 June	5153
Unit 2	Met Ed Cell #2603 B train, Fuel Handling	18 June	5154

The samples of new nuclear carbons are listed in Table II.

Table II. New nuclear carbons.

Carbon		Date	NRL Sample
NUSORB KITEG II Lot 024	Nuclear Consulting Services, Inc.	26 April	5145
BC 727	Barnebey Cheney Co.	New Stock	
NACAR G615	North American Carbon Co.	New Stock	
MSA (463563)	Mine Safety Appliance Co.	New Stock	

III. Thermal Desorption of the Impregnated Iodine in an Air Flow

Thermal analyses of the carbons were made in order to observe possible changes in iodine bonding to the base carbon.(2) The temperature gradient was 3°C/min and the upward air flow through a carbon bed 2.54 cm diameter and 2.54 cm depth was 5 L/min (10 meters/minute). The effluent elemental iodine from the heated carbon sample was determined and in an independent second determination, the effluent was converted completely to elemental iodine by passage through a hot quartz tube before the iodine measurement (Mast Instrument).

The thermal emission of elemental iodine (mg/M^3) from a typical nuclear carbon is given in Figure 1 as a function of the exit gas temperature. A comparison was made with a sample of new MSA 463563 carbon. The emission from the TMI carbons exceeded that from the new MSA carbon; also, the rate increased more rapidly in the lower temperature range (see Figure 1). It was concluded that the iodine in the KI_x impregnation of the service-aged carbons had been degraded by chemical change into a less thermally-stable entity.

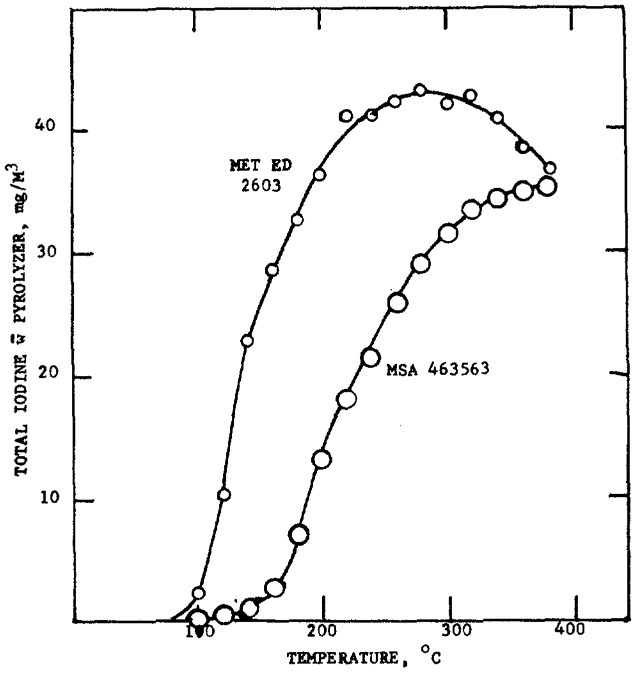

Figure 1.
Rate of Iodine (I_2) Emission with Temperature Programmed at 3°/minute for MET ED 2603 (NRL Test 3343)

The TMI carbons are aligned (Table III) in sequence of decreasing penetration of methyl iodine-131 in order to determine whether these KI_x impregnated carbons might be correlated with the iodine emission increase integrated to a specified temperature. The total iodine (mg) desorbed at 180°, 221°, and at the spontaneous ignition temperature (SIT) are given. The sequence does have the same alignment at the SIT as do the corresponding determinations of methyl iodide-131 penetration. The only anomaly is the A train (2759) of the Fuel Handling Building, but the available data do not allow a detailed analysis to be determined. There is a possible correlation between $^{131}ICH_3$ trapping and the thermal stability of the carbon.

The iodine emission with increase of temperature reached an approximately steady rate between 200 and 250°C (see Figure 1). The rate then decreased somewhat to the same level as, or somewhat lower than, the new MSA material. A determination was made of the total iodine impregnated in each carbon and the fractional loss of iodine during the oxidation up to the SIT was estimated.

Table III. Comparison of $^{131}ICH_3$ Penetration with Iodine Desorption.

Carbon	CH_3 I-131 Penetration	Total Iodine Desorption, mg 180°C	211°C	SIT
2603	50.9	2.71	4.50	18.5
2525	44.0	2.03	3.56	-
2652	30.5	0.63	2.0	14.0
2759	24.5	1.60	3.07	13.9
New MSA 463563	0.13	0.24	0.82	9.7

The results (Table IV) indicate that the iodine emission was significantly increased as a consequence of service. Since at most only 6% of the total iodine was emitted at the SIT, the leveling-off of the rate between 200-250°C is not a result of exhausting the iodine in the impregnated carbon.

Table IV. Thermal emission of the iodine impregnated in the Carbon.

Carbon	Wt.% I	Impregnated Iodine (mg)	Total Iodine Emitted at SIT mg	%
2603	4.8	306	18.5	6.0
2525	2.3	147	-	-
2652	5.8	334	14.0	4.2
2759	5.0	287	13.9	4.8
MSA 463563	4.2	276	9.7	3.5

Organic iodine compounds were present in the evolved fraction during the early stages of the heating cycle, but the amounts were small relative to elemental iodine. This observation agrees with previous results with a KI_x impregnated carbon (2701).(3)

In all of the samples analyzed, the iodine emission was accompanied by carbon oxidation to form CO and CO_2. The amount of carbon burn-off (i.e., that contained in the CO + CO_2) was qualitatively proportional to the elemental iodine evolved. The behavior has been explored in some detail since it would suggest the rupture of a strong bond between impregnated iodine and the carbon networks of a nuclear carbon. The lower section of Figure 2 compares the total iodine released from the various carbons as a function of the total carbon burn-off (CO and CO_2) up to 180°C using the four carbons of Table IV. The same is shown in the upper section of Figure 2 when integrated up to 360°C. There is a good correlation trend in both cases.

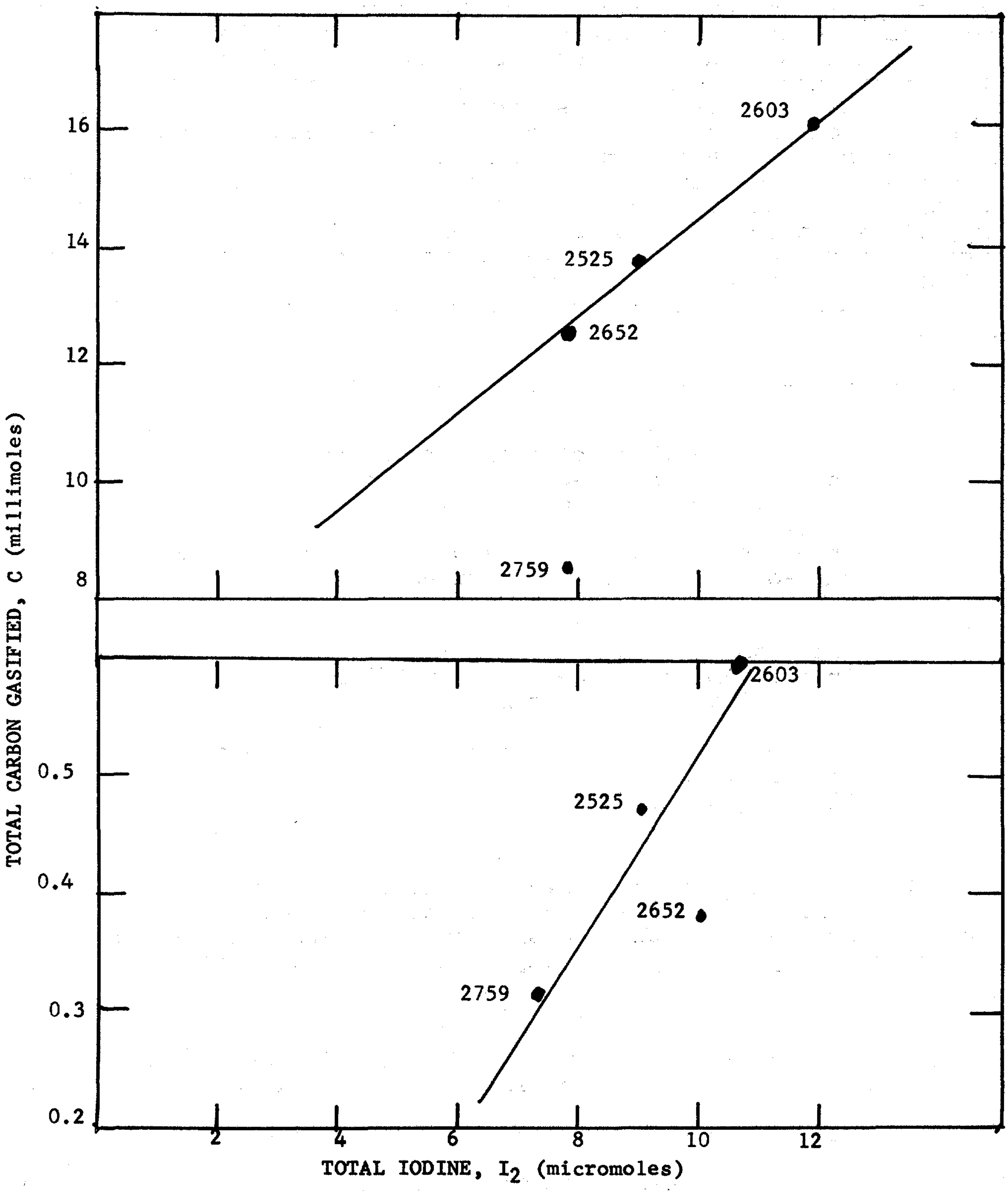

Figure 2.
Correlation using Four TMI Carbons Between the Total Carbon Gasified with the Total Iodine Emitted Integrated to 180°C (bottom) to 360°C (top).

The total carbon gasified using the MET ED 2603 sample is plotted in Figure 3 as a function of the elemental iodine emission. The X-coordinate is turned back after 3 decades and then continued in the upper section of Figure 3 to cover three additional decades. The ratio for these measurements of carbon gasified to iodine desorbed, expressed in stoichiometric quantities, first decreased as the temperature increased to about 160°, and then increased at higher temperatures. The behavior is compatible with a model in which there is a threefold increase of I_2 up to 160°C and less than a onefold increase in total carbon; this could be termed desorbed or easily decomposed iodine. In the temperature range 160-400°C, there is an additional threefold increase of I_2 and about a threefold increase in gasified carbon; this behavior indicates decomposition of a stronger iodine-carbon bond. The dotted line in Figure 3 indicates a 1 to 1 mole ratio of carbon to iodine. In all cases, a service-aged carbon resulted in an enhancement of the emitted impregnated iodine.

The above measurements may be summarized as follows:

1. The service ageing of a KI_x carbon decreases the thermal stability of the impregnated iodine in that a greater emission of elemental iodine takes place at lower temperatures.

2. When the carbon is gasified as CO and CO_2, the burn-off carbon is proportional to the iodine evolved.

3. The magnitudes of the $^{131}ICH_3$ penetrations are in the same sequence as the total iodine emissions compared at the same temperature.

4. The total iodine emitted up to the SIT is considerably less than that contained in the impregnation formulation.

The dominant iodine species in the thermal emission from KI_x carbons is the I_2 molecule. The formation of I_2 can result from several reactions. If the KI_x species is present on the carbon as a molecular entity, a simple decomposition could take place:

$$KI_x \rightarrow KI + I_2$$

If the iodine in KI is oxidized, another species on the carbon must be reduced. The process may be formulated as follows:

$$KI + \text{carbon-oxidized} \rightarrow I_2 + \text{carbon-reduced.}$$

However, it is more likely that the actual mechanism is quite complex due to the great heterogeneity of the impregnated carbon surface composition. The heat of dissociation of the carbon-iodine bond varies with the molecular species; for example, D(C-I) = 50 kcal/mole for CH_3I and that for t-C_4H_9I is 60 kcal/mole. The decomposition of all

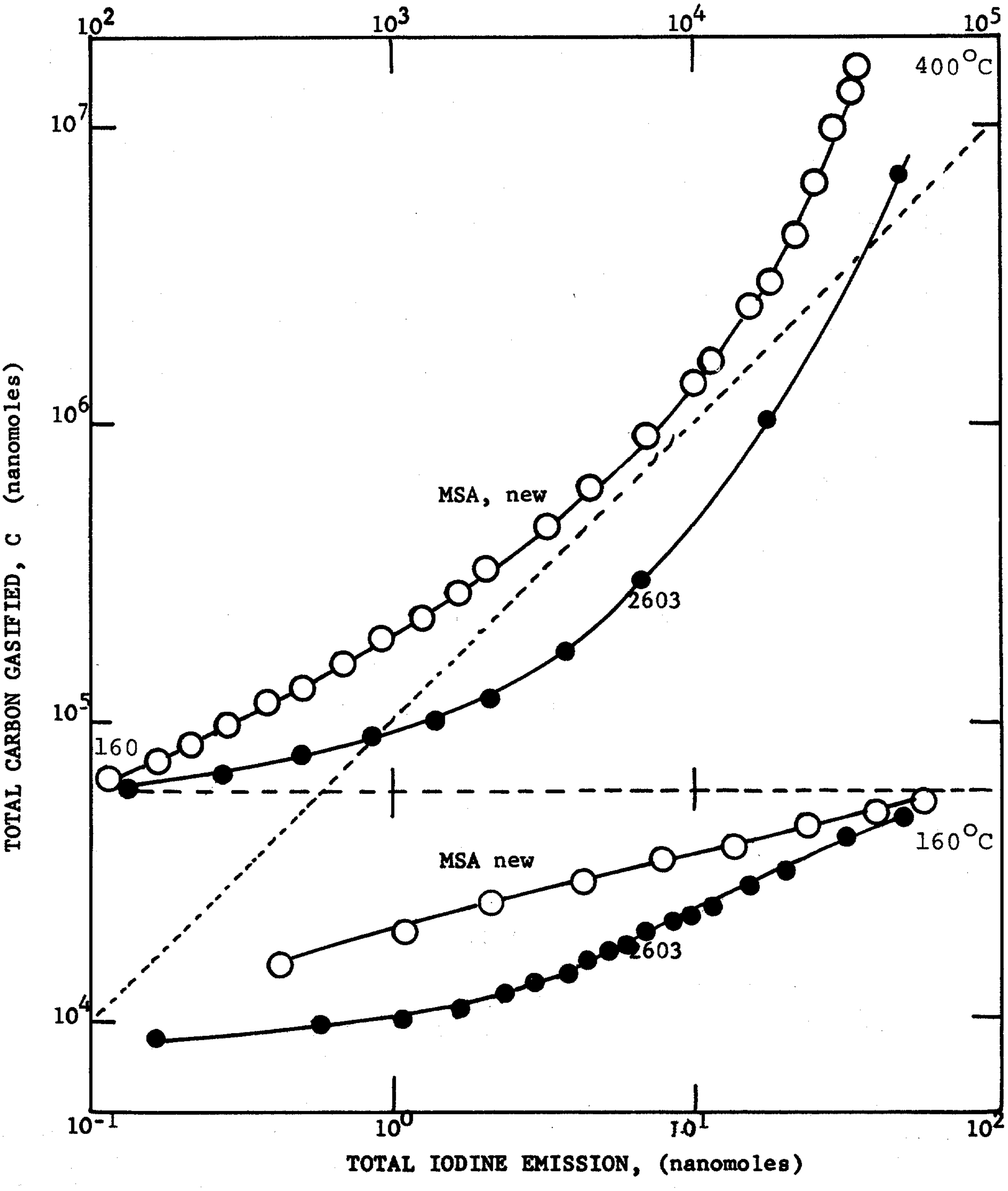

Figure 3.
Correlation of Carbon Gasified with the Emission of I_2 for MET ED 2603

iodine species on a nuclear carbon leads to a continuous emission of iodine.

IV. Penetration of $^{127}ICH_3$ Used as the Test Gas

Non-radioactive methyl iodide can be used to supplement the test results with radioactive methyl iodide-131.(3) Testing with methyl iodide-127 has several advantages: frequent on-line measurements can be made, a continuous record can be obtained of the inlet and exit gas concentrations, and the pattern of breakthrough can be observed any time including the subsequent purge with air. Thus, the presence of iodine bleeding through a carbon bed can be detected until background concentrations are reached.

Methyl iodide was found to be present at low levels in the air flow before the dose was added. The data for MET ED sample 250 (Unit 1) are given in Table V. Within the first 20 minutes of flow, the concentration was 3 to 4 ppb, and the concentration subsequently decreases to about 0.06 ppb in 200 minutes. It may be noted that another nuclear carbon service-aged in a different facility also gave methyl iodide emission (about 0.06 ppb) before the dose of methyl iodide-127 was added. These measurements served as a control in establishing the base line before addition of the dose and demonstrate that some methyl iodide is generated or purged off the carbon in the presence of the moving air stream.

Table V. Initial purge of sample (no dose) for Met Sample 250, Unit 1 (NRL sample 5149)

Time (min)	MeI (mg/M^3)	ppb
1	0.018	3.1
6	.026	4.5
20	.0181	3.1
40	.00832	1.44
80	.00387	0.67
111	.00248	0.43
135	.00231	0.40
210	.00033	0.057
405	.00050	0.088
1400	.00026	0.045

Air flow at V_1 = 12 m/sec, 0.25 sec residence time, 90% RH

The results for the above sample of MET ED 250 using a 120-minute dose of methyl iodide-127 are shown in Figure 4. The average dose concentration was 4.5 mg/M^3. Within 25 minutes of starting, corresponding to about 20% of the total dose, the effluent concentration reached that of the inlet and remained so until the dose was completed. The carbon obviously had a very poor removal efficiency. The marked decrease of methyl iodide concentration during the subsequent air purge is characteristic of a well-exhausted carbon in that there was little to desorb because there had been little adsorbed. The reported penetration of methyl iodide-131 for MET ED 250 was 49.7%.(4)

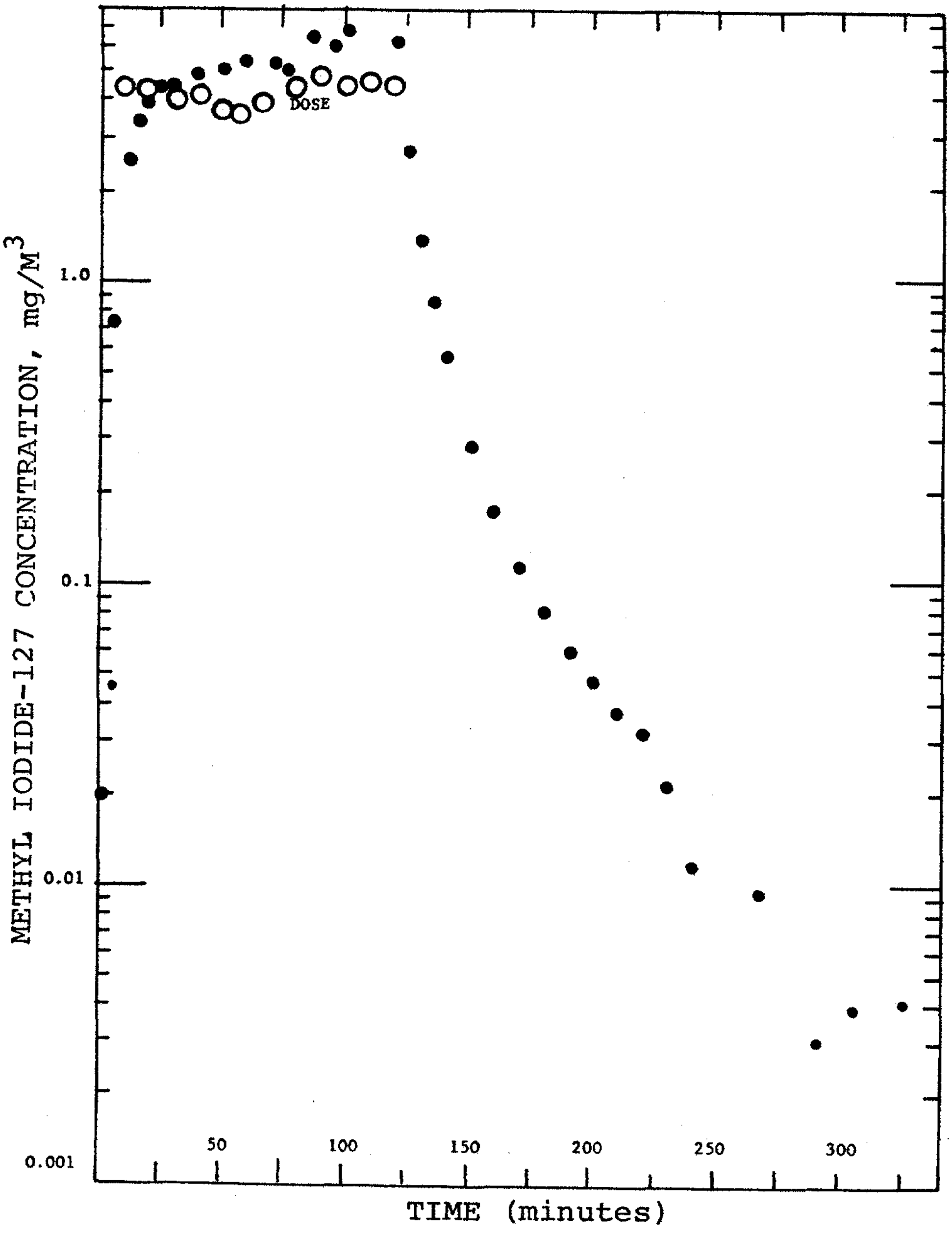

Figure 4.
On-line Measurements of Inlet and Outlet Concentrations of Methyl Iodide-127 in 90% RH Air for MET ED 250 (NRL 5149) with no Prehumidification.

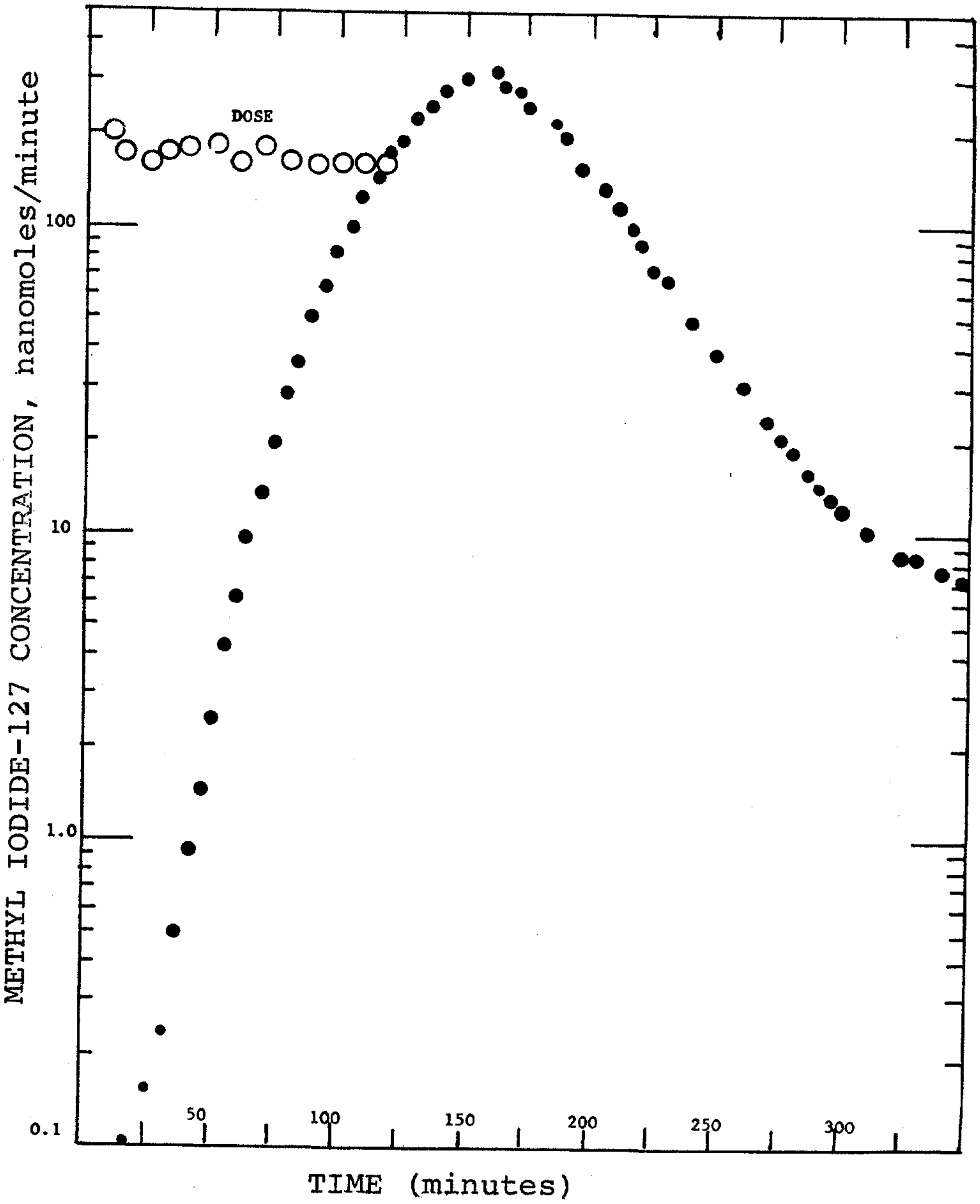

Figure 5.
On-line Measurements of Inlet and Outlet of Methyl Iodide-127 in 90% RH Air for MET ED 2759 (NRL 5153) with no Prehumidification.

The results for the sample MET ED 2759 from Unit 2 are shown in Figure 5. The effluent concentration of methyl iodide-127 did not reach that of the dose until the end of the dose period of 120 minutes. The subsequent air purge, however, continued to release methyl iodide.

The above behavior of a KI_x impregnated carbon at 25°C was observed for all of the samples from Three Mile Island. The penetration of the methyl iodide-127 was calculated from a summation of the effluent to time t divided by the dose after summation up to the same value of t and the results for the seven carbons are summarized in Table VI.

Table VI. Penetration of methyl iodide-127, No Prehumidification.

			Dose		Penetration	
	MET ED	NRL Sample	Time min.	Concn. mg/M^3	Methyl I-127 %	Methyl I-131* %
Unit 1	C217	5144	120	5.0	13.7	11.5
	76-217	5147	120	4.6	1.1	11.48
	250	5149	120	4.5	112.0	49.69
Unit 2	2652	5148	120	4.2	58.0	30.46
	2525	5152	120	4.5	69.0	44.02
	2759	5153	120	4.1	18.0	24.45
	2603	5154	120	4.4	80.0	50.92

* The penetration with methyl iodide-131 was reported by Nuclear Consulting Services, Inc.

In all measurements reported in Table VI, no pre-humidification was undertaken and the temperature and relative humidity of the air flow were maintained at 25°C and 95%, respectively.

The air purge, that immediately follows the completion of a test dose of 120 minutes, shows a very interesting and significant behavior. Many new carbons, and those with limited service life, exhibit a sharp drop in $^{127}ICH_3$ concentration immediately after the dose is completed. In contrast, the effluent concentration from service-aged carbons continues to increase after the dose period, reaches a maximum, and then slowly decreases in concentration until it approaches the initial base line of the detector.

As an example, the purge behaviors for three tests with the same carbon (NRL 5144, Table I) are shown in the graphs of Figure 6 and at 30% RH in Figure 7. The effluent concentration continued to rise and the maximum for Test 3318 (subjected to prehumidification) was 3.2 mg/M^3 at 170 minutes. The maximum for Test 3332 (without prehumidification) was 2.60 mg/M^3 at 230 minutes. The emission of

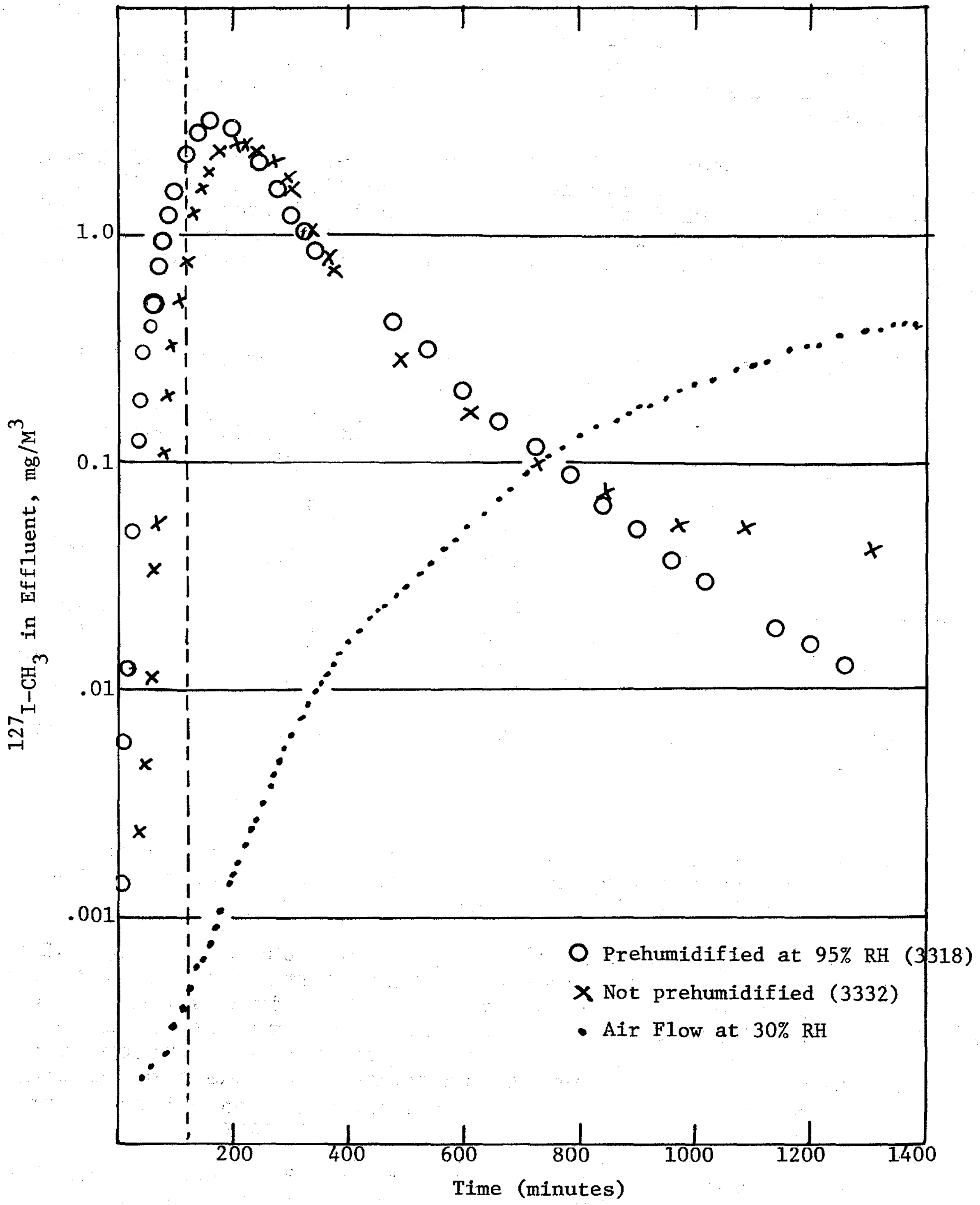

Figure 6.
Effluent $^{127}ICH_3$ (mg/M^3) for the three tests with NRL 5144

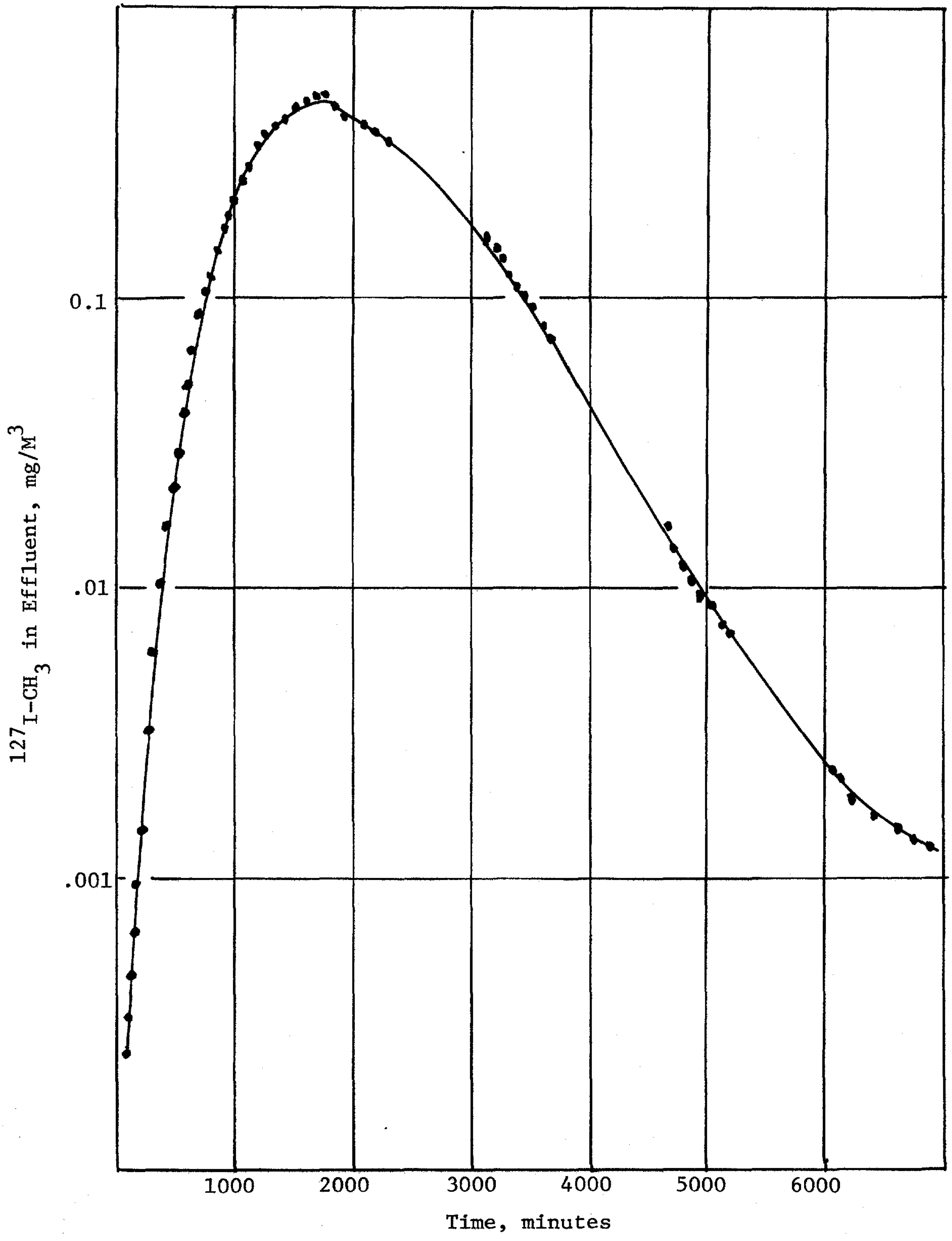

Figure 7.
$^{127}ICH_3$ in Effluent (mg/M^3) for the Prolonged (5 days) Purge at 30% RH with NRL 5144

^{127}ICH$_3$ in the air purge in Test 3339 at 30% RH was small, but reached a maximum of 0.50 mg/M^3 at 1700 minutes.

It may be concluded that a service-aged carbon filter behaves like a chromatographic column. The retention was least in the air flow of 95% RH and was most in air at 30% RH. The average concentration during the purge at 30% RH is least due to the large volume of air (43 M^3) required to attain the base line reading.

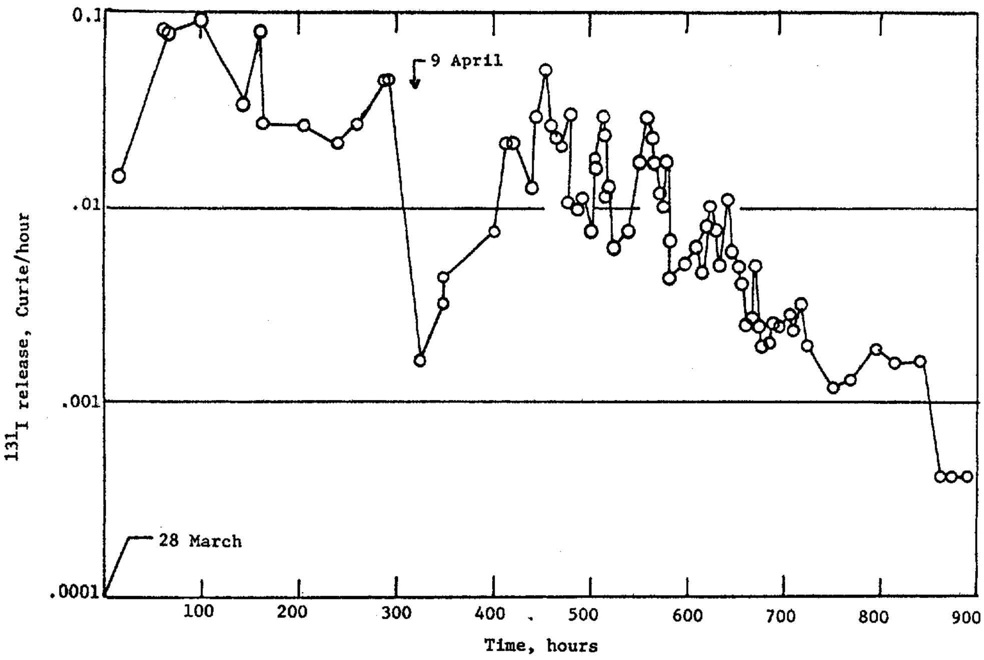

Figure 8.
The Rate of Release of ^{131}I in the Three Mile Island Accident as a Function of the Total Time after 28 March 1979.

The time-dependent release of ^{131}I is tabulated in the Rogovin Report(5) for the Three Mile Island accident of 28 March. The source data were those given in the TDR-TMI-116 Report of 31 July 1979. The monitor at Three Mile Island was located in the stack of Unit 2 which received all effluent from the iodized carbon filters until it was capped in early May 1979. The data in the Rogovin Report can be used to calculate the rates (Curie/hour) of ^{131}I release and the rates are plotted in Figure 8 as Y-coordinate with the total elapsed time from 28 March as X-coordinate. The initial iodine release and the 25-hour period thereafter may be considered as a dose period to the carbon filters; the subsequent 900 hours to 2 May is the air purge at a total flow of 10^5 CFM. There is a striking resemblance between the ^{131}I penetration at Three Mile Island and the behavior observed (Figure 7) in the laboratory with $^{127}ICH_3$ for which the air purge was followed for five days.

The interpretation of the observations and measurements in the early days after March 28th is not too straightforward. In a careful analysis, Cline, Voilleque, Pelletier and Thomas (6) concluded that the bulk of the iodine was evolved and deposited during the early part of the accident, possibly during the first day. Their thesis agrees with the above postulate that the major insult to the carbon filters could have taken place during this period. The subsequent "bleeding" of ^{131}I, set forth in Figure 8, was augmented to a lower degree by the added dose due to the slow gasification of ^{131}I from the surfaces of the various items in the containment space and duct work. There has been a major drop in ^{131}I emission after the stack was sealed, which directed all effluent through the new carbon beds installed on the roof.

V. The pH of Water Extracts

The pH of the water extracts for the seven TMI carbons is given in Table VII. The carbons from Unit 2 gave a pH significantly lower than those from Unit 1. The carbon from both units had weathered considerably and the pH values were significantly lower than those of new carbons. While the magnitude of pH is only a qualitative index of the possible efficiency of a carbon, it does signify an accumulation of inlet contaminants.(7) Evidently carbon from Unit 2 had received a larger quantity of contaminants than that in Unit 1 at the point in time when the samples were collected.

Table VII. The pH of water extracts and the water content as received

Met Ed	pH	Water Content, % (Weight Loss on Drying)
	Unit 1	
C217	7.56	5.0
76-217	7.63	5.2
250	5.58	4.1
	Unit 2	
2652	5.02	4.9
2525	4.80	8.3
2759	5.0	6.7
2603	4.3	8.0
	New Carbons	
KITEG II	10.1	
BC 727	9.5	
G 615	10.1	
MSA 463563	8.5	

VI. Concluding Remarks

1. The dominant iodine species in the thermal emission from KI_x carbons is the I_2 molecule, but appreciable organic iodides were detected in the service-aged carbons.

2. The TMI carbons had a decreased thermal stability in air flows relative to new KI_x impregnated carbons in that a greater desorption of elemental iodine took place at lower temperatures.

3. When the carbon is oxidized to CO and CO_2, the iodine evolved is proportional to the carbon burn-off.

4. The use of $^{127}ICH_3$ in penetration tests established a definite degradation of a KI_x impregnated carbon during service ageing.

5. The instability of a KI_x impregnated service carbon is responsible for the slow bleeding of the iodine during the air purge.

6. The chromatographic holdup of $^{127}ICH_3$ by service-aged carbons, together with the accompanying slow desorption step, could result in a dilution of the penetrating iodine to acceptable levels.

VII. References

1. Collins, J.T., Travers, W.D., and Bellamy, R.R., "Report on preliminary radioactive airborne release and charcoal efficiency data: Three Mile Island Unit 2," June 1979, U. S. Nuclear Regulatory Commission, Washington, D.C. 20555.

2. Deitz, V.R. and Romans, J.B., "Iodine emission during a programmed heating of impregnated charcoals in carrier air," Proc. 14th ERDA Air Cleaning Conference, Vol. 2, 844-851.

3. Romans, J.B. and Deitz, V.R., "A non-radioactive determination of the penetration of methyl iodide through impregnated charcoals during dosing and purging," Proc. 15th DOE Nuclear Air Cleaning Conference, Vol. 1, 313-334 (1978).

4. "Analysis of the adsorbers and adsorbent from Three Mile Island Unit 2," NUCON 6MT611/04, 11 June 1979, Nuclear Consulting Services, Inc.

5. Rogoven, M. and Franpton, Jr., G.T., "Three Mile Island," Vol. II, Part 2, Nuclear Regulatory Commission Special Inquiry Group (January 1980).

6. Cline, J.E., Voilleque, P.G., Pelletier, C.A. and Thomas, C.D., "^{131}I studies at TMI Unit 2," Nuclear Environmental Services, Research Project 274-06 for EPRI.

7. Evans, A.G., "Confinement of Airborne radioactivity," Progress Report, January-December 1976. DP-1463. Savannah River Laboratory, Aiken, S.C. 29801 (August 1977).

DISCUSSION

KOVACH: Would you elaborate a little more on the first part of the data on methyl iodide chromatography and the last curve, which is the iodine-131 released from TMI. I do not see the correlation.

DEITZ: In our laboratory measurements, the carbons were challenged with a dose of methyl iodide-127 of the same magnitude as in other tests (a total of 5 milligrams). After the 2-hour dose, the concentration of methyl iodide in the subsequent air purge was followed for four or five days. We found a slow desorption of methyl iodide-127 with the formation of a peak concentration which resembles a straightforward conventional chromatographic process.

KOVACH: But would not that be true only if, in fact, we were looking at the same isotopic species? As soon as there is a different isotopic species, there is absolutely no correlation.

DEITZ: The laboratory testing employed methyl iodide-127. In the release of iodine-131 from TMI, the counting measurements were, of course, blind to volatile I-127 species which are formed in the weathering of a KI_x impregnated charcoal. A charcoal responds in the same way to all iodine species, since the surface chemistry is independent of isotopic species of such large atomic weights. In the RDT laboratory testing with a challenge of methyl iodide-131, with methyl iodide-127 as a carrier, there is a 10^9 excess of I-127 relative to I-131 and only the latter is determined. In an accident, the I-131 can be in large concentration and the charcoal would then respond in similar fashion as when the I-127 is in large concentration. I would like to inquire of Dr. Kovach of the basis for his strong statement, "As soon as there is a different isotopic species, there is absolutely no correlation."

KOVACH: I just hope that the NRC will not set limits for I-127 after this.

PARISH: In your tests, where you ran for five days after injecting methyl iodide, were you using a trace of stable methyl iodide in the purge gas? If you did not inject stable methyl iodide, I am curious what the exchange mechanism was.

DEITZ: The laboratory experiments that were reported at 30% RH and the dose was introduced at a constant rate into the air stream for 120 minutes. The dose was then discontinued but the measurements of the concentration of methyl iodide-127 in the purge stream were continued. The curve that I showed indicated a peak concentration after a day or two, and then the concentrations slowly leveled off to base line.

PARISH: Are you saying that methyl iodide has a holdup time of approximately five days?

DEITZ: Yes, I think that is a good thing. You really might be able in special cases to use a carbon bed as a holdup bed and let the volatile iodide bleed slowly at a concentration that is below the allowable level.

PARISH: I am going on memory now, but as I recall, there is reason to believe that the holdup time for methyl iodide (for physical adsorption and running at 40 fpm, or so) in a couple of inches of charcoal would give much shorter times than this: on the order of minutes. This is based on some information that I saw a long time ago, and maybe I have forgotten the details.

DEITZ: You were probably dealing with new material. Nobody can use new charcoal for a significant period of time, since it steadily becomes a weathered service carbon.

PARISH: I am just saying that if the methyl iodide had not undergone ion exchange, but was simply physically adsorbed and eluted as you indicated, I think that the elution time would have been much shorter than the five days or so. Therefore, it would appear to me that ion exchange did occur.

DEITZ: We are not prepared to propose a detailed surface mechanism. There are many important facts to take into account. If one takes a clean base carbon and introduces elemental iodine, the carbon will, in the course of a temperature programming, desorb both elemental iodine and organic iodide. But, if one used the same charcoal and introduces organic iodide, then both elemental iodine and organic iodide are also desorbed. There is quite a free exchange of iodine possible on the surface of a charcoal.

CLOSING REMARKS OF SESSION CHAIRMAN:

If I can briefly summarize the papers we heard this morning, I think it is safe to say that we do not really understand everything that occurred at TMI. We need a lot of additional research effort to understand the effects of removing iodine in charcoal and, certainly, we have to pursue this research if we are to have confidence in the systems which are intended to mitigate the consequences of an accident.