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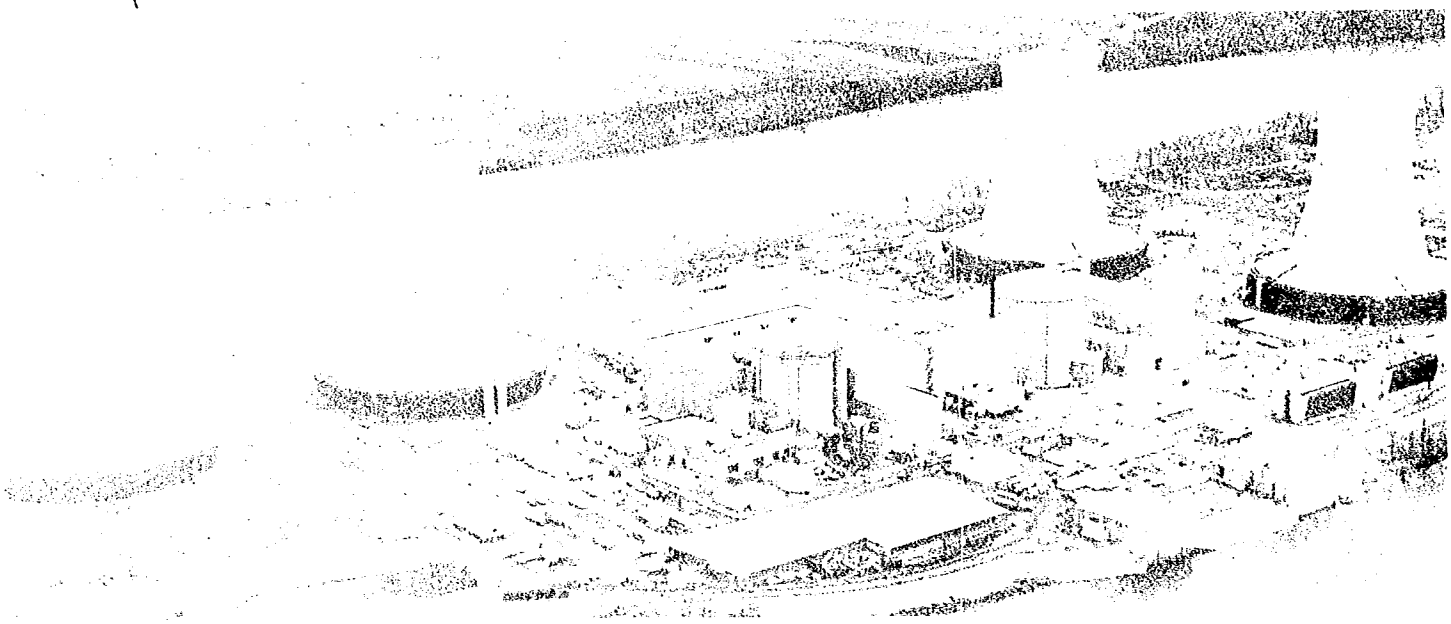

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Characterization of the Three Mile Island Unit-2 Reactor-Building Atmosphere Prior to the Reactor-Building Purge

J. K. Hartwell
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May 1981

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

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CHARACTERIZATION OF THE THREE MILE ISLAND UNIT-2 REACTOR-BUILDING ATMOSPHERE PRIOR TO THE REACTOR-BUILDING PURGE

DISCLAIMER

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Published May 1981

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ABSTRACT

The Three Mile Island Unit-2 reactor building atmosphere was sampled prior to the reactor building purge. Samples of the containment atmosphere were obtained using specialized sampling equipment installed through penetration R-626 at the 358-foot (109-meter) level of the TMI-2 reactor building. The samples were subsequently analyzed for radionuclide concentration and for gaseous molecular components (O_2, N_2, etc.) by two independent laboratories at the Idaho National Engineering Laboratory (INEL). The sampling procedures, analysis methods, and results are summarized in this report.

The concentration of ^{85}Kr present was found to be 1.02 ± 0.05 μCi/cm^3 at STP or 0.88 ± 0.04 μCi/cm^3 at TMI-2 reactor building conditions. Samples of suspended particulate contained ^{90}Sr, ^{137}Cs, and ^{134}Cs at levels of $1.9 \times 10^{-10} \pm 0.3 \times 10^{-10}$ μCi/sm^3, $8.4 \times 10^{-10} \pm 0.9 \times 10^{-10}$ μCi/cm^3, and $1.3 \times 10^{-10} \pm 0.1 \times 10^{-10}$ μCi/cm^3, respectively. Tritium and ^{14}C activity levels were $4.7 \times 10^{-5} \pm 0.8 \times 10^{-5}$ μCi/cm^3 and $3.5 \times 10^{-7} \pm 0.9 \times 10^{-7}$ μCi/cm^3. Measured concentrations or detection limits for these and 26 other radionuclides are reported. Additionally, the relative concentrations of the stable gaseous components, determined by mass spectrometry, are reported.

ACKNOWLEDGMENTS

This sampling program was conducted as a joint effort of EG&G Idaho, Inc., Exxon Nuclear Idaho Co., Inc., General Public Utilities/Metropolitan Edison, and personnel of other technical companies providing support to General Public Utilities/Metropolitan Edison. The authors wish to acknowledge the radiochemical analysis work performed by C. W. Filby and her laboratory staff and the staff of the Mass Spectrometry Laboratory under G. W. Webb. The neutron activation analyses were performed by C. P. Willis and F. A. Hohorst. Very special thanks are extended to Michael Pavelek and Mildred Boyer (RAD Services) without whose assistance the sampling described herein could not have been completed. Additionally, the authors wish to acknowledge the direct assistance of Ed Walker and Pete Wood (Bechtel), Jack Daniel (GPU), Elizabeth Scholmer and Lee Porter (Babcock & Wilcox), and Chip Chretian and Matt Bolanis (Nuclear Support Services).

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CHARACTERIZATION OF THE THREE MILE ISLAND UNIT-2--REACTOR BUILDING ATMOSPHERE PRIOR TO THE REACTOR BUILDING PURGE*

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I. INTRODUCTION

During the period from April 29, 1980, to May 2, 1980, the atmosphere of the Three Mile Island (TMI) Unit-2 Reactor Building (RB) was sampled and subsequently analyzed to provide characterization prior to the containment purge. Samples were obtained via a sampling probe installed through penetration R-626 at the 358-ft (109-m) level of the RB. Specialized sampling equipment, developed for the NRC Source Term Project, was used to obtain samples for the determination of ^{129}I (both total and iodine species), ^{14}C, ^{3}H, ^{85}Kr, and radionuclide activity of suspended particulates, and for molecular analysis of the atmospheric gaseous components. Analyses were performed by two different analytical laboratories at the Idaho National Engineering Laboratory (INEL).

This work was initiated to provide data requested by the TMI Examination Task 2.1 Planning Group relating to fission-product transport and deposition and environments characterization.[1] As the work progressed, an ancillary objective of providing data for a comprehensive evaluation of the source term for the TMI-2 RB purge developed.

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* Work supported by the U.S. Department of Energy under DOE Contract No. DE-AC07-76ID01570.

II. SAMPLING

1. Equipment

Samples were obtained using specialized sampling equipment installed at the R-626 penetration at the 358-ft (109-m) level of the TMI Unit-2 RB. The R-626 penetration is equipped with a glove box incorportating a double gate-valve-protected passthrough to the RB interior. The sampling equipment consisted of a 4.6-m by 0.64-cm diameter stainless steel sampling probe, a 2.4 liter/sec rotary vane sampling pump, flow and pressure/vacuum measurement equipment, and a variety of sampling devices. A moisture dropout trap was placed between the sampling probe and the sample media to protect the media from moisture condensation; however, no liquid collected in the moisture dropout during any of the sampling operations. All sample lines from the sampling probe inlet to the inlet of the last sample device were stainless steel. Figure 1 presents a schematic of the sampling configuration used. Note that sample gas was drawn directly from containment and exhausted into the glove box.

The sampling devices employed have been used extensively at nuclear power plants by INEL scientists on the NRC Source Term Measurement Program. The sample devices are shown schematically in Figures 2 through 4 and are discussed individually in the following sections.

1.1 Iodine Samplers

Two types of iodine samplers were employed: one for collection of a total iodine sample and another for determination of the iodine chemical species. Both sample devices are composed of five separate aluminum cups containing the desired sampling medium. The cups are O-ring sealed, and a stack of five are assembled for use in a sampling tower. In both the total iodine and the iodine species samplers, the first sample cup contains three (3) high efficiency particulate activity (HEPA) filters to remove particulate radionuclides.

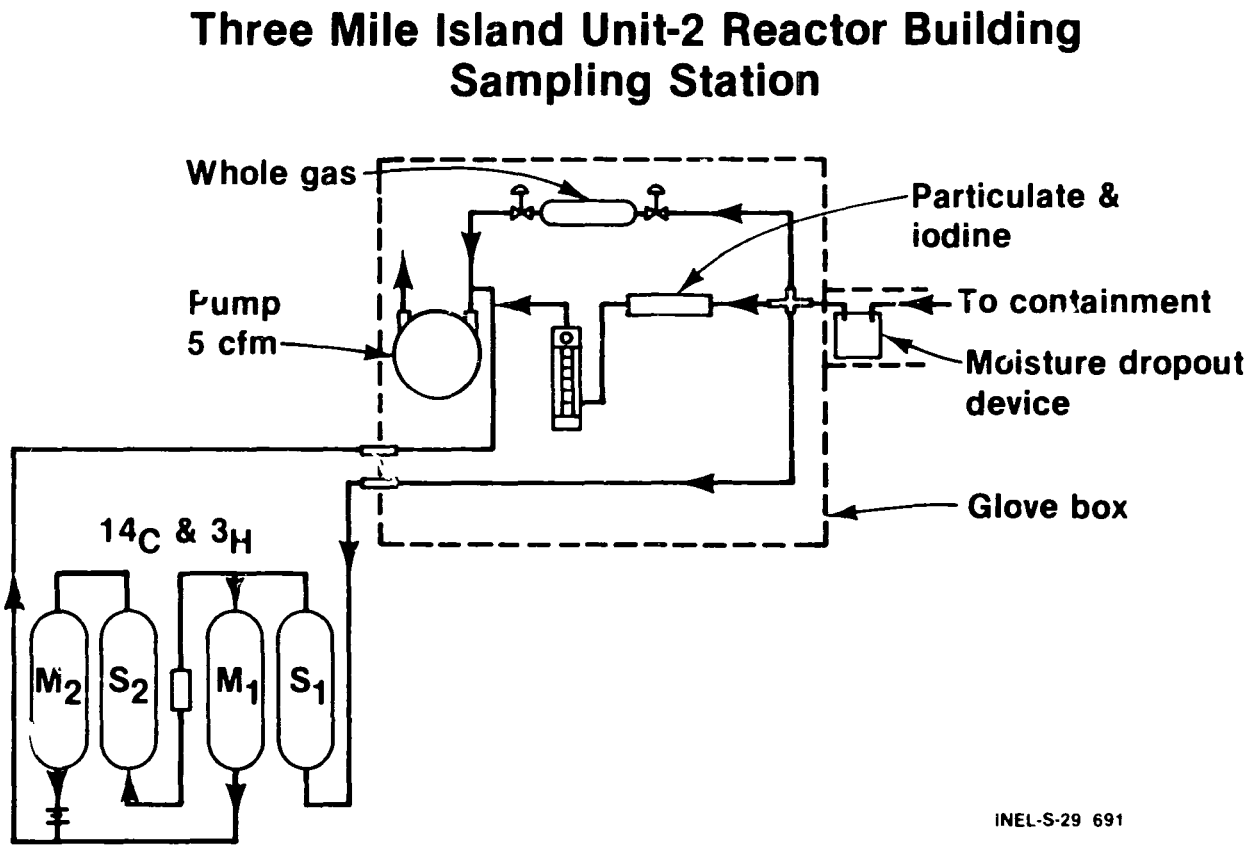

Figure 1. Three Mile Island Unit-2 Reactor Building R-626 Sampling Station Schematic

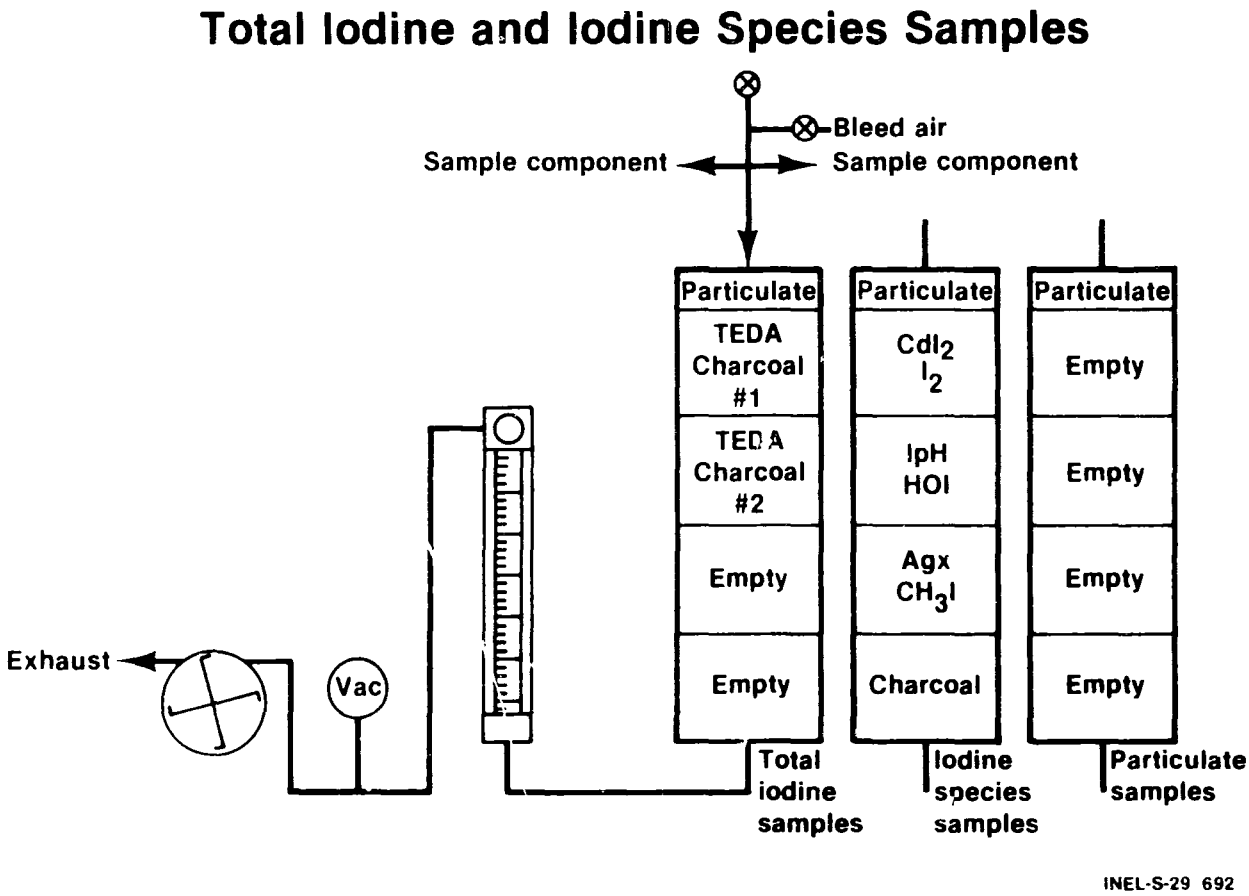

Figure 2. Sampler Tower Schematic, Showing Setup for Total Iodine, Iodine Species, and Particulate Sampling

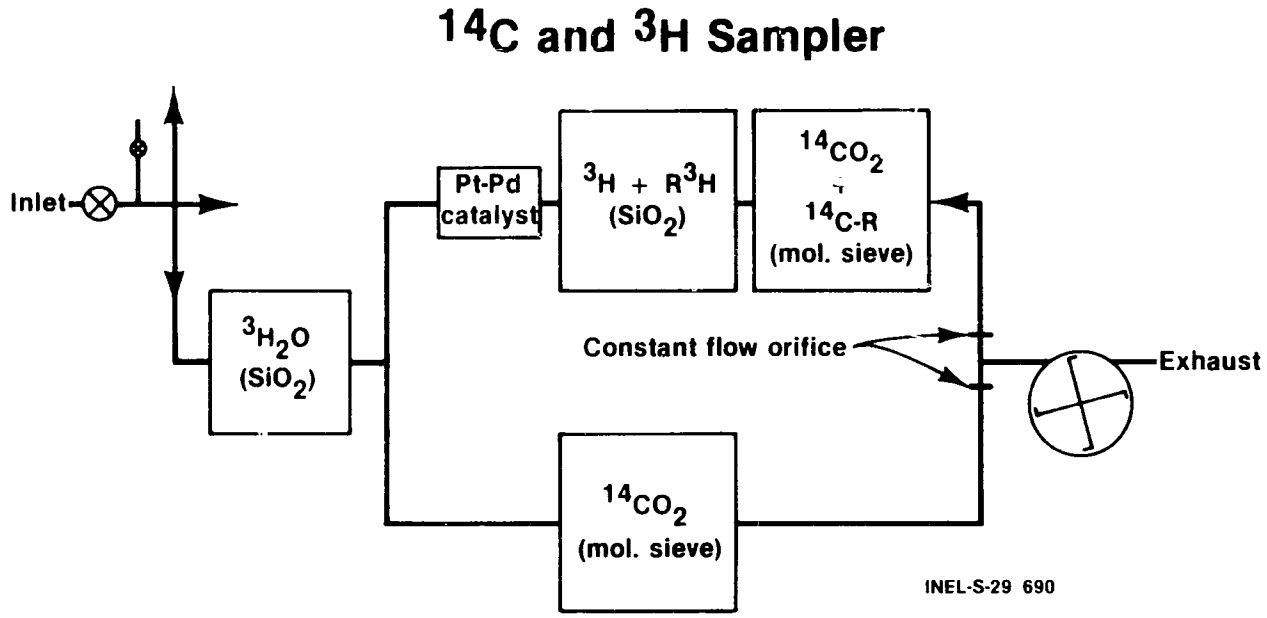

Figure 3. Carbon-14 and Tritium Sample Schematic

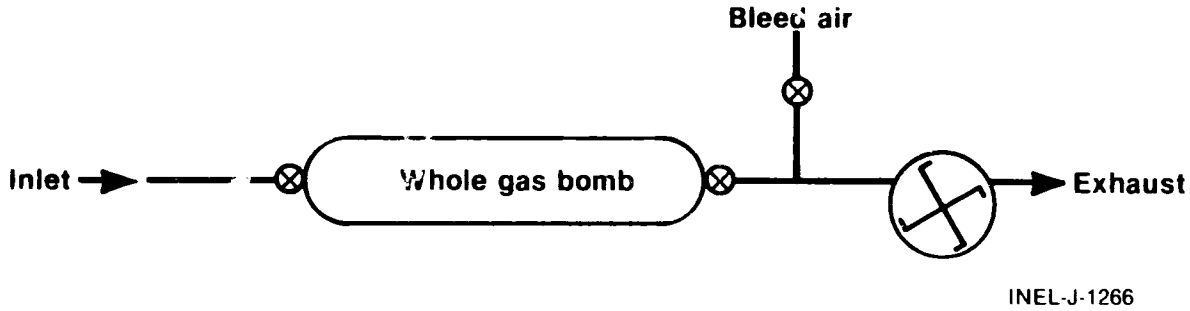

Figure 4. Whole Gas Sampler Schematic

The total iodine samplers consisted of the three HEPA filters followed by two triethylenediamine (TEDA) impregnated charcoal absorption beds. The remaining two cups in the sampler tower were empty (blank) cups.

The iodine species samplers use a variety of sampling media arranged to optimize the selective adsorption of particulate iodine, elemental iodine (I_2), hypoiodous acid (HOI), and organic (e.g., CH_3I) iodine. The first sampler stage contains three (3) HEPA filters. The second stage is a cadmium iodide (CdI_2) on Chromosorb (60/80 mesh) absorption bed for collection of elemental iodine (I_2). Stage 3 contains 4-iodophenol (IPH) on alumina (30/60 mesh) for hypoiodous acid (HOI) collection. The following stage is a 20/40 mesh silver zeolite (AgX) adsorption bed that collects the remaining organic iodines. The final sampler stage is a backup bed of TEDA-impregnated charcoal to ensure total sample collection.

1.2 Carbon-14 and Tritium

The ^{14}C-^{3}H sampler, shown schematically in Figure 3, is a sampler package developed to ensure collection of both oxidized (HTO, $^{14}CO_2$) and unoxidized (HT, hydrocarbon, etc.) species. The sampler consists of two silica gel (SG) columns, two molecular sieve (MS) columns, a heated platinum/palladium catalyst, and two flow control orifices. The inlet stream first passes through a silica gel column that removes water (H_2O and HTO). The sample stream is then split and one-half passes directly through a molecular sieve column that collects CO_2. The other half of the sample stream passes through the heated catalyst bed where unoxidized hydrocarbons, etc., are oxidized and the generated water and CO_2 are trapped in the remaining silica gel and molecular sieve towers, respectively. Tests on the catalyst oxidizing efficiency have been performed at the fixed sampler flow rate of 200 cm^3/minute. Oxidation was complete and collection was quantitative.[2]

1.3 Whole Gas Samples

Samples of the RB atmosphere were collected in standard 150 cm^3 stainless steel gas sample bombs equipped with needle valves for sample

isolation. During sampling, the whole gas sample bombs were flushed with more than 20 volume changes of RB air prior to sample isolation.

1.4 Particulate Radionuclide Samples

Samples of particulate material were collected on the package of three HEPA filters that form the entrance stage of the total iodine and iodine species samplers. Additionally, three series of particulate samples were collected by configuring the iodine sampling towers with a stack of three HEPA filters as the inlet stage, with blank cups as the backup media.

2. Methodology

With the containment isolation valves closed, the required sampling equipment was installed in the glove box at penetration R-626. The glove box was then sealed, pressure tested, and opened to the Unit-2 RB, following the approved GPU procedure for insertion of items through penetration R-616. The 4.6-m stainless steel sampling probe was inserted through the penetration and made up inside the glove box to the sampling components (see Figure 1). The distance from the glove box to the inner containment wall is an estimated 3.0 m, and thus the probe tip extended about 1.5 m into the RB. The RB air pressure was subatmospheric, and the differential pressure gauge installed on the R-626 glove box measured a nominal differential of 29.9 mm of Hg.

The samples obtained are cataloged in Table 1. Note that all sample volumes have been corrected to standard temperature and pressure (760 mm Hg and 273°K). Sampler flow rates for particulate filter, total iodine, and iodine species samples were 0.12 liter/sec, and sampling proceeded for two hours. A total flow rate of 170 cm^3/min was used for the ^{14}C-^{3}H samplers, and the sampling duration was four hours.

TABLE 1. THREE MILE ISLANG UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES

Sample Identification	Date	Time		Sample Volume (Standard cm^3)
Particulate Filter #2	05/01/80	2002	2110	461,300
Particulate Filter #3	05/01/80	1910	2000	314,850
Particulate Filter #4	05/02/80	1145	1243	393,470
Total Iodine #1	04/29/80	1420	1520	342,500
Total Iodine #2	05/01/80	1616	1516	392,700
Total Iodine #3	05/01/80	1519	1621	405,790
Total Iodine #4	05/01/80	1757	1855	386,570
Iodine Species #1	05/02/80	1246	1345	--[a]
Iodine Species #2	05/02/80	1348	1448	350,280
^{14}C-^{3}H #1	05/01/80	1416	1621	--[b]
		1757	2048	
^{14}C-^{3}H #2	05/01/80	1145	1448	--[c]
Whole Gas #1	04/29/80	1430	1500	135.1
Whole Gas #2	04/29/80	1430	1500	120.1
Whole Gas #3	04/29/80	1430	1500	121.4
Whole Gas #4	04/29/80	1430	1500	114.1

a. Due to insufficient sampler components, the CDI_2 and IPH sampler cups of the Iodine Species #1 sample were used as backup beds for the #3 particulate filter sample. Consequently, the total flow through the Iodine Species #1 sampler media cups were, respectively, 367,800 standard cm^3 for the HEPA filter and the AgX media, and 682,700 standard cm^3 for the CdI_2 and IPH. In the calculations of activity per cm^3, the respective volumes were applied.

b. Total sample flow through the silica gel adsorber upstream from the catalytic oxidizer was 44,400 standard cm^3; total flow through the other ^{14}C-^{3}H sampler components was 22,200 standard cm^3.

c. Total sample flow through the silica gel adsorber upstream from the catalytic oxidizer was 19,000 standard cm^3; total flow through the other ^{14}C-^{3}H component was 9,500 standard cm^3.

III. ANALYSIS PROCEDURES

1. General

All sample analyses were performed at the INEL by EG&G Idaho, Inc., and Exxon Nuclear Idaho Co., Inc., (ENICO). The analytical facilities of these two companies are separate and independent, and each maintains a completely independent quality assurance and standards program. In the following discussions, the EG&G and ENICO laboratories will be referred to as Lab A and Lab B, respectively. As far as practicable, analyses were duplicated in the two company facilities. Practical considerations precluded complete duplication. Lab B does not maintain a gamma spectrometer calibrated for gas sample bombs, and therefore, the determination of the radionuclide content of the whole gas samples obtained was performed solely by Lab A. Conversely, Lab A maintains a very limited capability for destructive radiochemical analyses, and so the assays of the ^{14}C-^{3}H samples, and the analyses of the particulate filters for beta-only radionuclides, transuranics, and uranium were performed solely by Lab B. Gamma spectral measurements and gross beta and alpha counting on the particulate filters as well as analyses for ^{129}I in the total iodine cartridges were performed by both laboratories. Rather than performing duplicate analyses on identical samples, analytical duplication was, in most cases, effected by dividing the samples obtained between the two laboratories. Table 2 details the assignments of the RB atmosphere samples and the analyses performed.

The four whole gas samples were actually split for analysis. The gas transfers were performed by Lab B. Prior to any gas transfer operations, the stainless steel sample bombs were counted in the same position on a gamma spectrometer. The relative counting rates in the 514-keV gamma peak of ^{85}Kr measured on the four sample bombs were compared as an initial check on sample integrity. The measured count rates were examined for statistically significant differences that might indicate if a sample cylinder had leaked or was otherwise compromised. No such differences were detected. After this initial integrity check, a sample bomb was mounted on a calibrated gas transfer rack that incorporated a manifold, a calibrated stainless steel archive bomb, a calibrated glass counting bomb, and the required pump and pressure/vacuum gauges. The system was evacuated, system pressure

TABLE 2. ASSIGNMENT OF SAMPLES FOR ANALYSIS

Lab B		Lab A	
Particulate Filters		**Particulate Filters**	
#1 Iodine Species	Gamma Spectrometry	#1 Total Iodine	Gamma Spectrometry
#2 Iodine Species	Gross beta (dissolved)	#2 PF	Gross beta (direct)
#2 Total Iodine	Gross alpha (dissolved	#3 PF	Gross alpha (direct)
#3 Total Iodine	^{129}I (direct count)	#4 PF	
#4 Total Iodine	Radiochemistry for ^{55}Fe, ^{63}Ni, ^{89}Sr, ^{90}Sr, ^{91}Y, ^{235}U, ^{238}U, ^{238}Pu, $^{239/240}Pu$		
^{14}C and Tritium			
#1 $^{14}C/^{3}H$	Liquid Scintillation		
#2 $^{14}C/^{3}H$	for ^{3}H and ^{14}C		
Charcoal Cartridges		**Charcoal Cartridges**	
#3 Total Iodine	^{129}I (direct count)	#1 Total Iodine	^{129}I (neutron activation)
#4 Total Iodine	$^{129}I/^{127}I$ (neutron activation)	#2 Total Iodine	
Iodine Species			
#1 Iodine Species	^{129}I (direct count)		
#2 Iodine Species			

TABLE 2. (continued)

Lab B		Lab A	
Particulate Filters		Particulate Filters	
Whole Gas[a]		Whole Gas[a]	
#1 Whole Gas	Mass Spectrometry	#1 Whole Gas	Gamma Spectrometry
#2 Whole Gas		#2 Whole Gas	
#3 Whole Gas		#3 Whole Gas	
#4 Whole Gas		#4 Whole Gas	

a. Whole gas samples were split by quantitative gas transfer techniques (see text).

measured, and the sample contents expanded into the gas transfer apparatus. The resulting system pressure was recorded and the bombs valved off. This procedure was then repeated for all four whole gas samples. The counting bombs were transferred to Lab A for gamma spectrometric analyses. The archive bombs were stored. The remaining contents of the original sample bombs were analyzed by mass spectrometry. After analysis of the original sample bomb contents, these sample bombs were volume calibrated, and the contents and pressures of all of the sample, archive, and counting bombs determined. The results of these determinations are shown in Table 3.

TABLE 3. VOLUMES, PRESSURES, AND STANDARD CUBIC CENTIMETERS OF THE COUNTING, ARCHIVE, AND ORIGINAL SAMPLE CONTAINERS

Counting Bombs	Volume (cm^3)	Pressure (mm Hg at 24°C)	Standard cm^3 of Sample
Counting Bomb #1	349.5	183	77.4
Counting Bomb #2	358.4	160	69.4
Counting Bomb #3	357.0	163	70.4
Counting Bomb #4	381.2	146	67.3
Archive Bomb #1	81.4	183	18.0
Archive Bomb #2	83.4	160	16.1
Archive Bomb #3	81.0	163	16.0
Archive Bomb #4	86.6	146	15.3
Original Bomb #1	152.1	735[a]	135.1[a]
Original Bomb #2	151.6	655	120.1
Original Bomb #3	150.6	667	121.4
Original Bomb #4	151.1	624	114.1

a. The pressures and standard cubic centimeters listed represent the original pressures and standard cubic centimeters.

2. Lab A Analytical Procedures

2.1 Gamma Spectrometry

Gamma spectrometry measurements on the particulate filters and on the whole gas counting bombs were performed by Lab A using equipment installed in a mobile gamma-ray acquisition and analysis system.[3] This system, which was developed for use on the NRC Source Term Measurements Program,

incorporates two independent lithium-drifted-germanium [Ge(Li)] spectrometer systems. Each Ge(Li) detector is equipped with special high-count-rate electronics with pulser-based gain compensation developed at the INEL. Data acquisition tasks are performed by a Data General NOVA 1200 minicomputer. The acquired spectral data are transmitted to a Data General Eclipse S/230 system for data analysis. The gamma spectral analysis routines used are basically those of GAUSS VI,[4] modified and optimized for nuclear power plant monitoring.

Since the mobile system is routinely used for nuclear power plant monitoring, the detectors have been calibrated for a variety of sample types at various source-to-detector distances. Among these are the particulate filters and gas counting bombs used in the TMI work.

The particulate filters were counted by insertion in a manila envelope, which in turn was placed in a special sample holder positioned in front of the Ge(Li) detector. Efficiency curves at various source-to-detector distances have been determined by repetitive counting of standard sources. The sources were prepared by depositing National Bureau of Standards (NBS) Standard Reference Materials on the particulate filter matrix and were then counted in the same source holder at the desired source-to-detector distances. The efficiency curves thus determined have an estimated overall error of less than ±5%.

Gas counting bombs were counted by placing them in a specially designed sample holder and positioning at the desired source-to-detector distance. Calibration of this geometry has been performed by a special gas calibration technique.[5] In this technique, small quantities of a volatile radionuclide-bearing organic liquid with a high activity/cm^3 are encapsulated in small quartz bulbs. The activity content of each glass bulb is determined by counting as a point source at a source-to-detector distance of more than 10 cm from a Ge(Li) detector calibrated with reference to NBS point source standards. After the total activity content of a quartz bulb has been determined, the bulb is placed in an evacuated gas counting bomb and shattered. The volatile contents expand quantitatively to fill the gas counting bomb, providing a gas standard that can be counted for accurate efficiency

curve determinations. The overall error in efficiency curves determined by this method is estimated to be ±10%, and arises primarily from variations in the dimensions of the vendor-supplied counting bombs.

Sample count results were software corrected for ambient background, random summing, and sample volume. Isotopes were identified by computer reference to an isotope library, and quantitative results or minimum detectable levels were calculated in $\mu Ci/cm^3$.

2.2 Gross Beta

Gross beta activities on the particulate filters were determined by direct counting on a calibrated end-window beta proportional counter. The proportional counter had been previously calibrated by counting ^{90}SrY deposited on stainless steel planchets. The sample count results were not corrected for filter matrix attenuation or for differences in backscattering between the filters and the standard sources.

2.3 Gross Alpha

Gross alpha activities on the particulate filters were measured directly by counting on a 2π alpha scintillation detector. Each filter was placed on a special sample pedestal, collection side up, and covered with a ZnS-coated plastic wafer, which in turn was covered by a thin mylar sheet. The sample stack was then locked together by means of a locking ring and placed on the face of a photomultiplier tube in a light-tight box. Alpha particles impacting the ZnS screen cause scintillations that are detected by the photomultiplier tube and counted by the NIM electronics attached. The detection efficiency is 50%. Sample count results were not corrected for filter matrix attenuation.

2.4 Determination of Iodine-129 by Neutron Activation Analysis (NAA)

The determination of ^{129}I by NAA is based on the reaction $^{129}I(\nu,q)^{130}I$, with determination of the induced ^{130}I activity by gamma spectroscopy. To provide analyses with the required sensitivity, a radiochemical separation of iodine from the charcoal sample matrix and its contaminants was

employed. The separation procedures used by both analytical laboratories take advantage of the water solubility of the TEDA that serves as the primary collection media for atmospheric iodine. However, the Lab A and Lab B procedures differ somewhat in methodoloy.

In Lab A, the charcoal samples were disassembled and the contents of each cartridge blended. Each sample consists of a primary cartridge and a backup cartridge. These were analyzed separately. Thus, the two total iodine samples to be analyzed resulted in four samples of blended charcoal. A weighted aliquot (approximately 5 grams) of each charcoal sample was placed in a polyethylene capsule for irradiation. A blank capsule was prepared by placing 5 grams of unexposed charcoal in a similar polyethylene capsule. A standard containing a known activity NBS ^{129}I and a known quantity of potassium iodide (for ^{127}I) was prepared by pipetting standard solutions into 5 grams of blank charcoal in a polyethylene capsule.

The samples, standards, and blank were irradiated in the Coupled Fast Reactivity Measurement Facility (CFRMF). The CFRMF is a reactor facility designed for precise reactivity measurements and provides a fine facility for NAA. Two well-characterized irradiation ports are available, and both were used in this work. The CFRMF reactor incorporates a servo-controlled power system that controls the irradiation flux precisely at a preselected level. Thus, flux monitor standards are not required for NAA in the CFRMF.

Prior to the irradiations for ^{129}I determination, one sample capsule, the blank, and the standard were irradiated for 20 minutes in a flux of 5.5×10^{11} $n \cdot cm^{-2} \cdot s^{-1}$. The irradiated capsules were counted by gamma spectrometry and the quantity of ^{127}I in each material determined by usual NAA methods. These results allowed a later direct determination of the recovery efficiency of the separation procedure for ^{129}I. After this irradiation, the capsules were allowed to decay for 24 hours before further irradiations.

All capsules were then analyzed for ^{129}I content in the following manner. Each capsule was irradiated to a total fluence of about 3×10^{15} $n \cdot cm^{-2}$ in the CFRMF. The irradiated charcoal samples were returned to the chemical laboratory, cooled for one hour (to allow complete buildup of

^{130}I from ^{130m}I decay), and then washed to remove the activated iodine. The wash liquid was a hot aqueous solution containing potassium iodide as carrier and sodium sulfite as a reductant. The charcoal was washed twice in a batch mode and then slurried to a column. The column was washed with approximately 100 column volumes of the hot wash solution, and all the wash effluent passed through a Dowex-1 ion exchange column to trap I^-. The column was then washed with water to remove completely the radionuclidic contaminants (notably ^{24}Na). The radioiodine was then washed from the Dowex column with a solution of sodium hypochlorite, and the effluent was placed in a plastic bottle for which the Ge(Li) spectrometers are calibrated. The solutions were counted on calibrated Ge(Li) spectrometers and the quantity of ^{130}I determined from the gamma lines at 518 keV, 536 keV, 668 keV, and 739 keV. On the wash solutions from samples that had their ^{127}I content determined earlier, the activity of ^{128}I (443 keV) was noted.

The quantity of ^{129}I per wash solution was determined from the induced ^{130}I activity by standard activation analysis calculations. A cumulative yield cross section of 24 barns, which includes both the direct yield of ^{130}I and the contribution from ^{130m}I, was used. The known irradiation fluxes for the two irradiation ports at CFRMF were used directly. The calculated values were corrected for sample aliquoting, for the estimated chemical yield, and for the sampled air volume, and ^{129}I concentrations were calculated in $\mu Ci/cm^3$. No ^{129}I was detected in the backup cartridges. The reported values are from the primary cartridges only.

The principal unknown in this method is the recovery efficiency of the separation procedure. Recoveries of ^{127}I were measured directly on the sample and the blank on which ^{127}I was measured directly. The ratio of the values for ^{127}I determined by direct activation and counting to those determined through the separation procedure was established, and ^{127}I recovery efficiencies of 101% and 117% were obtained for the blank and the sample, respectively. Iodine-129 recovery from the standard was 95.5%; however, this excellent recovery may partially reflect the method of standard

synthesis (direct pipetting). Based on these results and the results of further testing performed on ^{131}I-laden TEDA charcoal cartridges, a recovery efficiency of 100 ± 30% was chosen for use in the ^{129}I calculations.

3. Lab B Analytical Procedures

3.1 Gamma Spectrometry

Gamma spectrometry measurements were performed on the particulate filters assigned to Lab B for analysis. The particulate filters were prepared for analysis by pelletizing in a hydraulic press to ensure a precise counting geometry. This procedure is applicable to filters of several types and sizes, obviating the need for a separate detector efficiency curve for each filter configuration. Each filter was compressed to a uniform size at about 10,000 psi in a polyethylene vial, and the vial and pelletized filters were removed from the press apparatus and capped. The filter was then gamma counted.

Gamma spectrometry measurements were performed using a Data General NOVA-1200-based Ge(Li) spectrometer. The pelletized filters were placed at a calibrated source-to-detector distance and counted for a selected live time. The analysis software incorporates peak area determinations and isotope identification, and reports quantitative results by reference to detector efficiency curves. Calibrations have been performed using NBS standard reference materials fixed in appropriate configurations.

3.2 Gross Alpha, Gross Beta, Beta-Only Radionuclides, and Uranium and Plutonium Analytical Procedures

The referenced analyses were performed on solutions obtained by complete dissolution of each particulate filter and division of the dissolution solution into three aliquots for analysis. On aliquot was analyzed for gross alpha and gross beta activity, one for both uranium and plutonium, and the last aliquot for beta-only radionuclides. The particulate filters were dissolved by initial fume-downs with HNO_3-HF followed by fume-downs with H_2SO_4-HNO_3. The result was a clear solution with no undissolved residue.

Gross beta and gross alpha determinations were performed on the designated solution aliquots by evaporating the solution onto stainless steel planchets and counting on a calibrated end-window proportional counter. Gross beta results in $\mu Ci/cm^3$ of sampled air were reported using a ^{137}Cs counting efficiency, the aliquoting volume ratios, and the total volume of sampled air. Gross alpha results were calculated using a ^{233}U counting efficiency.

For determination of the beta-emitting radionuclides (^{89}Sr, ^{90}Sr, ^{55}Fe, ^{63}Ni, and ^{91}Y), a sequential separation and analysis sequence was used. Known quantities of stable tracers were added to the dissolution aliquots. The designated sample aliquot was boiled to remove volatile elements. Next, the sample was divided into two parts, one for the $^{89,90}Sr$ analyses and the other for ^{63}Ni, ^{91}Y, and ^{55}Fe analyses. The strontium was first precipitated as the sulfate. It was further cleaned up by precipitation as strontium nitrate, which separates it from most other cations. The radiostrontium yield was determined from the weight of strontium sulfate. For the determination of ^{90}Y, the yttrium-90 daughter was allowed to grow-in and was then separated and counted. The ^{90}Sr content was calculated from the ^{90}Y value. The strontium sulfate, which contains both ^{90}Sr and ^{89}Sr, was counted to determine total radiostrontium content. The difference between the total radiostrontium and the ^{90}Sr determined from the ^{90}Y was the ^{89}Sr.

Iron, yttrium, and nickel were gathered along with other cations by precipitating their hydroxides. Iron was separated from all other cations using a Cl^- anion exchange column. Iron-55 was determined by counting in a liquid scintillation counter. The yield of iron was determined spectrophotometrically using the orthophenanthroline complex of iron.

Nickel was separated from yttrium by precipitating yttrium hydroxide using ammonia, which forms a soluble complex with nickel. It was cleaned up by precipitation with dimethylglyoxime and extracting into chloroform. Nickel-63 was determined by liquid scintillation counting. The yield of nickel was determined spectrophotometrically using the dimethylglyoxime complex of nickel.

Yttrium was cleaned up by extraction into di-(2-ethylhexyl) phosphoric acid. Yttrium-91 was determined by counting yttrium oxalate in a gas proportional counter. The chemical yield was determined by weighing the yttrium oxalate.

Plutonium and uranium anlyses on the filter dissolution solutions were performed by procedures developed at the INEL.[6,7] These procedures use a $BaSO_4$ carrier precipitation for the initial separation of uranium and transuranic elements from the filter solutions. The carrier precipitates are dissolved, and uranium and plutonium are separated by liquid-liquid extraction. The uranium and plutonium fractions are electroplated and counted on an alpha spectrometry system. Chemical yields are determined by the addition of ^{236}Pu and ^{233}U for tracer recovery measurements.

3.3 Carbon-14

Carbon-14-bearing CO_2 was removed from the molecular sieve (MS) sampling towers using a gas transfer apparatus and procedure. An MS tower was positioned in the transfer apparatus, and the trapped CO_2 was removed by heating the tower to 350° for four hours. Gases and volatiles evolved during the bake-out process were carried through liquid nitrogen-cooled spiral traps by a dry helium purge. Carbon dioxide was retained in the spiral traps After the bake-out step, the trapped CO_2 was quantitatively transferred to a volume-calibrated bulb. The CO_2 was allowed to expand into the known system volume, and the system pressure was measured manometrically. The known CO_2 volume was then absorbed in ethanolamine, and the ^{14}C carbamate product was incorporated into an aqueous liquid scintillation gel cocktail. The vial was wiped clean and counted in a liquid scintillation counter which had previously been calibrated using known standards. Quench correction was by the external standard method.

3.4 Tritium

The tritium content of the silica gel towers was determined by direct counting in a liquid scintillation counter of an aliquot of the silica gel medium. The sample towers were dismantled, and the silica gel contents

from a sampler unit were decanted into a beaker. While monitoring the total weight gain from pick-up of atmospheric water, the silica gel was blended and three aliquots removed for liquid scintillation counting. The aliquots were mixed directly with an aqueous liquid scintillant cocktail and counted in a precalibrated liquid scintillation counter. Quench correction was effected via the external standard ratio method.

3.5 Determination of Iodine-129 by Direct Counting

In addition to the neutron activation analysis (NAA) for ^{129}I and ^{127}I performed on the charcoal total iodine cartridges, Lab B analyzed the total iodine samples and the iodine species cartridges by direct counting on a low-energy photon spectrometer (LEPS). Direct counting was required on the iodine species media since the media components (CdI_2, IPH, etc.) do not lend themselves to NAA. Direct counts on the charcoal cartridges were performed as an adjunct to NAA.

The LEPS consists of a planar germanium gamma-ray detector with a thin entrance window, and the electronics and the multichannel analyzer required to configure a spectrometer system. Particulate filters from the iodine species samplers were pelletized and counted on the detector face. The CdI_2, IPH, AgX, and charcoal media were transferred from the sample cartridges to counting vials, blended, and counted on the detector face. Typical counting times were 60,000 sec. The spectra were analyzed for the xenon x-rays and the 39.5 keV gamma ray associated with the decay of ^{129}I. The count rates obtained were quantified by replicate counting of standards prepared by blending measured aliquots of NBS ^{129}I standard reference material with the appropriate sampler media.

3.6 Determination of Iodine-129 and Iodine-127 by Neutron Activation Analysis (NAA)

The analytical scheme for NAA of ^{129}I and ^{127}I on charcoal is based on the activation of ^{129}I and ^{127}I to ^{130}I and ^{128}I, respectively. Iodine-127 was determined directly by irradiation of the charcoal media in the CFRMF reactor and gamma counting of ^{128}I. The charcoal media was then

washed to remove the iodine and the wash effluent passed through a Bio Rad AG1-X8 resin. The iodine is removed onto the ion exchange column. The ion exchange resin was then irradiated in the CFRMF and the amount of ^{127}I retained on the ion exchange column determined. From these two NAA results, the chemical yield of the iodine recovery technique was directly determined. After cooling, the ion exchange resins were reirradiated for six hours in the higher flux environment of the Advanced Test Reactor (ATR). A cobalt flux monitor wire was included with each sample in order to quantify the total neutron fluence. The irradiated ion-exchange resin samples were allowed to cool for at least 7 hours, and then gamma spectra were obtained to determine the ^{130}I content. Detector calibrations for the resin column geometry were affected by irradiation and counting of ^{127}I and ^{129}I standards loaded onto similar columns. The ^{127}I and ^{129}I sample contents were then determined by usual NAA calculation methods.

IV. RESULTS

Tables 4 through 10 present the results of the analyses performed by Lab B. Lab A analytical results are presented in Tables 11 through 13. All radionuclide concentrations are presented in microcuries per cubic centimeter at standard temperature and pressure (760 mm of Hg and 273°K). In these and all following tables, read 5.3 ± 1.4(-7) as $5.2 \times 10^{-7} \pm 1.4 \times 10^{-7}$.

Unless otherwise noted, the quoted uncertainties are one sigma errors estimated by propagation of the statistical net counting error, the estimated error in the detector calibration, and a ±10% uncertainty in the sampler flow rate. Where radiochemical analyses, sample aliquoting, or isotope separation techniques are employed, the uncertainties introduced by these operations are included in the overall error propagation if these uncertainties are large enough to affect the quoted error by 10% or more. These cases are footnoted.

Lab A and Lab B use different methods of calculating and reporting "less-than values" for nuclides that are below the minimum detectable level of a measurement technique. Lab B tests the net response obtained versus the estimated standard deviation of the net (σ). If the net is less than σ, a less-than value is reported as $<\sigma$. Lab A tests the net response versus twice the standard deviation of the net (2σ). If the net (R) is less than 2σ then a "less-than value" calculated as $<R + 2\sigma$ is reported.

TABLE 4. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (carbon-14 and tritium)

Sampler Identification	#1 ^{14}C ^{3}H	#1 ^{14}C ^{3}H	#2 ^{14}C ^{3}H	#2 ^{14}C ^{3}H
Sampler Component	Molecular Sieve	Silica Gel	Molecular Sieve	Silica Gel
Sample Data	5/1/80	5/1/80	5/2/80	5/2/80
Sample Time	1416-1621 1756-2048	1416-1621 1756-2048	1145-1448	1145-1148
Sample Volume (cm^3)	44,400	44,400	19,032	19,032
Concentration ($\mu Ci/cm^3$)				
^{3}H	--[a]	4.2 ± 1.0(-5)	--[a]	6.0 ± 1.6(-5)
^{14}C	5.2 ± 1.4(-7)[b]	--[a]	3.0 ± 0.8(-7)	--[a]

a. Not analyzed for radionuclide.

b. Read 5.2 ± 1.4(-7) as $5.2 \times 10^{-7} \pm 1.4 \times 10^{-7}$.

TABLE 5. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (particulates)

Sampler Identification	Iodine Species-1	Iodine Species-2	Total Iodine-2	Total Iodine-3	Total Iodine-4
Sampler Component	Paritculate Filter	Particulate Filter	Particulate Filter	Particulate Filter	Particulate Filter
Sample Date	5/2/80	5/2/80	5/1/80	5/1/80	5/1/80
Sample Time	1348-1448	1246-1345	1416-1516	1519-1621	1757-1855
Sample Volume (cm^3)	350,280	367,800	392,700	405,790	386,570
Concentration ($\mu Ci/cm^3$)					
^{134}Cs	1.4 ± 0.1(-10)	3.2 ± 0.4(-10)	1.1 ± 0.1(-10)	9.8 ± 1.0(-11)	1.6 ± 0.2(-10)
^{137}Cs	9.0 ± 0.9(-10)	1.3 ± 0.1(-9)	6.8 ± 0.7(-10)	5.8 ± 0.6(-10)	1.0 ± 0.1(-9)
^{58}Co	<9.1(-12)	2.4 ± 0.5(-11)	<3.4(-12)	<8.0(-12)	<6.1(-12)
^{60}Co	<2.5(-12)	5.4 ± 0.7(-11)	<5.0(-12)	2.7 ± 0.8(-12)	<2.5(-12)
^{110m}Ag	<6.1(-11)	4.6 ± 0.6(-11)	<6.5(-12)	<7.7(-12)	<5.5(-12)

TABLE 6. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (gross alpha, gross beta, and beta-only emitters)

Sampler Identification	Iodine Species-1	Iodine Species-2	Total Iodine-2	Total Iodine-3	Total Iodine-4
Sampler Component	Paritculate Filter	Particulate Filter	Particulate Filter	Particulate Filter	Particulate Filter
Sample Date	5/2/80	5/2/80	5/1/80	5/1/80	5/1/80
Sample Time	1348-1448	1246-1345	1416-1516	1519-1621	1757-1855
Sample Volume (cm^3)	350,280	367,800	392,700	405,790	386,570
Concentration ($\mu Ci/cm^3$)					
Gross alpha	<5(-11)	<5(-11)	<5(-11)	<5(-11)	<6(-11)
Gross beta	1.2 ± 0.1(-9)	1.6 ± 0.2(-9)	9 ± 1(-10)	7.3 ± 0.9(-10)	1.6 ± 0.2(-9)
^{55}Fe	<7(-11)	<6(-11)	<6(-11)	<6(-11)	<5(-11)
^{63}Ni	<3(-11)	<2(-11)	<2(-11)	<2(-11)	<2(-11)
^{89}Sr	1.2 ± 0.8(-10)	2.5 ± 0.8(-10)	<4(-11)	6 ± 3(-11)	<7(-11)
^{90}Sr	2.6 ± 0.5(-10)	2.9 ± 0.3(-10)	1.7 ± 0.2(-10)	1.2 ± 0.2(-10)	2.8 ± 0.3(-10)
^{91}Y	<4(-11)	<4(-11)	<8(-11)	<3(-11)	<3(-11)

TABLE 7. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (uranium and plutonium)

Sampler Identification	Iodine Species-1	Iodine Species-2	Total Iodine-2	Total Iodine-3	Total Iodine-4
Sampler Component	Paritculate Filter	Particulate Filter	Particulate Filter	Particulate Filter	Particulate Filter
Sample Date	5/2/80	5/2/80	5/1/80	5/1/80	5/1/80
Sample Time	1348-1448	1246-1345	1416-1516	1519-1621	1757-1855
Sample Volume (cm^3)	350,280	367,800	392,700	405,790	386,570
Concentration ($\mu Ci/cm^3$)					
^{235}U	<7.0(-13)	<2.0(-11)	<1.0(-12)	<1.0(-12)	<1.0(-12)
^{238}U	<4.0(-11)	<2.0(-11)	<2.0(-11)	<7.0(-13)	<4.0(-11)
^{238}Pu	<8.0(-12)	<9.0(-12)	<8.0(-12)	<7.0(-12)	<8.0(-12)
$^{239/240}Pu$	<2.0(-12)	<2.0(-12)	<2.0(-12)	<2.0(-12)	<2.0(-12)

TABLE 3. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (mass spectrometry)

Sample I.D. Number	Relative Concentration (Mole %)							
	H_2	He	N_2	O_2	Ar	CO_2	Xe	Organics as CH_4 Equivalents
1	0.70	0.06	83.50	14.60	0.98	0.075	0.009	0.066
2	0.89	0.07	83.90	14.00	0.98	0.105	0.009	0.071
3	0.93	0.07	83.90	14.00	0.98	0.095	0.009	0.069
4	0.92	0.07	83.80	14.00	0.99	0.073	0.008	0.070

NOTE: Other Gas Analyses

a. Neon: none over background (<100 ppm).

b. Sulfur Dioxide (SO_2): none detected (<100 ppm).

c. Krypton: none detected (<100 ppm).

d. Carbon Monoxide (CO): none detected (<500 ppm).

TABLE 9. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (total iodine-127 and iodine-129)

Sampler Identification	#3 Total Iodine	#4 Total Iodine
Sampler Component	Charcoal	Charcoal
Sampler Date	5/1/80	5/1/80
Sample Time	1519-1621	1757-1855
Sample Volume (cm^3)	405,790	386,570
Concentration ($\mu Ci/cm^3$)		
^{129}I (direct count)	6 ± 2(-11)	8.6 ± 2.2(-11)
^{129}I (NAA)[a]	5.7 ± 0.7(-11)	7.4 ± 0.9(-11)
Concentration ($\mu g/cm^3$)		
^{127}I[b]	<1.3(-4)	<1.4(-4)

a. Quoted uncertainties include the estimated contribution from the recovery procedure.

b. Iodine 127 results were below the minimum detectable level due to high blank concentrations.

TABLE 10. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB B (iodine-129 species)

Sampler Identification	Iodine Species-1	Iodine Species-2	Total Iodine-2	Total Iodine-3	Total Iodine-4
Sampler Component	Paritculate Filter	CdI_2 (Elemental I_2)	IPH (HOI)	AgX & Charcoal (Organic I)	Total
Sample Date	5/2/80	5/2/80	5/2/80	5/2/80	5/2/80
Sample Time	1348-1448	1348-1448	1348-1448	1348-1448	1348-1448
Sample Volume (cm^3)	350,280	350,280	350,280	350,280	350,280
Concentration ($\mu Ci/cm^3$)					
^{129}I (direct count)	<5(-11)	1.2 ± 0.5(-11)	1.0 ± 0.4(-11)	4.0 ± 1.3(-11)	6.2 ± 1.4(-11)
Percentage of Detected Activity		19 ± 9%	16 + 7%	65 ± 26%	

Sampler Identification	#2 Iodine Species	#2 Iodine Species	#2 Iodine Species	#2 Iodine Species	#2 Iodine Species
Sampler Component	Paritculate Filter	CdI_2 (Elemental I_2)	IPH (HOI)	AgX & Charcoal (Organic I)	Total
Sample Date	5/2/80	5/2/80	5/2/80	5/2/80	5/2/80
Sample Time	1246-1345	1246-1345	1246-1345	1246-1345	1246-1345
Sample Volume (cm^3)	367,800	367,800	367,800	367,800	367,800
Concentration ($\mu Ci/cm^3$)					
^{129}I (direct count)	<5(-11)	<1.0(-11)	<1.2(-11)	2.9 ± 1.0(-11)	2.9 ± 1.0(-11)
Percentage of Detected Activity				100%	

TABLE 11. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB A (krypton-85)

Sample Identification	#1	#2	#3	#4
Sample Type	Whole Gas	Whole Gas	Whole Gas	Whole Gas
Sample Date	4/29/80	4/29/80	4/29/80	4/29/80
Sample Time	1430-1500	1430-1500	1430-1500	1430-1500
Sample Volume (cm^3)[a]	77.4	69.4	70.4	67.3
Concentration ($\mu Ci/cm^3$)[b]				
^{85}Kr	0.98 ± 0.10	1.07 ± 0.11	1.03 ± 0.11	1.00 ± 0.11

a. Listed volumes are cubic centimeters of sample at standard temperature adn pressure contained in the sample counting bomb. Nominal sample bomb volumes were 150 cm^3.

b. Quoted errors include the contribution from a ±5% uncertainty in the sample volume, in audition to the statistical net counting error and the detector calibration error.

TABLE 12. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB A (particulate radionuclides)

Sample Identification	#1 Total Iodine	#2 PF	#3 PF	#4 PF
Sample Type	Particulate Filter	Particulate Filter	Particulate Filter	Particulate Filter
Sample Date	4/29/80	5/1/80	5/1/80	5/1/80
Sample Time	1420-1520	2002-2110	1910-2000	1145-1243
Sample Volume	342,500	461,300	314,850	393,470
Concentration ($\mu Ci/cm^3$)				
^{134}Cs	1.3 ± 0.2(-10)	2.3 ± 0.3(-10)	2.4 ± 0.3(-10)	1.6 ± 0.2(-10)
^{137}Cs	6.6 ± 0.8(-10)	1.3 ± 0.2(-9)	1.2 ± 0.1(-9)	9.6 ± 1.1(-10)
^{51}Cr	<6E-10	<3E-10	<3E-10	<4E-10
^{54}Mn	1.6 ± 0.8(-11)	1.3 ± 0.6(-11)	<3E-11	<2E-11
^{59}Fe	<5E-11	<1E-10	<1-10	<3E-11
^{57}Co	<1E-11	<1E-11	<1E-11	<2E-11
^{58}Co	2.4 ± 1.1(-11)	<1E-11	<2-E-11	<2E-11
^{60}Co	<3E-11	<4E-11	<5E-11	<4E-11
^{95}Zr	<5E-11	<2E-11	<3E-11	<4E-11
^{103}Ru	<4E-11	<1E-11	<4E-11	<4E-11
^{106}Rh	<2E-11	<1E-10	<1E-10	<1E-10

TABLE 12. (continued)

Concentration ($\mu Ci/cm^3$) (continued)				
^{110m}Ag	<2E-11	<2E-11	<2E-11	<1E-11
^{124}Sb	<1E-11	<1E-10	<2E-10	<1E-10
^{125}Sb	<3E-11	<2E-11	<6E-11	<4E-11
^{129}Te	<4E-10	<4E-10	4.8 ± 1.9(-10)	4.3 ± 1.3(-10)
^{141}Ce	<6E-11	<1E-10	<9E-11	<7E-11
^{144}Ce	<1E-10	<1E-10	<9E-11	<9E-11
^{152}Eu	<3E-11	<6E-11	<4E-11	<3E-11
^{154}Eu	<3E-11	<2E-11	<9E-11	<2E-11
^{155}Eu	<5E-11	<3E-11	<5E-11	<4E-11
Gross beta (direct)	4.8 ± 1.2(-10)	8.2 ± 2.0(-10)	8.8 ± 2.1(-10)	7.8 ± 1.9(-10)
Gross alpha (direct)	<4E-11	<2E-11	<2E-11	<2E-11

TABLE 13. THREE MILE ISLAND UNIT-2 REACTOR BUILDING ATMOSPHERE SAMPLES: LAB A (iodine-129)

Sample Identification	#1 Total Iodine	#2 Total Iodine
Sample Type	Charcoal	Charcoal
Sample Date	4/29/80	5/1/80
Sample Time	1420-1520	1416-1516
Sample Volume (cm^3)	342,500	392,700
Concentration ($\mu Ci/cm^3$)[a]		
^{129}I	9.9 ± 3.2(-11)	7.8 ± 2.6(-11)

a. Quoted uncertainties include the estimated contribution from the recovery procedure.

V. DISCUSSION

1. Whole Gas Analyses

An error-weighted average ($1/\sigma^2$ weighting) of the four values for ^{85}Kr concentration obtained yields a result of 1.02 ± 0.05 μCi/cm^3 ^{85}Kr. The volume units are cubic centimeters at standard temperature and pressure (760 mm Hg and 0°C). Samples drawn by other investigators[8] have not been corrected to standard temperature and pressure (STP). At the time of this sampling effort, the containment building pressure was 716 mm of Hg and containment temperature was 299°K. Using these values and the measured sample volumes presented in Table 3, the ^{85}Kr concentration measurements were corrected to actual containment conditions. The result is an average ^{85}Kr concentration of 0.88 ± 0.04 μCi/cm^3 at the existing RB conditions.

The utility-accepted value for the prepurge ^{85}Kr concentration in the TMI-2 RB is 0.80 ± 0.02 μCi/cc at reactor building conditions.[8] This value is in agreement with the results reported herein.

The mass spectrometric results reported in Table 8 indicate an atmosphere depleted in oxygen and high in nitrogen, helium, hydrogen, xenon, and carbon dioxide. Normal mole percentages of O_2, N_2, He, H_2, Xe, and CO_2 in the atmosphere are 20.95%, 78.09%, 5.24×10^{-4}%, 5.0×10^{-5}%, 8×10^{-6}% and 0.03%, respectively.[9]

It is interesting to note the detection of organic (hydrocarbon) constituents in the sampled air. The mass spectrometric analyses did not determine the organic species present but only reported total organics as CH_4 equivalents. Hydrocarbons were detected in all four samples at an average level of 0.069% CH_4 equivalents. Initial sampling plans included direct sampling for organic hydrocarbons by adsorption onto charcoal tubes. These samples were not taken due to practical considerations concerning the sampling time required. Lacking this information on specific hydrocarbons present, one can only speculate concerning the origin of the organic materials noted in the whole gas analyses. Some possibilities are volatile components of pump or lubricating oils, off-gasing of epoxy paints or plastics,

or possibly some combustion products produced by the charring of plastics, rubber, or epoxy materials during the hydrogen ignition that occurred during the March 28, 1979, incident. Other explanations must consider the possibility that organic materials were introduced by the sampling methodology. The stainless steel probe may have had machining oil on its interior. The water drop-out was sealed with silicone caulk which evolves solvent fumes and acetic acid during its curing process.

2. Particulate Radionuclides

The measurements of gamma-active particulate radionuclides (Tables 5 and 12) exhibit certain characteristics that highlight the inherent variability of particulate sampling and the nature of low-level gamma-ray spectrometry. The only gamma-active radionuclides detected on all filters were ^{134}Cs and ^{137}Cs. The pooled results from both labs for these isotopes are consistent, with no statistical outliers. The error-weighted average concentrations determined for ^{134}Cs and ^{137}Cs, respectively, are 1.32 ± 0.1(-10) $\mu Ci/cm^3$ and 8.4 ± 0.9(-10) $\mu Ci/cm^3$. Certain other radionuclides were detected occasionally on certain filters. Note the Lab B results obtained on the Iodine Species #2 particulate filter. This filter showed relatively high activity levels of ^{134}Cs and ^{137}Cs along with abnormally high levels of ^{58}Co, ^{60}Co, and ^{110m}Ag--nuclides not normally detected on similar filters. This is probably due to the collection of an isolated "hot" particle on this sample filter Results on the other particulate filters include isolated positive results for ^{54}Mn, ^{58}Co, ^{60}Co, and ^{129m}Te. These results are consistent with the lessthan values reported and may be due to statistical variations in the calculated net response.

The radiochemical results for the beta emitters (Table 6) show consistent detection of ^{90}Sr only. Strontium-89 was detected on three of the five filters analyzed. Note that although all results are statistically consistent, the highest values determined for both ^{90}Sr and ^{89}Sr were found on the Iodine Species #2 filter. This is the same filter that gave a high response for the gamma-emitting radionuclides. The error-weighted average ^{90}Sr concentration determined is 1.9 ± 0.3(-10) $\mu Ci/cm^3$. Calculation of an average ^{89}Sr value is complicated by the mixture of positive

("detected") results and less-than values. An error-weighted average of the positive results only yields a concentration of 8.7 ± 4.3(-11) $\mu Ci/cm^3$. An alternative method of calculation is to include the less-than values in the weighted average. In this mode, it is probably most appropriate to use an uncertainty equal to the entered value. This method of calculation yields an average ($1/\sigma^2$ weighted) of 7.3 ± 2.6(-11) $\mu Ci/cm^3$. This value is probably a better estimate of the ^{89}Sr concentration.

The theoretical activity ratio of ^{89}Sr to ^{90}Sr has been calculated from the estimated core inventories determined by the computer program ORIGEN,[10,11] and the known decay constants and sampling date. The theoretical ratio thus determined is 0.41. The sample results reported herein, using the ^{89}Sr value of 7.3 ± 2.6(-11) μCi/cc, yield a $^{89}Sr/^{90}Sr$ reactivity ratio of 0.38 ± 0.15.

The particulate filters were also analyzed specifically for ^{55}Fe, ^{63}Ni, ^{91}Y, ^{235}U, ^{238}Pu, and $^{239/240}Pu$. None of these nuclides were detected.

3. Iodine

The pooled results obtained by both laboratories for ^{129}I on the total iodine samplers were tested for statistical outliers, and none were found. An error-weighted average ($1/\sigma^2$ weighting) of the individual results yields a value of 6.6 ± 0.5(-11) $\mu Ci/cm^3$ ^{129}I. While the ^{129}I content of the TMI Unit-2 RB atmosphere has not previously been measured, the expected concentration has been estimated from the ORIGEN calculated $^{131}I/^{129}I$ core inventory ratios and measured ^{131}I concentration measurements.[12] The estimated concentration thus determined was 6.7(-11) $\mu Ci/cm^3$. The estimated concentration is in excellent agreement with our measured value. Attempts to quantify the concentration of stable ^{127}I failed due to a high ^{127}I content of the TEDA-charcoal media.

The iodine species sample results indicate that the predominant radioiodine species is the organic form. This is consistent with the behavior of "aged" radioiodine noted in studies at other nuclear power plants and the measurements of Cline et al.[12] at TMI.

4. Tritium and Carbon-14

Concentrations of ^{3}H and ^{14}C in the TMI-2 RB atmosphere were determined to be 4.7 ± 0.8(-5) μCi/cm^3 and 3.5 ± 0.9(-7) μCi/cm^3, respectively. These results are an error-weighted ($1/\sigma^2$ weighting) average of results on the two sets of ^{3}H-^{14}C samples obtained. These radionuclides have not previously been measured using equipment that samples both oxidized and unoxidized species.

5. Results Correlation

The radionuclide concentrations determined during this sampling program are presented in Table 14. The following methodology was used in calculating the final concentration values. In cases where a given radionuclide was detected in all samples analyzed, the individual results were tested for statistical outliers using a standard gap-over-range test. No outliers were detected in any of this work. The final concentrations were then determined by averaging the individual values with a weighted average program using $1/\sigma^2$ weighting. Measurement uncertainties were propagated through the averaging routine, and both the internal and external error on the averages were calculated. The quoted error is the larger of the two error components. For cases in which the analytical measurements resulted in some "positive" results and some "less-than values," the same weighted average program was used. The less-than values were considered to be "positive" results with a relative standard deviation of 100%. Error propagation was performed in the same manner as before. These cases are footnoted in Table 14. When all analytical measurements resulted in less-than values, the result of the most sensitive measurement (the smallest less-than value) was accepted as the upper concentration limit.

The average results determined per cubic centimeter at standard temperature and pressure (STP) were converted to the actual reactor building conditions of 716 mm of Hg and 299°K using the ideal gas law. The resultant concentrations in μCi/cm^3 of actual reactor building free volume are presented in Table 14 along with the estimated reactor building atmospheric inventory at the time of our measurements. A free building volume of 5.58×10^{10} cm^3 was used for these estimates.[8]

TABLE 14. RADIONUCLIDE CONTENT IN THE TMI UNIT-2 REACTOR BUILDING ATMOSPHERE
April 29 - May 2, 1980

Isotope	$\mu Ci/cm^3$ at STP	$\mu Ci/cm^3$ of Building Volume	Estimated[c] Reactor Building Inventory (μCi)
^{3}H	4.7 ± 0.8(-5)	4.0 ± 0.7(-5)	2.2 ± 0.4(6)
^{14}C	3.5 ± 0.9(-7)	3.0 ± 0.8(-7)	1.7 ± 0.5(4)
^{51}Cr	<6 E-10	<5 E-10	<3 E+1
^{54}Mn	2 ± 2(-11)	2 ± 2(-11)	1 ± 1(0)
^{55}Fe	<5 E-11	<4 E-11	<3 E+0
^{59}Fe	<3 E-11	<3 E-11	<2 E+0
^{57}Co	<1 E-11	<9 E-12	<5 E-1
^{58}Co	1.0 ± 0.3(-11)[a]	9 ± 3(-12)	5 ± 2(-1)
^{60}Co	3 ± 2(-12)[a]	3 ± 2(-12)	2 ± 1(-1)
^{63}Ni	<2 E-11	<2 E-11	<1 E+0
^{85}Kr	1.02 ± 0.05(0)	8.8 ± 0.4(-1)	4.9 ± 0.2(10)
^{89}Sr	7 ± 3(-11)[a]	6 ± 3(-11)	3 ± 1(0)
^{90}Sr	1.9 ± 0.3(-10)	1.6 ± 0.3(-10)	9 ± 2(0)
^{91}Y	<3 E-11	<3 E-11	<2 E+0
^{95}Zr	<2 E-11	<2 E-11	<1 E+0
^{103}Ru	<4 E-11	<4 E-11	<2 E+0
^{106}Rh	<1 E-10	<9 E-11	<5 E+0
^{110m}Ag	1.6 ± 0.6(-11)[b]	1.4 ± 0.5(-11)	8 ± 3(-1)
^{124}Sb	<1 E-10	<9 E-11	<5 E+0
^{125}Sb	<2 E-10	<2 E-10	<1 E+1
^{129m}Te	4 ± 2(-10)[a]	3 ± 2(-10)	2 ± 1(+1)
^{129}I	6.6 ± 0.5(-11)	5.7 ± 0.4(-11)	3.2 ± 0.2(0)
^{134}Cs	1.3 ± 0.1(-10)	1.1 ± 0.1(-10)	6.1 ± 0.6(0)
^{137}Cs	8.4 ± 0.9(-10)	7.2 ± 0.8(-10)	4.0 ± 0.4(+1)
^{141}Ce	<6 E-11	<5 E-11	<3 E+0
^{144}Ce	<9 E-11	<8 E-11	<5 E+0
^{152}Eu	<3 E-11	<3 E-11	<2 E+0
^{154}Eu	<2 E-11	<2 E-11	<1 E+0
^{155}Eu	<3 E-11	<3 E-11	<2 E+0
^{235}U	<7 E-13	<6 E-13	<4 E-2
^{238}U	<7 E-13	<6 E-13	<4 E-2
^{238}Pu	<7 E-12	<6 E-12	<4 E-1
$^{239/240}Pu$	<2 E-12	<2 E-12	<1 E-1

a. Calculated from a mixture of positive resutls and less-than values.

b. The only positive value found for ^{110m}Ag was on the "hot" filter Iodine Species #2. Eight other analyses were negative. The value presented has been calculated using the methodology discussed. A more realistic value for the overall containment average is probably <6 E-12 $\mu Ci/cm^3$.

c. Building inventory was estimated by multiplying the measured concentration times a building free volume of 5.58×10^{10} cm^3.

VI. REFERENCES

1. The TMI Examination Task 2.1 Planning Group, J. W. Mandler, Chrmn., "Recommended Data Acquisition Tasks at TMI-2 Relating to Fission Product Transport and Deposition and Enviornments Characterization," to be published, Draft Report (12/10/79).

2. N. C. Dyer, E. B. Nieschmidt, J. H. Keller, B. G. Motes, "Procedures, Source Term Measurements Program," TREE-1178, October 1977.

3. E. W. Killian and R. A. Morneau, "Advanced Mobile Multi-Processor Gamma-Ray Acquistiion and Analysis System," NUREG/CR-1668, July 1980.

4. J. E. Cline, M. H. Putnam, and R. G. Helmer, "GAUSS VI: A computer Program for the Automatic Batch Analysis of Gamma-Ray Spectra from Ge(Li) Spectrometers," ANCR-1113, June 1973.

5. R. J. Gehrke, J. I. Anderson, and D. H. Meikrantz, "Measurements of the Ge(Li) Dectector Efficiency for Gas Bomb Geometries," Nuc. Tech., 49, p. 165, June 1980.

6. C. W. Sill, F. D. Hindman, J. I. Anderson, "Simultaneous Determination of Alpha-Emitting Nuclides of Radium through Californium in Large Environmental and Biological Samples," Anal. Chem., 51, 1307 (1979).

7. C. W. Sill, F. D. Hindman, K. W. Puphel, "Simultaneous Determination of Alpha-Emitting Nuclides of Radium through Californium in Soil," Anal. Chem., 46, 1725 (1974).

8. P. J. Babel and T. C. Menzel, "Reactor Building Purge - Anaysis of the Measurement of Vented Activity," General Public Utilities Technical Data Report 182, September 1980.

9. CRC Handbook of Chemistry and Physics, 47th Edition, R. C. Weast, Editor, The Chemical Rubber Company, Clevelan, Ohio, 1967.

10. M. J. Bell, 'ORIGEN: ORNL Isotope Generation and Depletion Code," ORNL-4628, 1973.

11. ORYX-E, "ORIGEN Yield and Cross Sections - Nuclear Transmutation and Decay Data from ENDF/B," RSIC-DLC-38, 1975.

12. J. E. Cline, P. A. Roy, J. W. Hollcroft, J. Hobaugh, Jr., T. L. McVey, C. D. Thomas, Jr., C. A. Pelletier, and P. G. Voilleque, "Measurements of ^{129}I and Radioactive Particulate Concentrations in the TMI-2 Containment Atmosphere During and After the Venting," Science Applications Inc., SAU-139-80-573-LJ, October 1980.