Iodine Species in Reactor Effluents and in the Environment

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Science Applications, Inc.
Rockville, Maryland

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ELECTRIC POWER RESEARCH INSTITUTE

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Research Project 600-1

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NOTICE

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EPRI PERSPECTIVE

PROJECT DESCRIPTION

Determining the public's radiation exposure from iodine-131 emitted to the atmosphere by operating nuclear plants involves many variables, one of which is the chemical form. Iodine-131 can exist as a particulate, or as one of the following gaseous forms: elemental, organic, and inorganic. These forms behave differently in their impact on the food-milk ingestion chain. The elemental form is the most reactive and has an impact greater than the others. The chemical forms of iodine-131 and their stability and persistence in the environment affect the projected radiation exposure to humans and power plant effluent treatment requirements.

PROJECT OBJECTIVE

The primary objective of this project was to determine the persistence of the various chemical forms of iodine-131 in the environment subsequent to discharge from operating nuclear power plants. Of specific interest was whether chemical reactions would tend to drive the iodine to the elemental or organic forms.

PROJECT RESULTS

This report contains information relevant to determining the radiation exposure to the public. It should be of interest to operations and design engineers who are involved with the control of radioactive emissions.

The detailed measurements of radioiodine in gaseous effluents conducted in this study are the most extensive set of effluent species data to date. These measurements, in conjunction with the environmental air and vegetation concentration measurements, permitted an evaluation of the dispersion and disposition models in use for dose assessments. Details of the evaluation are given in this report. The study also concluded that the organic forms of iodine-131 persist in the environment and that the elemental form is rapidly converted to a less reactive form after release to the environment.

Henry Till, Project Manager
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ABSTRACT

The air-to-vegetation transfer velocity, an important parameter in evaluating dose to man from I-131 in the environment, has been used to establish an environmental reactivity ranking for the radioiodine species. In these terms, the hierarchy of reactivities is I_2 > particulate > HOI > organic iodides. Previous measurements showing that the least reactive forms, HOI and organic iodides, comprise 50% or more of the total I-131 release from boiling water reactors were confirmed in this study. Measurements of the chemical forms of I-131 and stable iodine in the environment were made using radioiodine species sampler media and a stable iodine species sampler. The compound HOI-131 was observed in the environment 1.5 km from the plant release point. Organic forms were prevalent in environmental samples. Organic iodide residence times calculated using the measured variability of ambient concentrations are > 80 days. Theoretical turnover time estimates are shorter (hours to days) but still long enough to preclude significant conversion to more reactive forms prior to arrival of the plume at the nearest real or potential pasture. In spite of the extremely short photochemical half-life estimated for I_2, a fraction of the released elemental radioiodine does deposit on vegetation.

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SUMMARY

Principal findings and conclusions of this study are briefly summarized in several topical subsections below. The study consisted of: (a) detailed measurements of I-131 in effluents and in the environs at two boiling water reactor (BWR) sites (Oyster Creek and Quad Cities), (b) measurements of stable iodine at three principal locations, two near the reactor sites, and (c) reviews and evaluations of published information on the sources and behavior of iodine in the environment. The measurement and evaluation of stable iodine behavior in the environment were considered reflective of the results of the same forces which would influence the fate of discharged radioiodine and were therefore made an integral part of the study. Two types of conclusions resulted from the work. The first relate directly to the release, behavior, and radiation dose assessment of I-131 in the environs of operating reactor facilities--the focal point of the study. The second type bear on the radioiodine dose assessment problem but are derived from evaluations of the environment behavior of iodine from sources other than nuclear reactors, namely natural sources and nuclear weapons testing.

RADIOIODINE IN BWR EFFLUENTS

The detailed measurements of radioiodine in gaseous effluent streams at the Oyster Creek and Quad Cities plants produced the most extensive set of I-131 effluent species data reported to date. These measurements confirm previous observations that a significant fraction of the I-131 discharged in all major BWR effluent streams is not elemental iodine (I_2). The average radioiodine species distributions measured at Oyster Creek and Quad Cities are presented in Table S-1.

Data obtained at Oyster Creek permitted calculation of normalized I-131 release rates (the ratio of the absolute I-131 release rate (Q,μCi/sec) to the concentration of I-131 in reactor water (C_w, μCi/g), both averaged over the same time period). The average normalized release rates for seven major effluent streams during the nearly 6-month measurement period at Oyster Creek are shown in Table S-2. Normalized release rates could not be computed for Quad Cities because of the variability of and uncertainties in the measurements of I-131 in reactor water. It should be noted that the SJAE delay line discharge has been virtually eliminated

Table S-1

SUMMARY OF MEASURED I-131 SPECIES FRACTIONS IN BWR GASEOUS EFFLUENTS

	Average Species Fractionation (%) (a)			
Source	Particulate	I_2	HOI	Organic
OYSTER CREEK				
Ventilation Air				
Reactor Building	9±1	28±1	33±2	29±3
Radwaste Building	<0.5 (b)	16±2	22±2	62±4
Turbine Building	17±1	43±2	32±2	8±1
SJAE Delay Line	<0.1 (b)	<1	12±2	87±2
Gland Seal and Mech. Vac. Pump Exhaust	<1 (b)	8±1	38±3	54±3
Feedwater and Cond Pump Room	8±2	57±7	28±8	6±3
Reheater Protection System & Lube Oil Storage Area Exh.	5±1	15±3	14±2	66±5
QUAD CITIES				
Ventilation Air Reactor Buildings	5±1	18±2	36±5	41±3
Main Chimney Exhaust (c)	8±2	40±5	24±2	29±7

(a) One standard deviation of the mean is given with the mean.

(b) Sample collected downstream of HEPA filter.

(c) Exhaust contained ventilation air from turbine and radwaste buildings, gland seal and mechanical vacuum pump discharges, and the treated off-gas from the steam jet air ejector.

Table S-2

AVERAGE NORMALIZED RELEASE RATES FOR I-131 AT OYSTER CREEK

Source	Average Normalized Release Rate[a] (Q/C_w) [(μCi/sec)/(μCi/g)]
Ventilation Air	
Reactor Building	0.22 ±.04
Radwaste Building	0.52 ±.16
Turbine Building	8.4 ±.7
Feedwater and Condensate Pump Room	0.092±.011
Reheater Protection System and Lube Oil Storage Area	0.59 ±.11
SJAE Delay Line	230±30
Gland Seal and Mechanical Vacuum Pump Exhaust	0.69 ±.11

(a) One standard deviation of the mean is given with the mean.

at most BWRs by the addition of augmented off-gas (AOG) treatment systems. The tabulated values are in reasonable agreement with other measurements at BWRs. Combining the data in Tables S-1 and S-2 indicates that approximately half of the radioiodine released is in particulate or elemental form.

RADIOIODINE DISPERSION AND DEPOSITION MODELS

Simultaneous measurements of radioiodine release rates and environmental air and vegetation concentrations permitted evaluation of the dispersion and deposition models used by the Nuclear Regulatory Commission in dose assessments of I-131 releases from light water reactors. The results of these evaluations were markedly different at the two measurement locations (one coastal and one inland in open terrain) and are discussed separately below.

Comparisons of Measured to Predicted Concentrations at Oyster Creek

The measured air concentrations (ϕ) were compared with predicted values (X) by evaluating the ratios of ϕ to X at five locations between 2.4 and 9.6 km from the Oyster Creek Plant. Ratios of ϕ to X were evaluated for total I-131 concentration (ϕ_T/X_T), inorganic I-131 concentrations (ϕ_{23}/X_{23}), and organic iodide-131 concentrations (ϕ_4/X_4). All comparisons showed the measured concentrations to be less than those predicted by the models. The lowest analytical detection limits were for organic iodide-131, so the ratios (ϕ_4/X_4) provided the most definitive set of comparisons. The observed values ranged from 0.16 to 0.33. The measured vegetation concentrations were also found to be only 1/6 to 1/3 as large as those predicted by the model.

Comparisons of Measured to Predicted Concentrations at Quad Cities

Predicted concentrations of I-131 in the Quad Cities environs were quite low, frequently below the analytical detection limits for the analyses. This fact limited the number of definitive comparisons which could be made. However, there was a tendency for observed concentrations to exceed those predicted. Minimum long term average ratios of ϕ_T/X_T were 2-5. This finding was supported by concurrent measurements of Xe-133 by another group, which found evidence of plume recirculation.

ENVIRONMENTAL PERSISTENCE OF IODINE SPECIES

The environmental reactivity of the iodine species can be defined in terms of the air-to-vegetation transfer velocity of each form. In these terms, the sequence of reactivities is I_2> particulate > HOI > CH_3I. The reactivity of other organic forms is presumably comparable to that of CH_3I. It is therefore important to study the environmental persistence of the iodine species. Several lines of evidence are summarized below.

Photochemistry

Photochemical considerations lead to the conclusion that both elemental iodine (I_2) and organic iodides (CH_3I and others) will be subject to photodissociation in sunlight. Estimated residence times for I_2 are less than a minute during the day, much shorter than the residence times for CH_3I and other alkyl iodides in plant effluents ($\sim$60 hours in sunlight). Estimated atmospheric residence times which consider lattitudinal and diurnal variations are $\lesssim$5 minutes for I_2 and $\lesssim$200 hours for CH_3I. The fate of I atoms released by photodissociation is not well understood; IO, IO_2, HOI, and organic forms are possible reaction products. Of all the potential atmospheric iodine compounds, CH_3I is the only one which has been definitively identified.

Field Measurement Results

A number of studies have been conducted of the distribution of stable iodine in the atmosphere. The operational distinction between inorganic iodine concentrations (ϕ_i) and organic iodide concentrations (ϕ_o) has been the separation achieved by tetrabutyl ammonium hydroxide (TBAH)-coated filters; the inorganic forms I_2,HI, and HOI are trapped by these filters and CH_3I is not. Note that lower case letter subscripts are used for the various forms of stable iodine and numeric subscripts are used when discussing radioiodine species. This convention is followed because the sampling systems used to operationally define the species are different. Table S-3 summarizes the results of stable iodine species measurements made as part of this study and other such measurements. The particulate concentration (ϕ_p) is generally a small fraction of the total (ϕ_t), averaging 0.16--0.18. The fraction of the gaseous in organic form has been determined at 8 locations. The ratio $\phi_o/(\phi_i+\phi_o)$ is somewhat variable with a mean between 0.57 and 0.60. (The mean ratio cannot be uniquely determined because some measured component concentrations were below analytical detection limits). Gas chromatographic measurements near Atlantic ocean sources by Lovelock and inland from the Pacific coast by Singh have shown high concentrations of methyl iodide, suggesting that it is a major constituent of the gaseous iodine.

Table S-3

STABLE IODINE SPECIES DISTRIBUTION RATIOS

Location	Particulate Iodine to Total Iodine (ϕ_p/ϕ_t)	Organic Iodides to Total Gaseous Iodine $[\phi_o/(\phi_i+\phi_o)]$
Hawaii(a)	0.22	ND(b)
Antarctica(a)		
McMurdo	0.29	ND(b)
South Pole	0.16	ND(b)
Bermuda(a)	0.078	0.62
Boston(a)	0.33	ND(b)
New York City(a)	0.0 --0.13(c)	0.25
Oyster Creek New Jersey(d)	0.21--0.33	0.32--0.50
Northwest Territories(a)	0.056	0.88
Arizona(a)	0.075	0.31
Kansas(a)	0.12	0.71
Quad Cities Illinois(d)	0.18--0.20	0.71--.74
Front Range Colorado(d)	0.15--0.18	0.73--.82

(a) Ratios of the reported mean component concentrations. See Table 2-1 for References.

(b) No data on gaseous iodine chemical forms.

(c) When the ratio could not be uniquely determined the range of possible values is given.

(d) Mean value of ratios for individual samples obtained in this study.

Wet and dry removal process are much less effective for organic forms than for elemental and particulate forms. Residence times were estimated using Junge's empirical relationship

$$\sigma_r \tau \approx 0.14 \text{ yr} \qquad \text{(S-1)}$$

in which σ_r is the relative standard deviation of the air concentration of the gas and τ is its residence time (yr). The estimated organic iodide residence time using the Quad Cities and Colorado Front Range was 120 days. This agrees well with the residence time (estimated the same way) for CH_3I in Yosemite and with similar estimates of the total gaseous iodine residence times made using the data from Hawaii. The estimates for Yosemite and Hawaii are based on σ_r for short sampling times ($<$24 hr), which indicates that the Illinois-Colorado result, based on (7-10)-day samples, is not due to the sampling duration. The organic iodide residence time in the Oyster Creek environment was estimated using Equation (S-1) to be 60 to 80 days.

Environmental concentrations of inorganic and organic I-131 were measured using the same media employed in the standard radioiodine species sampler. The inorganic forms I_2 and HOI were identified in the environment by that technique. HOI was first observed in an environmental air sample during the 1977 monitoring at Quad Cities. Observations of I_2 in environmental air samples are consistent with previous observations that a fraction of the effluent I-131 enters the milk food chain via deposition on vegetation. At Oyster Creek organic I-131 was frequently observed in the atmosphere. However, the ratio of organic iodide-131 to total gaseous I-131 was frequently indeterminate, due to less than detectable amounts of the inorganic forms. The ratios predicted from effluent monitoring and meteorological modeling (discussed above) ranged from 0.7 to 0.9 were in some cases distinctly higher than individual observed values, but analysis of individual cases indicated that variations in the release rate and uncertainties in the meteorological model or other mechanisms must account for the differences. A simple methyl iodide photolysis scenario could not explain the observations and would be inconsistent with the residence half-times estimated for stable organic iodide.

Observations of airborne I-131 from a nuclear explosion above ground suggest that organic iodides are the most persistent gaseous species. A large fraction of the I-131 in such debris is associated with particulates. However, the gaseous fraction changed from principally inorganic to principally organic during the 2 months following the detonation. This finding is consistent with (a) the species data for stable iodine, (b) the estimates of residence time obtained using Eq. (S-1), and (c) other residence time estimates based on deposition velocity and wet scavenging parameters. Transport of iodine from air to vegetation by precipitation scavenging was again found to be important for fallout I-131. As noted, most of fallout radioiodine was initially associated with particulates. In contrast, no I-131 from plant releases was detected in precipitation samples.

Although the organic iodine residence time observed at Oyster Creek was shorter than for other locations (60 to 80 days as opposed to 120 days or more) it is sufficiently long that no significant conversion of organic iodide to more reactive forms would occur between the release point and the nearest site boundary, or even within 50 miles of the plant. Theoretical turnover time estimates are shorter (hours to days) but still long enough to preclude significant conversion prior to arrival of the plume at the nearest real or hypothetical dairy farm pasture. It is also clear that, in spite of the extremely short photochemical half-life estimated for I_2, a fraction of the released elemental radioiodine does deposit on vegetation and enter the milk food chain.

Section 1

INTRODUCTION

BACKGROUND

The assessment of radiation doses likely to be received from radioiodine released from light water reactors (LWRs) is a difficult and complex task. There are many variables which affect the behavior of radioiodine released to the environment and which should be considered in models used to project radiation dose commitments. Principal among these are the chemical form of the released radioiodine and the meteorological variables which affect the dispersion of the released radioiodine species, their chemical stability, and their transfer to vegetation. Simultaneous measurements of these variables at an operating LWR provide data which can be used to evaluate the transport models and to determine, or at least bound, the values of important parameters. The variables which affect the subsequent transfer steps in the milk food chain, while also important to radiation dose assessment, are beyond the scope of this discussion.

The radioiodine in gaseous effluents may be associated with particulate material or in one of the several possible gaseous forms. Any discussion of gaseous iodine species must be related to the method used to obtain the concentrations of those species. The 4-component radioiodine effluent species sampling cartridge (1)* used in this study and its performance are discussed in detail in Appendix A and in reports referenced in Section 3; however, a brief summary is appropriate here.

The sequence of sampling media is designed to remove particulates first. Then the most reactive gaseous form (I_2) is removed from the sampled air. In the third section, a less reactive form (HOI) is extracted and finally the least reactive forms (organic iodides) are trapped. From the point of view of environmental dose assessment, the "reactivity" of these forms is most suitably defined by the air-to-vegetation transfer velocity. This quantity, whose only resemblance to a velocity is its dimensions (2), is the ratio of the activity deposited per unit area of vegetation to the time-integrated air concentration at a reference height (usually

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*References are listed at the end of each section of this report.

one meter) above the vegetation. Thus the transfer (or deposition) velocity is

$$V_d = \frac{\text{areal concentration of activity on vegetation } (\mu\text{Ci/m}^2)}{\text{time-integrated air concentration } (\mu\text{Ci-sec})/\text{m}^3}$$

and has dimensions L/T. Table 1-1 shows the relationships among sampler components, chemical forms, and air-to-vegetation transfer velocities. It should be noted that the transfer velocities listed for particulates and elemental iodine are, respectively, long term average and typical values for those forms. As indicated in the footnotes to the table, individual short term values for I_2 can vary substantially with meteorological conditions and the characteristics of the vegetation. The dependence on these variables of the transfer velocities for HOI and organic iodides is presumably similar, but too few experimental results are available to draw firm conclusions.

Since the air-to-vegetation transfer of radioiodine is a critical step for its entry into the milk food chain, the transfer velocities shown in Table 1-1 are indicative of the contribution of each species to doses from ingestion of contaminated milk. Those performing environmental dose assessments formerly assumed that all of the radioiodine releases from LWRs were of elemental iodine, the most environmentally significant form. However, measurements made during recent years have repeatly shown that a significant fraction of the radioiodine released was in other forms (11, 12, 13, 14, 15). The degree of overestimation of dose commitments from radioiodine entering the milk food chain would depend on the actual species distribution at the location of concern (*i.e.*, the nearest real or potential dairy farm), generally within a mile of the LWR's release point.

Thus, the distribution of the released iodine species and their persistence during transport to the dairy location in the environment are factors important to the calculation of the dose commitment attributable to the release. The environmental conversion of less reactive forms to elemental iodine (or another form with a comparable deposition velocity) would nullify any credit assumed for these species at the discharge point. On the other hand, conversion of discharged elemental iodine to less reactive forms would result in much lower doses to the consumers of milk from the nearby dairy. Since both I_2 and organic iodides are subject to photodissociation (16) these possibilities must both be considered. The nonreactive species, HOI and organic iodides are also the most difficult to remove from effluent air streams, so knowledge of their fate in the environment can also affect the capital and operating costs of LWR effluent treatment systems. If these less

Table 1-1

RELATIONSHIP BETWEEN RADIOIODINE EFFLUENT SPECIES SAMPLER COMPONENTS AND AIR-TO-VEGETATION TRANSFER VELOCITY

Effluent Sampler Component	Principal Radioiodine Species Trapped	Air-to-Vegetation Transfer Velocity of Species (cm/sec)
HEPA Filter	Particulate	0.2[a]
Cadmium Iodide Bed	Elemental (I_2)	1[b]
4-Iodophenol Bed	Hypoiodous Acid (HOI)	≤0.1[c]
Charcoal or Silver Zeolite Bed	Organic Iodides (CH_3I and others)	0.0001--0.005[d]

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[a] Based on the behavior of radioactive fallout particles. Long term average value from Reference (3). Short term values depend on wind speed, grass height, atmospheric stability, and particle size (4).

[b] Typical value observed in short term field experiments conducted under "average conditions". Individual values vary substantially owing to differences in wind speed, grass height and density, and atmospheric stability (4, 5, 6).

[c] Based on the results of a single laboratory experiment (7).

[d] Range of values from three field experimental studies. Other determinations may vary due to differences in wind speed, atmospheric stability, and vegetation canopy characteristics (8, 9, 10).

reactive forms persist in the environment the effluent treatment costs could be reduced without decreasing the level of protection afforded the nearby population.

PURPOSE AND SCOPE OF THE STUDY

The principal objective of the study was to determine the persistence of the various chemical forms of I-131 in the atmospheric environment of operating LWRs. It was necessary to measure the radioiodine species fractionation and the release rates of I-131 in gaseous effluents from the facility and to measure radioiodine concentrations in the atmosphere near the plant. These measurements, together with the continuous meteorological monitoring performed by the plant, permitted the comparison of the measured and predicted dispersion of discharged I-131. To provide further data on environmental iodine species and on air-to-vegetation transport by wet and dry processes, vegetation and rainfall sampling were also undertaken. A third approach to the problem was to obtain information on the distribution of stable iodine species in the environment. This stable iodine species distribution presumably is in approximate equilibrium with the environmental processes which would control the environmental species distribution of I-131 in LWR effluents. During the course of the study, a fourth approach to the problem became possible when an I-131 source of opportunity, fallout radioiodine, presented itself. Trends in the behavior of fallout I-131 species should reflect the same processes controlling the environmental behavior of stable iodine and I-131 in LWR effluents. Measurements of fallout I-131 were conducted to observe iodine species behavior. To complement the measurement program described above, literature reviews were undertaken, by the author and by consultants in the field of atmospheric iodine chemistry, to summarize relevant information from other theoretical and experimental studies and to consider the expected behavior of iodine species in the atmosphere.

PLAN OF PRESENTATION

The results of the literature reviews are presented in Section 2. Sections 3 and 4 contain the results and discussion of the measurements of I-131 in and near two operating LWRs--the Oyster Creek Nuclear Generating Station and the 2-unit Quad Cities Nuclear Power Station. The stable iodine measurements are presented in Section 5. The final section summarizes the results of all the various approaches to the problem and contains the conclusions reached. Detailed analytical procedures and measurement results are contained in the Appendices.

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Section 2

THEORETICAL CONSIDERATIONS AND PREVIOUS MEASUREMENTS

IODINE IN GASEOUS EFFLUENTS FROM REACTORS

The radioiodine effluent species sampler (1), briefly described above, has been used to ascertain the chemical forms of radioiodine being discharged in nuclear reactor effluents. The sampling system and the corresponding identification of the iodine species are both the result of a long period of testing and evaluation of radioiodine behavior in nuclear reactor systems (2). The sampling system represents the current state-of-the-art of airborne radioiodine measurements, but evaluation of I-131 profiles in segmented cadmium iodine sampling beds suggests there may be a third species, less reactive than elemental iodine, in reactor effluents which has not been identified (3). Identification of the "known" radioiodine species has generally been based on their reactions with sampling media. For example, the elemental iodine fraction was formerly determined by its reaction with metallic copper or silver (4) and the identification of HOI was based in part on its reaction with 4-iodophenol (5). In contrast, organic iodine forms in simulated reactor containments and in effluents have been more directly identified using gas chromatography (6,7). From the point of view of iodine chemistry, it would be desirable to have definitive data on the molecular properties of the forms trapped by the cadmium iodide and 4-iodophenol beds in the radioiodine species sampler. The measured radioiodine species distributions in reactor effluents have consistently indicated the presence of significant fractions of non-elemental iodine. From a dose assessment viewpoint, the relationship between the chemical form as determined using the sampler, and the air-to-vegetation transfer velocity (Table 1-1) is the crucial one.

There are nine principal iodine isotopes produced in nuclear reactors, those with mass numbers between 127 and 135. Iodine-127 is stable. The rest are radioactive; most of the isotopes have half-lives of less than 1 day. Iodine-129 and I-127 account for ∿90% of the total fission product iodine mass and I-131 accounts for most of the remaining 10%. The total mass of iodine produced in a 3000-MWt reactor during 1 year of operation is ∿5.5 kg (8). The amount of iodine released from a reactor can be estimated using a simple model relating the iodine inventories in the fuel (I_f), fuel pin plenum (I_p), and coolant (I_c). The model is described by three equations:

$$\frac{dI_f}{dt} = P - (\lambda + \lambda_1) I_f \tag{2-1}$$

$$\frac{dI_p}{dt} = \lambda_1 I_f - (\lambda + \lambda_2) I_p \tag{2-2}$$

$$\frac{dI_c}{dt} = \lambda_2 I_p - (\lambda + \lambda_3) I_c \tag{2-3}$$

in which

I_f, I_p, and I_c are the number of atoms of a given isotope of iodine in the fuel, fuel pin plenum, and coolant, respectively.

P is the average net production rate (atoms/sec) of the isotope from fission of U and Pu in the fuel, considering removal by neutron capture.

λ is the radioactive decay rate constant (sec^{-1}) for the isotope.

λ_1 is the fuel-to-plenum escape rate constant (sec^{-1}) for iodine.

λ_2 is the plenum-to-coolant escape rate constant (sec^{-1}) for iodine.

λ_3 is the coolant cleanup rate constant (sec^{-1}) for iodine.

At equilibrium, the coolant inventory would be

$$I_{ce} = \frac{\lambda_2 I_p}{(\lambda+\lambda_3)} = \frac{\lambda_1 \lambda_2 I_f}{(\lambda+\lambda_3)(\lambda+\lambda_2)} = \frac{\lambda_1 \lambda_2 P}{(\lambda+\lambda_3)(\lambda+\lambda_2)(\lambda+\lambda_1)} \tag{2-4}$$

For the two principal isotopes, I-127 and I-129, $\lambda \sim 0$, so

$$I_{ce} \simeq P/\lambda_3 \text{ for I-127 and I-129.} \tag{2-5}$$

The reactor coolant cleanup rate constant is

$$\lambda_3 = \frac{F\ r}{M} \tag{2-6}$$

where F is the cleanup flow rate (g/sec)

r is the average removal factor for iodine in water passing through the filtration system

M is the mass of reactor coolant (g).

For Oyster Creek, the first plant at which measurements were made, $M = 2.0 \times 10^8$ grams and the design cleanup flow rate (F) is 2.5×10^4 g/sec (9). During the measurement period (see Section 3) the actual average cleanup flow rate was 2.34×10^4 g/sec. The average removal factor r is between 0.9 and 1.0 and is taken as ∿1.0 for this calculation. Hence, $\lambda_3 \simeq 1.3 \times 10^{-4}$ sec^{-1}. For a power level of 1810 MWt, P(I-127)≃7.2×10^{16} atoms/sec and P(I-129)≃ 3.5×10^{17} atoms/sec (8), so the equilibrium coolant inventories would be about 0.12 grams I-127 and about 0.60 grams I-129. The average total iodine concentration in reactor coolant would be about 4×10^{-9} grams iodine per gram of water. The data for I-131 indicate an average normalized release rate of 100 g/sec for the facility (9). Assuming the I-127 and I-129 behavior is similar to that observed for I-131, the estimated total iodine release rate is ∿400 ng/sec. The average concentration of reactor produced iodine in plant ventilation air would be (400 ng/sec)(78 m^3/sec) or ∿ 5 ng/m^3. The estimated concentration, which could be ∿10 ng/m^3 at larger reactors, is perhaps 40-80% of typical stable iodine air concentration in the air in the United States (see below). While the concentration of iodine in plant effluents may be higher than ambient levels, a typical dispersion factor for reactor effluents is ∿10^{-8} (sec/m^3), so site boundary concentrations of reactor produced iodine would be in the fg/m^3 range and well below ambient levels. The iodine released from the reactor should approach the same equilibrium composition of chemical forms exhibited by the existing stable iodine in the atmosphere.

PREVIOUS MEASUREMENTS OF AIRBORNE STABLE IODINE

In earlier measurements of airborne stable iodine concentrations the only distinction made was between iodine associated with particulates and gaseous iodine compounds (10,11,12). More recent measurements, including those reported in Section 5, have utilized samplers designed to separate the airborne iodine into particulate, inorganic gas, and organic gas fractions (13,14). The previously observed stable iodine concentrations are summarized in Table 2-1. Gaseous iodine compounds are the principal constituents with variable distributions of inorganic and organic iodine. Reported concentrations of methyl iodide along the east coast are quite high, up to 2×10^4 ng/m^3 for Bayonne, N.J. in 1973. Actual mean or median concentrations cannot be determined from the data as reported, but the high concentrations are not consistent with the data in Table 2-1 unless one postulates local sources among the many chemical discharges near the coast between Baltimore and New York City. While high concentrations were also measured over local oceanic sources (16), the average Atlantic

TABLE 2-1

PREVIOUS STABLE IODINE CONCENTRATION MEASUREMENTS

Location	Average Stable Iodine Concentration (ng/m^3)				Reference
		Gaseous Forms			
	Particulate	Total	Inorganic	Organic	
Hawaii	2.4	8.3	ND[a]	ND	(10,11)
Antarctica McMurdo	0.9	2.2	ND	ND	(12)
South Pole	0.5	2.7	ND	ND	(12)
Northwest Territories	0.2	3.4	0.4	3.0	(13)
Bermuda	3.8	45	17	28	(13)
Arizona	1.3	16	11	5	(13)
Kansas	2.4	17	5	12	(13)
New York City	1	8	6	2	(14)
Boston	6.8	14	ND	ND	(15)

(a) No data on gaseous iodine chemical forms.

concentration of CH_3I and the total gas concentrations data from Hawaii suggest much lower levels would be found in east coast locations. Total iodine concentrations as high as 7000 ng/m^3 were reported for the United Kingdom (21). The range in concentrations was wide; the average given was 100 ng/m^3. It was stated that the iodine was principally in gaseous form, but no data on chemical species were obtained.

While the oceans are almost certainly the principal source of atmospheric iodine, the release mechanisms and chemical forms released are uncertain (see References (11, (16), and (22) through (27). Equally uncertain are the interactions and fates of the various components of the injected into the atmosphere. As noted, only methyl iodide has been identified directly as an airborne species. As is the case for the radioiodine effluent species sampler, the inorganic stable iodine species are distinguished by their reactions with sampling media, now typically TBAH-coated filters. These filters were initially tested in the laboratory to determine the collection efficiency for the inorganic forms I_2 and HI (13,14). A preliminary evaluation of the HOI collection efficiency of these filters was conducted as part of the study being reported (Section 5). This may be important, as there is now some evidence for the existence of HOI (as indirectly determined by radioiodine species sampling) in the environment (Section 4). However, theoretical atmospheric chemistry considerations, discussed below, raise as yet unanswered questions about environmental iodine behavior.

PREDICTED CHEMICAL BEHAVIOR OF IODINE IN THE ATMOSPHERE

Both the elemental iodine and organic iodides released to the atmosphere from nuclear facilities and the oceans are subject to photodissociation. Zafiriou (28) estimated a minimum half-life of I_2 in bright sunlight of less than 10 seconds and has since suggested that an average daylight half-life of 15-300 seconds (except near sunrise and sunset or in thick fog) is appropriate for a wide variety of locations and conditions (29). Thus photolysis, Reaction (2-7), would be an important process for released I_2.

$$I_2 \xrightarrow{\text{sunlight}} 2\ I_{\text{atomic}} \qquad (2\text{-}7)$$

Three possible subsequent reactions were suggested (28) which could control the behavior of the atomic iodine produced by photodissociation:

$$I + O_3 \longrightarrow IO + O_2 \qquad (2\text{-}8)$$

$$IO + CO \longrightarrow CO_2 + I \qquad (2\text{-}9)$$

$$IO + N_2O \longrightarrow IO_2 + N_2 \qquad (2\text{-}10)$$

Thus I-131 released as I_2 may exist in the environment as radioactive I, IO or IO_2.

Alternative fates of I_2 during periods of darkness are adsorption, on particles and reactions with trace gases. Zafiriou (29) concludes:

> ...in clean marine air the diffusion time to particles is of the order of 1500's, and such reactions are unlikely to be extremely fast. On the other hand, in continental and especially in polluted air the concentration and size-distribution of the atmospheric aerosol both favor gas-particle interaction by large and highly variable factors. Unsaturated compounds, aromatics, amines, and sulfides are types of organic compounds that occur in air particulates and would be expected to react with iodine or its hydrolysis product, HOI, to give some organoiodine materials. Under laboratory conditions, iodine reacts with known or suspected trace-gas constituents of the atmosphere, including but not limited to: hydrogen peroxide, ozone, nitric acid, and NO_x (30). Iodine may be thermodynamically unstable in air with respect to oxides, especially I_2O_5. As mentioned previously, in the presence of near-neutral water, the thermodynamically stable form of the element I in the atmosphere is exclusively as iodates dissolved in aqueous solution (Wong, 1976, pers. comm.). It is reported that radiolysis of I_2+O_2 in an organic solvent (CCl_4) produces I_2O_5 (s) as the product (31). Similar reactions leading to other organic and inorganic forms might take place in the atmosphere.

Iodine adsorbed on particles would exhibit a deposition velocity about 1/ that attributed to elemental iodine. Iodine converted to organic forms would have an even lower V_d (see Table 1-1).

However, any organic iodine forms produced from I_2 dissociation would themselves be subject to photolysis and other chemical reactions which could take place in the atmosphere. Zafiriou has pursued the possible fates of methyl iodide and other organic iodine compounds in the atmosphere (29). His findings are summarized below. The several types of chemical, including photochemical, reactions identified by Zafiriou (29) are listed in Table 2-2. Reaction R3 has the fastest rate of those listed in Table 2-2 and was previously considered in detail by Zafiriou (28) who found a turnover time of 2.3 days for continuous exposure, which is in good agreement with the 50-hour estimate of Eggleton and Clough cited in Reference (16). Zafiriou has made a new calculation of the rate of CH_3I photolysis and found a maximum excitation rate for clear skies of about 0.12 per calendar day, corresponding to a minimum turnover time of 8 calendar days.

TABLE 2-2

MAJOR REACTION TYPES FOR METHYL IODIDE (29)

R1. S_N2 Displacement

$$X^- + CH_3I \longrightarrow CH_3X + I^-$$

R2. Dissociative Electron Capture

$$CH_3I + e^- \longrightarrow CH_3I^- \longrightarrow CH_3\cdot + I^{-\,(a)}$$

R3. Homogeneous C-I Bond Cleavage to Free Radicals

$$CH_3I \xrightarrow{\text{heat or light}} \cdot\ CH_3^{\overset{*}{+}} + I_{atomic}{}^{(b)}$$

R4. Formation of Organometallic Compounds

$$CH_3I + (Mg^{o} \text{ or } Li^{o}) \longrightarrow (CH_3Mg \text{ or } CH_3Li) + I^-$$

R5. Loss of H by Free Radical Abstraction

$$CH_3I + OH \longrightarrow .CH_2I + HOH$$

R6. Possible New Reaction Type$^{(c)}$

$$\underset{\text{(or other oxidants)}}{CH_3I + O_3} \longrightarrow \underset{\text{(or other products)}}{CH_310 + O_2}$$

R7. Possible Alternative Interaction With Very Strong Lewis Acids$^{(d)}$

$$CH_3\text{-}I + A \xrightarrow{?} CH_3\text{-}I\text{-}A \text{ or } {}^{+}CH_2I + HA^-$$

(a) The electron may also be loosely bound to a reductant.

(b) The * indicates that CH_3 formed in light reaction is highly energetic.

(c) Based on the report in Reference (32).

(d) Leads to identified products. It has no firm basis in literature for CH_3I, but may be significant for alkyl iodides in general.

The estimated maximum photolysis rate (sun directly overhead) is estimated to be about 0.4 per day. Table 2-3 summarizes estimates of probable reaction rates of CH_3I in the atmosphere. Possible rates for unsuspected reactions are included in addition to those given in Table 2-2. Ignoring the unsuspected reactions leads to estimates of turnover times between 2 hours and 200 hours. If all the maximum rates (including those for unsuspected reactions) prevail, then the minimum turnover time is ∿0.3 hour. However, the unsuspected rapid reactions with particles appear very unlikely in view of the small fraction of iodine (∿0.1) found in particulate form (Table 2-1), so a more realistic lower limit of the turnover time is ∿1 hour. Zafiriou (29) concludes that the photochemistry of alkyl iodides would be similar to that of CH_3I. Thus the limiting estimates on turnover time based on Table 2-3 would also apply to the organic iodides identified by Haller and Perkins (7) in the Hanford Purex Plant off-gas stream. Photolysis of aromatic iodine compounds is expected to be considerably faster than for CH_3I (29).

Reactions (2-8--(2-10), from Reference (28), are also important for the I atoms released by photolysis of methyl iodide. Thus I atoms and the IO and IO_2 molecules may be important airborne iodine forms subsequent to release of either I_2 or CH_3I. However, these forms may themselves react to form I_2 or HOI, HI or an organic form, and estimates of turnover times are very uncertain (28).

IMPLICATIONS FOR ENVIRONMENTAL SAMPLING AND DOSE ASSESSMENT

Photochemical considerations thus abandon us in an environment of largely unfamiliar iodine species whose stability and behavior are not understood. As noted above, both the 3-part stable iodine sampler and the 3- or 4-part radioiodine species sampler have been tested in the laboratory using iodine species generated for that purpose. Neither of the samplers used has been exposed to atomic iodine, IO, or IO_2 to determine their behavior in the sampling systems.

Zafiriou has speculated that I, IO, or reaction products from their interaction with organic material collected on the particulate filter, would be unlikely to penetrate the IPh bed. On the other hand, organics other than CH_3I could penetrate the cadmium iodide and iodophenol beds to reach the charcoal bed (29). It seems quite unlikely that any of the possible iodine compounds would penetrate the charcoal beds of the radioiodine species samplers. The intended separation of environmental radioiodine into particulate, inorganic gas, and organic gas components using the particulate filter, IPh bed, and charcoal bed may in fact be achieved. However, the inorganic fraction may be an assortment of reactive

TABLE 2-3

SUMMARY OF ESTIMATES OF PROBABLE METHYL IODIDE REACTION RATES IN THE TROPOSPHERE (29)

REACTION	ESTIMATED PROBABLE TURNOVER RATE (T, %/hr)	Remarks
R1	0	Reliable
R2	0	Insufficient information on $O_2(H_2O)_n^-$ reactivity
R3 - thermal	0	Reliable
R3 - Photochemical	$0.005 < T < 1.5$	Reliable
R4	0	Reliable in metal-free environments
R5	$0.05 < T << 50$	Insufficient information on radicals in air
R6	0?	Speculative, experimentally tractable
R7	0	See text, hypothetical reaction type
Unknown Reactions on Particles	0-200	Very variable, probably slow. Insufficient information.
Unknown Trace Gas Reactions	0-30	

species, with varying deposition velocities instead of the expected I_2/HOI mixture. The organic fraction will consist of the less reactive components including (but not necessarily limited to) the expected organic iodides. The interactions of I, IO, and IO_2 with TBAH-coated filters are not known either, so the validity of the inorganic/organic separation of the gaseous stable iodine is also somewhat uncertain. The sum of the concentrations determined using TBAH-impregnated filters and charcoal beds in series will, in any case, reflect the total gaseous iodine concentrations.

The projected atmospheric iodine forms and their behavior must be consistent with observations of the behavior of radioiodine, stable iodine, and specific chemical forms in the environment. These observations are rather limited and our understanding of the iodine cycle is correspondingly incomplete. It is known from measurements in the vicinity of nuclear facilities that some of the radioiodine released to the atmosphere deposits on vegetation and appears in cows' milk or animal thyroids (33), 34, 35, 36). It is also known from environmental radioiodine measurements that less reactive components exist in the environment. Comparisons of air and grass concentrations have indicated mean I-131 deposition velocities of 0.2--0.3 cm/sec. Most of the I-131 was in gaseous form; the observed deposition velocity is intermediate between those measured for I_2 and CH_3I (33). The ratio of the airborne I-129 concentration to its bomb fission yield has been found to be greater than the comparable ratio for Cs-137 (37). This implies that the I-129 has a longer atmospheric residence time than particulate Cs-137, which in turn indicates (a) that the I-129 is not in elemental form, (b) that the deposition velocity of the I-129 is less than ∿0.2 cm/sec, and (c) that the I-129 washout ratio is smaller than ∿300. Observed washout ratios for stable iodine were found to be only 10-50 (21), intermediate between those expected for I_2 and CH_3I and well below that measured for particulate fallout (38). Section 5 contains a more detailed discussion of estimated residence times for iodine in the atmosphere.

As was indicated in Section 1, the name of an airborne iodine species is less important for dose assessment than its deposition velocity. It appears that the reactive atmospheric forms thought to result from photolysis of I_2 and organic forms would be trapped on the IPh bed in the environmental radioiodine sampler and would be called inorganic. In the current absence of data on the deposition velocity of these species, treating them as I_2 is clearly a cautious approach. On the other hand, the unreactive products will have correspondingly lower deposition velocities which make them much less significant in terms of the milk food chain.

The data collected during the study, discussed in Sections 3-5, provide information which helps to clarify the problem of dose assessment for radioiodine and adds to our knowledge of the behavior of iodine in the atmosphere.

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Section 3

MEASUREMENTS OF I-131 AT OYSTER CREEK

The Oyster Creek Nuclear Generating Station is located in Ocean County in central New Jersey. The station is about 3 km west of Barnegat Bay, from which it obtains condenser cooling water, and 9 1/2 km from the Atlantic Ocean. The proximity of the plant to the coast results in a mixed climatic regime, with both marine and continental influences. The relatively flat coastal plain on which the plant is sited is typical of the New Jersey pine barrens. Only a small amount of land in this area is devoted to agriculture. The boiling water reactor has a thermal power rating of 1930 MW and the plant's rated electrical output is 640 MW. Prior to the present study the principal source of I-131 discharged to the atmosphere had been found to be the steam jet air ejector (SJAE) delay line and the I-131 in that effluent stream was mostly in organic form (1). A new facility to provide additional treatment for the SJAE exhaust gas was under construction during the study.

Subsections which follow describe the measurements of I-131 in gaseous effluents, the predicted transport of the I-131 in the plant's environment, measured concentrations of I-131 in environmental media, and comparisons of the predicted and measured concentrations. The behavior of airborne I-131 from a second source, fallout from a nuclear device exploded in the atmosphere, was also observed in the Oyster Creek environs during the study. The results of the measurements of the much higher concentrations of fallout I-131 are discussed in a separate section.

I-131 IN GASEOUS EFFLUENTS

Sampling Locations and Discharge Flow Rates

An important part of the study was to measure the I-131 species and release rates in all gaseous effluents discharged from the plant. The principal I-131 discharge point was the 112-meter stack which receives the gases from the steam jet air ejector delay line and the gland seal and mechanical vacuum pump delay line as well as the ventilation air from the reactor, turbine, and radwaste buildings.

Because it was not possible to locate the radioiodine species sampler in the stack sampling line, each of the five sources was monitored separately. Two additional discharge points were monitored; these were the feedwater and condensate pump room exhaust and the exhaust from the reheater protection system and lube oil storage area. Both of these discharges enter the atmosphere about 10 meters above the ground.

Table 3-1 contains the discharge flow rates for each of the effluent streams monitored during the study. Except for the gland seal and mechanical vacuum pump exhaust line, for which the design flow rate is given, the tabled flow rates are based on pitot tube or helium dilution measurements made by AEC contractors (1), Nuclear Environmental Services personnel (2), or Oyster Creek plant personnel. Previously established and validated sampling points (1,2) were used for the first six locations shown in the table. A new sampling point was established in the duct exhausting the reheater protection system and lube oil storage area. Measurements showed the exhaust flow of this short duct to be quite uniform with an average exhaust velocity of 20 m/s.

Table 3-1

OYSTER CREEK GASEOUS EFFLUENT FLOW RATES

Effluent Stream	Flow Rate ft^3/min	 cm^3/sec
Reactor Building Ventilation Air	70,500	$3.33x10^{7}$
Radwaste Building Ventilation Air	22,475	$1.06x10^{7}$
Turbine Building Ventilation Air	72,500	$3.42x10^{7}$
Steam Jet Air Ejector Delay Line	115	$5.41x10^{4}$
Gland Seal and Mechanical Vacuum Pump Exhaust	632	$2.98x10^{5}$
Feedwater and Condensate Pump Room Exhaust	40,500	$1.91x10^{7}$
Reheater Protection System and Lube Oil Storage Area Exhaust	14,500	$6.84x10^{6}$

Sampling and Analytical Methods

The chemical forms and release rates of I-131 discharged to the atmosphere were measured continuously throughout the study. Normally, five of the samples were changed weekly and two, the feedwater and condensate pump room and the reheater protection system and lube oil storage area, were changed every other week. The sampled air was drawn through a standard NES radioiodine species sampling assembly. It consists of (1) a high efficiency particulate filter, (2) a bed of cadmium iodide (CdI_2), with excess I, adsorbed on Chromosorb-P, (3) a bed of 4-iodophenol (IPh) adsorbed on alumina, and (4) two beds of TEDA-impregnated activated charcoal or silver zeolite (AgX). The sampling system is designed to separate the I-131 in the air stream into four components: iodine attached to particulate material, elemental iodine (I_2), hypoiodous acid (HOI), and organic iodides (CH_3I and others). The development and initial testing of the sampling have been described by Keller et al. (3). Emel et al. have recently reevaluated the device and found collection efficiencies in close agreement with those reported previously (4). The sample loading, sample exchange, and gamma spectrometric analysis procedures used for the effluent monitoring portion of the study are described in detail in Appendix A. Also in Appendix A are the species sampler collection efficiencies and the procedure used to determine the species distribution from the distribution of I-131 activities on the sampling media.

Plant Performance Data

Table 3-2 gives the average reactor power level (MW_e) for the Oyster Creek station for each of the 23 measurement periods. These averages were compiled from a daily plant log sheet which contains the power level at the time (usually about 8 a.m.) when the daily reactor water sample was taken by plant personnel. As can be seen from the table, the average power varied significantly in only three sampling periods. The most important of these was Period 7 during which the reactor was shut down for approximately five days. Also shown in Table 3-2 are the average concentrations of I-131 in reactor water for each of the sampling periods. The average concentrations, computed from the daily measurements made by plant personnel, were also relatively constant except during Period 7 when the I-131 concentration peaked at 273 nCi/g. The reactor water cleanup system operated at an average flow rate of 2.34×10^4 g/sec (371 gpm) during power operation. This system operated at reduced flow for three days and was turned off for two days during the outage in Period 7.

TABLE 3-2

AVERAGE REACTOR POWER LEVELS AND CONCENTRATION OF I-131 IN REACTOR WATER DURING MONITORING PERIODS AT OYSTER CREEK

PERIOD	DATES	AVERAGE REACTOR POWER LEVEL (MW_e)	AVERAGE CONCENTRATION OF I-131 IN REACTOR WATER (nCi/g)
1	6-16/6-23	578±15(1)	1.43±.27(1)
2	6-23/6-30	584±3	1.47±.27
3	6-30/7-7	575±20	1.29±.17
4	7-7 /7-13	590±4	1.36±.18
5	7-13/7-20	585±21	1.49±.19
6	7-20/7-27	516±208(2)	1.52±.61
7	7-27/8-3	211±292(2)	45±94
8	8-3 /8-10	588±23	1.53±.39
9	8-10/8-17	614±10	1.71±.32
10	8-17/8-24	616±16	2.17±.52
11	8-24/8-30	619±6	1.47±.34
12	8-30/9-7	613±18	1.35±.46
13	9-7 /9-16	624±9	1.61±.45
14	9-16/9-23	534±113	1.14±.24
15	9-23/9-30	621±11	1.19±.34
16	9-30/10-7	597±33	1.93±.47
17	10-7/10-14	617±7	1.81±.48
18	10-14/10-21	604±31	2.18±.32
19	10-21/10-28	636±7	2.05±.31
20	10-28/11-4	619±31	3.1 ±1.9
21	11-4 /11-11	639±7	2.36±.48
22	11-11/11-18	624±41	2.36±.25
23	11-18/11-30	612±43	2.05±.23

---

(1) Sample standard deviation is given.

(2) Plant shut down for five days between 26 July and 1 August.

Chemical Forms of I-131

The average I-131 concentrations in the seven effluent streams were measured during each sampling period. The concentrations are tabulated in Appendix B (Tables B-1 through B-7). Two measurements yielded unusually low I-131 concentrations: the reactor building ventilation air during Period 15 and the gland seal exhaust during Period 9. The sampling and analytical data for these measurements were reviewed and could not be faulted. Similarly, the data for the radwaste building exhaust measurements for Periods 14-16, periods of high concentrations, were reviewed and found to be valid.

The fractions of each of the chemical forms of I-131 in these effluent streams were derived from the air sampling data using the technique specified at the end of Appendix A. The resulting distributions are shown by period for each effluent stream in Appendix B (Tables numbered B-8 through B-14). The average species fractionations for the seven sources are summarized in Table 3-3. The average values shown include data from Period 7, when the outage occurred. Elimination of the data from that sampling period would not substantially alter the means.

Table 3-3

SUMMARY OF OYSTER CREEK RADIOIODINE SPECIES MEASUREMENTS

	Average Percent Of Discharged I-131 In Each Form(1)			
Source	Particulate	I_2	HOI	Organic
Reactor Building Ventilation Air	9±1	28±1	33±2	29±3
Radwaste Building Ventilation Air	<0.5	16±2	22±2	62±4
Turbine Building Ventilation Air	17±1	43±2	32±2	8±1
SJAE Delay Line	<0.1	<1	12±2	87±2
Gland Seal and Mechanical Vacuum Pump Exhaust	<1	8±1	38±3	54±3
Feedwater and Condensate Pump Room Exhaust	8±2	57±7	28±8	6±3
Reheater Protection System and Lube Oil Storage Area Exhaust	5±1	15±3	14±2	66±5

---

(1) One standard deviation of the mean is given with the mean. The total for a source may differ from 100 percent due to rounding of individual entries.

The only previous BWR radioiodine species data for a period of comparable duration were obtained at the Quad Cities station during 1974 (5). Ventilation air streams from the Quad Cities reactor buildings were sampled individually, but the other sources were combined upstream of the third sampling point at the Quad Cities plant. During a four-month period, mostly power operation, the average species distributions were 10±2% on particulate, 22±2% I_2, 19±1% HOI, and 50±4% organic iodides in the Unit 1 reactor building ventilation air and 10±1% on particulates, 31±3% I_2, 23±1% HOI, and 36±4% organic iodides in the reactor building ventilation air from Unit 2. As in Table 3-3 the standard deviation of the mean is given with the means of twelve measurements for Unit 1 and thirteen measurements for Unit 2. The major difference between the Quad Cities and Oyster Creek reactor building data is in the distribution of I-131 between the two less reactive forms, HOI and the organic iodides. The amounts of I-131 attached to particulates and in elemental form in the exhausted air are quite similar. Since conversion of elemental iodine to the less reactive forms in buildings appears to take place in the various available surfaces (2), differences in the HOI and organic iodide fractions may well be related to different surface coatings and the residence time of radioiodine deposited on those surfaces.

Previous short-term radioiodine species sampling at Oyster Creek yielded results which may be compared with those obtained during this study. Radioiodine species at the end of the SJAE delay line were determined in single brief samples obtained in 1971 (1) and again in 1973 (6). In 1971 the I-131 in this effluent was found to be 0.3% I_2, 10.7% HOI, and 89% organic iodides. In 1973, the distribution was found to be 10.5% I_2, 22.9% HOI, and 66.4% organic iodides. No more than 0.2% of the I-131 was attached to particulate material in either sample. The organic iodide fraction found in 1973 is unusually low; however, Table 0-11 shows that a similar organic fraction was observed once (Period 13) during the 1976 sequence of measurements. The elemental iodine fraction measured in 1973 is seen, in view of that sequence of data, to be very unusual.

The chemical forms of radioiodine in ventilation air were also determined at Oyster Creek as part of a detailed study of radioiodine sources in boiling water reactors (2). Two to four I-131 species measurements (depending on the source) were made during a two and one half-month period of power operation late in 1975. The average particulate, elemental, HOI, and organic fractions observed were, respectively: 18, 45, 11, and 27 percent for the turbine building ventilation exhaust air; 21, 26, 22, and 32 percent for the reactor building ventilation exhaust air; and <1, 14, 24, and 63 percent for the radwaste building exhaust air. These results are in substantial agreement with the body of data on I-131 species in ventilation air summarized in Table 3-3.

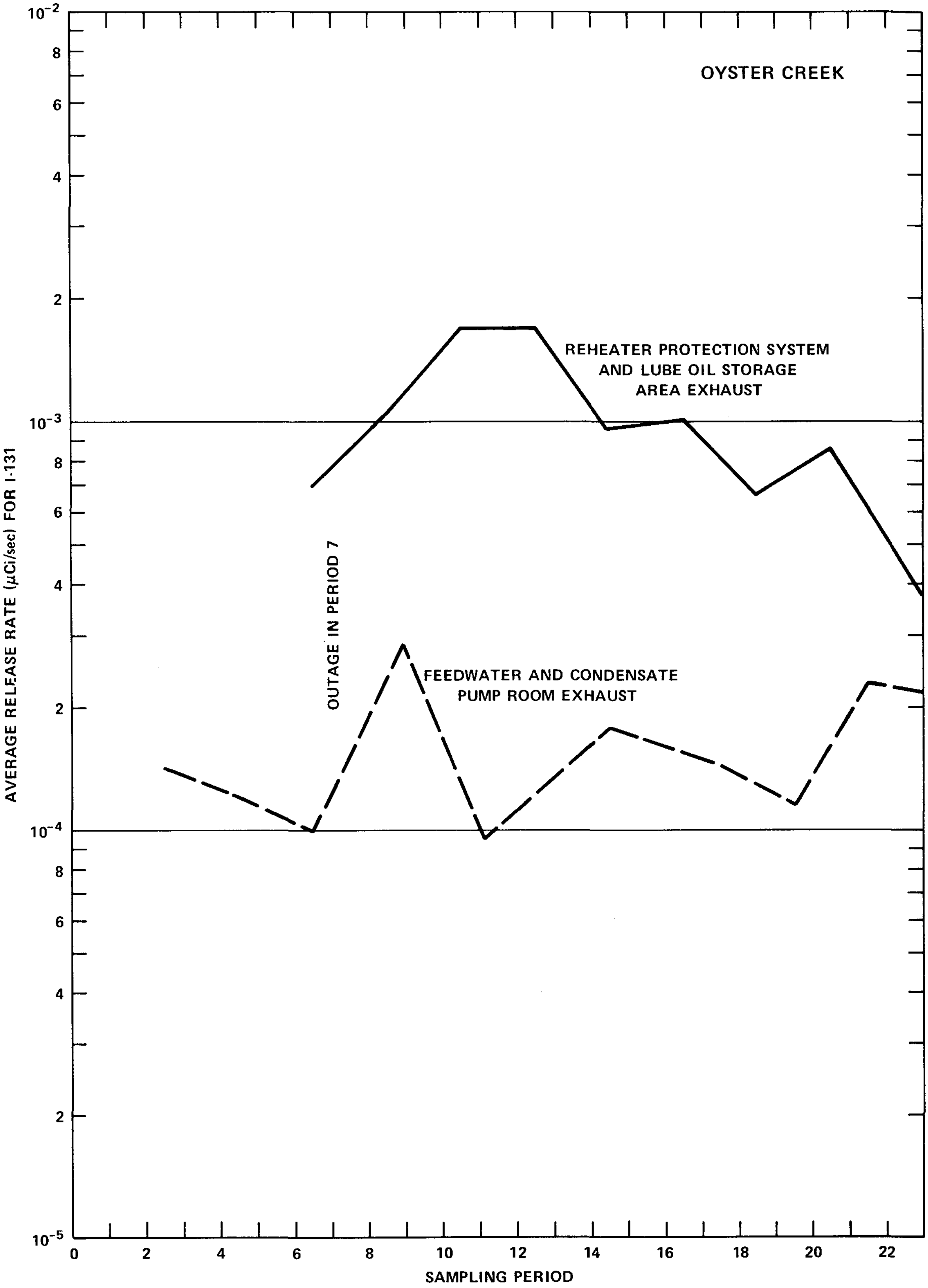
I-131 Release Rates

The average I-131 release rate for a particular effluent stream during a given monitoring period was obtained by multiplying the average I-131 concentration measured in that stream by the appropriate flow rate from Table 3-1. Figure 3-1 shows the variations with time of the average release rates for the three principal turbine side discharges. (Although the individual values in this and other figures are plotted as points, it should be remembered that they are averages of (1-2)-week periods.) The large increase in release rate for the gland seal and mechanical vacuum pump exhaust line during Period 7 is believed to be principally the result of operation of the mechanical vacuum pump during the outage, although part of the increase may have resulted from carryover of higher I-131 concentrations in the gland seal steam.

The average release rates for the reactor and radwaste building ventilation air are shown in Figure 3-2. Both sources exhibit peaks during the period which includes the outage and other variations not readily assigned to operational occurrences. The two components of the radioiodine effluent not discharged via the tall stack are shown in Figure 3-3. As indicated previously, these average release rates are for 2-week (and occasionally longer) periods.

The normalized release rate for I-131 was obtained by dividing the average release rate (Q, μCi/sec) for each period by the average concentration (Cw, μCi/g) of I-131 in reactor water for that period. The quotient (Q/Cw), is plotted for the several I-131 sources in Figures 3-4 through 3-6. Figure 3-4 shows the normalized release rates for three discharge pathways involving reactor steam. Qualitatively, the three normalized discharge rates are similar and it is clear that there are time variations in the release rate which are not eliminated by normalization. The similarity in time-dependent behavior of the normalized release rates for steam discharges suggests that the partition coefficient may have been changing during the course of the study. Figures 3-5 and 3-6 show the time dependence of the normalized release rates for the other four sources.

The normalized release rates can be compared with some previous measurements made at Oyster Creek, principally those made during the EPRI study of specific radioiodine sources at BWRs (2). Average normalized release rates for power operation


OYSTER CREEK
REHEATER PROTECTION SYSTEM AND LUBE OIL STORAGE AREA EXHAUST
FEEDWATER AND CONDENSATE PUMP ROOM EXHAUST
OUTAGE IN PERIOD 7
AVERAGE RELEASE RATE (μCi/sec) FOR I-131
SAMPLING PERIOD
10^-2
8
6
4
2
10^-3
8
6
4
2
10^-4
8
6
4
2
10^-5
0
2
4
6
8
10
12
14
16
18
20
22

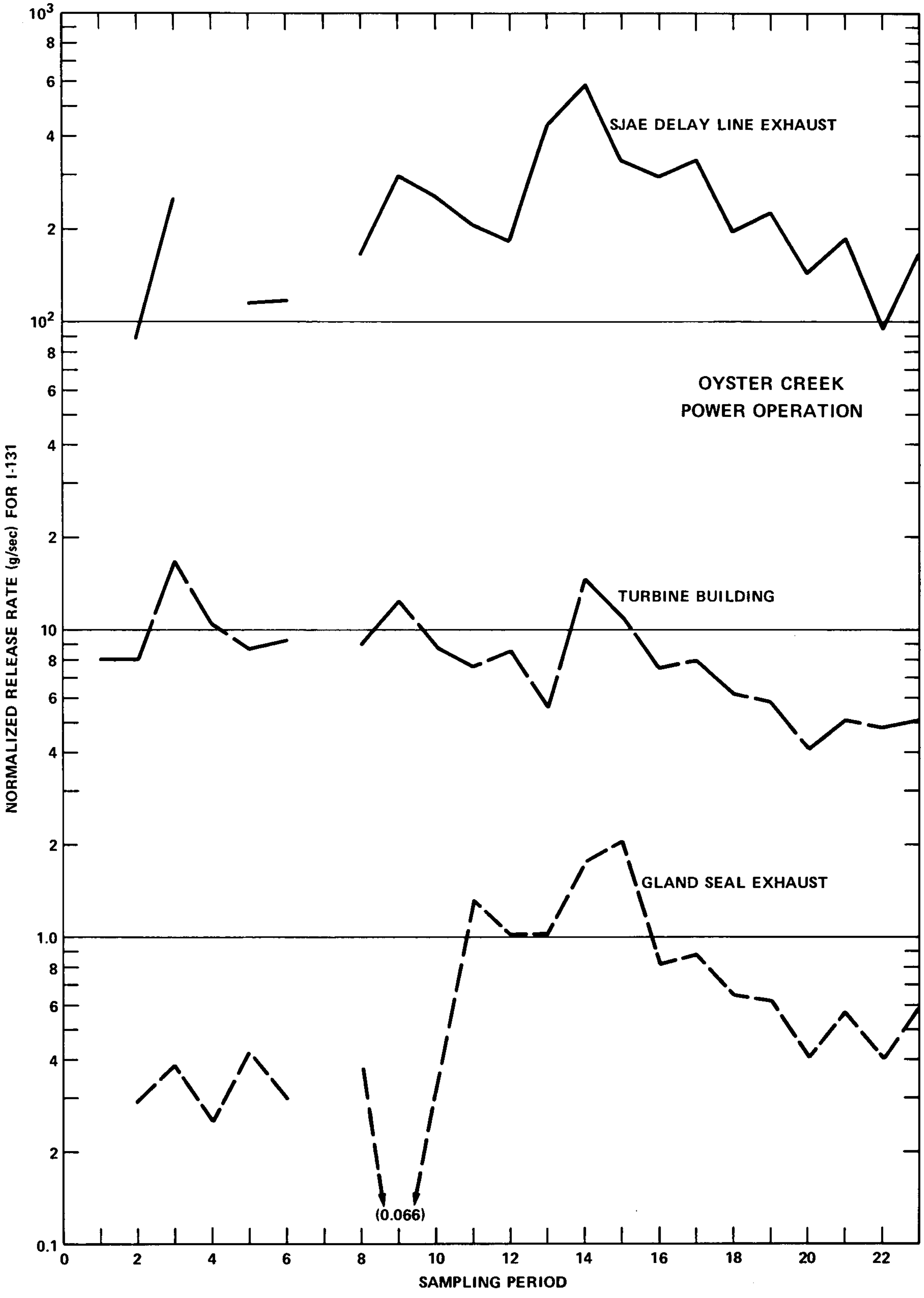
were computed from the individual values plotted in Figures 3-4 through 3-6. The cumulative distribution of normalized release rates for each source were plotted on log probability paper. The median and geometric standard deviation were obtained for each distribution of normalized release rate from the plots. The mean normalized release rate with the sample standard deviation and the median normalized release rate with the associated geometric standard deviation are shown for all sources in Table 3-4. Also shown in the table are the normalized release rates for power operation from earlier (1975-76) measurements reported by Pelletier *et al*. (2) and some rather uncertain estimates derived from data obtained in 1971-72 (1,7). The uncertainty in the latter estimates arises from the fact that reactor water I-131 concentrations were reported for only a limited number of dates during the measurement period.

There are a number of possible reasons for the differences in normalized release rates measured at different times. In the turbine building increases could be associated with increased steam leakage, an increase in the partition coefficient, an increase in the amount of leakage of condensate enriched in I-131, or a combination of these factors. As noted above, there is at least a qualitative indication that the partition coefficient may have changed during the measurement period, but other factors may be of equal or greater importance.

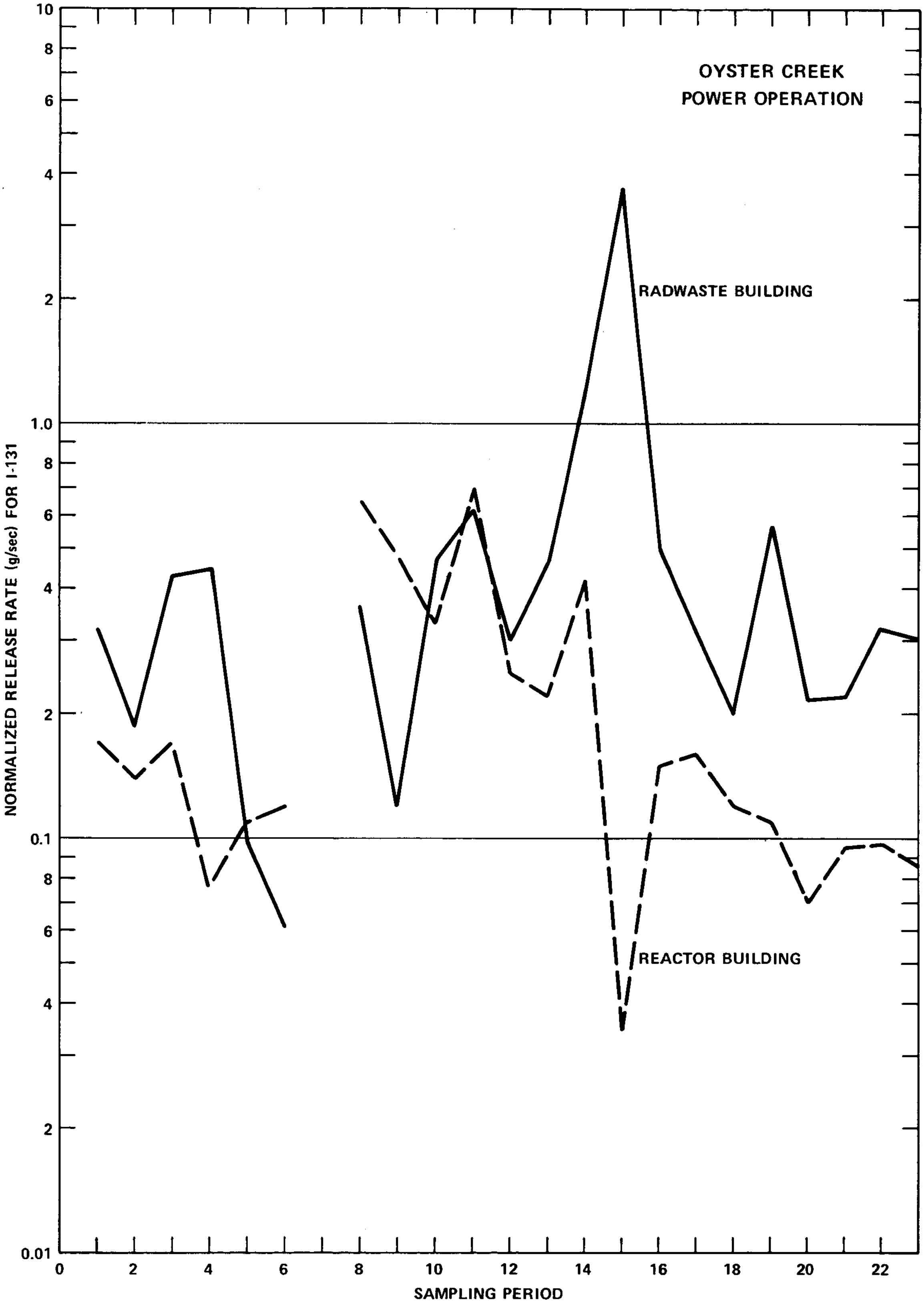
PREDICTED ATMOSPHERIC TRANSPORT AND DEPOSITION OF I-131

The methodology outlined by the Nuclear Regulatory Commission (8) for estimating atmospheric transport and dispersion of I-131 released during normal operation of light water reactors was used to predict concentrations of radioiodine in the air and on vegetation in the Oyster Creek environs. The revised curves describing plume depletion and deposition were used in the computations. The straight-line airflow model (9) was employed with no corrections for terrain effects or recirculation of the plume. Predictions were made for an elevated release, the 112-m stack, and for a ground level release, since the vent releases were below the top of the building complex.

Meteorological data routinely collected for the Oyster Creek/Forked River site by Pickard, Lowe, and Garrick were used to calculate (a) average dispersion factors for non-depositing effluents, (X/Q) (sec/m^3), (b) average dispersion factors for depositing effluents, (X/Q)^d (sec/m^3), and (c) relative deposition factors for depositing effluents, δ (m^{-2}). These parameters were computed for both release points for each monitoring period during the study. Computations were made for the



OYSTER CREEK
POWER OPERATION
SJAE DELAY LINE EXHAUST
TURBINE BUILDING
GLAND SEAL EXHAUST
(0.066)
NORMALIZED RELEASE RATE (g/sec) FOR I-131
10^3
8
6
4
2
10^2
8
6
4
2
10
8
6
4
2
1.0
8
6
4
2
0.1
0
2
4
6
8
10
12
14
16
18
20
22
SAMPLING PERIOD


OYSTER CREEK
POWER OPERATION
RADWASTE BUILDING
REACTOR BUILDING
NORMALIZED RELEASE RATE (g/sec) FOR I-131
SAMPLING PERIOD
10
8
6
4
2
1.0
8
6
4
2
0.1
8
6
4
2
0.01
0
2
4
6
8
10
12
14
16
18
20
22

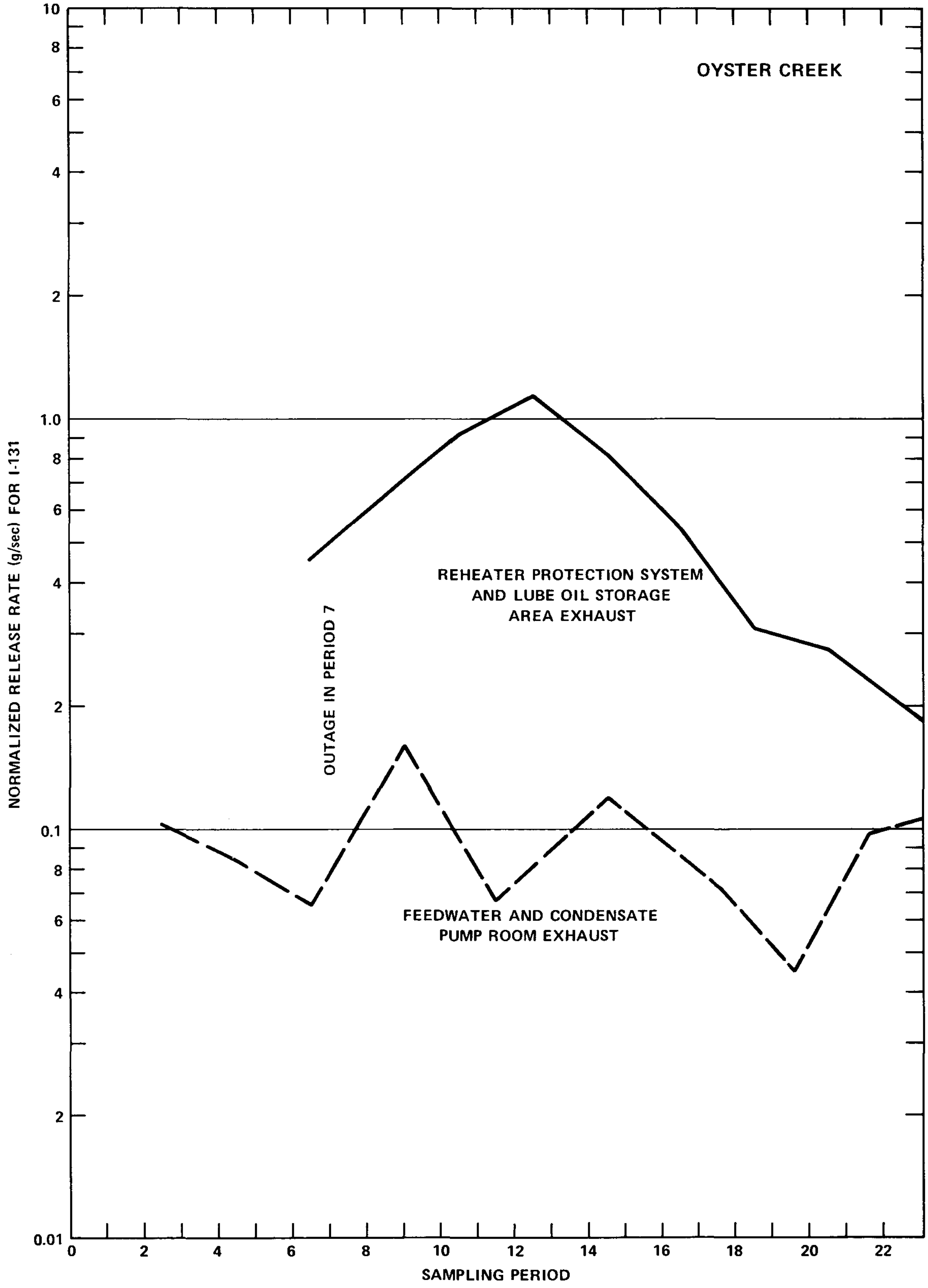

OYSTER CREEK
NORMALIZED RELEASE RATE (g/sec) FOR I-131
10
8
6
4
2
1.0
8
6
4
2
0.1
8
6
4
2
0.01
REHEATER PROTECTION SYSTEM
AND LUBE OIL STORAGE
AREA EXHAUST
OUTAGE IN PERIOD 7
FEEDWATER AND CONDENSATE
PUMP ROOM EXHAUST
0
2
4
6
8
10
12
14
16
18
20
22
SAMPLING PERIOD

TABLE 3-4

MEAN AND MEDIAN NORMALIZED RELEASE RATES FOR I-131 OYSTER CREEK GASEOUS EFFLUENTS, POWER OPERATION

	Normalized Release Rate(Q/Cw)[(µCi/sec)/(µCi/g)]for I-131				
Source	Arithmetic Mean	Sample Standard Deviation	Median	Geometric Standard Deviation	Previous Measurements
Reactor Building (22)(1) Ventilation Air	0.22	0.19	0.16	2.3	(0.38)(2), 0.22(3)
Radwaste Building (22 Ventilation Air	0.52	0.75	0.32	1.9	(0.11)(2), 0.079(3)
Turbine Building (22) Ventilation Air	8.4	3.2	8.0	1.4	(3.2)(2), 3.8(3)
Steam Jet Air Ejector (20) Delay Line	233	123	205	1.6	(60)(2)
Gland Seal and Mechanical (21) Vacuum Pump Exhaust	0.69	0.51	0.53	2.0	0.81(3)
Feedwater and Condensate (10) Pump Room Exhaust	0.092	0.033	0.089	1.4	0.058(3)
Reheater Protection System (9) and Lube Oil Storage Area Exhaust	0.59	0.32	0.53	2.0	-

---

(1) Number of measurements for which mean and median are derived is shown in parentheses.

(2) Estimated using data from References (1) and (7); only a few measurements of reactor water concentrations were reported.

(3) Measurements reported by Pelletier et al. (2).

environmental monitoring locations established for the program; the coordinates for each site are given in Table 3-5. Appendix C contains the calculated dispersion and deposition parameters for each monitoring period for sites 1, 2, 3, 4, and 6, the principal environmental monitoring locations.

Table 3-5

ENVIRONMENTAL SAMPLING LOCATIONS, OYSTER CREEK

Site	Name	Location r(m)	Location θ[(a)]
1	Forked River Marina	2440	24°
2	State Game Farm	3880	43°
3	Pinewalk Substation	8320	24°
4	Garden State Parkway	6490	352°
5	Sands Point	2870	109°
6	Island Beach State Park	9480	98°

(a) Direction from the plant; a location at 0° or 360° is due north of the plant, one at 90° is due east, and so on.

The dispersion and deposition parameters were used with the measured discharge rates for the various chemical forms of I-131 to predict environmental air concentrations of the various forms of I-131 and to predict the amounts of I-131 deposited on vegetation. The predicted concentrations of radioiodine in particulate form (X_1), radioiodine as I_2 or HOI (X_{23}), and radioiodine in organic form (X_4) were obtained by adding the contributions from the vent and stack releases. At a given location:

$$X_1 = Q_{1s}(X/Q)_s^d + Q_{1v}(X/Q)_v^d \tag{3-1}$$

$$X_{23} = Q_{23s}(X/Q)_s^d + Q_{23v}(X/Q)_v^d \tag{3-2}$$

$$X_4 = Q_{4s}(X/Q)_s + Q_{4v}(X/Q)_s \tag{3-3}$$

in which Q_1 is the release rate (Ci/sec) of I-131 attached to particulates, Q_{23} is the sum of the I_2 and HOI release rates (Ci/sec), and Q_4 is the release rate (Ci/sec) of organic iodides. The subscripts s and v denote release rates or dispersion parameters for the stack and vent respectively. Calculations of X_1, X_{23}, and X_4 at each location were made for each period using the I-131 concentration and chemical form data and the appropriate dispersion factors, tabulated in Appendices B and C. The total predicted concentration, X_T, the sum of X_1, X_{23}, and X_4. Predictions of the average deposition rate, D Ci/(m^2-sec), at a monitoring location were made using

$$D = (Q_{1s} + Q_{23s})\delta_s + (Q_{1v} + Q_{23v})\delta_v \qquad (3-4)$$

with the source terms and deposition parameters for each period. The expected concentration of radioiodine on vegetation, C (Ci/m^2), at the end of a period of duration T (sec) was computed using

$$C = C_o e^{-\lambda_e T} + \frac{D}{\lambda_e}(1 - e^{-\lambda_e T}) \qquad (3-5)$$

where C_o is the concentration (Ci/m^2) of I-131 on the vegetation at the start of the period and λ_e (sec^{-1}) is the effective rate constant for removal of I-131 from the vegetation. The effective half-life for I-131 on vegetation was assumed to be five days, a frequently observed value, which corresponds to a λ_e of 1.60×10^{-6} sec^{-1}.

The predicted environmental concentrations of I-131 attached to particulates, as I_2 or HOI, and as organic iodides are given for each principal monitoring location in Tables 3-6 through 3-10. Predicted deposition rates are given for the four locations where grass samples were exposed to the ambient air for various lengths of time during the study.

I-131 CONCENTRATIONS IN THE ATMOSPHERE

The environmental sampling program was centered on measurements of the chemical forms of I-131 in the atmosphere around the station to obtain data which could be related to the iodine species in plant effluents. Toward the end of the planned study period, I-131 concentrations in the Oyster Creek environment were increased (to 10-100 times those measured previously) by the arrival of radioactive fallout resulting from the detonation of a nuclear device in the atmosphere. Monitoring was continued during the period of radioactive fallout to observe the behavior

TABLE 3-6

PREDICTED AIR CONCENTRATIONS AND DEPOSITION RATES FOR SITE #1, FORKED RIVER MARINA

Period	Predicted Air Concentrations (fCi/m^3) Of I-131 In Various Forms		
	Particulate	I_2 + HOI	Organic Iodides
1	-	-	-
2(1)	0.37	4.7	15
3(1)	0.086	2.6	8.6
4(1)	0.087	1.8	7.1
5(1)	0.029	0.37	1.3
6	0.12	1.5	4.7
7	0.052	0.87	4.3
8	0.079	1.4	3.8
9	0.26	4.9	17
10	0.029	0.31	2.1
11	0.11	6.1	21
12	0.12	2.8	7.2
13	0.20	1.4	10
14	0.045	1.1	6.6
15	0.060	1.6	6.7
16	0.054	0.68	6.0
17	0.12	0.60	17
18	0.081	0.64	4.2
19	0.095	0.54	2.1
20	0.094	0.44	4.7
21	0.045	0.33	3.5
22	0.041	0.22	1.4
23	0.043	0.47	7.2

---

(1) Predicted values for this period do not include the contribution from vent releases.

TABLE 3-7

PREDICTED AIR CONCENTRATIONS AND DEPOSITION RATES FOR SITE #2, STATE GAME FARM

	Predicted Air Concentrations (fCi/m^3) Of I-131 In Various Forms			Predicted Deposition
Period	Particulate	I_2 + HOI	Organic Iodides	Rate ($aCi/m^2/sec$)
1	-	-	-	-
2⁽¹⁾	0.11	1.4	4.4	13
3⁽¹⁾	0.024	0.73	2.4	20
4⁽¹⁾	0.050	1.0	4.2	20
5⁽¹⁾	0.0070	0.090	0.33	5.4
6	0.094	1.3	4.1	26
7	0.023	0.36	1.7	3.9
8	0.033	0.55	1.5	0.62
9	0.22	4.5	16	37
10	$4.5x10^{-5}$	0.0012	0.0048	0.023
11	0.061	3.7	13	68
12	0.12	0.91	2.4	9.5
13	0.062	0.38	2.2	1.9
14	0.073	1.8	11	50
15	0.053	1.4	6.7	18
16	0.040	0.41	3.0	2.6
17	0.045	0.24	3.4	4.3
18	0.035	0.32	2.5	8.5
19	0.094	0.80	8.2	13
20	0.10	0.48	2.2	0.82
21	0.044	0.39	5.0	2.2
22	0.038	0.22	2.1	1.5
23	0.043	0.48	7.7	3.4

---

(1) Predicted values for this period do not include the contribution from vent releases.

TABLE 3-8

PREDICTED AIR CONCENTRATIONS AND DEPOSITION RATES FOR SITE #3, PINEWALD SUBSTATION

	Predicted Air Concentrations (fCi/m^3) Of I-131 In Various Forms			Predicted Deposition
Period	Particulate	I_2 + HOI	Organic Iodides	Rate ($aCi/m^2/sec$)
1	-	-	-	-
2(1)	0.17	2.2	7.5	16
3(1)	0.051	1.6	5.2	23
4(1)	0.066	1.3	5.5	15
5(1)	0.017	0.22	0.80	2.1
6	0.056	0.79	2.7	11
7	0.036	0.73	4.3	3.9
8	0.081	1.7	4.6	6.8
9	0.22	4.6	17	22
10	0.0058	0.085	0.51	0.054
11	0.048	3.1	11	46
12	0.041	1.7	4.6	17
13	0.032	0.28	2.8	5.6
14	0.035	0.77	5.0	8.4
15	0.037	0.94	5.0	10
16	0.013	0.23	2.4	1.9
17	0.045	0.23	9.5	3.5
18	0.021	0.25	2.4	4.1
19	0.016	0.13	1.2	0.58
20	0.048	0.24	6.3	1.4
21	0.0085	0.16	3.1	0.75
22	0.010	0.058	0.73	0.47
23	0.021	0.36	9.0	1.4

---

(1) Predicted values for this period do not include the contribution from vent releases.

TABLE 3-9

PREDICTED AIR CONCENTRATIONS AND DEPOSITION RATES FOR SITE #4, GARDEN STATE PARKWAY

Period	Predicted Air Concentrations (fCi/m^3) Of I-131 In Various Forms: Particulate	I_2 + HOI	Organic Iodides	Predicted Deposition Rate ($aCi/m^2/sec$)
1	-	-	-	-
2(1)	0.060	0.77	2.6	17
3(1)	0.053	1.6	5.3	23
4(1)	0.018	0.37	1.5	3.4
5(1)	0.071	0.92	3.4	16
6	0.033	0.47	1.6	7.1
7	0.045	0.90	5.3	7.1
8	0.21	4.5	12	42
9	0.15	3.0	11	28
10	0.022	0.57	2.3	0.014
11	0.027	1.8	6.1	24
12	0.030	0.98	2.6	26
13	0.018	0.25	3.2	3.4
14	0.073	1.8	12	46
15	0.038	0.98	5.4	21
16	0.0	0.0	0.0	0.0
17	0.082	0.42	16	4.9
18	6.1×10^{-6}	9.7×10^{-5}	0.0011	0.0010
19	0.035	0.48	7.6	3.7
20	0.073	0.36	9.9	2.8
21	0.010	0.097	1.4	0.94
22	6.3×10^{-4}	0.0030	0.0029	0.033
23	0.0075	0.077	1.1	0.10

---

(1) Predicted values for this period do not include the contribution from vent releases.

TABLE 3-10

PREDICTED AIR CONCENTRATIONS AND DEPOSITION RATES FOR SITE #6, ISLAND BEACH STATE PARK

Period	Predicted Air Concentrations (fCi/m^3) Of I-131 In Various Forms			Predicted Deposition
	Particulate	I_2 + HOI	Organic Iodides	Rate (aCi/m^2/sec)
1	-	-	-	-
2(1)	0.014	0.18	0.63	4.5
3(1)	0.031	0.96	3.3	15
4(1)	0.025	0.50	2.1	6.4
5(1)	0.035	0.45	1.7	5.4
6	0.017	0.24	0.85	5.0
7	0.029	0.57	3.4	2.5
8	0.025	0.48	1.3	1.3
9	0.040	0.85	3.1	6.1
10	0.020	0.53	2.3	12
11	0.012	0.66	2.4	8.0
12	0.044	0.94	2.5	8.3
13	0.042	0.41	4.4	3.0
14	0.072	1.8	11	5.7
15	0.018	0.49	1.5	0.90
16	0.0	0.0	0.0	0.0
17	0.025	0.22	6.0	1.6
18	0.010	0.12	1.2	2.6
19	0.032	0.52	8.6	2.5
20	0.050	0.24	5.8	4.4
21	0.018	0.45	9.7	4.6
22	0.026	0.16	1.9	1.4
23	0.024	0.41	10	2.9

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(1)Predicted values for this period do not include the contribution from vent releases.

of the chemical forms of radioiodine with time after detonation. The methods of air sampling and analysis and the measured I-131 concentrations are discussed below.

Sampling Methods and Locations

Air samples were obtained using a constant flow rate high volume sampler fitted with a special filter head. Detailed descriptions of the sampling system, its operation, and the sample changing technique are given in Appendix D. The filter head holds a particulate filter and two beds of gaseous iodine adsorbers, normally a bed of 4-iodophenol (IPh) adsorbed on alumina followed by a bed of activated charcoal impregnated with triethylenediamine (TEDA). This sequence of sampling media was designed to collect (1) iodine associated with particulate material, (2) the gaseous inorganic iodine species I_2 and HOI, and (3) organic forms of iodine. For a few samples, collected during the period of radioactive fallout, a bed of cadmium iodide (CdI_2), with excess I, adsorbed on Chromosorb P, was used in place of the IPh bed. This filtration sequence is designed to collect (1) iodine associated with particulates, (2) elemental iodine gas, I_2, and (3) HOI and organic iodine species. A third sampling arrangement was employed during some monitoring periods. Two samplers were operated at the same location: one as described above with a CdI_2 or IPh bed ahead of the charcoal bed and the second with only a charcoal bed behind the particulate filter. The second sampler was used to measure the total gaseous iodine concentration and the concentration of I_2 (or I_2 plus HOI) was determined by subtraction. Concentrations determined using either of the two latter methods are noted in the appropriate tables in the next section.

The particulate filters and beds of CdI_2, IPh, and charcoal were returned to the laboratory for radiochemical analysis. Particulate filters were counted using a shielded Ge(Li) detector and gamma spectrometric analysis system calibrated for the counting geometry employed. The CdI_2 beds were counted in Marinelli-type containers which fit within the shielding for the calibrated Ge(Li) spectrometer. Some IPh and charcoal beds were also counted in Marinelli-type containers. Most of the IPh and charcoal beds were leached to remove the I-131, which was subsequently extracted, precipitated, and counted. Details of the leaching procedures are given in Appendices E and F. The procedures for sample counting and analysis with the Ge(Li) spectrometer used for the environmental samples are given in Appendix G.

The air sampling sites were selected from available locations in desirable sampling areas which were determined by reviewing historic seasonal dispersion parameters for the Oyster Creek environs. The coordinates of the sampling locations were given above in Table 3-5 and are shown in Figure 3-7. The samplers at sites 2, 3, and 4 were operated continually from Period 2 through Period 23. Three samples at site 1 were lost due to pump and power failures. The sampler at site 6 operated continuously beginning with Period 4. Monitoring at site 5 was delayed until electrical power was available at that location. It was terminated shortly thereafter owing to theft of the sampling system.

Measured Concentrations of I-131 in Air

The measured concentrations of I-131 in various forms at monitoring sites 1, 2, 3, 4 and 6 are presented in Tables 3-11 through 3-15. As noted above the normal sampling configuration was designed to separate the I-131 activity into three components: particulate, inorganic gases I_2 and HOI, and organic gases. The symbol ϕ is used to denote measured air concentrations. The subscripts used to denote chemical form introduced in the previous section are employed: ϕ_1 is the measured concentration of I-131 associated with particulates, ϕ_{23} is the measured concentration of I_2 plus HOI, and ϕ_4 is the measured concentration of organic iodides. The 1-sigma counting uncertainties are included in the tables. If the net I-131 activity in a sample was less than the 2-sigma counting uncertainty for the measurement, a "less than" value was reported. As can be seen from the tables, the detection limit for organic forms was lower than for the other forms. The detection limit for inorganic gases was lowered by about a factor of two soon after the start of the monitoring.

The most striking feature of the data is the relative magnitudes of (a) the airborne I-131 attributable to plant operations (Periods 1-15) and (b) the fallout I-131 which dominates Periods 16-23. Radioiodine associated with particulate material was a small component of plant discharges and was detected only once in environmental air samples taken before the first arrival of fallout radioactivity. Iodine-131 attached to particulates was the largest fraction of fallout radioiodine. Inorganic iodine gas was detected in about 1/5 of the pre-fallout samples analyzed. The concentrations of these species increased markedly during Period 16. Organic iodides were detected in about 1/2 of the samples analyzed prior to Period 16 and their concentrations also increased after arrival of the fallout.

TABLE 3-11

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #1 FORKED RIVER MARINA

Measured Concentration (±1σ, fCi/m^3) of I-131

Period	Φ_1	Φ_{23}	Φ_4
1	No Sample		
2	NA(a)	4.4 ± 1.2	NA
3	NA	<2.2	NA
4	<1.7	<1.9	<3.4
5	<1.5	<2.0	<0.31
6	Pump Failure		
7	Pump Failure		
8	<0.65	<1.2	0.71 ± .12
9	<0.70	2.7 ± .7	0.84 ± .12
10	<1.4	<1.5	1.11 ± .12
11	<1.0	3.4 ± 1.3	8.36 ± .29
12	1.03 ± .43	<2.1	3.07 ± .23
13	<0.62	<0.98	<0.19
14	<0.46	<1.4	1.06 ± .12
15	<0.36	<1.4	6.47 ± .25
16	34.7 ± .7	22.5 ± 1.4	5.49 ± .24
17	49.3 ± 1.0	NA	8.66 ± .27
18	15.8 ± .5	8.57 ± .90	9.25 ± .32
19	19.7 ± 1.4	2.76 ± .61	5.92 ± .26
20	11.0 ± .8	(b)	(b)
21	3.60 ± .39	(c)	(c)
22	<0.60	(d)	(d)
23	Power Failure		

(a) NA means no analysis or sample lost during analysis; the same designation is used in subsequent tables.

(b) $\phi_2 = 5.53 \pm .58$; ϕ_3 and ϕ_4 not measured.

(c) $\phi_2 < 1.8$; $\phi_{34} = 0.42 \pm .14$.

(d) $\phi_2 < 2.2$; $\phi_{34} < 0.34$.

TABLE 3-12

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #2
STATE GAME FARM

Period	Measured Concentration (±1σ, fCi/m^3) of I-131		
	Φ_1	Φ_{23}	Φ_4
1		No Sample	
2	NA	<2.3	5.9±1.2
3	NA	<3.0	<3.9
4	<2.3	<3.7	<4.2
5	<0.93	<1.8	<0.25
6	<0.65	<2.0	0.51±.11
7	<0.74	<1.3	<0.21
8	<0.86	<1.7	0.44±.13
9	<0.81	<1.7	0.41±.13
10	<1.5	2.54±.87	0.84±.14
11	<1.1	<1.5	2.33±.22
12	<0.43	NA	0.92±.15
13	<0.64	0.64-0.85(a)	<0.21
14	<0.54	1.28±.20(a)	0.68±.13
15	<0.36	0.95±.30(a)	3.06±.21
16	28.0±.7	17.2±2.6	8.17±.30
17	39.8±1.2	3.63±.42(a)	8.50±.29
18	13.2±.6	6.8±1.1	3.55±.24
19	8.43±.85	2.74±.34(a)	3.22±.23
20	6.82±.75	7.76±.72	0.72±.16
21	3.43±.43	(b)	(b)
22	<0.63	(c)	(c)
23	<0.43	(d)	(d)

(a) The inorganic gas concentration was determined by subtracting the organic iodide concentration from the total gaseous iodine concentration measured with a second sampler.

(b) Φ_2 <1.7; Φ_{34}= 0.34 ± .14

(c) Φ_2 = 1.2--1.6 by difference method; Φ_{34} <0.38.

(d) Φ_2 <0.58; Φ_{34} <0.25.

TABLE 3-13

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #3 PINEWALD SUBSTATION

	Measured Concentration (±1σ, fCi/m^3) of I-131		
Period	Φ_1	Φ_{23}	Φ_4
1		No Sample	
2	NA	<2.5	<2.6
3	NA	<3.5	<2.4
4	<1.6	<2.1	NA
5	<0.94	3.4±1.0	<0.27
6	<0.58	<1.1	<0.21
7	<0.72	<1.3	<0.20
8	<0.84	<1.5	0.57±.13
9	<0.72	<1.1	1.40±.14
10	<1.3	2.27±.65	1.82±.15
11	<1.0	<2.1	6.35±.25
12	<0.47	<1.6	0.59±.14
13	<0.70	<0.86	<0.21
14	<0.54	<1.2	0.39±.12
15	<0.39	<1.9	3.18±.21
16	34.3±.7	21.6±1.1	8.07±.29
17	41.8±1.2	14.0±1.2	8.76±.29
18	14.3±.5	7.2±1.1	7.27±.30
19	9.70±.75	<2.9	5.50±.26
20	9.79±.77	<1.2	7.21±.28
21	2.74±.41	<1.4	2.55±.18
22	<0.57	<1.6	1.13±.21
23	0.97±.20	<0.44	1.81±.09

TABLE 3-14

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #4 GARDEN STATE PARKWAY

	Measured Concentration (±1σ, fCi/m^3) of I-131		
Period	Φ_1	Φ_{23}	Φ_4
1		No Sample	
2	NA	<2.4	<2.7
3	NA	<2.4	5.7±1.8
4	<0.87	<2.2	<2.5
5	<0.98	1.73±.62	1.96±.15
6	<0.54	<1.3	<0.19
7	<0.57	<1.1	2.15±.15
8	<0.73	<1.2	0.84±.12
9	<0.77	<1.3	0.58±.12
10	<1.4	<1.0	2.96±.17
11	<0.92	<1.3	0.89±.16
12	<0.42	<1.6	0.56±.14
13	<0.68	2.62±.40(a)	1.19±.13
14	<0.50	2.44±.95(a)	0.54±.11
15	<0.34	1.45±.26(a)	1.70±.17
16	31.8±.7	18.9±.5 (a)	7.36±.28
17	65.0±1.3	24.1±.9	12.2 ±.3
18	15.8±.5	6.2±.9	5.27±.26
19	8.31±.64	0.50±.35(a)	5.32±.26
20	8.93±.81	1.41±.33(a)	3.99±.23
21	<0.90	0.58±.24(a)	1.46±.16
22	2.37±.32	~0(a,b)	0.94±.21
23	<0.46	<0.88	0.56±.06

(a) The inorganic gas concentration was determined by subtracting the organic iodide concentration from the total gaseous iodine concentration measured with a second sampler.

(b) Concentration by the difference method was = 0.12±.28 fCi/m^3.

TABLE 3-15

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #6
ISLAND BEACH STATE PARK

	Measured Concentration (±1σ, fCi/m^3) of I-131		
Period	Φ_1	Φ_{23}	Φ_4
1		No Sample	
2		No Sample	
3		No Sample	
4	<0.83	<2.2	<3.1
5	<0.98	1.64±.65	<0.21
6	<0.56	<1.1	<0.18
7	<0.70	<1.4	<0.19
8	<0.56	<1.1	<0.21
9	<0.79	<1.4	<0.20
10	<1.4	<1.3	1.22±.13
11	<0.98	<1.3	<0.29
12	<0.44	<0.98	<0.25
13	<0.74	<1.3	<0.21
14	<0.57	<1.4	1.26±.15
15	<0.39	<1.4	1.24±.16
16	13.4±.5	20.7±1.0	4.29±.24
17	43.1±1.2	17.0±1.1	6.25±.25
18	16.2±.5	31.9±2.8	5.07±.28
19	9.2±1.0	7.51±.69	3.76±.24
20	10.5±.8	<1.3	3.10±.21
21	3.52±.39	<1.7	4.65±.23
22	<0.63	2.9±1.0	1.62±.23
23	0.98±.24	<0.47	0.70±.05

COMPARISON OF MEASURED AND PREDICTED AIR CONCENTRATIONS

Analysis of the fate of the released I-131 species is linked to the predictions of environmental concentrations at increasing distances from the plant. The validity of the dispersion model's predictions is itself an important question in the area of environmental dose assessment. For both reasons, it is important to compare the measured concentrations (ϕ) with predicted concentrations (X) of I-131 in the atmosphere. Since the predictive model is designed to estimate long term averages, its application for prediction of weekly average concentrations may be questioned. However, if the predicted short term average concentrations are consistently lower or higher than measured values then the long term average prediction will be similarly biased.

The measured total concentration of I-131, ϕ_T, is the sum of three components: iodine associated with particulates, ϕ_1; the inorganic forms I_2 and HOI, ϕ_{23}; and organic iodides, ϕ_4. The ratios of ϕ_T to X_T, shown in Table 3-16, are all indeterminate because at least one of the three component concentrations was below the analytic detection limit during each period. One ϕ_T/X_T ratio is quite large (Site #2, Period 10). The predicted concentration was quite small (0.0060 fCi/m^3) and the measured value was 3.4-4.9 fCi/m^3, but probably closer to the lower limit since ϕ_1 would be expected to be small. The presence of activity where little or none was expected has been observed previously (5,6). Such observations may be indicative of recirculation of dispersed material which is not considered in the basic single station windrose model. In only 4 of 56 comparisons does the upper limit of the ratio (ϕ_T/X_T) exceed three, indicating that either (a) such recirculation occurs infrequently at Oyster Creek, or (b) the effect of recirculation (which would increase measured concentrations) is more than balanced by overestimation of the concentrations resulting from straight line transport. That the model overestimates the environmental air concentration is clear from the ratios in Table 3-16. The ratios of time weighted average concentrations for all periods, $\bar{\phi}_T/\bar{X}_T$, indicate the model's performance over a 2.5- to 3-month period. The true ratio for each site undoubtedly lies closer to the minimum value given because the less than values of ϕ_1 and ϕ_2 markedly bias the maximum values of ϕ_T/X_T.

The four maximum ratios of ϕ_T/X_T which are greater than 3 occur at Sites #2 and #3 during Periods 5 and 10. In one case (Site #2, Period 5) the maximum ϕ_T is the sum of three concentrations each less than the appropriate detection limit. As noted in the footnote to the table the two lower level release points were not

TABLE 3-16

RATIOS(a) OF MEASURED TO PREDICTED CONCENTRATIONS OF TOTAL I-131

	ϕ_T (fCi/m^3)/X_T (fCi/m^3)				
Period	Site #1	Site #2	Site #3	Site #4	Site #6
1-3	—	—	—	—	—
4	0.0--0.78(b)	0.0--1.9(b)	—	0.0--2.9(b)	0.0--2.3(b)
5	0.0--2.2(b)	0.0--7.0(b)	3.3--4.4(b)	0.84--1.1(b)	0.75--1.3(b)
6	—	0.09--.57	0.0--.54	0.0--.97	0.0--1.7
7	—	0.0--1.1	0.0--.44	0.34--.61	0.0--.57
8	0.13--.48	0.21--1.4	0.09--.46	0.05--.17	0.0--1.0
9	0.16--.19	0.02--.14	0.06--.15	0.04--.19	0.0--.60
10	0.46--1.7	560--810	6.8--9.0	1.0--1.9	0.43--1.4
11	0.44--.47	0.14--.29	0.45--.67	0.11--.39	0.0--.84
12	0.41--.62	—	0.09--.42	0.16--.71	0.0--.48
13	0.0--.15	0.25--.65	0.0--.57	1.1--1.3	0.0--.46
14	0.14--.38	0.15--.19	0.07--.37	0.21--.25	0.10--.25
15	0.77--.98	0.49--.53	0.53--.92	0.49--.54	0.62--1.5
4-15(c)	0.26--.50	0.18--.57	0.26--.56	0.27--.53	0.12--.73

(a) When the ratio was indeterminate the range of possible ratios is shown.

(b) Predicted concentration (X_T) does not include the contribution of vent releases, so the actual ratio or range of ratios is lower than the tabulated ratio or range.

(c) Ratio of the time weighted average concentrations ($\bar{\phi}_T/\bar{X}_T$), including all periods when measurement data were available. Averaging periods ranged from 10 to 12 weeks.

monitored during Periods 1-5 so the actual ratios of ϕ_T/X_T are necessarily less than the tabulated values. During Period 10 very little inorganic I-131 gas was expected at Sites #2 and #3 and detectable amounts were found. Thus the ratios of ϕ_{23}/X_{23} for Period 10, shown in Table 3-17, are very large for those locations. (The data in this table clearly illustrate the cumulative effect of the many less than values for ϕ_{23} on the maximum value of the time weighted average concentrations.) As noted above, recirculation of dispersed effluents or deviations from straight line transport are possible causes of unexpected air concentrations. In Period 10, the model predicted no contribution from low level releases to the concentration at Site #3 and little from the elevated release. However, X/Q values in adjacent sectors were higher so it is quite possible that I-131 from nearby areas was transported to the sampling location. A second factor which may influence the predicted values is the assumption that the measured release rate was constant during the period. If the release rate were high while the wind was blowing toward Site #3 and lower at other times, then the observed concentration would reflect that, but the predicted value would not. The fact that both the inorganic iodine and the organic iodide were found in higher concentrations than had been predicted is consistent with either (or a combination) of those mechanisms.

A third possible explanation is that organic iodides released from the plant were converted to more reactive forms which could be trapped by the IPh bed. Figure 3-8 shows the relative excitation rate by ultraviolet (UV) light (wavelengths between 292.5 and 342.5 nm) during the entire monitoring period at Oyster Creek. The excitation rates were calculated following Zafiriou (10) using the average sea level UV radiation levels from Reference (11) and the absorption cross sections from Reference (12). The correction for cloud cover suggested by Johnson et al. (11) was made using data from the National Weather Service station at Atlantic City, N.J. (13), the coastal station nearest to the Oyster Creek plant. It is interesting to note that the maximum excitation rate occurred in Period 10; however, the peak value is only about 30% higher than the baseline value, which is ∿0.6 during the first 12 monitoring periods. The fraction of the total gaseous iodine which was in organic form did not decrease with distance from the plant during Period 10, as can be seen in Table 3-18.

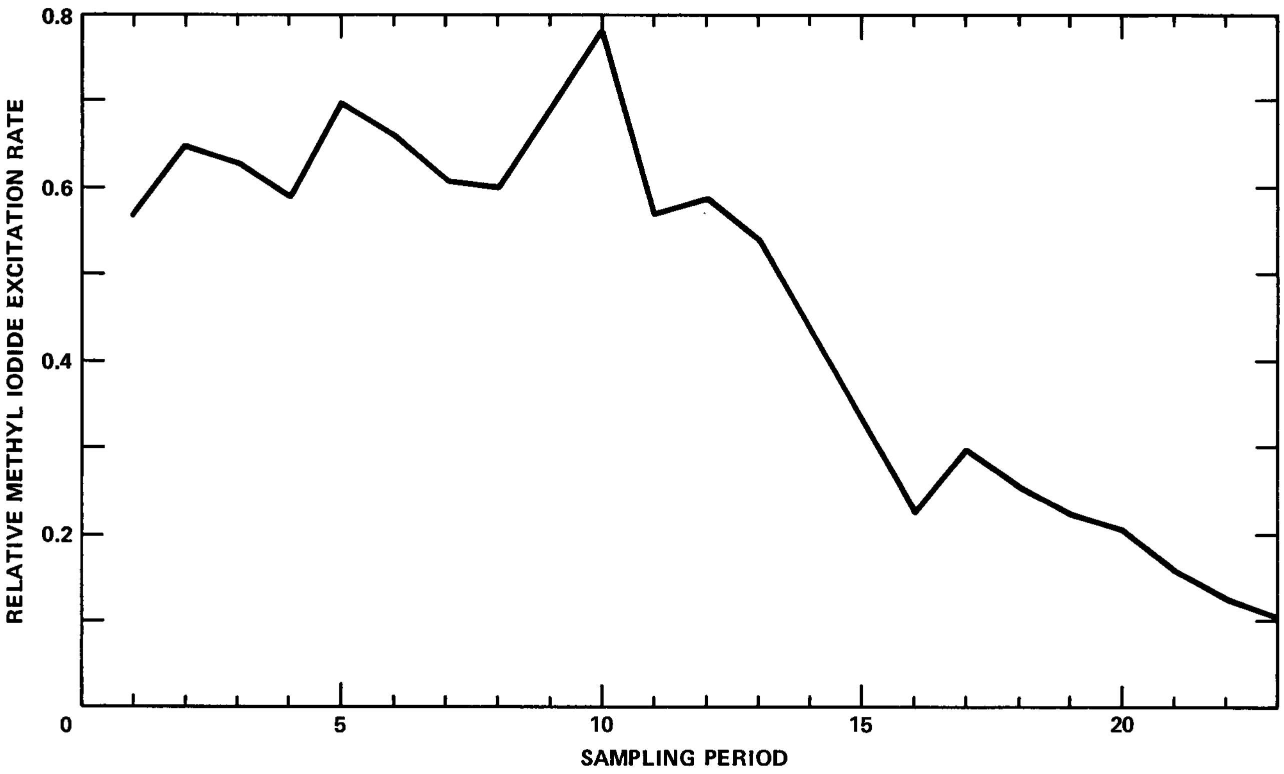

0.8
0.6
0.4
0.2
0
5
10
15
20
RELATIVE METHYL IODIDE EXCITATION RATE
SAMPLING PERIOD

TABLE 3-17

RATIOS$^{(a)}$ OF MEASURED TO PREDICTED CONCENTRATIONS OF INORGANIC I-131 GAS

	ϕ_{23} (fCi/m^{3}X$_{23}$ (fCi/m^3)				
Period	**Site #1**	**Site #2**	**Site #3**	**Site #4**	**Site #6**
1	—	—	—	—	—
2	0.93$^{(b)}$	0.0--1.7$^{(b)}$	0.0--1.1$^{(b)}$	0.0--3.1$^{(b)}$	—
3	0.0--8.4$^{(b)}$	0.0--4.1$^{(b)}$	0.0--2.2$^{(b)}$	0.0--1.5$^{(b)}$	—
4	0.0--1.1$^{(b)}$	0.0--3.7$^{(b)}$	0.0--1.6$^{(b)}$	0.0--6.0$^{(b)}$	0.0--4.4$^{(b)}$
5	0.0--5.4$^{(b)}$	0.0--20$^{(b)}$	16$^{(a)}$	1.9$^{(a)}$	3.6$^{(a)}$
6	—	0.0--1.6	0.0--1.4	0.0--2.8	0.0--4.6
7	—	0.0--3.6	0.0--1.8	0.0--1.2	0.0--2.5
8	0.0--.84	0.0--3.1	0.0--.87	0.0--.27	0.0--2.3
9	0.55	0.0--3.8	0.0--.24	0.0--.44	0.0--1.6
10	0.0--4.8	2100	27	0.0--1.8	0.0--2.4
11	0.55	0.0--.40	0.0--.67	0.0--.72	0.0--2.0
12	0.0--.74	—	0.0--.92	0.0--1.6	0.0--1.1
13	0.0--.72	1.7--2.2	0.0--3.1	10	0.0--3.1
14	0.0--1.2	0.70	0.0--1.6	1.3	0.0--.78
15	0.0--.85	0.67	0.0--2.0	1.5	0.0--2.9
2-15$^{(c)}$	0.35--.87	0.33--1.4	0.29--1.3	0.47--1.3	0.21--2.1

(a) When the ratio was indeterminate the range of possible ratios is shown.

(b) Predicted concentrations do not include the contribution of vent releases, so the actual ratio or range of ratios is lower than the tabulated ratio or range.

(c) Ratio of the time weighted average concentrations ($\overline{\phi}_{23}/\overline{X}_{23}$), including all periods when measurement data were available. Averaging periods ranged from 12 to 14 weeks.

Table 3-18

OBSERVED RATIO OF ORGANIC IODIDE-131 TO TOTAL GASEOUS I-131 DURING PERIOD 10

Site	Distance From Plant (m)	$\phi_4/(\phi_{23}+\phi_4)$
#1	2440	0.43-1.0
#2	3880	0.25±.08
#4	6490	0.75-1.0
#3	8320	0.44±.08
#6	9480	0.48-1.0

Considering only the three locations to the northeast of the plant (#1, #2, and #3), the smallest organic fraction was found at the intermediate distance. However, the organic iodide fraction at Site #2 was about 180 times that predicted by the model. These observations are not consistent with a simple methyl iodide photolysis scenario. It should also be noted that higher than expected concentrations of inorganic gases were detected during Periods 13-15 at locations #3 and #4 when the average UV excitation rate was about 30% lower than during the first 12 monitoring periods. Thus, other factors, such as time variations in the I-131 source term or transport of material from adjacent sectors, must account for the observed inorganic iodide concentrations in Period 10.

A better evaluation of the predictive model can be made using the ratios of measured to predicted organic iodide concentrations. Because the detection limit for organic iodides was better than for other components, more of the ratios could be uniquely determined. Table 3-19 contains the ratios ϕ_4/X_4 for each location by sampling period and the long term average values. The model over-predicts the long term average concentrations at the sampling locations by factors of 3-6. The predictions for the two most distant locations (#3 and #6) are somewhat worse than for the other three locations.

As can be seen from the table, there is a rather broad range of the weekly ϕ_4/X_4 ratios for a particular location but most values are concentrated between 0 and 1. This suggested that an alternative analysis would be to consider the possibility of a log-normal distribution of the ratios. The distribution of ratios at each location was plotted on log-probability paper and the median and geometric

Table 3-19

RATIOS(a) OF MEASURED TO PREDICTED CONCENTRATIONS OF ORGANIC I-131 GAS

	Ratios of Measured to Predicted Concentrations of Organic I-131				
Period	Site #1	Site #2	Site #3	Site #4	Site #6
1	-	-	-	-	-
2	-	1.4(b)	0.0--.35(b)	0.0--1.0(b)	-
3	-	0.0--1.6(b)	0.0--.46(b)	1.1(b)	-
4	0.0--.48(b)	0.0--1.0(b)	-	0.0--1.6(b)	0.0--1.5(b)
5	0.0--.23(b)	0.0-- .76(b)	0.0--.34(b)	0.58(b)	0.0-- .12(b)
6	-	0.13	0.0--.078	0.0-- .12	0.0-- .21
7	-	0.0-- .12	0.0--.047	0.41	0.0-- .057
8	0.19	0.30	0.12	0.069	0.0-- .16
9	0.050	0.026	0.084	0.054	0.0-- .064
10	0.54	175	3.6	1.3	0.54
11	0.40	0.18	0.59	0.15	0.0-- .12
12	0.43	0.39	0.13	0.21	0.0-- .099
13	0.0--0.019	0.095	0.0--.075	0.37	0.0-- .048
14	0.16	0.061	0.078	0.044	0.11
15	0.97	0.46	0.63	0.31	0.81
2-15 (c)	0.26--.30	0.22--.34	0.19--.28	0.26--.33	0.10--.22

(a) When a ratio was indeterminate, the range of possible ratios is shown.

(b) Predicted concentrations do not include the contribution of vent releases so the actual ratio or range of ratios is lower than the tabulated ratio or range.

(c) Ratio of the time weighted average concentrations ($\bar{\phi}_4/\bar{X}_4$), including all periods when measurement data were available. Averaging periods ranged from 10 to 14 weeks.

standard deviation were determined graphically. In this evaluation an indeterminate ratio (there are many for Site #6) was assumed to be equal to the maximum of the range of possible values. Table 3-20 contains the median and geometric standard deviations for each location. The median ratios are comparable to the ratios of the long term average concentrations. Again, the more distant locations showed the largest differences between measured and predicted values, with the predicted concentration about 6 times that which was measured.

Table 3-20

DISTRIBUTION PARAMETERS FOR RATIOS OF
MEASURED TO PREDICTED CONCENTRATIONS OF ORGANIC I-131

Site Number and Name	Range of Ratios[a]	Median Ratio	Geometric Std. Dev.
1. Forked River Marina	0.019 - 0.97	0.25	2.3
2. State Game Farm	0.026 - 175	0.32	4.7
3. Pinewalk Substation	0.047 - 3.6	0.17	5.3
4. Garden State Parkway	0.044 - 1.6	0.33	3.7
6. Island Beach State Park	0.048 - 1.5	0.17	3.6

(a) In cases when the measured concentration was less than the detection limit, the concentration was assumed equal to the detection limit for computation of the ratio.

I-131 CONCENTRATIONS IN PRECIPITATION

Sampling Methods

Both total precipitation and precipitation rate were measured at a location southwest of the plant on the Oyster Creek-Forked River site. Precipitation samples were collected at the same location and analyzed according to the method in Appendix H. The collector contained 1 M mercaptoacetic acid to assure retention of any organically bound iodine scavenged from the atmosphere by the precipitation.

Results

The nominal detection limit for I-131 in rainwater (procedure in Appendix H) was 2 pCi/liter. No I-131 was detected in rain samples collected at the Oyster Creek Site.

Since the principal component of releases from the plant was organic iodine, this is not surprising. Although, organic iodides have been detected in precipitation (14), the expected removal of organic iodides by rain is a small fraction of that expected for elemental iodine (15). While transport of fallout I-131 by rain to the ground surface has been frequently observed, the similarity of washout ratios (16) strongly suggests that the scavenged I-131 was attached to particulate material. Data in a subsequent subsection show that about half of the fallout I-131 was in particulate form for nearly two months after the above ground detonation during the field monitoring period at Oyster Creek. Eggleton et al. (17) also observed a distinct difference between washout ratios for fallout I-131, which ranged between 300 and 1500, and those for the principally gaseous stable iodine (ratios between 10 and 50).

I-131 CONCENTRATIONS IN VEGETATION

Sampling Methods and Locations

Prepared grass flats were exposed to the atmosphere for two-week periods at four locations (Sites #2, #3, #4, and #6) in the Oyster Creek environment. At the end of the exposure period the grass was clipped and placed in double polyethylene bags at the field monitoring location. The samples were appropriately labeled and returned to the laboratory for radiochemical analysis. The exposed grass flats were also returned to the laboratory and replaced with fresh grass flats. Prior to field exposure the grass was watered on a regular schedule under greenhouse operator supervision. Grass flats exposed at the field monitoring locations were not given special watering or other attention. The radiochemical analysis procedure for the grass samples is contained in Appendix I and, as previously indicated, the gamma spectrum analysis procedures are described in Appendix G.

Measured Concentrations of I-131 in Vegetation

Table 3-21 contains the results of the measurements of I-131 in grass exposed to the atmosphere in the vicinity of the plant. The two-week exposure period was selected (instead of the typical 1-week period) to improve chances of detecting I-131 in the vegetation. Even so, only 1/3 of the samples collected prior to the arrival of the fallout radioactivity contained detectable amounts of I-131. The highest measured concentration attributable to plant releases was 4.2 pCi/m^2 at Site #2. That concentration was less than 1/100 the peak fallout I-131 concentration measured during the study.

Table 3-21

MEASURED CONCENTRATIONS OF I-131 ON GRASS

Exposure Periods	Areal I-131 Concentration (pCi/m^2)			
	Site #2	Site #3	Site #4	Site #6
6 & 7	-	3.3±1.4	3.2±1.7	-
7 & 8	<0.9	-	-	<0.7
8 & 9	-	1.9± .6	<0.9	-
9 & 10	4.2± .7	-	-	1.6± .6
10 & 11	-	<1.5	<2.1	-
11 & 12	<2.3	-	-	<1.7
12 & 13	-	<1.4	1.7± .8	-
13 & 14	<1.4	-	-	<1.3
14 & 15	-	<1.6	<1.6	-
15 & 16	373±6	-	-	667±10
16 & 17	-	392±7	395±7	-
17 & 18	353±7	-	-	290±6
18 & 19	-	277±6	165±6	-
19 & 20	127±4	-	-	102±4

COMPARISON OF MEASURED AND PREDICTED VEGETATION CONCENTRATIONS

An evaluation of the combined atmospheric transport and deposition models can be made by comparing the measured concentrations of I-131 in grass with those predicted by the models. Table 3-22 shows the results of such a comparison for each of the four monitoring locations prior to the arrival of the fallout. Again the ratio or the measured concentration (C_m) to the predicted concentration (C) was used as a measure of the validity of the model.

Table 3-22

RATIOS OF MEASURED TO PREDICTED GRASS CONCENTRATIONS

Exposure Periods	C_m (pCi/m^2)/C (pCi/m^2)			
	Site #2	Site #3	Site #4	Site #6
6 & 7	-	1.1	0.84	-
7 & 8	<1.1	-	-	<0.81
8 & 9	-	0.21	<0.054	-
9 & 10	0.79	-	-	0.28
10 & 11	-	<0.095	<0.26	-
11 & 12	<0.20	-	-	<0.39
12 & 13	-	<0.15	<0.036	-
13 & 14	<0.07	-	-	<0.48
14 & 15	-	<0.39	<0.11	-
6 - 15	-	0.26--.39[(a)]	0.7--.26[(a)]	-
7 - 14	0.20--.54[(a)]	-	-	0.07--.49[(a)]

(a) Since the average value is indeterminate, the range of possible average values is shown.

The range of ratios of C_m/C was from <0.036 to 1.1. The graphically determined median of the 18 values was 0.26; the geometric standard deviation was 3.1. In the determination of the median ratio, all measured concentrations less than the detection limit were assumed equal to the detection limit. The true median value is no doubt lower than 0.26 since it is quite unlikely that all the unknown concentrations lie just below the detection limits for those analyses.

The results of the comparison of measured and predicted grass concentrations are consistent with the air concentration comparisons. In both cases the comparisons show that the models predict concentrations higher than those measured by factors of 3 to 6. The fact that the average measured to predicted vegetation concentration ratio, which reflects both the dispersion and deposition models, is similar to that observed for the dispersion model above suggests that the deposition model may be valid and that the dispersion model is the source of the overprediction of the I-131 concentrations in vegetation. The time periods covered, significant fractions of a grazing season, are long enough to provide a valid test of the models.

BEHAVIOR OF FALLOUT RADIOIODINE SPECIES

Monitoring was continued after the beginning of the weapons test fallout to observe the behavior of airborne I-131 from that source. The high fallout concentrations permitted detection of fallout iodine species for about two months. During that time the iodine concentrations and species were measured at ground level at the monitoring locations described previously. Downward migration and lateral dispersion of fallout from the detonation were taking place throughout the monitoring period. The sampled air therefore contained a mixture of fallout radioiodine on particles and as gases which had resided in the lower troposphere for various time periods.

Initially, a large fraction of the total airborne I-131 was associated with particulate material. Table 3-23 shows that half to two-thirds of the I-131 was associated with particulates during the first six sampling periods. However, the fraction ϕ_1/ϕ_T declined to 1/4--1/3 in the last two monitoring periods, which is closer to the particulate-to-total ratio for stable iodine in the atmosphere.

Table 3-23

FRACTION OF TOTAL FALLOUT I-131 ASSOCIATED WITH PARTICULATES

	Observed Fraction ϕ_1/ϕ_T					
Period	Site #1	Site #2	Site #3	Site #4	Site #6	Mean
16	0.55±.02[a]	0.52±.03	0.54±.02	0.55±.01	0.35±.02	0.50±.04[b]
17	-	0.77±.03	0.65±.03	0.64±.02	0.65±.02	0.68±.04[b]
18	0.47±.02	0.56±.04	0.50±.03	0.58±.03	0.31±.02	0.48±.05[b]
19	0.69±.06	0.59±.07	0.54--.64[c]	0.59±.06	0.45±.06	0.57--.59[c]
20	-	0.45±.06	0.54--.58	0.62±.07	0.70--.77	0.58--.61
21	0.62--.90	0.62--.90	0.41--.52	0.0--.31	0.36--.43	0.40--.61
22	-	0.0--.29	0.0--.17	0.72±.13	0.0--.12	0.18--.28
23	-	0.0--.34	0.30--.35	0.0--.45	0.46--.58	0.19--.43

---

[a] Standard deviation of the ratio is shown.

[b] Standard deviation of the mean of the ratios.

[c] When the fraction or the mean was indeterminate, the range of possible values is shown.

The character of the gaseous iodine fraction changed during the period when measurements were made. Table 3-24 contains the fractions of the gaseous iodine in organic form, $\phi_4/(\phi_{23}+\phi_4)$, for each location and sampling period. The mean ratios are plotted as a function of time after detonation in Figure 3-9. While the average ratio can only be bounded for the later monitoring periods, a trend is distinguishable; the organic fraction increased with time after detonation from 1/4 to ∿3/4 of the gaseous iodine. Since the rate of injection of the iodine species into the lower troposphere during the period is not known, it is not possible to estimate the conversion rate of inorganic forms to organic iodides or to speculate about the variability of the organic fraction (for example, the lower fractions during Period 22).

Table 3-24

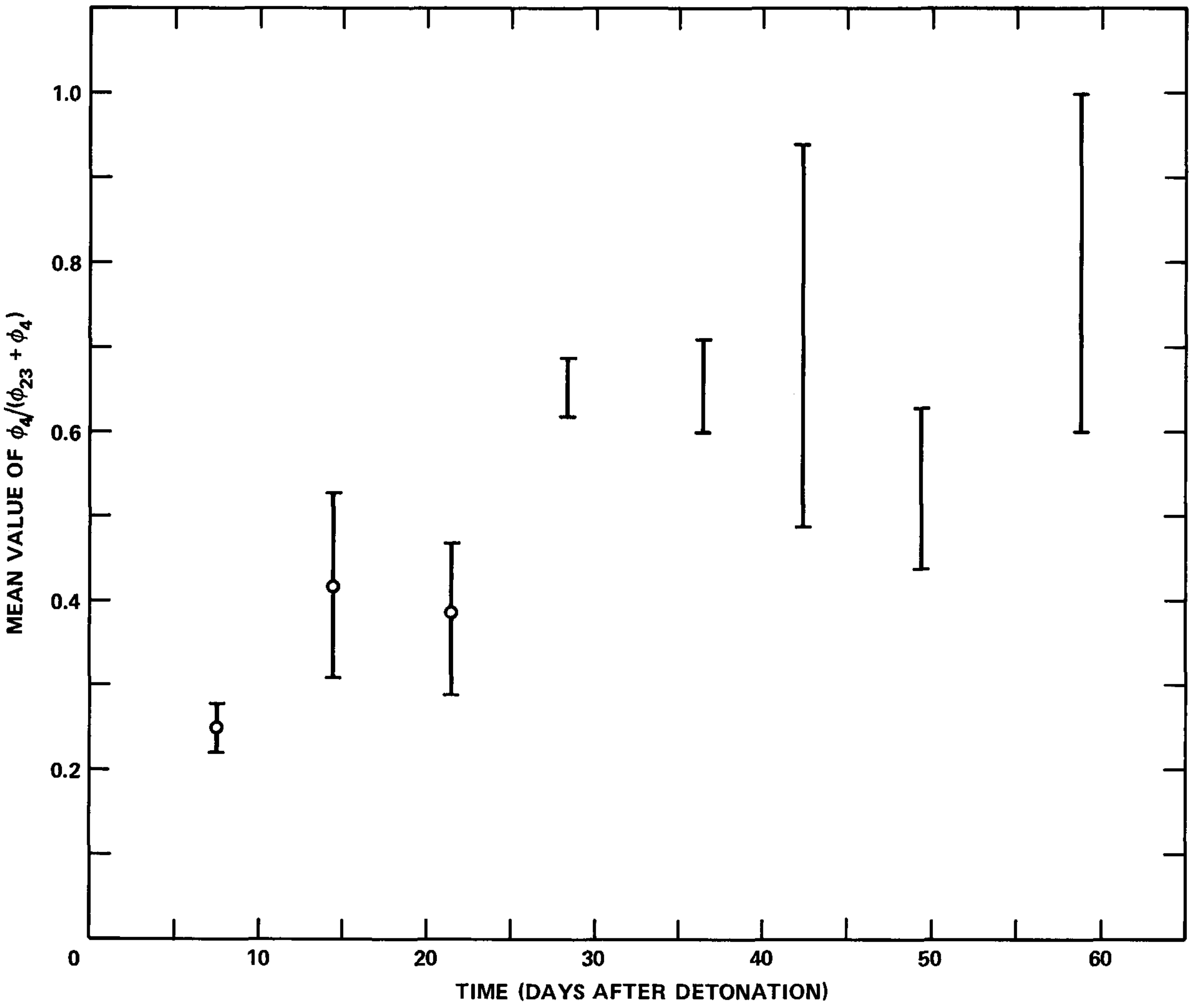
FRACTION OF GASEOUS FALLOUT I-131 IN ORGANIC FORM

	$\phi_4/(\phi_{23}+\phi_4)$					
Period	**Site #1**	**Site #2**	**Site #3**	**Site #4**	**Site #6**	**Mean**
16	0.20±.01(a)	0.32±.03	0.27±.01	0.28±.01	0.17±.01	0.25±.03(b)
17	-	0.70±.04	0.38±.02	0.34±.01	0.27±.02	0.42±.11(b)
18	0.52±.03	0.34±.04	0.50±.04	0.46±.04	0.14±.01	0.39±.08(b)
19	0.68±.06	0.54±.05	0.65--1.0(c)	0.91±.08	0.33±.03	0.62--.69
20	-	0.085±.020	0.86--1.0	0.74±.07	0.70--1.0	0.60--.71
21	0.19--1.0	0.17--1.0	0.64--1.0	0.72±.13	0.73--1.0	0.49--.94
22	-	0.0--.15	0.41--1.0	1.0 ±.2	0.36± .09	0.44--.63
23	-	-	0.80--1.0	0.39--1.0	0.60--1.0	0.60--1.0

(a) Standard deviation of the ratios shown.

(b) Standard deviation of the mean of the ratios.

(c) When the ratio was indeterminate, the range of possible ratios is shown.


MEAN VALUE OF $\phi_4/(\phi_{23} + \phi_4)$
1.0
0.8
0.6
0.4
0.2
0
10
20
30
40
50
60
TIME (DAYS AFTER DETONATION)

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Section 4

MEASUREMENTS OF I-131 AT QUAD CITIES

The Quad Cities Nuclear Power Station is located in western Illinois on the east bank of the Mississippi River. The climate is distinctly continental with local influences of the large river. The area surrounding the plant site is relatively flat with some rolling hills and is principally devoted to agriculture. A major metropolitan area containing the adjacent cities of Moline and Rock Island, Illinois and Davenport and Bettendorf, Iowa, from which the plant name is derived, is located 25-35 km to the south. The Station contains two boiling water reactors (BWRs), each having a thermal power rating of 2450 MW and an electrical output of 800 MW. The steam jet air ejector exhaust gas, formerly a principal source of radioiodine in BWRs, normally passed through an augmented off-gas (AOG) system at the Quad Cities plant during the study, in the summer and fall of 1977.

During the same period a second field study was undertaken at the Quad Cities Station by Allied Chemical Corporation (ACC) under contract with the Nuclear Regulatory Commission. (1) That study was also concerned with environmental transport of plant effluents and the behavior of I-131 in the milk food chain. The programs were complementary. The effluent discharge rate and radioiodine species measurements (described below) were used by both groups. The ACC group made additional measurements of noble gas, tritium, and C-14 gaseous discharge rates. Noble gas concentrations were also monitored in the field to provide data for evaluating the meteorological dispersion model. The ACC I-131 studies concentrated on measurements of wet deposition, concentrations of I-131 in vegetation, and concentrations of I-131 in milk of cows grazing at two field locations. Field monitoring by ACC continued to the end of October. The measurement data and analysis are contained in Reference (1).

Subsections which follow describe the measurements of I-131 in gaseous effluents, the predicted transport of I-131 in the plant's environs, measured concentrations of I-131 in environmental air samples, and comparisons of predicted and measured concentrations. Some of the descriptions are rather brief since the procedures have been previously discussed in Section 3 and are described in detail in the Appendices.

IODINE-131 IN GASEOUS EFFLUENTS

Sampling Locations and Discharge Flow Rates

There were two principal I-131 discharge points: a 94-meter chimney located just west of the north end of the building complex and a 50-meter vent stack near the center of the turbine and reactor building complex. The vent stack rises only about 8 meters above the two reactor buildings whose ventilation air streams are discharged through it. The tall chimney discharges ventilation air from the turbine and radwaste buildings and the exhaust from the steam jet air ejector (SJAE) and turbine gland seal lines. When the mechanical vacuum pump is operated its exhaust is also discharged through the tall chimney. The tall chimney and vent stack discharges were monitored continuously during the study. The discharge flow rates were measured at the beginning of the study using the helium dilution technique. The measured flow rates were 270,200 ft^3/min (1.28×10^8 cm^3/sec) for the tall chimney and 110,200 ft^3/min (5.20×10^7 cm^3/sec) for the vent stack. The flow rates varied from day to day depending upon the discharge fan combinations. The measured flow rates were used in combination with plant records to determine the daily flow rates.

Sampling and Analytical Methods

The release rates and chemical forms of I-131 discharged to the atmosphere were monitored continuously throughout the study. The release rates from the two reactor buildings and from the tall chimney were measured on a daily basis using the SAI CP-200 impregnated charcoal cartridges. These were first counted at the plant and then returned to the laboratory for analysis by gamma spectrometry (see Appendix A). Radioiodine species sampling for the two discharges was normally on a (7-10)-day schedule. These samples were used to determine the chemical forms of I-131 in the combined reactor building vent discharge and in the combined discharge of turbine building, radwaste building, SJAE, and gland seal exhausts. The radioiodine species sampler and the radiometric analysis procedures are described in Appendix A.

Plant Performance Data

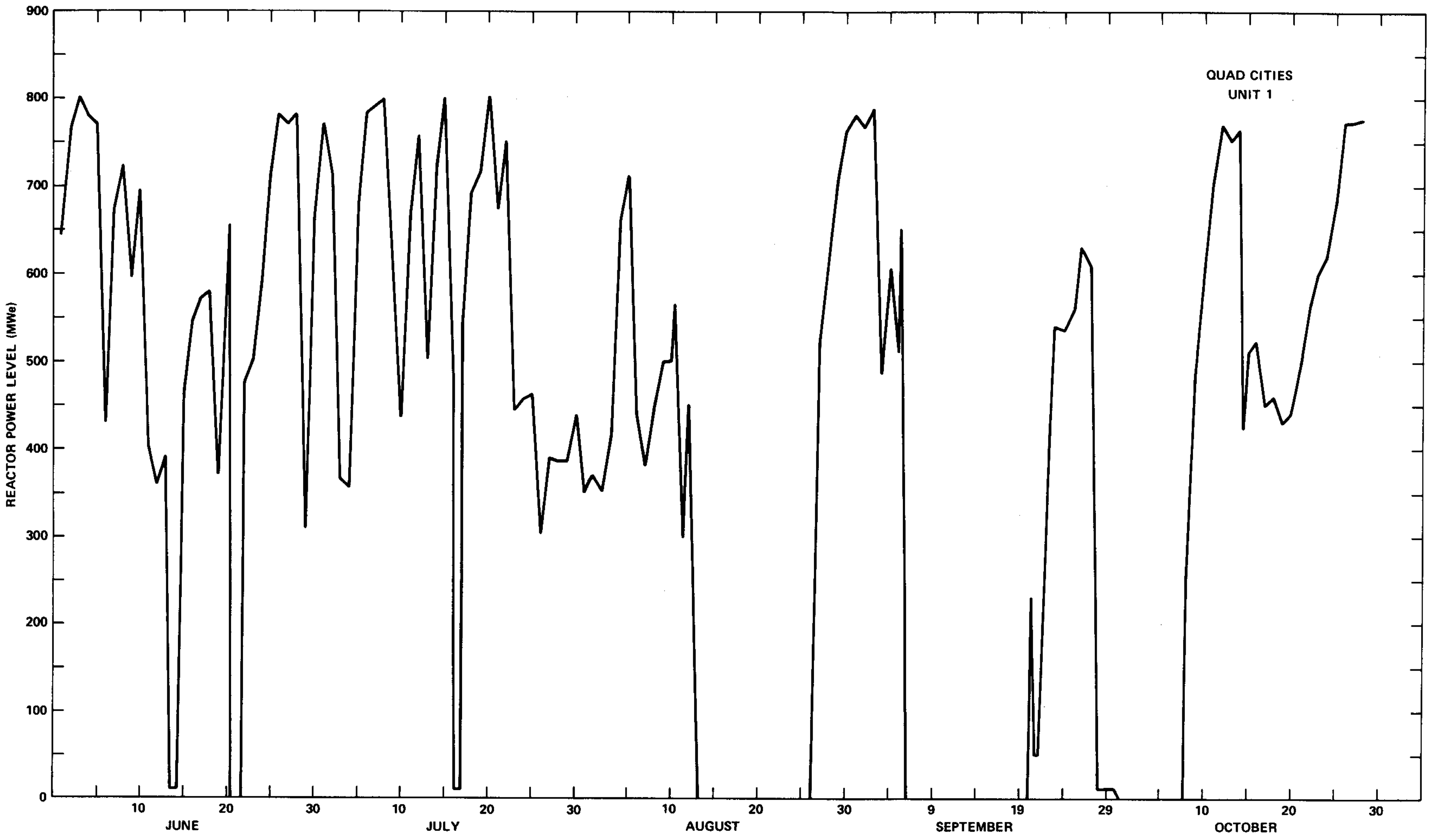
The electrical power outputs for the two Quad Cities reactors are plotted in Figures 4-1 and 4-2 for the period June--October, 1977. These data were obtained from plant logsheets of hourly power levels. It is clear from the plots that the power levels of both plants varied widely throughout the monitoring period. It was not possible to obtain detailed data on the variations of the I-131 concentration reactor water during the period. Daily reactor water samples were taken by plant personnel but those were analyzed promptly using a NaI(Tl) detector so the I-131 concentration was often undeterminable. Weekly composite samples were analyzed for I-131 by plant personnel. The results of those analyses have been plotted in Figure 4-3 for Unit 1 and in Figure 4-4 for Unit 2 (points with counting uncertainties indicated). Also shown in the figures are estimates of the I-131 concentrations obtained from the summaries of the daily sample counting results. It must be emphasized that these values (plotted as points) are uncertain and, perhaps more important, the record is incomplete since there were many days for which a concentration could not even be estimated. The data in Figures 4-3 and 4-4 are only indicative of the variability of the I-131 concentration in reactor water.

Chemical Forms of I-131

The data on radioiodine species fractionation are tabulated by sampling period in Appendix J. The radioiodine species fractionation for each sample was derived using the procedure given in Appendix A. The species distributions varied considerably from period to period. The source of the increased organic iodine releases from the tall chimney during Periods 11-14 is not known. Table 4-1 contains the average species distributions observed during the 14 monitoring periods. Included for comparison are the reactor building species distributions measured at the Quad Cities plant in 1974 (2) and the reactor building concentrations measured at Oyster Creek (from Section 3). There are no data comparable to those for the combination of sources released through the Quad Cities main chimney. The 1977 data for the Quad Cities reactor building vent are in reasonable agreement with previous long term measurements, although the average particulate and elemental iodine fractions are lower than those observed previously.

I-131 Release Rates

Average daily I-131 release rates were measured for the two reactor building ventilation air streams and the discharge of the main chimney. The results of


QUAD CITIES
UNIT 1
REACTOR POWER LEVEL (MWe)
900
800
700
600
500
400
300
200
100
0
10
20
30
JUNE
10
20
30
JULY
10
20
30
AUGUST
9
19
29
SEPTEMBER
10
20
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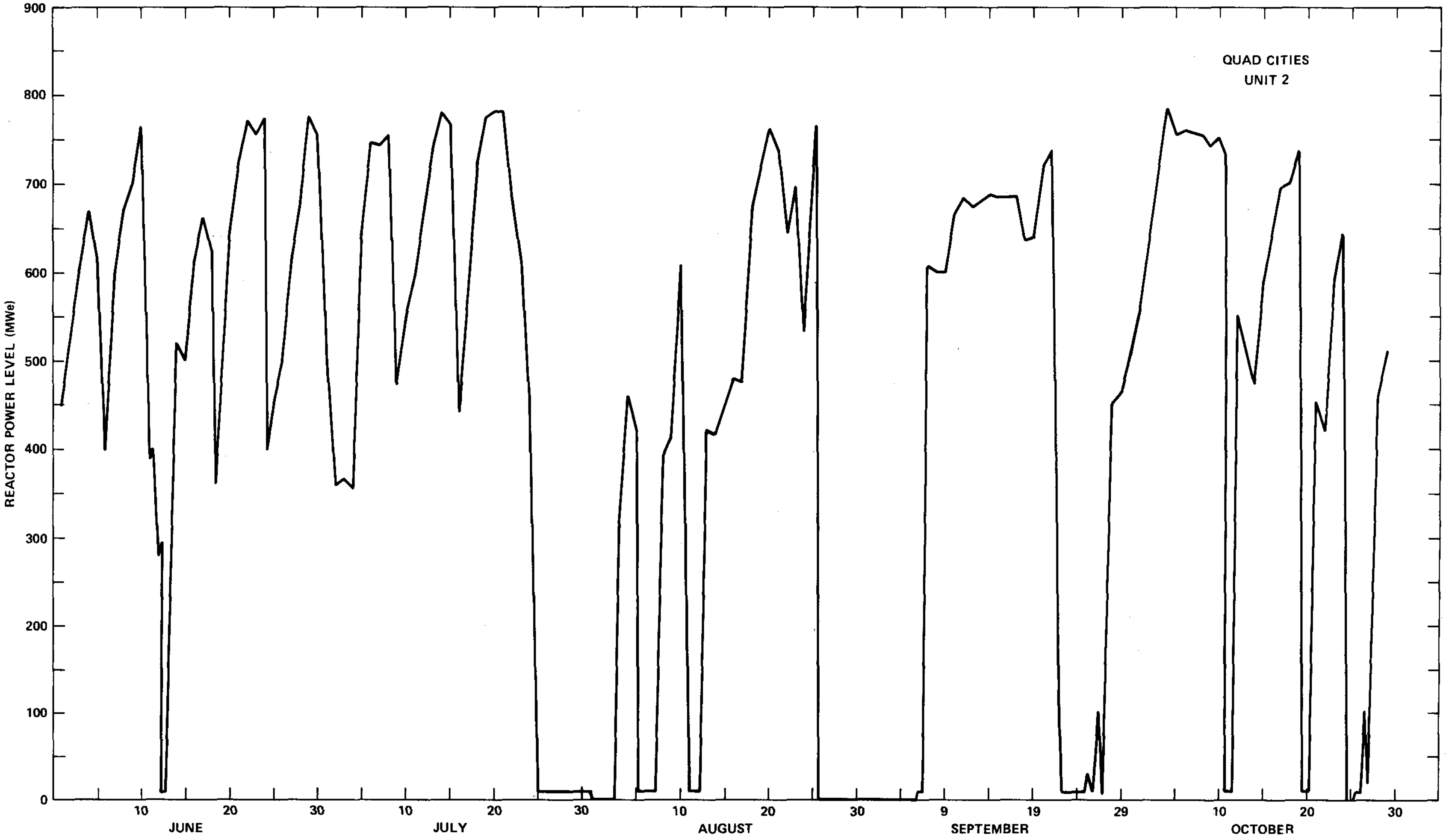
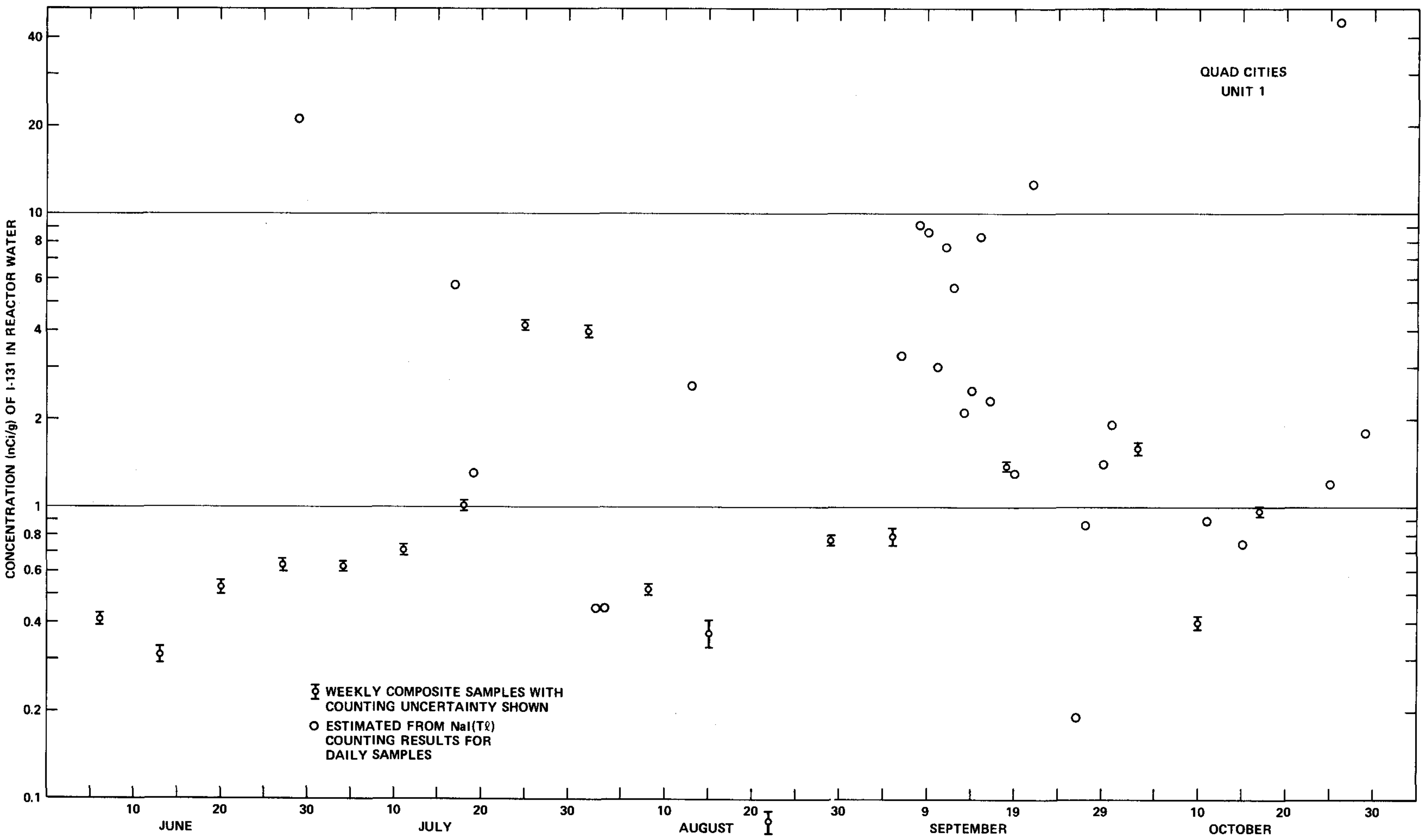
OCTOBER


QUAD CITIES
UNIT 2
REACTOR POWER LEVEL (MWe)
900
800
700
600
500
400
300
200
100
0
10
20
30
10
20
30
10
20
30
9
19
29
10
20
30
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER


QUAD CITIES
UNIT 1
CONCENTRATION (nCi/g) OF I-131 IN REACTOR WATER
40
20
10
8
6
4
2
1
0.8
0.6
0.4
0.2
0.1
10
20
30
JUNE
10
20
30
JULY
10
20
AUGUST
30
9
19
29
SEPTEMBER
10
20
30
OCTOBER
WEEKLY COMPOSITE SAMPLES WITH COUNTING UNCERTAINTY SHOWN
ESTIMATED FROM NaI(Tℓ) COUNTING RESULTS FOR DAILY SAMPLES

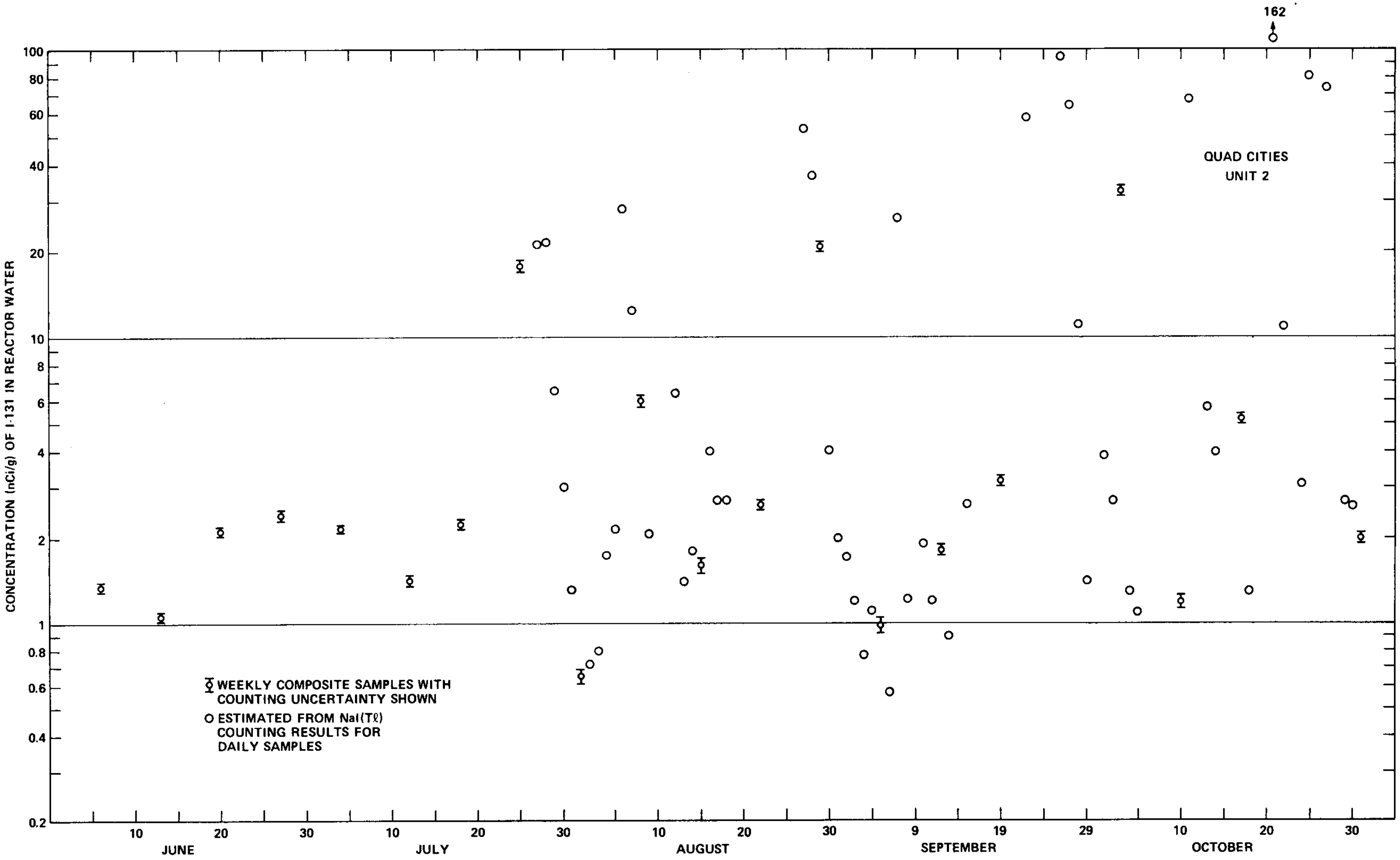

162
QUAD CITIES
UNIT 2
CONCENTRATION (nCi/g) OF I-131 IN REACTOR WATER
100
80
60
40
20
10
8
6
4
2
1
0.8
0.6
0.4
0.2
WEEKLY COMPOSITE SAMPLES WITH COUNTING UNCERTAINTY SHOWN
ESTIMATED FROM NaI(Tℓ) COUNTING RESULTS FOR DAILY SAMPLES
10
20
30
10
20
30
10
20
30
9
19
29
10
20
30
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER

Table 4-1

AVERAGE QUAD CITIES RADIOIODINE SPECIES FRACTIONATIONS AND COMPARABLE DATA FOR REACTOR BUILDING VENTILATION AIR

Source	Number of Samples	Average Percent of Discharged I-131 in Each Form[a]			
		Particulate	I_2	HOI	Organic
Quad Cities Main Chimney[b]	14	8±2	40±5	24±2	29±7
Quad Cities Reactor Building Vent[c]	14	5±1	18±2	36±5	41±3
Quad Cities Reactor Bldgs.					
Unit 1 (1974)	12	10±2	22±2	19±1	50±4
Unit 2 (1974)	13	10±1	31±3	23±1	36±4
Oyster Creek Reactor Building (1976)	23	9±1	28±1	33±2	29±3

(a) One standard deviation of the mean is given with the mean. The total for a source may differ from 100 percent due to rounding of individual entries.

(b) Includes ventilation air from the turbine and radwaste buildings, the steam jet air ejector exhaust, the gland seal exhaust, and the mechanical vacuum pump exhaust.

(c) Includes the ventilation air from both reactor buildings.

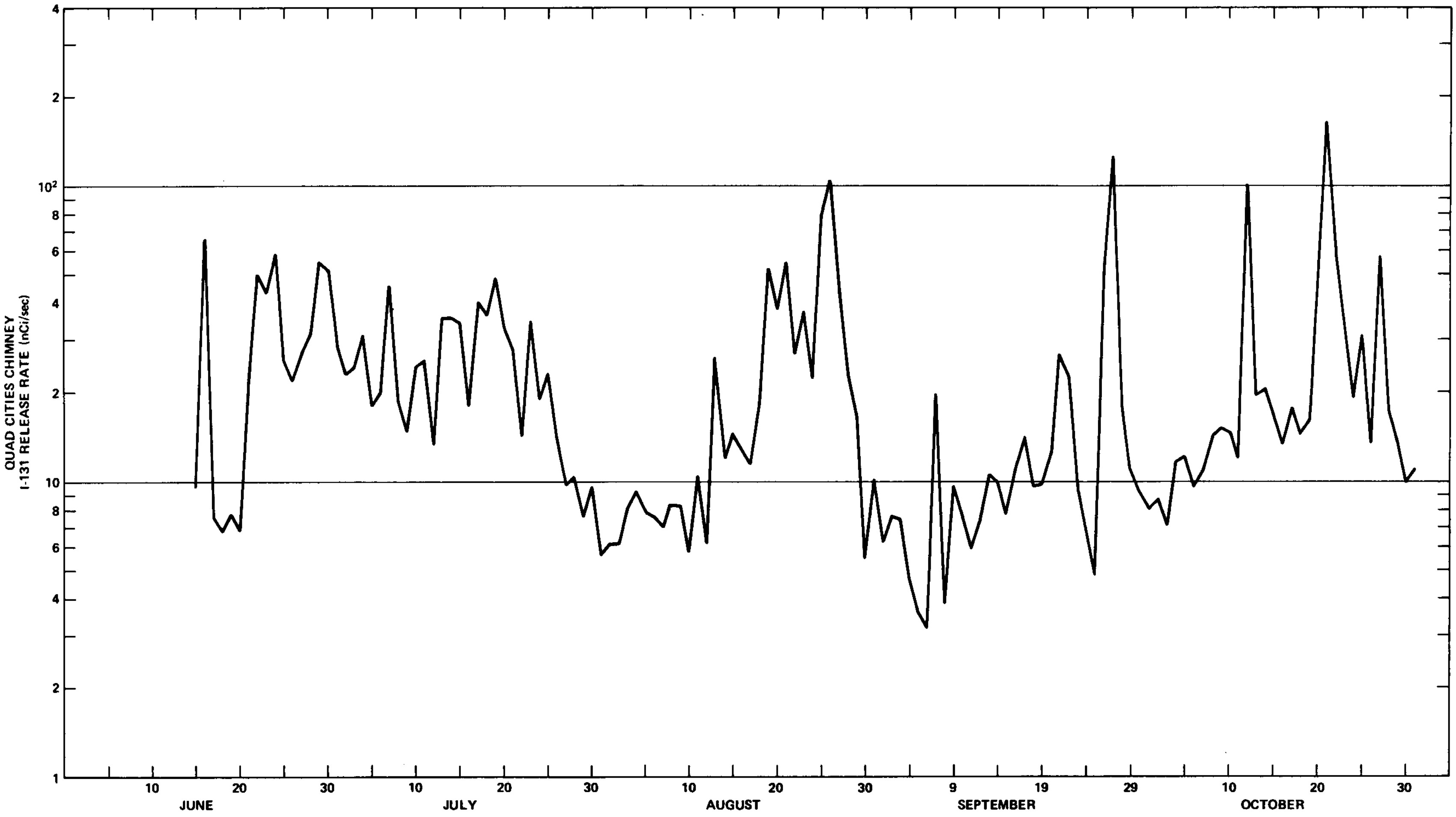
the measurements are shown in Figures 4-5 and 4-6. Tabulations of the daily release rates are contained in Appendix J. The daily release rates reflect the large variability in reactor power level shown in Figures 4-1 and 4-2. Because of the uncertainties in and incompleteness of the data on I-131 concentration in reactor water, it was not possible to compute normalized release rates for the reactor buildings or the combined sources discharged via the tall chimney.

The daily release rates and radioiodine species data were used to calculate average release rates for each sampling period. These average values, shown in Table 4-2, and the species data for each period were used to compute expected concentrations of I-131 in the environment as described in the next section.

PREDICTED ATMOSPHERIC TRANSPORT AND DEPOSITION OF I-131

The methodology outlined by the Nuclear Regulatory Commission (3) for estimating atmospheric transport and dispersion of I-131 released during normal operation of light water reactors was used to predict concentrations of radioiodine in the air and on vegetation in the Quad Cities Station environment. The revised curves describing plume depletion and deposition were used in the computations. The straight line airflow model (4,5) was employed with corrections for plume recirculation for a site in open terrain (6). Predicted concentrations for the tall chimney and the vent were summed, as described in Section 3, to obtain the total predicted concentration. The nomenclature used in this section is the same as that defined in Section 3.

Meteorological data routinely collected for the Quad Cities site by Murray and Trettel Inc. were used as input for the calculations of dispersion and deposition factors. Computations were made for each of the environmental monitoring locations established for the study. These monitoring locations and their coordinates are given in Table 4-3, together with the open terrain correction factors used in the calculations.


QUAD CITIES CHIMNEY
I-131 RELEASE RATE (nCi/sec)
4
2
10²
8
6
4
2
10
8
6
4
2
1
10
20
30
10
20
30
10
20
30
9
19
29
10
20
30
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER

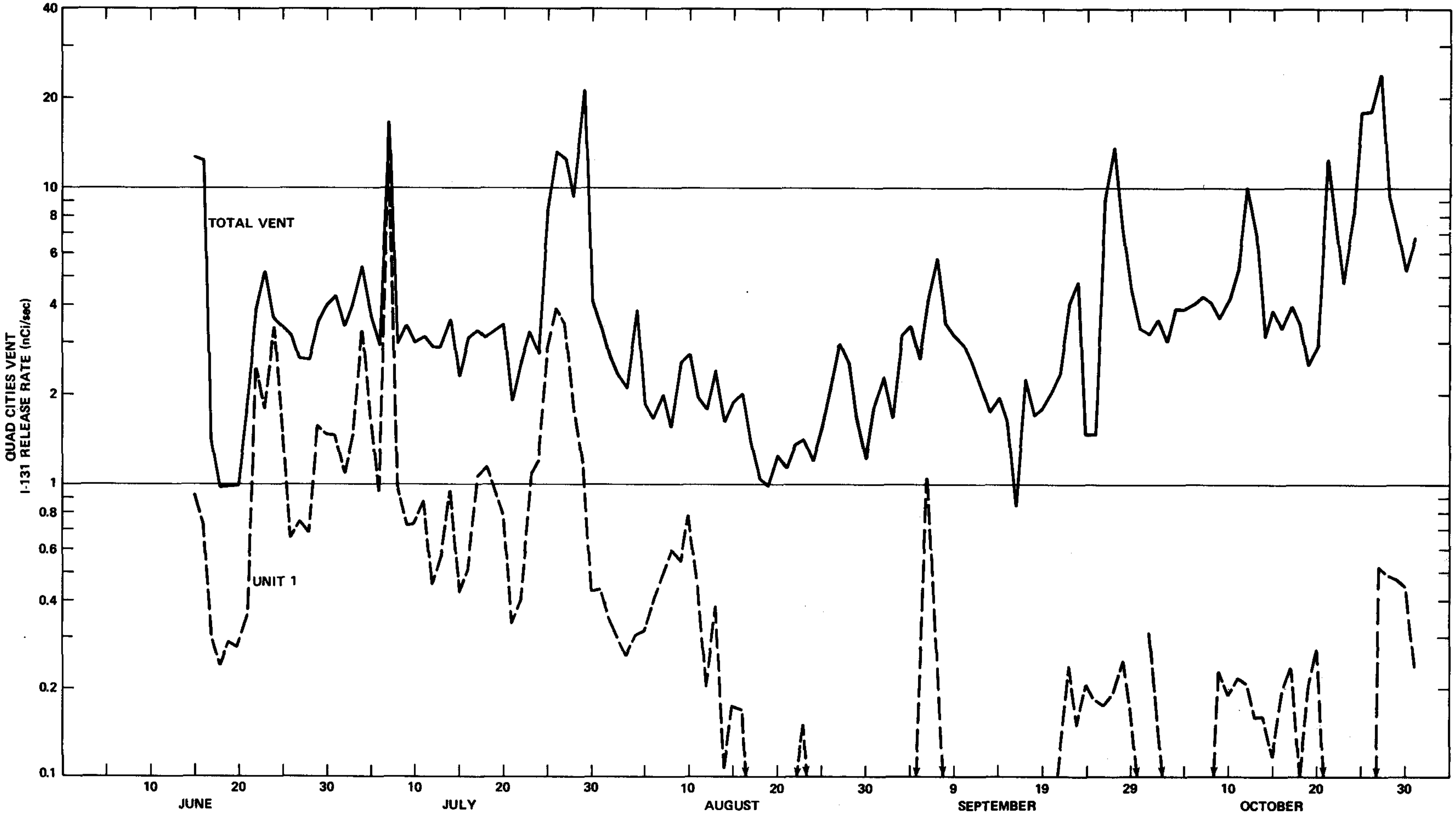

40
20
10
8
6
4
2
1
0.8
0.6
0.4
0.2
0.1
QUAD CITIES VENT
I-131 RELEASE RATE (nCi/sec)
TOTAL VENT
UNIT 1
10
20
30
10
20
30
10
20
30
9
19
29
10
20
30
JUNE
JULY
AUGUST
SEPTEMBER
OCTOBER

Table 4-2

AVERAGE I-131 RELEASE RATES DURING THE STUDY

Period	Average I-131 Release Rate (nCi/sec)[a] Main Chimney	Average I-131 Release Rate (nCi/sec)[a] Reactor Building Vent
1	27.5±7.1	4.3±1.4
2	31.1±3.5	4.6±1.1
3	27.5±3.9	4.6±1.6
4	28.4±3.3	4.2±1.0
5	8.7±0.8	7.5±2.1
6	10.4±1.6	2.1±0.2
7	25.2±1.6	1.4±0.1
8	20.7±9.3	2.2±0.2
9	8.1±1.6	3.1±0.4
10	13.3±2.1	2.3±0.4
11	29.1±15.4	5.7±1.7
12	17.9±7.1	4.4±0.5
13	43.5±16.5	5.1±1.1
14	53.3±20.5	12.7±2.5

(a) One standard deviation of the mean is given with the mean release rate.

Table 4-3

ENVIRONMENTAL SAMPLING LOCATIONS, QUAD CITIES

Site	Name	Location r(m)	Location θ[a]	Open Terrain Recirculation Factor
1	Commonwealth Edison	1450	173°	3.40
2	Decker Farm	8370	92°	1.13
3	Wherry Farm	4500	101°	1.49
4	Nitrin Plant	2720	42°	2.05
5	George Farm	4780	59°	1.44

(a) Direction from the plant; a location at 0° or 360° is due north of the plant, one at 90° is due east, and so on.

Predicted environmental concentrations of I-131 at the environmental sampling locations during the field monitoring periods are presented in Tables 4-4 through 4-8. (1)

Table 4-4

PREDICTED AIR CONCENTRATIONS FOR SITE #1

COMMONWEALTH EDISON

Period	Predicted Air Concentrations (fCi/m^3) Particulate (X_1)	I_2+HOI (X_{23})	Organic Iodides (X_4)
1	0.14	1.4	0.48
2	0.23	2.0	0.67
3	0.091	1.3	1.4
4	0.20	1.1	0.68
5	0.011	0.90	0.50
6	0.0064	0.068	0.017
7	0.84	3.8	0.71
8	0.053	0.74	0.24
9	0.012	1.2	0.69
10	0.0090	0.23	0.20

Table 4-5

PREDICTED AIR CONCENTRATIONS FOR SITE #2

DECKER FARM

Period	Predicted Air Concentrations (fCi/m^3) Particulate (X_1)	I_2+HOI (X_{23})	Organic Iodides (X_4)
1	0.034	0.33	0.11
2	0.078	0.67	0.24
3	0.11	0.35	0.14
4	0.019	0.11	0.040
5	0.0021	0.15	0.092
6	0.092	0.22	0.060
7	0.11	0.45	0.057
8	0.0061	0.26	0.077
9	0.0044	0.11	0.052
10	0.0068	0.15	0.085

Table 4-6

PREDICTED AIR CONCENTRATIONS FOR SITE #3

WHERRY FARM

	Predicted Air Concentrations (fCi/m^3)		
Period	Particulate (X_1)	I_2+HOI (X_{23})	Organic Iodides (X_4)
1	0.072	0.72	0.25
2	0.095	0.91	0.43
3	0.068	0.44	0.25
4	0.015	0.085	0.040
5	0.0061	0.45	0.27
6	0.038	0.41	0.10
7	0.21	0.87	0.13
8	0.044	0.71	0.22
9	0.014	0.32	0.14
10	0.0082	0.18	0.11

Table 4-7

PREDICTED AIR CONCENTRATIONS FOR SITE #4

NITRIN PLANT

Period	Predicted Air Concentrations (fCi/m^3) Particulate (X_1)	 I_2+HOI (X_{23})	 Organic Iodides (X_4)
1	0.20	1.9	0.63
2	0.23	2.6	1.7
3	0.12	0.81	0.49
4	0.18	0.99	0.55
5	0.015	0.97	0.54
6	0.16	1.7	0.45
7	0.26	1.2	0.26
8	0.077	1.1	0.36
9	0.0057	0.34	0.19
10	0.0041	0.098	0.083

Table 4-8

PREDICTED AIR CONCENTRATIONS FOR SITE #5

GEORGE FARM

Period	Predicted Air Concentrations (fCi/m^3) Particulate (X_1)	 I_2+HOI (X_{23})	 Organic Iodides (X_4)
1	0.088	0.90	0.30
2	0.12	1.1	0.49
3	0.20	0.60	0.25
4	0.12	0.68	0.29
5	0.0041	0.27	0.16
6	0.073	0.76	0.20
7	0.16	0.69	0.11
8	0.026	0.41	0.13
9	0.0029	0.19	0.10
10	0.014	0.32	0.18

I-131 CONCENTRATIONS IN THE ATMOSPHERE

The environmental air sampling program and the measurement results are described in this section. The sampling locations were given in Table 4-4 in the previous section. The air sampling equipment was the same as that described in Section 3 and the same environmental sampling media were employed. More measurements using cadmium iodide beds were made at Quad Cities. This permitted better definition of the inorganic forms. The analytical procedures used were the same as those used for the Oyster Creek samples and are described in Appendices B-E. Sampling was initiated at all five locations in June and was continued until the arrival of fallout from nuclear weapons testing in late September. At Site #2, the Decker Farm, problems with the electrical power supply for the sampling equipment resulted in the loss of the first four samples at that location.

Measured Concentrations of I-131 in Air

The measured concentrations of I-131 in various forms are given in Tables 4-9 through 4-13. The 1-sigma counting uncertainties are included in the tables for the detectable concentrations. If the net I-131 activity was less than the 2-sigma counting uncertainty, a "less than" value is given. The designations used for measured concentrations in Section 3 are also employed here.

Several features of the data should be noted. Radioiodine associated with particulates was detected during only one period prior to the arrival of weapons testing fallout. In Period 7, I-131 associated with particulates was detected at the two locations northeast of the plant. The absence of particulate activity attributable to plant releases was again strongly contrasted with the fallout radioiodine. The samples analyzed by ACC (4-1) show that during Periods 11 and 12 the mean ratio ϕ_1/ϕ_T was 0.55, which is comparable to that measured in 1976 at Oyster Creek.

A second notable aspect of these results is the detection of HOI in the environment, during Period 7 at Site #1 and probably during Period 8 at Site #5. The ratio ϕ_3/ϕ_{23} was 0.17 at Site #1 during Period 7. As noted, the organic iodide component

Table 4-9

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #1

COMMONWEALTH EDISON

Period	Measured Concentration (±1σ, fCi/m^3) of I-131		
	ϕ_1	ϕ_{23}	ϕ_4
1	<0.30	NA(a)	<0.33
2	<0.30	2.35±.52	<0.26
3	<0.48	2.23±.83	<0.50
4	<0.26	<0.71	<0.23
5	<0.42	3.05±.34	0.12±.05
6	<0.39	NA	<0.58
7	<0.49	(b)	(b)
8	<5.1	<6.7	5.54±.56
9	<0.49	(c)	(c)
10	<0.46	2.13±.35	2.76±.15
11(d)	27±5	8.1±1.5	15±3
12(d)	15±3	6.4±1.7	6.2 ±1.9
13(d)	<4.4	<3.0	<7.6

(a) NA means no analysis or sample lost during analysis; the same designation is used in subsequent tables.

(b) ϕ_2=2.03±.23; ϕ_3=0.42±.15; ϕ_4 was not measured.

(c) ϕ_2<0.50; ϕ_3 <0.56; ϕ_4 was not measured.

(d) These samples were collected and analyzed by Allied Chemical Corporation (<u>1</u>).

Table 4-10

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #2

DECKER FARM

Period	Measured Concentration (±1σ, fCi/m^3) of I-131		
	ϕ_1	ϕ_{23}	ϕ_4
1		Power Failure	
2		Power Failure	
3		Power Failure	
4		Power Failure	
5	<0.61	<0.90	0.25
6	<0.54	<0.60	<2.0
7	<0.53	<0.58	2.45±.46
8	<0.63	(a)	(a)
9	<0.60	<0.57	<0.84
10	<2.1	<2.6	<1.8
11 (b)	32±7	12 ±3	15 ±3
12 (b)	22±5	6.8±1.6	8.8±2.0
13 (b)	<4.8	<4.2	<5.4

(a) ϕ_2 <0.32; ϕ_{34}=1.00±.32.

(b) These samples were collected and analyzed by Allied Chemical Corporation (1).

Table 4-11

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #3

WHERRY FARM

Period	Measured Concentration (±1σ, fCi/m^3) of I-131		
	ϕ_1	ϕ_{23}	ϕ_4
1	<0.86	NA	<0.88
2	<0.65	1.28±.59	<0.41
3	<0.59	<1.5	<1.7
4	<0.45	<0.74	<1.9
5		Sampler Failed	
6	<0.42	<0.53	0.27±.08
7	<0.50	<0.75	3.89±.14
8	<0.51	<0.47	1.16±.30
9	<0.54	<0.56	<0.78
10		Sampler Failed	

Table 4-12

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #4

NITRIN PLANT

	Measured Concentration (±1σ, fCi/m^3) of I-131		
Period	ϕ_1	ϕ_{23}	ϕ_4
1	<0.70	NA	<0.50
2	<0.46	1.72±.55	<0.32
3	<0.53	2.03±.76	<0.82
4	<0.43	<0.72	<0.54
5	<0.44	<0.57	<0.95
6	<0.36	<1.1 (a)	(a)
7	1.38±.25	(b)	(b)
8	<0.47	(c)	(c)
9	<0.51	(d)	(d)
10	<0.48	(e)	(e)

(a) $\phi_2 < 0.42$; $\phi_3 < 0.61$; ϕ_4 was not measured.

(b) $\phi_2 = 1.13 \pm .24$; $\phi_{34} = 3.17 \pm .46$.

(c) $\phi_2 = 0.57 \pm .17$; $\phi_{34} = 0.90 \pm .25$.

(d) $\phi_2 < 0.40$; $\phi_{34} < 0.72$.

(e) $\phi_2 < 0.41$; $\phi_{34} = 1.37 \pm .36$.

Table 4-13

MEASURED CONCENTRATIONS OF I-131 IN AIR AT SITE #5

GEORGE FARM

Period	Measured Concentration ($\pm 1\sigma$, fCi/m^3) of I-131		
	ϕ_1	ϕ_{23}	ϕ_4
1	<0.44	NA	<0.45
2	<0.36	1.63±.53	<0.63
3	<0.58	<1.4	<0.85
4	<0.60	<0.81	<1.2
5	<0.49	<0.72	<0.45
6	<0.41	5.19±.37	<0.87
7	(a)	(a)	(a)
8	<0.53	<0.54 (b)	<0.57 (b)
9	<0.56	(c)	(c)
10	<0.52	(d)	(d)

(a) Sample volume could not be determined. The I-131 activity (A) fractionation was: A_1=0.10; A_2=0.22; A_{34}=0.68.

(b) Two samplers were operated to obtain more detail. The results were ϕ_2 <0.36; ϕ_{23} <0.54; ϕ_{34}=0.86±.28; ϕ_4 <0.57.

(c) ϕ_2 <0.36; ϕ_{34} <0.79.

(d) ϕ_2 <0.44; ϕ_{34}=0.98±.38.

was not measured so the overall gaseous iodine distribution is not known for that period. The possible range of the ratio $\phi_3/(\phi_2+\phi_{34})$ for Site #5 during Period 8 was 0.15--0.44. Eleven samples were taken, principally at Sites #4 and #5, which divided the total gaseous iodine into elemental and non-elemental components. Nine of these measurements yielded detectable amounts of non-elemental iodine; elemental iodine was detected in three samples. The mean of the nine ratios $\phi_2/(\phi_2+\phi_{34})$ lies between 0.13 and 0.28; the maximum observed ratio $\phi_2/(\phi_2+\phi_{34})$ was 0.39.

COMPARISON OF MEASURED AND PREDICTED AIR CONCENTRATIONS

The ratios of the measured to predicted concentrations of total I-131, ϕ_T/X_T, are shown in Table 4-15. As was the case with the Oyster Creek comparisons, most of the ϕ_T/X_T ratios are indeterminate because one or more of the measured concentrations was below the detection limit. The range of possible values of the ratio is given when the ratio is indeterminate. However, the ratio of the long term average values $(\bar{\phi}_T/\bar{X}_T)$ indicates that the model generally underpredicts the concentration for the 8- to 12-week averaging periods. The minimum $(\bar{\phi}_T/\bar{X}_T)$ was less than one for only one site (#4) and ranged from 0.9--2.8. Correction factors for open terrain recirculation were included in the calculation of X_T (See Table 4-4). If these factors are not included, the minimum ratios $(\bar{\phi}_T/\bar{X}_T)$ are 5.4, 3.2, 1.8, and 2.0 for sites #1 through #5 respectively. The noble gas measurements reported by Keller *et al.* (1) also indicated that recirculation of the plume occurred as did earlier measurements at the Quad Cities plant (2).

None of the data on radioiodine species provides more insight into the period to period variability of measured to predicted concentrations. The values of ϕ_1 are nearly all below the detection limit, which is generally greater than X_1, so the ϕ_1/X_1 ratios are not particularly meaningful. The ratios ϕ_{23}/X_{23} range from <0.59 to <17; the highest value for a positive determination of ϕ_{23} was 9.3 (Site #1, Period 10). Too few data points are available to determine a median ratio for each site; however, the median of all the values appears to lie between 1 and 2. This is lower than the median of all ratios ϕ_T/X_T, which lies between 3 and 4. The range of ratios ϕ_4/X_4 was from 0.19 to 78. The median of all values was approximately 4.

As noted earlier, there were 7 measurements which permitted evaluation of $\phi_2/(\phi_2/\phi_{34})$. The measured ratios can be compared with those predicted at the monitoring locations, $X_2/(X_2+X_{34})$. The average ratio of measured to predicted values $[\phi_2/(\phi_2+\phi_{34})]/[X_2/(X_2+X_{34})]$ lies between 0.25 and 0.67 for the seven measurements. This is consistent with the lower ratios of ϕ_{23}/X_{23} and suggests the possible transformation of I_2 to less reactive species.

Table 4-14

RATIOS OF MEASURED TO PREDICTED CONCENTRATIONS OF TOTAL I-131

Period	$\phi_T(\text{fCi/m}^3)$ / $X_T(\text{fCi/m}^3)$ Site #1	Site #2	Site #3	Site #4	Site #5
1	--	--	--	--	--
2	0.82--1.0	--	0.89--1.6	0.38--0.55	0.93--1.5
3	0.71--1.0	--	0.0--5.0	1.4--2.4	0.0--2.7
4	0.0--0.59	--	0.0--22	0.0--0.98	0.0--2.4
5	2.3--2.6	0.0--7.3	--	0.0--1.3	0.0--3.9
6	--	0.0--9.0	0.49--2.8	--	5.0--6.3
7	--	7.2--9.0	3.2--4.3	3.3	--
8	5.3--17	2.9--5.7	1.2--2.2	0.94--1.2	1.5--3.1
9	--	0.0--12	0.0--4.0	0.0--3.0	0.0--5.9
10	11--12	0.0--27	--	7.6--13	1.8--5.5
2-10[(a)]	1.6--3.0	2.8--11	1.2--3.5	0.87--1.5	1.4--3.3
2-10[(b)]	5.4--10	3.2--12	1.8--5.2	1.8--3.1	2.0--4.8

(a) Ratio of the time-weighted average concentrations $(\bar{\phi}_T/\bar{X}_T)$ (Tables 4-5 --4-9) including all periods when measurement data were available. Averaging periods ranged from 59 to 80 days.

(b) Ratio of the average time-weighted average concentration $(\bar{\phi}_T/\bar{X}_T)$ to the average concentration computed using single station windrose technique with correction for open terrain recirculation.

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Section 5

MEASUREMENTS OF STABLE IODINE SPECIES

BACKGROUND

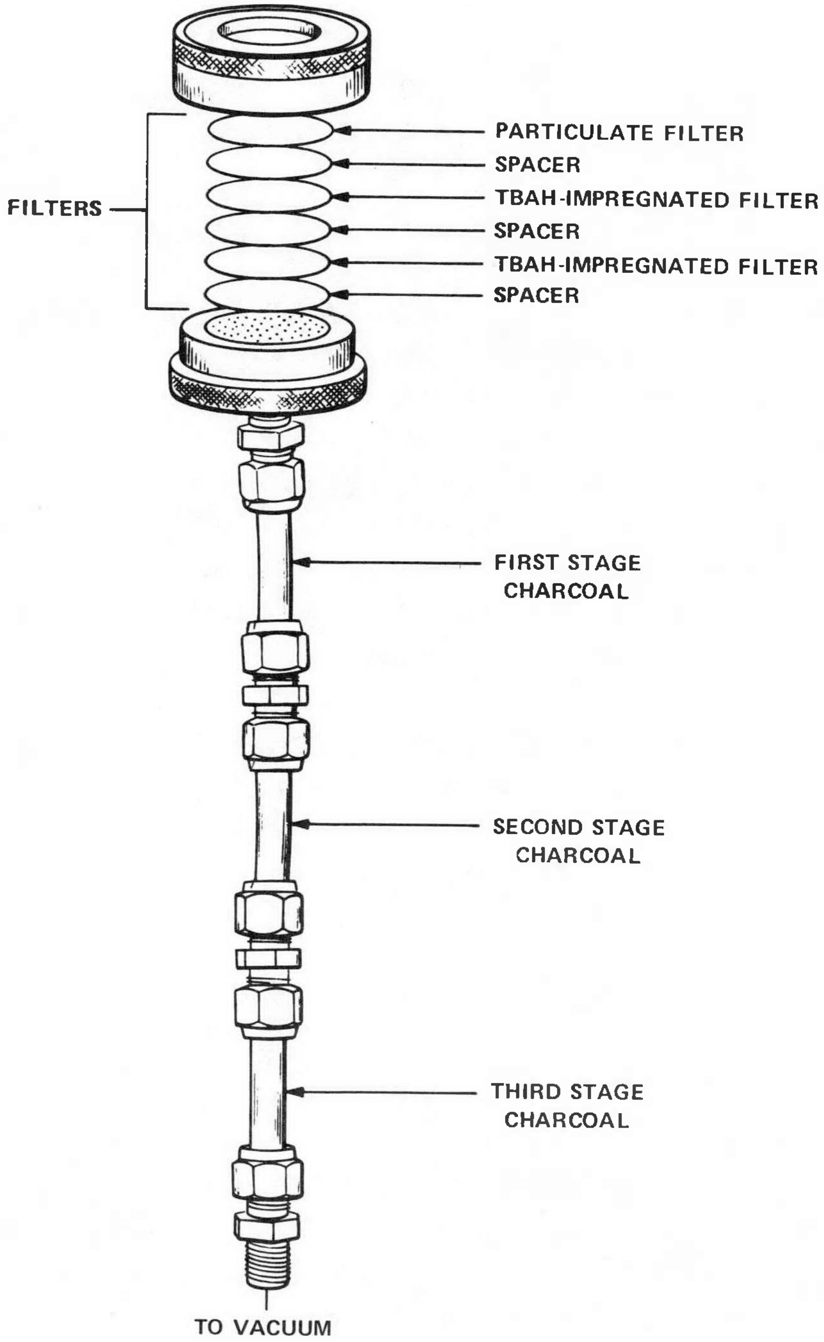
It is not currently possible to obtain stable iodine species data which are comparable to those from the radioiodine effluent species sampler. Indeed, since HOI is partially identified by its specific reaction with 4-iodophenol (1), it may only be possible to measure radiolabeled HOI. However, the sampling system reported by Moyers and Duce (2) and Rahn et al. (3) does permit a breakdown of stable iodine species into categories comparable to those obtained using the radioiodine field sampler with a particulate filter, IPh bed, and charcoal bed in sequence. The resultant species can be characterized as iodine associated with particulates, inorganic forms, and organic forms.

It was desirable to use a stable iodine system which could be used to obtain reliable samples containing detectable concentrations of stable iodine collected during sampling periods of 7-10 days. The required materials and sample analysis were provided by the University of Arizona Analytical Center (UAAC) under the direction of Dr. Jarvis L. Moyers. The detection limit for the neutron activation analysis technique used was 1-5 ng of iodine. The next section discusses the design and testing of the sampling system.

SAMPLING APPARATUS AND MEDIA PERFORMANCE

The stable iodine sampling apparatus is shown in Figure 5-1. It consists of a 47-mm openfaced aluminum filter holder, which contains the particulate and inorganic iodine filters and the Teflon^R filters used as spacers, and a series of three 0.5-gram activated charcoal beds packed in 3/8" o.d. fluorocarbon tubing. Air was drawn through the sampling apparatus by a small sampling pump.

The preparation of the sampling media is described in detail in Appendix K. Polycarbonate filters with a pore size of 0.4 μm were used to collect iodine associated with particulate materials. The two inorganic iodine gas collectors were cellulose filters which had been impregnated with tetrabutyl ammonium hydroxide (TBAH). The charcoal beds used were prepared from activated coconut shell charcoal (8-12 mesh).


PARTICULATE FILTER
SPACER
TBAH-IMPREGNATED FILTER
SPACER
TBAH-IMPREGNATED FILTER
SPACER
FILTERS
FIRST STAGE CHARCOAL
SECOND STAGE CHARCOAL
THIRD STAGE CHARCOAL
TO VACUUM

The polycarbonate filters were found in laboratory experiments by Rahn et al. (4) to have a very small collection efficiency for I_2, 0.4%. The efficiency for collection of HI was given as 14% and that for CH_3I was 0.01%. The only filter tested by them having a lower I_2 collection efficiency (0.06%) was the Delbag Microsorban filter. The same authors reported the results of field testing with multiple TBAH-impregnated filters in series. The first TBAH filter was found to contain slightly more than 97% of the total iodine collected on the TBAH-impregnated filters during a 1-week exposure period. Reference (4) also provides information on CH_3I collection by the activated charcoal. The data show that penetration does occur at higher flow rates (i.e. 20-30 lpm). The authors suggest methods for evaluating the extent of penetration and correction techniques to analytically account for this.

To evaluate the penetration of organic iodine through the activated charcoal, chromatographic tests were designed to determine the retention volume of methyl iodide on the activated charcoal to be used. These tests were conducted at the UAAC according to the procedure described by Butler and Burke (5). Retention volumes of CH_3I on activated charcoal were measured as a function of temperature which allowed evaluation of the thermodynamic properties of the gas-surface interaction and the determination of the volume required for penetration under field sampling conditions. Once these volumes were determined, frontal analysis chromatography was used to check the suitability of the selected sampling rates and volumes. A CH_3I permeation tube was used to generate known concentrations in air which passed through the charcoal sampling system and into an electron capture detector. No breakthrough of CH_3I was seen in the sampling volumes selected for this work. The tests suggested an optimum sampling volume of 10 m^3 per gram of charcoal. As a result of limitations in sample volumes for the analytical work (neutron activation analysis), $\sim$10 m^3 was settled upon for the field work. Field evaluation of organic iodide was made possible by division of the charcoal into three sections which were analyzed individually.

Additional evaluations of the filtration media were conducted using I-131 and the filter testing apparatus described by Emel et al. (6). Filters were exposed to radioiodine in various forms for a period of $\sim$2 hours and were then purged with laboratory air for 1 week to simulate field exposure. As in the laboratory tests reported in Reference (4) the stable iodine concentrations were $\sim 10^3$ greater than those expected in the field sampling locations. The expected field collection efficiencies for I_2 were <10% for the particulate filters 92% for the TBAH-impregnated filters. The initial collection efficiency of the TBAH-impregnated filters for HOI was 75%; however, approximately 23% was removed during the 1-week purge period. The collection efficiency of the TBAH-impregnated filters for methyl iodide was found to be less than 2%. Less than 5% of the methyl iodide-131 penetrated the three fresh beds of charcoal used to collect organic iodides. This was also found to be true for beds which had been purged for 1 week prior to testing with I-131. However, in both cases the I-131 activity was found in all three beds, indicating penetration through the beds. This result may be due to the high methyl iodide mass level used in the testing compared with ambient concentrations for which the system was designed.

SAMPLING AND ANALYTICAL METHODS

Stable iodine samplers were operated at four different locations during the course of the study. Both shore Island Beach State Park #6) and inland Garden State Parkway, #4) locations were sampled near the Oyster Creek plant. The sampling location near the Quad Cities station was the Commonwealth Edison site (#1), about 1.5 km south of the plant. The fourth sampling location was located about 2440 meters above sea level, along the front range of the Rocky Mountains west of Golden, Colorado. The Illinois and Colorado sampling locations were operated together (with a two-day sampling time offset), in an attempt to detect differences in iodine species for these two continental locations.

At all locations the sampling apparatus was located about 1 meter above the ground. The apparatus was connected to the pump with thick walled vacuum tubing. A critical orifice was used to provide a constant flow rate of about 1 liter per minute. The sampling apparatus was placed in a cylindrical open ended weather shield during operation in the field and the pump was enclosed in a ventilated metal weather shield. The procedures for loading, exchange and unloading of the sampling apparatus were described in Appendix L.

As indicated previously, the samples were analyzed using neutron activation analysis. The detailed analytical procedures are presented in Appendix M.

RESULTS OF MEASUREMENTS IN OYSTER CREEK ENVIRONS

Table 5-1 contains the results of the stable iodine measurements at the two locations near the Oyster Creek plant. Most of the measurements were made at Site #6, between the Atlantic Ocean and Barnegat Bay. Three samples were obtained at Site #4, approximately 10 km from the coastline.

Table 5-1

STABLE IODINE CONCENTRATIONS NEAR THE OYSTER CREEK NUCLEAR GENERATING STATION

Location	Sampling Period	Sample Volume (m^3)	Stable Iodine Concentration (ng/m^3)		
				Gaseous Forms	
			Particulate	Inorganic	Organic
#6	14	6.0	0.8 ±0.1	<3	(a)
	15	6.0	4.5 ±0.4	3.3±1.5	4.0±1.0
	16	6.1	4.3 ±0.4	6.7±1.5	3.3±1.0
	17	11.1	2.0 ±0.2	(a)	(a)
	18	10.6	1.5 ±0.1	1.7 ±.5	<1
	19	11.1	0.42±0.05	<1	1.8±0.4
	20	10.9	0.58±0.05	<1.5	<1
	21	11.1	0.62±0.05	5.9±0.9	2.3±0.4
	22	11.1	1.2 ±0.1	3.3±0.9	<0.8
	23	22.3	0.71±0.07	4.5±0.5	3.3±0.3
#4	21	6.0	1.1 ±0.1	<2	<2
	22	6.0	0.20±0.05	<2	<1
	23	12.1	0.61±0.05	<1	2.8±0.5

(a) Sample lost.

As was stated earlier, the organic iodine collector was divided into three stages to check penetration through charcoal bed. With one exception, iodine was only found on the first sampling stage. The one case which did show penetration to the second charcoal stage (but not to the third) was the 22-m^3 sample collected over a two-week period (23) at location #6. For this sample, the second charcoal collector contained approximately 40% of the total organic iodine. These data (although limited) suggest that the selected sampling volumes are perhaps close to the optimum for the flow rate and sampling time used and the environmental conditions encountered in this program.

For the inorganic iodine sampler apparent penetration to the second collector was only observed for the first three samples collected at Site #6. For all other samples the amount of iodine on the second TBAH-impregnated filter was within one standard deviation of the blank value. At the present time, it is uncertain if this apparent penetration is the result of a change in the distribution of iodine species (possibly the presence of HOI) or the result of some unknown contamination. It is pointed out, however, that these first three samples were stored in double plastic bags, whereas the remaining samples were packaged and stored in strict accordance with the procedure described in Appendix L. It was assumed that the iodine found on the second filter was due to sample contamination. The tabulated concentrations are those calculated on the assumption that equal contamination of both filters has occurred.

The particulate stable iodine concentration (ϕ_p) averaged 1.7 ng/m^3 at Site #6 and 0.64 ng/m^3 at Site #4; however, the variability in the concentration is large at both locations. There is not a significant difference in ϕ_p at the two locations nor is there any indication of a systematic relationship between them.

The mean ratio of the particulate component to the total stable iodine concentration (ϕ_p/ϕ_t) was 0.23-0.26 at the beach location and was indeterminate, between 0.23 and 0.41, at the inland location. The inorganic iodine component ϕ_i was generally predominant at Site #6; the average concentration was 3.2-3.3 ng/m^3. The mean value of ϕ_i for location #4 lies between 0 and 1.7 ng/m^3 and there appears to be a significant difference between the two locations. Considering the residence times suggested by Rahn _et al_. (_3_) for the three iodine species (particulate, 14 days; inorganic gas, 10 days; and organic gas, 17 days) and the proximity of the two sampling locations, one would not expect to see large changes in either the absolute or relative species concentrations at these sites. It is possible that the shore location is a source for the inorganic gaseous iodine.

The organic iodide concentration ϕ_o averaged 1.9-2.0 ng/m^3 at Site #6 and 1.5-1.8 ng/m^3 at Site #4 and there was no significant difference between locations. The organic iodide to total gaseous iodine ration $\phi_o/(\phi_i+\phi_o)$ was larger at the inland location owing to the small amounts of inorganic iodine. The mean ratios for the two locations can only be bounded (0.32-0.41 for Site #6 and 0.34-0.59 for Site #4) and are not significantly different.

The stable iodine species concentrations measured in the Oyster Creek environs are in close agreement with those reported for New York City (4). Both the particulate and gaseous iodine concentrations are relatively low compared to those measured at other coastal or marine locations. These results may suggest interactions between the iodine species and pollutant materials which effectively reduce the residence times of iodine species in the atmosphere.

RESULTS OF MEASUREMENTS IN QUAD CITIES AND COLORADO FRONT RANGE ENVIRONMENTS

The results of stable iodine concentration measurements at the Commonwealth Site south of the Quad Cities Nuclear Power Station and at the Robinson Hill, Colorado sampling location in the front range of the Rocky Mountains are presented in Table 5-2. As indicated previously the Colorado location was selected to obtain stable iodine species data for an air mass which could subsequently be transported to the Quad Cities plant site in western Illinois. However, the Quad Cities environs was most frequently influenced by air parcels from the south during the sampling period, so the two sets of samples were largely independent.

The mean concentrations of each species were very similar for the two locations and the small differences between them are not statistically significant. The mean particulate concentration, ϕ_p, was 2.9 ng/m^3 near the Quad Cities Plant and 2.8 ng/m^3 at the Colorado location. The mean of the ratio ϕ_p/ϕ_t was between 0.18 and 0.20 in Illinois and between 0.15 and 0.18 in Colorado. Inorganic iodine species were detected in only two of twelve samples. The estimates of the mean concentration are 1.1-2.2 ng/m^3 for the Commonwealth Edison Site and 0-2 ng/m^3 for the Robinson Hill, Colorado location. Organic iodide concentrations predominated at both locations averaging 14.3 and 14.2 ng/m^3 in Illinois and Colorado, respectively. The ratio $\phi_o/(\phi_o+\phi_i)$ was correspondingly high at both locations; the average value was between 0.71 and 0.74 at Quad Cities and between 0.73 and 0.82 in Colorado.

The measured organic iodide concentrations were much higher in Colorado and Illinois than those measured in New Jersey nine months earlier. The analytical detection

Table 5-2

STABLE IODINE CONCENTRATIONS NEAR THE QUAD CITIES NPS AND
AT THE FRONT RANGE OF THE ROCKY MOUNTAINS IN COLORADO

Location	Sampling Period	Sample Volume (m^3)	Stable Iodide Concentrations (ng/m^3)		
				Gaseous Forms	
			Particulate	Inorganic	Organic
QC #1	6-15/6-30	23.7	5.3±0.5	<2	7±1
	6-30/7-7	(a)	--	--	--
	7-7 /7-14	11.8(b)	1.9±0.2	<2	(c)
	7-15/7-25	16.6	2.7±0.3	<4	23±2
	7-25/8-4	15.4	1.1±0.1	<2	13±2
	8-4 /8-15	17.8	3.3±0.3	<3	14±2
Colorado	6-15/6-25	8.6(b)	10±1(d)	54±2(d)	116±12(d)
	6-25/7-4	7.8(b)	1.7±0.2	<2	13±3
	7-4 /7-12	6.9	3.3±0.3	<2	15±3
	7-12/7-23	9.1	2.8±0.3	<2	24±2
	7-23/8-2	9.0	3.7±0.4	<2	8±2
	8-2 /8-13	9.1	2.7±0.3	<2	11±2

(a) Sampler disturbed; volume unknown. Quantities of stable iodine found in sampling media were: particulate, 19±2 ng; inorganic gas, <20 ng; and organic gas, 80±8 ng.

(b) Some flow rate uncertainty for these samples.

(c) Sample lost.

(d) The unusually high iodine concentrations for this sample indicate either sample contamination or a local source of airborne iodine. Results for this sample are not included in averages or the evaluation of results as discussed in text.

limits for the inorganic iodine concentration are close to the values measured in New Jersey, so, while the Island Beach State Park concentrations are higher, it is not clear whether the differences are statistically significant. The average particulate concentrations are slightly higher for the inland samples but within the range of values seen for the New Jersey samples.

It is also noted that the particulate and organic iodine species values obtained for the Colorado and Illinois samples are in close agreement with the results for Kansas (3) (see Table 2-1). The apparent elevations of organic iodine in inland locations relative to that seen for coastal regions is somewhat surprising. This may suggest an inland source of organic iodine high relative to the normal marine source; however, such a suggestion is contrary to the generally accepted view of the sources of atmospheric iodine. The inorganic gaseous iodine is lower than for most previously reported values (3,4). The reason for this difference is not known. It is possible that the differences are within the normal variations of the gaseous inorganic iodine concentrations since the geometric standard deviation for inorganic iodine concentration is large (3). It is also possible that variations in the sampling flow rate and duration may affect the observed species distributions.

TROPOSPHERIC RESIDENCE TIMES FOR IODINE SPECIES

Presuming that the expression developed by Junge (7) applies, one can estimate the tropospheric residence times of the airborne iodine species. Junge's relationship between the residence time (τ) and relative standard deviation (σ_r) of the concentration is:

$$\tau \, \sigma_r = 0.14 \text{ yr} = 51 \text{ days} \qquad (5\text{-}1)$$

The relative standard deviation (σ_r) is the ratio of the sample standard deviation (σ) to the mean concentration:

$$\sigma_r = \sigma/\bar{\phi}.$$

Rahn et al. (3) employed the log-normal distribution to describe the highly variable tropospheric halogen concentrations. Median concentrations ($\hat{\phi}$) and the associated geometric standard deviations (σ_g) were determined by them and the values of σ_g were used in Equation (5-1) to estimate the residence times of particulate iodine, inorganic iodine gas, and organic iodide gas.

The data from the two locations in the Oyster Creek environs (September-November, 1976) were treated as one data set and the June-August, 1977 results for western Illinois and the Colorado Front Range were combined to form a second data set. The iodine data were found to fit log-normal distributions; medians and geometric standard deviations were determined graphically. The upper portion of Table 5-3 contains the number of measurements (N), the parameters for each distribution, and the residence times estimated using σ_g. Also shown are the residence time estimates obtained by Rahn et al. using 16 measurements from Arizona, Bermuda, Kansas, and the Northwest Territories (June-October, 1974).

Because the inorganic iodine concentration was nearly always below the detection limit in the 1977 samples, it was not possible to estimate the median concentration or geometric standard deviation. The inorganic iodine residence time in the Oyster Creek environs was more than double that estimated by Rahn et al. While the particulate iodine and organic iodine gas residence times estimated using the Oyster Creek data are comparable to those of Reference (3), they are somewhat less than half the corresponding residence times for those species estimated using the 1977 data from Illinois and Colorado. The later locations are both considered relatively remote with low levels of atmospheric pollution and are in that sense similar to the locations of Reference (3). The potential interaction of air pollutants with iodine species has been mentioned as a possible reason for the low iodine concentrations in New Jersey and New York City.

In the lower portion of Table 5-3 are the residence times obtained using Equation (5-1) as originally presented. The relative standard deviations of the concentration were small, all within the range 0.4--1.0. The residence times computed are 2--4 times larger than those computed using σ_g. They are also large with respect to (a) expectations based on previous particle size measurements and residence time correlations (3,8) and (b) expectations based on photochemical calculations (9), if the "organic gas" component label is chemically accurate. (Alternative labels and associated residence times are at this point highly speculative). If the true residence times are comparable to previous expectations and computations using σ_g, then the average sampling time employed in this study was a significant fraction of the residence time and the results are inappropriate for such residence time calculations. To evaluate the possible impact of sampling time on the result, other data were examined and used to compute residence times in a similar manner. The particulate and total gaseous iodine data for the Antarctic sites and for Hawaii (10,11,12) were obtained using sampling periods (<24 hours) which are short with respect to the previous residence time estimates. Table 5-4 contains the

Table 5-3

DISTRIBUTION PARAMETERS AND RESIDENCE TIME ESTIMATES FOR STABLE IODINE SPECIES

	Oyster Creek, New Jersey (1976)							Quad Cities, Illinois and Front Range, Colorado (1977)	
Parameter	Particulate	Inorganic Gas		Organic Gas		Total Gas		Particulate	Organic Gas
RESIDENCE TIMES COMPUTED USING GEOMETRIC STANDARD DEVIATION									
N	13	13		11		11		10	9
$\hat{\phi}$ (ng/m^3)	1.0	2.4		1.5		3.8		2.5	13
σ_g	3.8	2.0		3.7		2.7		1.5	1.5
τ (days	13	26		14		19		34	34
Estimates From Reference (3) Using σ_g									
τ (days	14	10		18		17			
RESIDENCE TIMES COMPUTED USING RELATIVE STANDARD DEVIATION									
N	13	12		11		11		10	9
		(a)	(b)	(a)	(b)	(a)	(b)		
$\bar{\phi}$ (ng/m^3)	1.4	2.4	2.9	1.8	2.0	5.6	6.3	2.9	14.2
σ (ng/m^3)	1.4	2.4	1.9	1.5	1.3	4.5	4.0	1.2	5.9
σ_r	1.0	1.0	0.68	0.83	0.64	0.81	0.62	0.41	0.41
τ (days	51	51 -- 75		62--80		63--82		120	120

(a) Computed assuming all undetectable concentrations were zero.

(b) Computed assuming all undetectable concentrations were just below the detection limit.

relative standard deviations and residence time estimates for particulate and gaseous iodine based on those data.

Table 5-4

ESTIMATED RESIDENCE TIMES FOR PARTICULATE AND
GASEOUS IODINE USING DATA FROM HAWAII AND ANTARCTICA

Location	Particulate			Total Gas		
	N	σ_r	τ	N	σ_r	τ
Antarctica						
McMurdo		0.42	120		0.27	190
South Pole		0.24	210		0.30	170
Hawaii	10	0.42	120	59	0.43	120

Recent data on methyl iodide concentrations in Yosemite (13) indicate a residence time of 120 days for that species, which is in close agreement with the total gas residence times in Table 5-4 and the organic gas residence times in the lower portion of Table 5-3. The sampling times were, in all three cases, much shorter than the estimated residence time.

The data obtained by Lovelock *et al.* (14) and the east coast data of Lillian *et al.* (15,16,17) may not be usable for residence time estimates because of the proximity of some of the sampling locations to natural and anthrogenic sources of methyl iodide. However, it would be of interest to examine the data obtained by Lillian *et al.* at locations remote from possible industrial sources.

If Junge's relationship can be applied to atmospheric iodine species and the limited data available are representative of the global distribution and temporal concentration variations, then (a) the residence time of methyl iodide is much longer than suggested by photochemical considerations and (b) the organic iodine gas component is probably largely methyl iodide.

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APPENDIX A

MONITORING OF RADIOIODINE SPECIES IN GASEOUS EFFLUENTS

DESCRIPTION OF RADIOIODINE SPECIES SAMPLER

The chemical forms of I-131 discharged to the atmosphere were monitored continuously in gaseous effluent streams. Sampling periods ranged from 7 to 14 days. The sampling train consisted of (1) a high-efficiency particulate filter, (2) a bed of cadmium iodide (CdI_2) with excess I, adsorbed on Chromosorb-P, (3) a bed of 4-iodophenol (IPh) adsorbed on alumina, and (4) two beds of TEDA-impregnated activated charcoal or silver zeolite (AgX). The sampler is designed to separate the I-131 in the air stream into four components: particulate, elemental iodine (I_2), hypoiodous acid (HOI), and organic iodides (CH_3I and others). The following sections describe sampler preparation, installation, removal, and preparation for analysis. Gamma spectrometric analyses were performed and the procedures for the analyses are also given in this appendix.

RADIOIODINE SPECIES SAMPLER PREPARATION

Radioiodine species samplers consist of five color-coded cups in a cylindrical body with two end caps. Rubber O-rings serve as seals between the cups and end caps, forcing all air flow through the sampling media. The sampling media containers are coded as follows: Particulate filter: red cup; CdI_2: blue cup; IPh: gold cup: AgX: silver cup; and charcoal: black cup.

PARTICULATE FILTER

The procedure for loading the particulate filter (red) cup is as follows:

- Insert a snap ring flush with notched end of red cup.
- Insert a teflon washer behind snap ring to protect the easily broken filter from the metal snap ring.
- Insert a Flanders F700 particulate filter with the lined side up.
- Insert another teflon washer behind the filter.
- Insert support screen and snap ring.
- Force inside and outside snap rings together to seal filters.

BEDS OF SAMPLING MATERIAL

The procedures for preparation of the CdI_2, IPh, Charcoal, and AgX beds are almost identical. Each bed is color-coded as described above.

- Insert a snap ring flush with notched end of the cup.
- Insert a teflon washer behind snap ring.
- Insert a filter behind teflon washer.
- Fill cup to within 3/16" of back of cup with the appropriate sampling medium.
- Tap lightly to pack and insert a filter, a wire screen for support, and a snap ring.
- Pack material (to prevent channeling of air flow) by forcing snap rings together until filter paper at front begins to bulge.

RADIOIODINE SPECIES SAMPLER ASSEMBLY

The procedure for assembling the set of cups, O-rings, sleeve, and end pieces is given below.

- Put a rubber O-ring on each notched end of each cup and on notched end cap stamped "OUT".
- Place successively, with the O-ring down, on inlet end cap (stamped "IN"), the red, blue, gold, and two silver or two black cups.
- Slide open-ended cylinder down over all cups.
- Place outlet end cap (stamped "OUT") with O-ring on top of cylinder and cups.
- Insert three long screws through holes of the inlet end cap and screw them into the outlet end cap. Tighten screws evenly and sufficiently tight to compress the O-rings.
- Attach a strip of tape to the cylinder for recording data on sampler location and operation.

INSTALLATION OF RADIOIODINE SPECIES SAMPLERS

Radioiodine species samplers were installed in ventilation ducts and pipes using glass or stainless steel probes. Any glass ball joints were secured with clamps and all fittings were tightened to prevent leakage. The following procedure was used for sampler installation.

- Connect the outlet end of sampler to the vacuum side of the air mover. Start air mover and measure the air flow (28-42 liters/min) by attaching a rotameter having a very low flow resistance to the inlet of the sampler. Remove the rotameter.
- Connect the inlet end of the sampler to the glass probe or stainless steel sampling line installed in the duct or pipe containing the effluent stream to be sampled.

- Record sampler location, date, time, pump head, pressure drop, and flow rate on sampling form.
- Record the location and time the sampler was turned on on a strip of tape attached to the cylindrical tube containing the sample cups.

REMOVAL OF RADIOIODINE SPECIES SAMPLERS

The following procedure was used at the time samplers were removed.

- At time of sampler removal, record the date, time, pressure drop, and pump head on the sampling form.
- Disconnect the sampler inlet from the glass probe. Measure the flow rate using a rotameter having a very low flow resistance and record the flow rate on the sampling form.
- Disconnect the outlet end of the sampler from the air mover.
- Record the time the sampler was turned off on the strip of tape attached to the cylinder.
- Close off the air flow at the end of the tube formerly attached to the outlet end of the sampler. Record the pressure drop at zero flow.
- When the sampler is returned to the laboratory, remove the individual cups from the holder and place them in plastic counting vials.
- Enter appropriate sampling information into the proper sample log book, assign sample numbers, and label each vial with its assigned number.
- The samples are ready for gamma ray analysis.

GAMMA-RAY SPECTROMETRIC ANALYSIS OF ACTIVITY COLLECTED BY RADIOIODINE SPECIES SAMPLER COMPONENTS

The two spectrometers used to analyze effluent samples are 4096-channel pulse-height analysis utilizing Ge(Li) detectors. Acquisition of data consists of a) placing the sample in the appropriate position for counting; b) clearing the analyzer memory; c) setting the desired counting "live" time; and d) beginning the acquisition. Since the energy scale for all of our data is approximately 0.7 keV/channel (∿ 2800 keV full scale), there is no need to adjust the energy scale prior to data acquisition, providing normal energy calibration procedures have been followed. The following sections describe the procedures in some detail.

Sample Alignment

Detectors used by NES to count effluent samples are mounted so that the axis of symmetry is horizontal. Samples are aligned with the detector about this axis. Many different sample configurations are used by NES in the sampling program. It is important that the sample be counted in the same alignment for which the detection efficiency calibration applies. At present, the following general rule applies: For all samples in which the sample material is tightly packed and contained (*e.g.*, iodine species sampler cups), the axes of symmetry of the sample and detector are aligned to coincide.

Normal counting distances used by NES are 1.0, 2.5, 5.0, 10.0, 20.0, and 30.0 cm from the front face of the cap surrounding the Ge(Li) detector to the nearest surface of the sampler container. Samples should be placed in the sample holder at the smallest of the distances listed above that yields an analyzer dead-time of less than 20%. This restriction is to minimize counting losses from pulse pile-up in the electronic system.

DATA ACQUISITION AND ANALYSIS

The analysis of data taken with the gamma-ray spectrometer to yield nuclide concentrations in the sample generally consists of a semi-automated analysis using the computer based analyzer system followed by an evaluation of the results by a competent experimenter.

The analysis functions are used to record the pulse-height spectra on magnetic tape or to analyze the data using the computer and to type out the analysis results. These functions are used following the acquisition of data or the reading of data from magnetic tape into the analyzer memory. The functions are:

a. search the spectrum for peaks;
b. calculate the energy of the peaks found;
c. calculate the gross and net counts in each peak;
d. identify the radionuclide giving rise to each peak;
e. determine the concentration ($\mu Ci/cm^3$) for each nuclide in the sample with decay corrections to the end of the sampling period;
f. print the results of the analysis in tabular form; and
g. store pulse-height spectrum on magnetic tape for permanent record.

The data acquisition and analysis procedures are all controlled through the teleprinter keyboard. Since the detailed procedures differ for the two spectrometers, they shall be given separately.

Tracor-Northern NS-600 System. This system is under computer (NOVA 1210) control. The functions are controlled through the teleprinter keyboard. Detailed operating discussions are contained in the analyzer manuals. The following refers to routine acquisition of the data. The commands for data acquisition are typed as follows: (- denotes the space bar.)

STEP	KEYBOARD COMMAND	OPERATION
a.	K_	Clears the Analyzer Memory
b.	YR_	Prints; Month, Day, Hour, Min., Sec., Count is begun.
c.	MPATXXXX	Acquires data for XXXX seconds, live time

d. While the data are being acquired, the teleprinter is switched to "local" operation and a title description of the sample being analyzed is to be typed. This information should be sufficient to properly identify the measurement. An example is:

"Oyster Creek Total Turbine Building, SN 4628-CdI Bed, Record #21370000"

Following the typing of the title, switch the teletypewriter back to "remote" operation.

e. Record the sample counted in the counting log book according to the format outlined in the log book procedures.

f. Following completion of the counting period, the teleprinter will perform a carriage return and line feed. The analyzer will be placed in the data display mode. At this point, record the spectrum on magnetic tape using the command:

LOT_XXXXXXXX_ Loads spectrum onto magnetic tape. XXXXXXXX is the proper eight-digit, octal tagword (next sequential tagword from log book)

This step requires that the correct magnetic data tape has been properly loaded on tape deck and positioned at the correct spot to record the data. The proper procedures for this are contained in the operating manuals.

The analysis programs in the NS-660 system require that certain initialization procedures be followed prior to the actual analysis of data. These procedures provide the following data tables, used in the spectral analysis.

a. FWHM Table. The peak search and integration techniques used in the NS-660 require a reasonably accurate table of peak width (expressed as full-width-at-half-maximum, FWHM) as a function of channel number.

b. Energy Calibration Table. In order that energy equivalents may be calculated for each peak found in a pulse-height spectrum, it is necessary that an accurate table of gamma-ray energy as a function of channel number be inserted into the computer.

c. Detection Efficiency Table. To convert from counts/second in the peaks of a pulse-height spectrum to gamma-rays/second emitted from the sample, it is necessary to have in the computer the detection efficiency vs. gamma-ray energy table that corresponds to the appropriate sample configuration at the distance it was counted.

d. Nuclide Table. To assign an observed gamma-ray energy to the decay of a specific nuclide in the sample to convert from gamma-rays emitted per second to disintegrations per second of that specific nuclide (and hence to micro-curies), and to make a correction to the observed intensities for radioactive decay between the time the sample was collected and when it was counted, it is necessary to have a table in the computer containing a) nuclides, b) principal gamma-rays emitted by the nuclides, c) gamma-rays emitted per decay for each gamma energy, and d) half-life of each nuclide.

The initialization procedures are performed routinely by NES counting laboratory personnel prior to the analysis of spectral data.

The analysis of a pulse-height spectrum, following the initialization, is controlled from the teleprinter keyboard and is detailed below. Detailed keyboard command descriptions are contained in the manuals for the system. Only the routine procedures are outlined below. In the nomenclature, _ denotes a space bar and a line under a letter denotes the fact that it has been typed by the computer. The symbol Ø denotes the number zero.

STEP	KEYBOARD COMMAND	OPERATION
a.	RFEXXXX Name of Efficiency Table	Place proper detection efficiency table into working region of memory.
b.	RFTXXXX Name of Nuclide Table	Places proper nuclide table into the working region of memory.
c.	0_R_Ø_	Types contents of channel 0, elapsed live time of data acquisition.
d.	0_R_1_	Types contents of channel 1, elapsed clock time of acquisition.
e.	RIEXXXX	Types name of efficiency table currently in working region.
f.	RIFXXXX	Types name of nuclide table currently in working region.
g.	Q	Provides entry into analysis subroutine.

STEP	KEYBOARD COMMAND	OPERATION
h.	S_C_	Provides entry into automatic peak search and energy calculation subroutines.
i.	/XX_YY_	Channel limits on peak search, XX = (lower limit) any channel from 0 to 4094. YY = (upper limit) any channel from 0 to 4094.
j.	DEL E ? X_	Maximum energy (keV) difference between calculated energy and energy in nuclide table for a nuclide assignment to be made. (X is generally made to be 3).
k.	DECAY TIME, HRS XX_	Time, in hours, between the end of the sampling period and beginning of data acquisition. Used for decay correction.
l.	VOLUME: XX_	Volume of sample to be used in calculating microcuries/unit volume. For air samples, convention has been established that the volume is put in units of 10^8 cm^3 so that the fixed point format of the answer fits the column.
m.	REJECT UN.%?XX_	Maximum value of statistical uncertainty desired for inclusion in list of found peaks (in %). A value of 70 usually results in the listing of all real peaks.
n.	UPPER HALF WIDTH =2__	Values of parameters used in peak search algorithm.
o.	LOWER HALF WIDTH =4__	Value of parameter used in peak search algorithm.

The analysis will proceed automatically from this point, yielding a list of energies and intensities for all peaks found in the spectrum and a list of all identified lines that includes the nuclide identified, a decay corrected intensity, and a conversion of microcuries/volume. This completes the computer-aided portion of the analysis of a gamma-ray pulse-height spectrum using this system. The decay corrected concentration computed for each of the I-131 lines identified was used to obtain a weighted average concentration using a technique described below. The weighted average concentration is corrected for decay of the I-131 during sampling. Both computations were performed using a small programmable calculator.

Canberra 8100 Quanta System. The data acquisition use of this system is performed under local control, with the analyzer portion in "stand-alone" operations. The procedure is as follows:

a. Place I/O Device Switch on 8100 chassis in "tape-out" position.
b. Depress both "memory reset" switches to clear memory and time channel.
c. Set live-time controller to proper value using "thumb-wheel" switches.
d. Press "collect" button until the light comes on to initiate count.
e. Set proper magnetic tape dump number using "thumb-wheel" switches.
f. Record the sample counted in the counting log book according to the format outlined in the log book procedures.
g. When count is over and the "collect" light goes out, press I/O button to record the spectrum on magnetic tape.

Analysis of spectral data using the Canberra System also requires that an energy calibration has been performed, that the detection efficiency tables are stored in the computer, and that the nuclide tables are stored in the computer.

The analysis is controlled from the keyboard and is initialized in the following example by the "RUN JEB" command. The arrow symbolizes a carriage return. The underlined statements are the actions which the operator performs. Once the required input information has been provided, the computer will analyze the spectrum and print the results automatically. Three output tables are printed:

a. a list of all peak channel positions, gamma-ray energies, counts/sec., gammas/sec., uncertainties, and possible nuclide assignments, listed in order of increasing gamma-ray energy.
b. a list, by nuclide, of all gamma-rays associated with that nuclide, the activity (μCi) per unit volume for each gamma-ray identified in the spectrum, and the associated uncertainty.
c. a list of weighted average intensities and a weighted 2σ uncertainty for each nuclide identified, calculated using the formulae given below.

Corrections for decay during sampling are made automatically by the Canberra System.

```
 RUN JEB ↵
TIME AND DATE OF ANALYSIS IS:   MARCH  25  11:17  '75

DETECTOR NUMBER: <TYPE IN DETECTOR NUMBER> ↵
DETECTION EFFICIENCY CURVE NAME: <TYPE IN NAME OF EFFICIENCY CURVE> ↵
SAMPLE NUMBER: <<TYPE IN SAMPLE NUMBER> ↵
MAGNETIC TAPE NAME: <TYPE IN NAME OF TAPE> ↵
TAPE RECORD NUMBER: <TYPE IN DUMP NUMBER> ↵
SAMPLE DESCRIPTION: <PLACE KEYBOARD IN LOCAL MODE. TYPE IN SAMPLE
                      DESCRIPTION. PLACE KEYBOARD IN LINE MODE> ↵
SAMPLE VOLUME: <TYPE IN VOLUME IN WHATEVER UNITS YOU DESIRE> ↵
VOLUME UNITS: <TYPE IN NAME OF UNITS USED FOR VOLUME> ↵
TIME BETWEEN SAMPLING AND COUNTING TIMES <DECAY TIME> <HRS>: <TYPE IN
               DECAY TIME IN HOURS> ↵
SAMPLING DURATION <HRS>: <TYPE IN SAMPLING TIME IN HOURS> ↵
NUMBER OF SIGMA UNITS ON PEAK FIND: <USE 3 TO FIND ALL REAL PEAKS> ↵
ENERGY SEPARATION FOR NUCLIDE ID <KEV>: <USUALLY 0.3 IDENTIFIES THE
                                         PEAKS IF THE ENERGY CALIBRATION
                                         IS CURRENT> ↵

MAKE CERTAIN THAT I/O DEVICE SWITCH IS ON REMOTE! TYPE RETURN

ANALYZER IS FREE TO ACQUIRE NEXT SPECTRUM


<AT THIS POINT THE 8100 UNIT CAN BE PLACED IN LOCAL OPERATION BY RETURNING
 THE I/O SWITCH TO TAPE OUT AND THE NEXT SPECTRUM CAN BE ACQUIRED>
```

Date Evaluation and Interpretation

As indicated above, the weighted average concentration is reported. The average is computed automatically by the Canberra System; it is obtained using data from the NS-660 System printout using a small programmable calculator. The calculational procedure is the same in either case. For I-131, the gamma-rays used and their absolute branching ratios were as follows:

TABLE A-1

I-131 PEAKS ENERGIES AND BRANCHING RATIOS

Peak Number	I-131 Gamma-Ray Energy (keV)	Absolute Branching Ratio
1	284.3	0.059
2	364.5	0.824
3	637.0	0.067
4	722.9	0.018

In determining the weighted average, each peak's contribution is weighted by the inverse of the square of its uncertainty. If X_i is the estimated concentration of I-131 based on data for the $i^{\underline{th}}$ peak and S_i is the uncertainty (standard deviation) of that result, the weighted average concentration for the sample is defined to be

$$\overline{X} = \frac{\sum_{i=1}^{k} (X_i/S_i^2)}{\sum_{i=1}^{k} (1/S_i^2)} \qquad \text{(A-1)}$$

where k is at least 1 (the 364-keV peak) and at most 4 (all the peaks in the previous table). The equation for the standard deviation of the weighted average concentration is

$$S = \left(\frac{\sum_{i=1}^{k} (X_i - \overline{X})^2/S_i^2}{(k - 1) \sum_{i=1}^{k} (1/S_i^2)} \right)^{1/2} \qquad \text{(A-2)}$$

This uncertainty is based on external consistency rather than internal consistency in order to include uncertainties in branching ratios and relative uncertainties in the efficiency curves. It does not take into the absolute accuracy of the efficiency curves or the positioning of the source in the source holder. If the uncertainty(s) calculated using the above equation is less than 5% of the average value ($\overline{X}$), a standard deviation of 5% of $\overline{X}$ is used in place of the calculated value.

The NS-660 System calculates the concentration without considering the duration of the sampling. Because the actual sampling times are significant with respect to the half-life of I-131 (8.07 days) an additional correction must be applied to account for the decay during sampling. The correction factor is given by $\lambda t_s/(1 - \exp(-\lambda t_s))$, where t_s = length of the sampling period (hours) and λ is the radiologic decay rate constant for I-131, 0.00358 hr^{-1}. This decay correction necessarily assumes that the concentration of I-131 in the effluent stream was constant during the sampling period. This decay correction is made automatically by the Canberra System and is reflected in the second table.

COMPUTATION OF RADIOIODINE SPECIES DISTRIBUTION

When the mean concentrations of I-131 collected by the species sampler particulate filter and adsorption beds have been determined as described above, the apparent radioiodine species distribution is known. The true species distribution is somewhat different owing to imperfect separation of the species by the adsorber beds. Let the Z_j ($j = 1,2,3,4$) be the observed concentrations on the particulate filter, cadmium iodide bed, iodophenol bed, and charcoal bed respectively, and the C_j be the true concentrations of particulate iodine, elemental iodine, HOI, and organic iodides. The following relationships are assumed to hold:

$$Z_1 = C_1 + aC_2 = \text{concentration on particulate filter} \qquad \text{(A-3)}$$

$$Z_2 = bC_2 + dC_3 = \text{concentration of } CdI_2 \text{ bed} \qquad \text{(A-4)}$$

$$Z_3 = eC_2 + fC_3 = \text{concentration in the IPH bed} \qquad \text{(A-5)}$$

$$Z_4 = gC_3 + C_4 = \text{concentration in AgX or charcoal bed} \qquad \text{(A-6)}$$

The above equations assume no organic iodides are trapped by the particulate filter, CdI_2 bed, or the IPH bed and that no HOI is collected on the particulate filter. The table of collection efficiencies for the sampling media is as follows:

TABLE A-2

RADIOIODINE SPECIES SAMPLER COLLECTION EFFICIENCIES

	Collection Efficiencies for Four Chemical Forms			
Medium	Particulate	I_2	HOI	Organic
Particulate Filter	1.0	0.05(a)	0.0	0.0
CdI_2 Bed	0.0	0.87(b)	0.04(d)	0.0
IPH Bed	0.0	0.08(e)	0.94(f)	0.0
Charcoal or AgX Bed	0.0	0.0	0.02(g)	1.0
Total Sampler	1.0	1.0	1.0	1.0

The letters in parentheses are the symbols used in the four equations given above. The equations can be solved and yield the following formulae for calculating the true species distribution.

$$C_3 = \frac{b}{(bf - de)} (Z_3 - \frac{eZ_2}{b}) = \text{concentration of HOI} \qquad \text{(A-7)}$$

$$C_4 = Z_4 - gC_3 = \text{concentration of organic iodides} \qquad \text{(A-8)}$$

$$C_2 = \frac{Z_2 - dC_3}{b} = \text{concentration of } I_2 \qquad \text{(A-9)}$$

$$C_1 = Z_1 - aC_2 = \text{concentration of I-131 associated with particulate material} \qquad \text{(A-10)}$$

APPENDIX B

TABULATIONS OF MEASURED CONCENTRATIONS AND CHEMICAL FORMS OF I-131 AT OYSTER CREEK

TABLE B-1

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK REACTOR BUILDING VENTILATION AIR

Sampling Period	Total I-131 Concentration (pCi/m^3)	Species Fractionation (%)			
		Part.	I_2	HOI	Organic
1	7.17±.18	< 1	24	12	64
2	6.29±.51	10	69		21
3	6.51±.23	17	62		21
4	3.15±.23	< 7	27	40	25
5	4.79±.25	17	24	35	24
6	5.63±.25	13	30	33	24
7	2.48±3	8	26	35	31
8	30.0 ±.7	2	25	46	27
9	24.4 ±.6	5	24	39	32
10	21.3 ±.4	6	19	38	37
11	30.9 ±.6	5	11	17	67
12	10.1 ±.3	11	24	41	24
13	10.7 ±.3	7	26	46	21
14	14.3 ±.3	9	26	39	25
15	1.22±.11	4	28	37	31
16	8.50±.28	6	35	34	25
17	8.41±.22	10	27	41	22
18	7.93±.25	11	33	29	27
19	6.90±.20	10	34	31	25
20	6.50±.29	13	35	27	25
21	6.74±.24	16	33	26	25
22	6.87±.24	15	40	21	24
23	5.30±.27	< 5	37	30	29

TABLE B-2

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK RADWASTE BUILDING VENTILATION AIR

<table>
<tr><th rowspan="2">Sampling Period</th><th rowspan="2">Total I-131 Concentration (pCi/m^3)</th><th colspan="4">Species Fractionation (%)</th></tr>
<tr><th>Part.</th><th>I_2</th><th>HOI</th><th>Organic</th></tr>
<tr><td>1</td><td>43.7 ± .9</td><td>< 0.2</td><td>9</td><td>18</td><td>73</td></tr>
<tr><td>2</td><td>25.6 ± .6</td><td>6</td><td colspan="2">49</td><td>45</td></tr>
<tr><td>3</td><td>52.4 ±1.8</td><td>< 0.2</td><td colspan="2">47</td><td>53</td></tr>
<tr><td>4</td><td>57.3 ±1.4</td><td>< 0.1</td><td>15</td><td>28</td><td>57</td></tr>
<tr><td>5</td><td>13.8 ± .3</td><td>< 0.5</td><td>13</td><td>26</td><td>61</td></tr>
<tr><td>6</td><td>8.77± .30</td><td>< 0.7</td><td>13</td><td>20</td><td>66</td></tr>
<tr><td>7</td><td>103 ±2</td><td>< 0.1</td><td>5</td><td>17</td><td>78</td></tr>
<tr><td>8</td><td>51.8 ±1.4</td><td>< 0.2</td><td>9</td><td>14</td><td>77</td></tr>
<tr><td>9</td><td>18.9 ± .3</td><td>< 0.3</td><td>41</td><td>43</td><td>16</td></tr>
<tr><td>10</td><td>96.0 ±1.8</td><td>< 0.1</td><td>7</td><td>21</td><td>72</td></tr>
<tr><td>11</td><td>85.7 ±1.7</td><td>0.3</td><td>8</td><td>17</td><td>75</td></tr>
<tr><td>12</td><td>38.3 ± .9</td><td>< 0.2</td><td>14</td><td>23</td><td>63</td></tr>
<tr><td>13</td><td>70.4 ±1.4</td><td>< 0.1</td><td>9</td><td>23</td><td>68</td></tr>
<tr><td>14</td><td>127 ±3</td><td>< 0.1</td><td>10</td><td>14</td><td>76</td></tr>
<tr><td>15</td><td>416 ±10</td><td>< 0.1</td><td>2</td><td>3</td><td>95</td></tr>
<tr><td>16</td><td>91.1 ±1.4</td><td>0.5</td><td>41</td><td>17</td><td>41</td></tr>
<tr><td>17</td><td>52.4 ± .8</td><td>< 0.2</td><td>29</td><td>20</td><td>51</td></tr>
<tr><td>18</td><td>41.1 ± .9</td><td>< 0.2</td><td>15</td><td>30</td><td>55</td></tr>
<tr><td>19</td><td>109 ±2</td><td>< 0.1</td><td>27</td><td>33</td><td>40</td></tr>
<tr><td>20</td><td>62.7 ±1.0</td><td>0.6</td><td>27</td><td>18</td><td>55</td></tr>
<tr><td>21</td><td>49.0 ±1.4</td><td>< 0.2</td><td>17</td><td>32</td><td>51</td></tr>
<tr><td>22</td><td>71.1 ±1.3</td><td>< 0.1</td><td>8</td><td>23</td><td>69</td></tr>
<tr><td>23</td><td>58.2 ±1.7</td><td>< 0.1</td><td>19</td><td>22</td><td>59</td></tr>
</table>

TABLE B-3

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK TURBINE BUILDING VENTILATION AIR

Sampling Period	Total I-131 Concentration (pCi/m^3)	Species Fractionation (%)			
		Part.	I_2	HOI	Organic
1	336±5	18	49	28	5
2	345±4	22	49	45	4
3	637±26	11	84		5
4	415±6	20	37	38	5
5	377±6	23	29	43	5
6	409±5	21	43	31	5
7	189±3	18	31	39	13
8	401±6	25	45	25	5
9	620±9	25	39	30	6
10	561±8	22	29	40	9
11	328±5	9	39	41	11
12	335±5	12	40	38	10
13	265±4	15	30	46	9
14	489±7	21	41	29	9
15	383±6	18	52	23	7
16	422±6	15	52	27	6
17	419±5	18	45	31	6
18	395±5	16	51	25	8
19	351±5	12	54	26	8
20	375±6	22	44	25	9
21	354±6	3	65	24	8
22	333±5	14	57	19	10
23	308±5	6	56	30	8

TABLE B-4

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK STEAM JET AIR EJECTOR DELAY LINE

Sampling Period	Total I-131 Concentration (μCi/m^3)	Species Fractionation (%)			
		Part.	I_2	HOI	Organic
1	No Sample	-	-	-	-
2	2.40±.07	< 0.1	1	18	81
3	5.96±.12	< 0.1	0.1	19	81
4	No Sample	-	-	-	-
5	3.20±.12	0.1	< 0.1	18	82
6	3.33±.10	0.1	3	16	81
7	3.99±.13	< 0.1	1	10	89
8	4.67±.13	< 0.1	1	24	75
9	9.35±.31	< 0.1	0.4	19	80
10	10.2 ±.2	< 0.1	1	17	82
11	5.61±.16	< 0.1	3	18	79
12	4.55±.12	< 0.1	1	25	74
13	12.9 ±.3	< 0.1	0.4	33	67
14	12.4 ±.3	< 0.1	0.4	12	88
15	7.25±.22	< 0.1	< 0.1	13	87
16	10.5 ±.4	< 0.1	0.2	6	94
17	11.1 ±.4	< 0.1	0.1	< 0.1	100
18	7.83±.26	< 0.1	0.2	6	94
19	8.52±.29	< 0.1	0.1	3	97
20	8.15±.29	< 0.1	< 0.1	1	99
21	8.09±.28	< 0.1	< 0.1	2	98
22	4.08±.14	< 0.1	< 0.1	1	99
23	6.27±.11	< 0.1	< 0.1	1	99

TABLE B-5

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK GLAND SEAL AND MECHANICAL VACUUM PUMP EXHAUST

<table>
<tr><th rowspan="2">Sampling Period</th><th rowspan="2">Total I-131 Concentration (nCi/m³)</th><th colspan="4">Species Fractionation (%)</th></tr>
<tr><th>Part.</th><th>I_2</th><th>HOI</th><th>Organic</th></tr>
<tr><td>1</td><td>No Sample</td><td>-</td><td>-</td><td>-</td><td>-</td></tr>
<tr><td>2</td><td>1.45± .04</td><td>< 0.1</td><td>9</td><td>12</td><td>79</td></tr>
<tr><td>3</td><td>1.64± .04</td><td>1</td><td>18</td><td>26</td><td>55</td></tr>
<tr><td>4</td><td>1.14± .03</td><td>1</td><td>15</td><td>19</td><td>65</td></tr>
<tr><td>5</td><td>2.08± .05</td><td>1</td><td colspan="2">36</td><td>63</td></tr>
<tr><td>6</td><td>1.49± .33</td><td>1</td><td>7</td><td>33</td><td>59</td></tr>
<tr><td>7</td><td>93.3 ±2.0</td><td>< 0.1</td><td>6</td><td>15</td><td>79</td></tr>
<tr><td>8</td><td>1.88± .03</td><td>1</td><td>22</td><td>38</td><td>39</td></tr>
<tr><td>9</td><td>0.38± .01</td><td>1</td><td>1</td><td>28</td><td>70</td></tr>
<tr><td>10</td><td>2.22± .05</td><td>3</td><td>9</td><td>42</td><td>46</td></tr>
<tr><td>11</td><td>6.47± .13</td><td>0.2</td><td>8</td><td>55</td><td>37</td></tr>
<tr><td>12</td><td>4.57± .08</td><td>0.4</td><td>5</td><td>55</td><td>40</td></tr>
<tr><td>13</td><td>5.51± .10</td><td>0.3</td><td>6</td><td>57</td><td>37</td></tr>
<tr><td>14</td><td>6.69± .11</td><td>0.1</td><td>8</td><td>50</td><td>42</td></tr>
<tr><td>15</td><td>8.20± .14</td><td>0.3</td><td>5</td><td>50</td><td>44</td></tr>
<tr><td>16</td><td>5.20± .09</td><td>1</td><td>6</td><td>43</td><td>50</td></tr>
<tr><td>17</td><td>5.30± .09</td><td>0.2</td><td>7</td><td>49</td><td>44</td></tr>
<tr><td>18</td><td>4.73± .09</td><td>0.3</td><td>6</td><td>43</td><td>51</td></tr>
<tr><td>19</td><td>4.31± .07</td><td>0.4</td><td>6</td><td>47</td><td>47</td></tr>
<tr><td>20</td><td>4.28± .08</td><td>0.3</td><td>5</td><td>44</td><td>51</td></tr>
<tr><td>21</td><td>4.52± .07</td><td>1</td><td>6</td><td>45</td><td>48</td></tr>
<tr><td>22</td><td>3.18± .07</td><td>0.4</td><td>10</td><td>6</td><td>83</td></tr>
<tr><td>23</td><td>4.12± .07</td><td>1</td><td>6</td><td>42</td><td>51</td></tr>
</table>

TABLE B-6

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK FEEDWATER AND CONDENSATE PUMP ROOM EXHAUST

Sampling Periods	Total I-131 Concentration (pCi/m^3)	Species Fractionation (%)			
		Part.	I_2	HOI	Organic
1	No Sample	-	-	-	-
2 & 3	7.46±.18	<1	35	62	3
4 & 5	6.26±.21	5	95		<1
6 & 7	5.18±.16	10	83		7
8-10	15.1 ±.4	8	87		4
11 & 12	4.98±.17	12	44	15	29
13-16	9.30±.36	6	41	49	4
17 & 18	7.53±.30	6	50	43	1
19 & 20	6.06±.17	7	82	8	3
21 & 22	12.0 ±.5	17	77	6	<1
23	11.4 ±.3	12	69	15	4

TABLE B-7

I-131 CONCENTRATION AND SPECIES DISTRIBUTION IN OYSTER CREEK REHEATER PROTECTION AND LUBE OIL STORAGE AREA EXHAUST

Sampling Period	Total I-131 Concentration (pCi/m^3)	Species Fractionation (%)			
		Part.	I_2	HOI	Organic
1	No Sample	-	-	-	-
2 & 3	No Sample	-	-	-	-
4 & 5	No Sample	-	-	-	-
6 & 7	101±3	3	9	19	69
8 & 9	154±5	1	6	7	86
10 & 11	246±8	1	2	5	92
12 & 13	247±8	6	12	14	68
14 & 15	139±4	0.3	17	12	71
16 & 17	147±5	10	28	15	47
18 & 19	96.0±1.5	9	21	14	56
20 & 21	110±2	9	20	13	58
22	No Sample	-	-	-	-
23	55.3±3.3	7	20	23	50

APPENDIX C

TABULATION OF PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR OYSTER CREEK FIELD MONITORING LOCATIONS

TABLE C-1

PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR SITE #1, OYSTER CREEK

	Dispersion and Deposition Parameters--Stack Releases			**Dispersion and Deposition Parameters--Vent Releases**		
PERIOD	$(X/Q)_s^d(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)_v^d(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
1	195.2	3790	199.9	1143	11110	1348
2	140.9	2099	145.0	884.3	5296	1043
3	31.69	1690	32.76	598.8	4005	706.4
4	30.15	1099	30.50	816.9	3632	963.6
5	9.095	238.3	9.289	492.2	1332	580.6
6	27.57	889.9	28.03	1040	5708	1227
7	17.38	542.9	17.62	747.0	2584	881.2
8	16.02	293.8	16.10	626.2	3237	738.7
9	36.42	606.4	36.61	1679	5943	1980
10	$2.36x10^{-4}$	0.1842	$2.36x10^{-4}$	1112	1277	1311
11	78.16	2834	79.75	794.6	4625	937.4
12	33.12	1071	33.58	663.2	3595	782.4
13	16.27	992.5	16.96	1491	2372	1759
14	10.50	369.2	10.64	346.5	2309	408.8
15	16.10	507.8	16.19	1430	4233	1687
16	10.40	205.6	10.82	288.1	1049	339.9

TABLE C-1 (cont.)

PERIOD	Dispersion and Deposition Parameters--Stack Releases $(X/Q)^d_s(\frac{\text{nsec}}{\text{m}^3})$	 $\delta_s(\text{m}^{-2})\text{x}10^{12}$	 $(X/Q)_s(\frac{\text{nsec}}{\text{m}^3})$	Dispersion and Deposition Parameters--Vent Releases $(X/Q)^d_v(\frac{\text{nsec}}{\text{m}^3})$	 $\delta_v(\text{m}^{-2})\text{x}10^{12}$	 $(X/Q)_v(\frac{\text{nsec}}{\text{m}^3})$
17	26.88	1004	27.51	413.8	2664	488.2
18	9.274	504.6	9.511	881.0	3313	1039
19	3.284	135.0	3.461	1312	1814	1548
20	9.912	233.5	9.960	738.1	3428	870.8
21	7.244	186.6	7.521	472.8	1523	557.8
22	5.472	103.4	5.505	384.3	1142	453.3
23	20.65	292.0	20.75	568.1	3964	670.1

TABLE C-2

PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR SITE #2, OYSTER CREEK

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)^d_s (\frac{nsec}{m^3})$	$\delta_s (m^{-2}) x10^{12}$	$(X/Q)_s (\frac{nsec}{m^3})$	$(X/Q)^d_v (\frac{nsec}{m^3})$	$\delta_v (m^{-2}) x10^{12}$	$(X/Q)_v (\frac{nsec}{m^3})$
1	65.86	588.4	67.81	154.0	1465	189.2
2	40.64	362.4	41.19	425.1	2399	522.4
3	8.812	232.9	9.159	435.1	1368	534.7
4	17.40	326.8	17.75	528.9	1771	649.9
5	2.208	122.3	2.287	395.8	928.3	486.4
6	24.57	524.5	25.08	620.6	2638	762.6
7	6.582	96.19	6.687	404.2	450.2	496.7
8	5.526	0.3698	5.526	369.6	1368	454.2
9	37.46	296.5	37.99	492.1	1449	604.7
10	0.01044	0.193	0.01044	0	0	0
11	48.22	876.6	50.77	268.9	1128	330.4
12	4.648	110.3	4.843	1025	1586	1260
13	3.144	30.67	3.170	498.0	1157	612.0
14	17.26	503.0	18.14	481.1	2047	591.1
15	17.51	260.5	17.81	712.2	1278	875.2
16	5.191	42.33	5.254	271.2	614.2	333.2

TABLE C-2 (cont.)

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)^d_s(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)^d_v(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
17	5.250	221.2	5.404	285.9	1021	351.4
18	5.765	209.8	5.917	321.8	908.9	395.4
19	16.78	457.0	17.18	1030	1770	1266
20	3.650	0.4131	3.650	1061	1672	1303
21	10.87	61.77	11.06	439.8	1950	540.5
22	9.050	76.04	9.181	273.5	928.3	336.1
23	21.98	192.4	22.26	539.8	1881	663.3

TABLE C-3

PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR SITE #3, OYSTER CREEK

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)_s^d(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)_v^d(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
1	91.22	783.0	97.31	129.8	1337	176.0
2	67.05	440.7	71.0	111.6	637.0	151.4
3	18.83	264.6	19.71	66.76	481.8	90.52
4	22.63	243.6	23.62	101.2	436.8	137.2
5	5.284	47.92	5.557	64.85	160.2	87.94
6	16.83	217.0	17.69	121.5	686.5	164.8
7	18.64	95.33	19.17	80.82	310.8	109.6
8	23.10	86.13	23.55	63.67	389.3	86.35
9	39.25	177.7	40.38	218.0	714.8	295.6
10	0.3435	0.05401	0.3435	166.0	153.6	225.1
11	40.96	590.5	43.42	90.85	556.3	123.2
12	22.73	215.4	23.84	78.95	432.4	107.1
13	4.769	149.7	5.219	208.3	285.3	282.4
14	7.980	84.54	8.309	35.91	277.7	48.68
15	13.64	148.8	14.17	187.1	509.2	253.7
16	4.173	36.08	4.426	35.29	126.2	47.85

TABLE C-3 (cont.)

PERIOD	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
	$(X/Q)^d_s(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)^d_v(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
17	14.85	206.6	15.65	56.17	320.5	76.17
18	5.662	101.4	5.924	121.2	398.4	164.4
19	2.371	17.65	2.439	183.6	218.2	248.9
20	13.90	68.45	14.18	95.15	412.4	129.0
21	6.968	32.77	7.113	57.92	183.1	78.53
22	3.020	30.31	3.215	53.97	137.4	73.18
23	25.74	85.60	26.35	62.22	476.8	84.36

TABLE C-4

PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR SITE #4, OYSTER CREEK

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)_s^d (\frac{nsec}{m^3})$	$\delta_s (m^{-2}) x10^{12}$	$(X/Q)_s (\frac{nsec}{m^3})$	$(X/Q)_v^d (\frac{nsec}{m^3})$	$\delta_v (m^{-2}) x10^{12}$	$(X/Q)_v (\frac{nsec}{m^3})$
1	133.4	1234	140.5	151.2	1825	198.1
2	22.96	468.0	24.39	164.9	800.8	216.1
3	19.71	265.4	20.11	46.47	516.1	60.89
4	6.286	56.49	6.495	75.72	376.0	99.22
5	22.52	368.7	23.68	101.9	525.5	133.5
6	10.21	145.9	10.56	37.35	331.8	48.94
7	23.11	176.3	23.94	96.54	475.7	126.5
8	60.37	531.4	62.09	102.7	1072	134.6
9	24.45	223.7	25.26	406.4	1607	532.5
10	4.994	0.1154	4.994	0	0	0
11	23.45	307.0	24.62	44.98	106.4	58.94
12	12.24	338.6	13.10	116.9	504.2	153.2
13	6.871	90.74	6.561	64.43	181.9	84.43
14	19.18	468.8	20.55	32.20	656.8	42.19
15	14.98	310.3	15.72	70.68	408.2	92.62
16	0	0	0	0	0	0

TABLE C-4 (cont.)

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)_s^d(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)_v^d(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
17	26.12	280.5	27.13	117.7	595.6	154.2
18	0.00266	0.0258	0.00266	0	0	0
19	15.98	130.6	16.75	181.3	296.9	237.5
20	21.91	147.2	22.48	115.7	552.2	151.6
21	2.972	43.50	3.117	96.99	140.1	127.1
22	0	0	0	7.054	70.07	9.243
23	3.044	0.686	3.044	106.1	256.4	139.0

TABLE C-5

PREDICTED DISPERSION AND DEPOSITION PARAMETERS FOR SITE #6, OYSTER CREEK

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)_s^d(\frac{nsec}{m^3})$	$\delta_s(m^{-2})x10^{12}$	$(X/Q)_s(\frac{nsec}{m^3})$	$(X/Q)_v^d(\frac{nsec}{m^3})$	$\delta_v(m^{-2})x10^{12}$	$(X/Q)_v(\frac{nsec}{m^3})$
1	0	0	0	0	0	0
2	5.506	125.8	5.961	123.7	262.7	171.4
3	11.59	170.2	12.57	233.6	500.7	323.9
4	8.535	104.7	9.089	101.8	399.3	141.1
5	11.12	123.1	11.76	226.6	564.0	314.1
6	5.243	103.6	5.738	13.43	139.1	18.62
7	14.06	61.43	14.93	130.2	222.2	180.5
8	5.791	15.98	5.876	121.8	139.9	168.8
9	7.305	49.83	7.674	15.81	189.2	21.92
10	4.641	105.0	4.990	0	0	0
11	8.323	102.0	8.770	119.1	428.3	165.1
12	11.54	107.5	12.42	138.5	237.8	189.3
13	8.062	75.11	8.398	242.5	457.6	336.2
14	18.48	57.35	18.68	125.4	272.1	173.9
15	5.215	12.19	5.314	360.2	224.0	499.3
16	0	0	0	0	0	0

TABLE C-5 (cont.)

	Dispersion and Deposition Parameters--Stack Releases			Dispersion and Deposition Parameters--Vent Releases		
PERIOD	$(X/Q)_s^d (\frac{nsec}{m^3})$	$\delta_s (m^{-2}) x10^{12}$	$(X/Q)_s (\frac{nsec}{m^3})$	$(X/Q)_v^d (\frac{nsec}{m^3})$	$\delta_v (m^{-2}) x10^{12}$	$(X/Q)_v (\frac{nsec}{m^3})$
17	9.339	87.89	9.859	160.5	334.8	222.5
18	2.706	65.19	2.896	64.40	210.9	89.29
19	18.88	88.16	19.16	69.74	205.3	96.69
20	12.04	241.5	13.03	180.6	436.5	250.4
21	21.30	216.4	22.37	92.47	458.2	128.2
22	8.066	93.34	8.458	151.7	352.5	210.3
23	28.57	191.3	29.93	99.39	615.1	137.8

APPENDIX D

ENVIRONMENTAL AIR SAMPLES

DESCRIPTION

Samples of airborne I-131 in the environment are collected using a special filter head through which ambient air is drawn at a constant rate between 570 and 850 liters/minute. Normally, air entering the special filter head passes, in sequence, through a particulate filter, a bed of iodophenol (IPh) adsorbed on alumina, and a bed of activated charcoal impregnated with triethylenediamine (TEDA). This 3-filter sequence is designed to collect (1) iodine attached to particulate materials, (2) inorganic iodine species, I_2 and HOI, and (3) organic iodides. In some cases the IPh bed was replaced with a bed of CdI_2, so the breakdown achieved was particulates, elemental iodine, and HOI plus organic iodides. A third arrangement, used only occasionally, separated the non-organic gaseous forms into the elemental and HOI fractions. In the latter case the air passed through a particulate filter, a CdI_2 bed, and an IPh bed. The procedure is written for the normal arrangement of particulate filter, IPh bed, and charcoal bed.

The filter head is rectangular in shape; the 20 cm by 25 cm HEPA filter (Flanders F700) is supported by a metal screen. The top bed is supported by a metal screen and particulate filter 19-cm by 24-cm and is 2.5-3.2 cm deep. The bottom bed is similarly supported by a particulate filter placed on a 18-cm by 23-cm metal screen and is also 2.5-3.2 cm deep. The material for the IPh beds is manufactured and tested by the Nuclear Environmental Services Division of SAI. The TEDA-impregnated charcoal is obtained from North American Carbon Company. Procedures for loading and unloading the filter heads are given below.

Operational procedures for the high volume air sampling systems are also described below. The systems are equipped with flow regulators to maintain a constant flow rate for the duration of the sampling period, from 7 to 14 days.

SAMPLER LOADING AND UNLOADING

The procedure for loading the sampler is given below. Sets of filters are precut in the required sizes (18 cm by 23 cm, 19 cm by 24 cm, and 20 cm by 25 cm) and the IPh and charcoal are bagged in premeasured amounts in the laboratory. To load the sampler:

- Insert the filter head into the sampler and take out the two removable metal screens.
- Turn the sampler on and position the 18 cm by 23 cm filter over the bottom fixed metal screen, taking care to assure that a good seal is achieved. Turn the sampler off.
- Pour in the TEDA-impregnated charcoal and smooth it into a uniform layer; slight vibration of the filter head and/or tamping will assist uniform settling of the charcoal.
- Replace the middle supporting screen and position the 19 cm by 24 cm filter, taking care to assure a good seal.
- Pour in the IPh and smooth it into a uniform bed using a straight edge. Tamp down the media, especially near the corners, to assure uniform settling.
- Replace the top supporting screen and position the 20 cm by 25 cm filter paper on the screen.
- Close and secure the top gasket of the filter bed.
- Restart the sampler and adjust the flow rate to ∿710 liters/minute. Reset the elapsed time meter to zero, and observe sampler behavior for a few minutes to verify that the flow control system is functioning properly.
- Record the sampler starting date and time, the pressure gauge reading, the elapsed time meter setting, and any comments pertinent to sampler operation.

As indicated the samplers will operate for 7-14 days before the media in the sampler head are changed. The procedure for unloading the head are as follows:

- Record the pressure gauge reading; turn the sampler off and record the date, time, and elapsed time meter reading.
- Release the top gasket of the filter head, remove the particulate filter, fold it in half twice, and seal it in a new plastic bag. Label the bag with the sampler location and the date and time the sampler was turned off. Remove the top support screen.
- Carefully scoop out the IPh and place it in a new plastic bag. Remove the support filter and add it to the contents of the bag. Seal the bag and label it with the sampler location and the date and time the sampler was turned off. Remove the middle support screen.
- Carefully scoop out the charcoal and place it in a new plastic bag. Remove the support filter and add it to the contents of the bag. Seal the bag and label it with the sampler location and the date and time the sampler was turned off.

SAMPLER OPERATION AND FLOW ADJUSTMENTS

Field Assembly

The procedure for assembling an air sampler in the field is as follows.

- Unpack sampler, legs, and the special sampling head. Check the sampler for loose nuts and connections.
- Mount the removable legs on the bolts protruding from the edges of the sampler body; tighten nuts securely.
- Open the sampler lid and insert the sampling head, taking care not to damage the O-rings and to obtain a good seal.
- Check the fuse.
- Plug the sampler into a 117 V, 60 Hz power source.

Flow Adjustment

The calibration curve for the sampler is mounted inside the sampler lid. This curve shows the relationship between the flow rate and the reading of the pressure gauge mounted on the front panel. There are two small red lights mounted near the pressure gauge; when lighted, these indicate that the sampler flow is being changed (automatically or as the result of adjustment by the operator). One light indicates increasing flow, the other bulb lights when the flow is being reduced. The following operations should be performed with the sampling head loaded and in place and the sampler running.

- Determine the pressure gauge reading which corresponds to the desired flow rate, normally $\sim$710 liters/minute (25 ft^3/min.).
- Locate the flow adjustment knob; it is a large black knob protruding from the bottom panel of the chassis.
- If the red indicator lights are both off, proceed with the adjustment; otherwise, wait for the unit to stabilize before continuing.
- If the gauge pressure is lower than the desired value, increase the flow rate by turning the flow adjustment knob clockwise (as viewed from beneath the knob). If the gauge pressure is higher than the desired value, decrease the flow by turning the flow adjustment knob counterclockwise. The flow rate is quite sensitive to changes in the valve opening, so small incremental adjustments are generally recommended.
- After adjusting the flow, allow the unit to stabilize (the indicator light will turn off) before making further adjustments.

Flow Sensing Switch Setting

The flow sensing switch is set at the factory and normally does not require adjustment. Access to the switch is obtained by removing the small circular cover on the front panel between the indicator lights. The switching point separation is adjusted with a screwdriver which is inserted into a plastic cap. (This cap is glued to an electrically "hot" metal component so care should be taken not to damage it).

If at any time both indicator lights are on, the point separation requires adjustment and the following procedure should be used, with a loaded sampling head in place.

- Turn the adjustment screw counterclockwise to the point at which both lights are no longer lighted simultaneously.
- Turn the adjustment screw an additional 1/4 turn counterclockwise.
- Monitor the behavior of the flow controller. If it rapidly cycles between increasing and decreasing the flow (as indicated by alternate lighting of the two indicator lights), a wider point separation is required. Turn the adjustment screw counterclockwise a small additional amount and again monitor the system's behavior. The flow controller should not adjust the flow more frequently than once per minute when the proper separation is achieved. If the controller continues to hunt for the set point, increase the separation again by a small counterclockwise turning of the adjustment screw.

RADIOCHEMICAL ANALYSIS AND COUNTING

The particulate filter, IPh bed, and charcoal bed from each environmental air sampling location were returned to the laboratory for processing and analysis. Each sample component was assigned a unique log number. Particulate filters were placed in sample counting vials and counted directly using the Nuclear Data 4410 (ND4410) gamma spectrometry system, shielded Ge(Li) detector, and automatic sample changer. Iodophenol beds were either (a) counted directly in calibrated Marinelli-type containers which fit within the lead shield around the Ge(Li) detector used with the ND4410 System or (b) leached to remove the I-131 which was subsequently extracted, precipitated and counted using the ND4410 System or a 12.5 cm by 12.5 cm NaI(Tℓ) crystal spectrometer. Charcoal beds were either (a) counted directly in a Marinelli-type container using the ND4410 System or (b) leached to remove the I-131 which was subsequently extracted, precipitated, and counted using the Beckman Wide-Beta II beta counter.

The leaching and analysis procedures for charcoal beds are described in detail in Appendix E. The leaching procedure for IPh beds is described in detail in Appendix F. Gamma-ray spectrum analyses using the ND4410 System are described in Appendix G.

APPENDIX E

LEACHING PROCEDURE FOR CHARCOAL BEDS

The following procedure was used to leach I-131 from TEDA-impregnated charcoal, collect the I-131 using an ion-exchange resin, elute the I-131 from the resin, extract it into CCl_4 and then into aqueous solution from which it was precipitated for counting.

Sample: 1.03 liters NAC G-618 TEDA-impregnated 8-16 mesh charcoal
Basis: HASL Milk Procedure
Reagent:

- Anion Resin - Bio-Rad A61 x 8, 100-200 mesh, CI^- form
- Sodium Hypochlorite - ~5% solution NaOCl (Purex)
- Hot distilled water - 80-90°, 6 liters
- Hydroxylamine Hydrochloride - $NH_2OH.HCl$-crystals
- Sodium Bisulfite - 1M $NaHSO_3$ 1.04g $NaHSO_3$/10 ml (fresh daily)
- Carbon tetrachloride
- Palladium chloride
- Sodium iodide - 1.0 mg I^-/ml, 1.18g NaI/100 ml
- Nitric Acid - 1.12M HNO (1:14)

Equipment:

- Leach column with stopcock
- Millipore filter assembly, 47 mm
- Ion exchange column - 42 mm i.d. x 80 mm deep
  - Separatory funnels - 250-ml and 500-ml capacity.

Procedure Steps	Remarks
1. Place the charcoal in a 2-liter glass beaker.	
2. Fill a 1000-ml graduate with hot water, ~85°C.	

Procedure Steps	Remarks
3. Add hot water slowly to the charcoal until the charcoal is covered by ~1/4 inch of water. Stir gently to remove trapped air.	
4. Let the charcoal-water slurry set for ~15 minutes. Stir gently for 30 seconds.	Prepare ion exchange column at this time.
5. Close the stopcock in the leach column, then transfer the slurry to the column. Wash the residue from the beaker to the column with the remainder of the hot water from step 3.	
6. Prepare ion exchange (Ix) column. Transfer 50 ml slurry of A61 x 8 Cl^- form resin to a modified vial (42 mm i.d. x 80 mm) with a glass wool mat in the bottom. Allow excess water to drain off.	Set up water aspirator vacuum line for use if necessary.
7. Set Ix column under leach column to receive the effluent. The effluent of the Ix column goes to the sink as waste.	
8. Drain the leaching solution at the rate of 100 ml/minute into the Ix column. Maintain the same flow through the Ix column with vacuum yet keep ~1 inch of liquid above the resin.	Do not allow either leach column or Ix column to run dry until the last one-liter wash.
9. When the level of liquid in the leach column reaches ~1 inch above the charcoal add 1 liter of hot water to the column. Maintain effluent rate at 100 ml/min.	Control flow to ±20 ml/min.
10. Repeat step #9 4 more times. All columns to drain dry on last liter.	A total of 6 liters H_2O including the load solution should be passed through the charcoal.
11. Remove Ix column.	
12. Pass 1 ml of NaI solution (10 mg I^-) through the Ix column. Discard effluent.	
13. Wash column with 50 ml of distilled water. Discard effluent. Apply vacuum to remove the last of the water.	

Procedure Steps	Remarks
14. Transfer resin to a 250-ml beaker with 50 ml sodium hypochlorite; heat in a hot water bath with stirring for 10 minutes.	
15. Filter resin-NaOCl mix through a #42 Whatman filter in a Buchner funnel with suction; transfer resin back to beaker; wash with 50 ml more NaOCl (as in step #14); filter through clean #42 filter; save filtrate; store resin.	
16. Add to filtrate 10 ml of concentrated nitric acid in 1 ml portions with stirring; heat to near boiling on hot plate; remove when chlorine evolution ceases and cool in ~20°C in a cold water bath.	
17. Put liquid in a 500-ml separatory funnel; add 50 ml CCl_4; add 1.5g hydroxylamine hydrochloride; shake slowly, relieve pressure; shake for several minutes. Let phases separate well; drain CCl_4 to a 250-ml funnel slowly, leaving interphase material behind.	
18. Add another 25 ml CCl_4 to 500-ml funnel, then add 1.0g hydroxylamine hydrochloride; shake slowly; relieve pressure; let phases separate well; add CCl_4 to previous wash, leaving interphase material behind.	
19. Add 25 ml water and 10-15 drops of fresh $1\underline{M}$ $NaHSO_3$ to the 250 ml funnel with the CCl_4; shake; if CCl_4 does not clear quickly, add more $NaHSO_3$, drain lower (CCl_4) phase to beaker with any interphase material present, drain water to a beaker or centrifuge tube.	
20. Return CCl_4 to separatory funnel with another 10 ml water; shake; discard CCl_4; combine water with previous wash.	

Procedure Steps	Remarks
21. Add 2 ml 1:14 HNO_3 to aqueous fraction, heat in water bath to expel drops of CCl_4; add 2 ml $PdCl_2$ solution, stir; cool to room temperature.	
22. Filter precipitate through a weighed ~5.0 cm Whatman GF/C glass fiber filter, wash with 10-20 ml of water. Dry, weigh, count.	

APPENDIX F

LEACHING PROCEDURE FOR IODOPHENOL BEDS

The following procedure was used to leach the 4-iodophenol containing I-131 from the beds of 4-iodophenol impregnated alumina used in the environmental radioiodine sampler. Approximately 1.2 kg of sampling material was processed in this manner. The sampling medium is 10% 4-iodophenol by weight.

Reagent:

Acetone 1.5--2.0 liters.

Equipment:

Leach column, 1-liter capacity with 1-liter reservoir
Distillation apparatus with 3-liter flask
Hot plate
2-liter Bottle
2 300-ml Beakers
2-liter Beaker
Balances

Procedure Steps

1. Weigh the sample and record the weight.
2. Place the sample in a closed container with one liter of acetone. Shake periodically, allowing about 1/2 hour for acetone to thoroughly soak through the sample.
3. Set up distillation apparatus. Transfer sample and all liquid to leach column with glass wool plug in bottom. Wash container with ∿0.5 liter acetone.
4. Start flow from leach column into distillation flask. After about 250 ml of leachate has accumulated in flask begin distillation.
5. Maintain flow from condenser at 10-15 ml/minute for at least 2 hours, transferring 200-ml batches of condensate back to leach column reservoir. The condensate should be colorless.

6. During last 1/2 hour of distillation do not recycle condensate. Wash contents of flask (∿100 ml) into counting vial with some of the condensate.
7. Let solvent evaporate at room temperatue or on warm (50°C) hot plate. The last traces of liquid can be removed by heating to 100°C for several minutes.
8. Determine yield from mass of iodophenol recovered.
9. Count the iodophenol using a calibrated Ge(Li) spectrometer.

APPENDIX G

GAMMA SPECTRUM ANALYSES USING MODIFIED ND 4410 SYSTEM WITH AUTOMATIC SAMPLE CHANGER

The gamma spectroscopy system used for the environmental samples is built around a Nuclear Data ND-4410 with a 4K memory extension and a Pertec seven-track tape transport and formatter, a Princeton Gamma Tech type LGC9SD Ge(Li) detector with a PGT R6-11 preamp and a Canberra 1416B amplifier, and an automatic sample changer. System software is based on programs supplied by Nuclear Data: IM41-1060-01 single parameter physics analyzer, IM41-1064-00 magnetic tape overlay, IM41-1108-01 peak search overlay, IM41-0085-00 Pertec magnetic tape copier, and IM41-1135-01 isotope identification overlay. Subroutines have been added to permit a channel dump from the automatic analysis routine and to identify analyses with tag and ID numbers.

PROGRAM LOADING

Programs with various configurations and isotope lists are stored on magnetic tape; select the program most suitable for desired operation. The program is loaded using the following procedure.

- Load program tape on the Pertec tape transport.
- Load Pertec tape copier program (IM-41-0085-00) into the computer memory field 3 from the paper tape reader; put teletype off line before starting program, memory location 3, 7700.
- Read in selected program.
- Rewind tape, unload; stop tape copier program.
- Ascertain that the following software modifications are in memory by examining the contents of memory locations listed below.

Memory Location	Correct Memory Content
0, 3123	3135
0, 3135	0000
0, 3136	0645
0, 3137	3674
0, 3140	6303
1, 3611	1400
1, 3771	5073
1, 3772	5473

These modifications allow peak search and nuclide ID from auto analyze mode and correct mistakes in the nuclide identification program. If not found in the memory, these changes should be loaded into these memory locations using the panel switches.

SYSTEM START OR RESTART

The following sequence is followed when starting or restarting the system.

- Press "STOP".
- Set front panel switches to 0, 0200 (octal); that is, switch 4 up, all others down.
- Press "LOAD AR".
- Press "START"; printer should respond with "*".

INITIALIZATION PROCEDURE

To initialize the system, perform the following:

- Set group width, normal value = 2048 (COMMAND G).
- Set display markers to channel 50 and channel 2048 using the display module pushbuttons.
- Set counting time, input in centiseconds (COMMAND C).
- Set peak search half-width, normal value = 3 (COMMAND L).
- Set energy tolerance on nuclide identification, normal normal value = 2 (COMMAND CTRL T).
- Set peak search sensitivity as follows:
  - -- Stop computer (press "STOP").
  - -- Examine memory location 0, 4552 (set panel switches to 0, 4552, press "LOAD AR"). If displayed content is not 0010, load that value (set panel switches to 0, 0010, press "LOAD MR").
- -- Restart system at location 0, 0200.
- Set decay time and sample size. Normal values for automatic analysis routines decay time = 0.0H and sample size = 100.0 (gives output in μCi/sample) (COMMAND CTRL N, L).
- Enter detector efficiency correction coefficients for sample type being analyzed (COMMAND CTRL E). The following are the values used for each sample type:
  - -- Particulate filter, folded and placed in sample vial: - - 1/slope = 0.8843, - offset = 1.9235.
  - -- Marinelli beaker containing IPH: - 1/slope = 0.8965, - offset = 2.9751.
  - -- Sample vial containing copper iodide precipitate from IPH leaching procedure: - 1/slope = , - offset =
- Ascertain that peak isotope library contains the following information (CTRL L).

```
MN-54 303.00D 100.00 834.80
CO-57 270.00D 85.00 122.06 136.47
CO-60 5.26Y 99.98 1173.23 1332.35
CO-58 71.30D 99.00 810.81
I-131 8.07D 82.40 364.49 284.31 637.01 722.92 80.16
I-132 2.30H 100.00 667.74 772.69 954.68 522.64 546.95 0.00
I-133 20.90H 89.00 529.89 706.65 856.29 875.36
I-134 52.00M 96.00 847.04 884.18 1072.52 1136.15
I-135 6.70H 35.00 1260.49 1131.58 1678.20
CS-134 2.05Y 98.00 604.23 569.35 795.84
CD-137 30.20Y 84.80 661.64
BE-7 53.37D 10.30 477.59
FE-59 45.10D 56.50 1099.20 1291.60
CR-51 27.80D 9.00 320.10
MO-99 66.69H 89.60 140.50 739.30 180.90
LA-140 40.26H 95.30 1596.20 487.00 815.70 328.70
TH-228 1.91Y 44.79 238.60 583.17
BA-140 12.79D 29.00 537.30
```

If not, see instructions under "APPEND COMMAND".

- Load data tape on Pertec tape transport, position past load point.
  Note: If the modifications to permit channel dump routine are in memory, the read-from-tape routine must be restored. Load paper tape marked "restore read from tape" into memory field 0 and restart the system. Set the tag or ID search numbers (COMMAND T) and perform the tape read/search operation (COMMAND H).

- Perform energy calibration by acquiring gamma spectrum from standard thorium source or NBS point source and completing the energy calibration sequence described in a subsequent section.

- Load into memory the program modifications which permit channel dump, print ID, print tag during auto-analysis sequence. These modifications are loaded into memory field using the paper tape marked "channel dump, print ID, print tag, print CR-LF". Load the parameters for the channel dump routine into memory using the panel switches. The number (octal) of the first channel to be dumped (normal value = 0) goes into location 0, 3246 and the number (octal) of channels to be dumped goes into location 0, 3247 (normal value = 2048 or 4000 octal). Restart the system.

- Set up auto-analysis string using COMMAND A, terminate the entry with the RETURN key. Briefly, typical commands and their functions are:

  - E Erases previous spectrum.
  - A Acquires spectrum for preset time.
  - I Increments ID number.
  - C Writes spectrum on tape after incrementing tag number.
  - L Prints ID number.
  - G Prints tag number.
  - J Performs peak search and nuclide identification.
  - B Performs channel dump.
  - K Spaces printer 2 lines.
  - H Repeats auto-analysis sequence until tag or ID equals preset value.

A normal command string would be: EAICKLKGKJKBKKKH

- Set the counting time in centiseconds (COMMAND C).
- Set the last ID or tag number for auto-analysis routine (COMMAND T).
- Load sample changer, put on line (see automatic sample changer operating procedures).
- Begin analysis (COMMAND X).

AUTOMATIC SAMPLE CHANGER OPERATING PROCEDURES

The "POWER" button is used to turn the power supply on and off. When the power is on the "POWER" button light will also be on. The "ON LINE" button is used to select manual or computer controlled operation of the sampler changer. When the system is under computer control, the indicator light will be on. When the system is in the manual operation mode, the indicator light will be off. The "CYCLE" button initiates insertion or withdrawal of the sample slide when manual operation is permitted. The button indicator light is turned on during the sample change operation. To operate the sample changer, do the following:

- Depress the "POWER" button to turn the power on.
- If the sample slide is not fully inserted to sample analysis configuration, depress the "CYCLE" button to insert slide.
- With the sample slide in the analysis configuration, load the samples into the vertical storage and feed apparatus in the order in which they are to be counted (first sample on bottom, last sample on top). Ascertain that the sample vial lids are properly and uniformly applied.
- Place one empty sample holder on top of the sample stack. Place one weighted sample holder on top of the empty sample holder.
- Close access panel and latch.
- Depress the "CYCLE" button; wait for sample slide to withdraw fully.
- Depress the "CYCLE" button; wait for the first sample to reach analysis configuration.
- Depress the "ON LINE" button; "POWER" and "ON LINE" indicator lamps should be lit. Samples will now change automatically at the end of each data acquisition period.
- Start the computer auto-analysis sequence. After all samples have been analyzed, the empty sample holder should be left in the analysis configuration. Remove the weighted holder from the storage and feed apparatus. Depress the "ON LINE" button to permit manual operation, then depress the "CYCLE" button to eject the empty holder from the sample changing mechanism. Turn the power off by depressing the "POWER" button.

DESCRIPTION OF ADDITIONAL SOFTWARE

Software modifications have been made to permit an analyzer channel dump and printout of tag and ID numbers from the auto-analyze mode. As noted above these changes prevent the read from tape operation unless that capability has been restored. The programs occupy memory locations: 0, 3121 through 0, 3146; 0, 3171 through 0, 3223; and 0, 3246 through 0, 3342. Procedures for using these additional capabilities are as follows:

- The channel dump routine is entered with command "B" in the auto-analyze command string. The channel number (in octal) for the start of the dump must be placed in memory location 0, 3246 and the total number of channels to be dumped is placed in 0, 3247. The output format for the channel dump is: C1 D1 D2 D3 D4 D5 D6 D7 D8 D9 D10 D11 etc. where C1 is the channel number of data point D1, D2-D8 are the contents of next seven channels, C9 is the channel number of the first channel in the second line of data, D9-D16 are the contents of that and the seven succeeding channels, and so on. All the output is decimal.
- The print ID number routine is entered with command "L" in the auto-analyze command string. The output is the current sample tag number.
- The print tag routine is entered with command "G" in the auto-analyze command string. The output is current sample tag number.
- The print carriage return-linefeed routine is entered with command "K" in the auto-analyze command string. The output is two "CR-LF" commands to printer.

HARDWARE ENTRY COMMAND SUMMARY

The hardware commands and their functions are summarized below.

- Direction ( ) switch: stipulates direction of display, marker, counts full scale, or group movement.
- Motion pushbutton: moves display horizontally in the direction specified.
- Width pushbutton: horizontally expands or contracts display.
- Mark POS pushbutton: moves markers in direction specified.
- Mark SPAN pushbutton: moves right marker in direction specified, increasing or decreasing the number of channels between the markers.
- CFS pushbutton: increases or decreases the counts full scale value in binary increments. Selects logarithmic display after reaching the maximum or minimum counts full scale value.
- Print (WRITE) pushbutton: increments the current tagword by one and writes the current group on magnetic tape.
- Status pushbutton: sequentially displays: (1) center pointer channel and content and left-right marker channels; (2) current group number and total number of groups, current group width, and

counts full scale value; (3) remaining/elapsed and present analysis time; or (4) current ID number/tagword number, elapsed acquisition time, and group size of last group read/group size of current group.

- Display pushbutton: alternately selects live or static display.
- Groups pushbutton: sequentially selects groups for data storage and display in direction specified.
- Expand pushbutton: expands the marker defined display to horizontal full scale.
- Acquire pushbutton: alternately starts or stops data acquisition.
- Return pushbutton: reinitializes the program and returns to display.
- Erase pushbuttons: clears currently displayed data group to zero.
- INTEG/DIFF (READ) pushbutton: reads the next file on magnetic tape into the current group.
- Total (REWIND) pushbutton: rewinds the magnetic tape to the beginning of tape (BOT) marker.

KEYBOARD ENTRY COMMAND SUMMARY

The functions which are controlled by keyboard commands are described below.

- Clock Set Command: permits user to enter a preset analysis time in centiseconds (0.01 second). Maximum analysis time is 2^{23}-1 centiseconds (83,886.07 seconds); entering 0 selects the maximum analysis time.
- Group Set Command: permits user to enter the number of channels per group. The group size can be any number from 1 to the maximum number of channels allllocated for data storage.
- L, Half-Width Command: permits entry of a number equal to the average number of channels at the full-width at half-maximum FWHM points of the peaks to be determined during the peak extraction/fit process. The peak width can be varied from 2 to 85.
- Peak Find/Fit Command: determines the centroid, FWHM, background, and intensity information for each peak contained within the marker-defined portion of the spectrum stored in the currently displayed group using a peak extraction process which uses the entered half-width parameter. Upon extraction of each peak, markers are displayed at the peak limits and the following information is printed at the teletype: the number of the extracted peak, an error code list number (if applicable), the channel location of the peak centroid, the number of channels at the full-width-half-maximum (FWHM) point, the background counts, the net total (the total number of counts within the peak limits minus the background counts), and the channel locations of the left and right limits of the peak. Upon completion of printout of the information determined for each peak during the peak extraction process, the peak fit process is initiated. The fit process first determines if the peak found is a single peak or doublet. If the peak is found to be a single peak, a least squares Gaussian fit process is performed using the centroid, FWHM and height values found by using the peak extraction process as initial guesses. Upon completion of the fit process,

markers are again displayed at the peak limits and values for the centroid, FWHM, height and area of the Gaussian are printed at the teletype. If the peak is found to be a doublet, pairs of input guesses are generated for the double peak which are based upon the skewness of the peak, the centroid, FWHM, height, left and right channel values found by the peak extraction process. The pairs of guesses are then used as input to the peak fit process which separates and fits the doublet, improving the initial guesses by an iterative least squares Gaussian fit process. Upon completion of the iterative fit process, markers are again displayed at the peak limits and new values for the centroid, FWHM, height and area for each peak arepprinted at the teletype. The number of iterations is also printed.

- Energy Calibrate Command: permits entry of the half-width and then performs the peak extraction/fit process, marker display and printout in the same manner as the Peak Find/Fit command. In addition, it permits entry of the known energy in eV for any two peaks and then calculates the values for the energy intercept in eV and the energy per channel (slope) in eV/channel.
- $, Smooth Command: performs a five-point coefficient smooth on the data spectrum contained in the currently displayed group a specified number (1 to 4095) of times.
- CTRL A: appends additional entries to the isotope library.
- CTRL D: deletes specified entry from the isotope library.
- CTRL E: enters slope and offset for detector efficiency correction curve.
- CTRL K: kills (deletes) entire isotope library.
- CTRL L: lists isotope library including any entries appended by CNTL A.
- CTRL N: performs peak search and printout functions for all peaks found; calculations and ID printouts are for representative isotope peaks identified from library list.
- CTRL T: defines allowable difference between a calculated peak energy and a library energy within which identification will still be made.
- Set ID Number Command, (*J): permits user to enter an identification number for a file on magnetic tape. Any number from 0 to 8,388,607 can be entered.
- Input Search Tag, ID Command (*T): permits user to enter a tagword and an identification number for comparison with the tagword and identification numbers read-in from magnetic tape during the Compare Tag, ID Command. Any number from 0 to 8,388,607 can be entered for either number. Entering 0 for either number causes that number to be ignored during the Compare Tag, ID Command.
- Compare Tag, ID Command (*H): initiates a magnetic tape read/search operation. During this operation, sequential files are read from magnetic tape into the current group and a comparison is made between the tagword and identification numbers entered for the Input Search Tag, ID Command and those being read from the identification record of each file. When the numbers match, read-in is terminated at the end of the file in which the match occurred.

- Erase, acquire, write command (*K): clears the current group to zero, initiates analysis in the current group, and, upon completion of the preset time, it increments the current tagword number by one and writes the current group on magnetic tape. The preset time is entered using the Clock Set Command.
- Backspace Command (*B): permits backspacing over one file on magnetic tape.
- Increment ID Number Command (*I): permits user to increment the current identification (ID) number by one.
- Execute Auto-Analyze Command (*X): initiates the sequence of operations specified for the Auto-Analyze Command.
- Auto-Analyze Command (*A): permits entry into the auto-analyze command mode. Upon entry into this mode any logical sequence of the following commands can be entered. After entry of the last command the sequence is terminated by depressing the RETURN key of the teletype.
  - Acquire Command (A): when this command is encountered in the auto-analyze sequence, analysis is inititated in the current group. Upon completion of the preset analysis time, the next command in the sequence is initiated. The preset analysis time is entered using the Clock Set Command.
  - Channel Dump Command (B): prints data in channels starting at channel number in memory location 0, 3246; number of channels printed at location 0, 3247.
  - Write Command (C): when this command is encountered in the auto-analyze sequence, the current tagword number is incremented by one and the current group is written on magnetic tape.
  - Rewind Command (D): when this command is encountered in the auto-analyze sequence the magnetic tape is rewound to the beginning of tape (BOT) marker.
  - Erase Command (E): when this command is encountered in the auto-analyze sequence, the current group is cleared to zero.
  - Filemark Command (F): when this command is encountered in the auto-analyze sequence, a filemark is written on the magnetic tape.
  - Compare Tag, ID Command (H): when this command is encountered in the auto-analyze sequence, the current tagword and identification numbers are compared with the tagwordaand identification numbers entered for the Input Search Tag, ID Command. If the numbers match, the next command in the sequence is initiated. If the numbers do not match, the entire sequence prior to the Compare Tag, ID Command is repeated.
  - Increment ID Number Command (I): when this command is encountered in the auto-analyze sequence, the current identification number is incremented by one.
  - Peak Search and ID Command (J): when this command is encountered in the auto-analyze sequence the system will perform the peak search and nuclide identification routines. (Perform command CNTRL N, L to set the decay time and sample size before execution of an auto-analyze string including the J command.)

-- Print CR-LF (K): prints 2 carriage return-linefeed characters.

-- Print ID (L): prints current sample ID number.

-- Print Tag Number (G): prints current sample tag number.

DESCRIPTION OF SOFTWARE OPERATIONS

Some important system software operations are described in more detail in this section.

- Energy Calibrate Command: the Energy Calibrate Command permits entry of the half-width and then performs the peak extraction/fit process, marker display and printout in the same manner as the Peak Find/Fit Command. In addition, it permits entry of the known energy in eV for any two peaks and then calculates the energy intercept (A0) and the energy per channel (A1) in eV/channel. Once the energy calibrate command has been executed, the centroid and FWHM values printed during any ensuing peak extraction/fit process, will be in KeV and will be based on the last two known energy values entered. The Energy Calibrate Command is specified by typing E after an asterisk (*) is typed. When E is typed, the routine causes the teletype to print ENERGY CALIBRATE, perform a carriage return/linefeed and print HW:. When HW: is printed, the routine waits for entry for the half-width. The half-width can be any number from 2 to 85. Entry of the half-width must be terminated by depressing the SPACE bar at the teletype. When the SPACE bar is depressed, the routine causes the teletype to perform a carriage return/linefeed, print the column headings NO., ECL, CENTROID, FWHM, BKG(HGT), AREA, LEFT and RIGHT, perform another carriage return/linefeed. The routine then performs the peak extraction/fit process for the first peak. Upon completion of the extraction/fit process for the first peak, the routine displays markers at the left and right limits of the peak and prints the respective information at the teletype. After the fit process value under the AREA column is printed, the routine causes the teletype to print E(EV):, and then waits for entry of a known energy value in eV. If the first peak is one of the known peaks for which an energy value is to be entered, enter the energy value after the E(EV): is printed. If not, enter 0. Entry of an energy value or zero (0) must be terminated by depressing the SPACE bar at the teletype. When the SPACE bar is depressed the routine causes the teletype to perform a carriage return/linefeed, and then performs the peak extraction/fit process for the next peak. Upon completion of the extraction/fit process for the next and each succeeding peak, the routine displays markers at the peak limits, prints values for each column heading and E(EV):, and then waits for entry of a known energy value. An energy value can be entered for any two peaks determined during the peak extraction/fit process with zeroes entered for all other peaks. The energy value can be any number from 1 to 8,388,607 eV. When the SPACE bar is depressed to terminate entry of an energy value for the second known peak, the routine causes the teletype to perform a carriage return/linefeed, print A0 = and the value of the energy intercept in eV, perform another carriage return/linefeed, print A1 = and the value of the slope (energy per channel) in eV/channel, performs another carriage return/linefeed, and type an asterisk (*), signifying return to the command mode. To return to the uncalibrated mode once the Energy Calibrate Command has been performed, change the contents of location 0, 5722_8 to 0000_8.

- Peak Find/Fit Command: (Note: Prior to performing the Peak Find/Fit Command, the Half-Width Command should be performed to enter the half-width. The half-width is initially set to 4 by the program.) The Peak Find/Fit Command determines centroid, FWHM and intensity information for each peak contained with the marker defined portion of the spectrum stored in the currently displayed group. Each peak is determined using a peak extraction process which is based upon the entry for the half-width parameter. During the peak extraction process, the display is disabled. Upon extraction of each peak, the display is re-enabled with markers at the left and right limits of the peak and the following information is printed at the teletype: the number of the extracted peak (from left to right), an Error Code List (ECL) number (if applicable), the channel location of the peak centroid, the number of channels at the Full-Width-Half-Maximum (FWHM) point, the background counts, the net total counts (the total number of counts in the area defined by the peak minus the background counts), and the channel locations of the left and right minimum points of the peak. The Error Code List number indicates three characteristics of peak extraction process. ECL #1 indicates that the slopes on either side of the peak do not meet the test criteria either because the half-width entered is smaller then the half-width of the extracted peak, the extracted peak represents a compton edge or the extracted peak represents one or more peaks. ECL #2 indicates the left portion of the extracted peak falls beyond the first channel of the group. ECL #3 indicates the right portion of the extracted peak falls beyond the last channel of the group. Upon completing printout of the information determined for each peak during the peak extraction process, the peak fit process is initiated. During the peak fit process, the display is disabled. The peak fit process first determines if the peak found during the peak extraction process is a single peak or doublet. If the peak is a single peak, a least squares Gaussian fit process is performed which uses the centroid, FWHM and height values found by the peak extraction process as initial guesses. Upon completing the fit process, the display is re-enabled with marker at the left and right limits of the peak and values for the centroid, FWHM, height and area of the Gaussian are printed at the teletype. If the peak is a doublet, pairs of input guesses are generated for the double peak which are based upon the skewness of the peak, the centroid, FWHM, height, left and right channel values found by the peak extraction process. The pairs of guesses are then used as input to the peak fit process which separate and fit the doublet, improving the initial guesses by an iterative least squares Gaussian fit process. Upon completing the iterative fit process, the display is re-enabled with markers at the left and right limits of the peak and fit values for the centroid, FWHM, height and area for each of the two peaks are printed at the teletype. The number of iterations is also printed. The Peak Find/Fit Command is specified by typing P after an asterisk (*) is typed. When P is typed, the routine causes the teletype to perform a carriage return/linefeed, print the column heading NO., ECL, CENTROID, FWHM, BKG(HGT), AREA, LEFT and RIGHT, and perform another carriage return/linefeed, and then performs the peak extraction process for the first peak. During the peak extraction process the displayed is disabled. Upon extraction of the first peak, the routine re-enables the display with markers at the left and right limits of the peak and prints the values under each column heading at the teletype. Upon completion of printout of the values determined during the peak extraction process for the first peak, the routine initiates the peak fit process. During the peak fit process the display is disabled. Upon completion of the peak fit process the routine re-enables the display with markers at the left and right limits of the peak and prints either one or two lines of values under the column heading: CENTROID, FWHM, BKG(HGT) and AREA. If the peak is a single peak only one line is printed. If the peak is a doublet, two lines are printed with the number of iterations (10, maximum) printed in the second line under the column heading: ECL. Upon completion of printout of the values determined for the first peak, the routine causes the teletype to perform a carriage return/linefeed, performs the peak extraction process for the next peak, displays markers at the peak limits, prints the values determined by the peak extraction process, performs the peak fit process, again displays markers at the peak limits, prints the values determined by the peak fit process and then causes the teletype to perform a carriage return/linefeed and type an asterisk (*), signifying return to the command mode.

- Peak Sensitivity Adjustment: the sensitivity level of the program is set at a nominal value for peak extraction using spectra with average background content. The nominal value is defined by the content of location 0, 4552_8 and is initially set to 0020_8. By altering the contents of this location, the sensitivity of the peak extraction process can be increased or decreased, accordingly. To increase sensitivity, i.e., to extract peaks that slightly exceed the average background content; the content of location 0, 4521_8 can be changed to any octal value less than 0020_8, but not 0000_8 since zero is an unacceptable value. Maximum sensitivity would be obtained using a value of 0001_8. To decrease sensitivity, i.e., to extract only large peaks; the content of location 0, 4552_8 could be changed to a value of 0021_8 or greater.

- Smooth Command: the Smooth Command permits performing a five-point coefficient smooth on the data spectrum contained in the currently displayed group to reduce the deviation of counts in the spectrum peaks or to compensate for poor statistics. The smooth operation can be performed from 1 to 4095 times. The following is an example of the entry required to perform a single smoothing operation on the spectrum contained in the currently displayed group: *$:1(SPACE). (Note: The Peak Find/Fit and Smooth Commands cannot be performed simultaneously. Once the smooth operation is initiated, it must continue until completed before entering the Peak Find/Fit Command, or vice versa.)

- Peak Search and Nuclide Identification: the Peak Search and Nuclide Identification can be performed only from the auto-analyze mode (COMMAND J in the auto-analyze string). Perform the command CNTRL N, L to define the decay time (in seconds, minutes, hours, days or years) and the sample size. The printout includes two sections: the peak find and fit section, and the isotope identification and calculation section. In the peak find and isotope identification sections the definitions of the headings are as follows:

- PK, Sequential number of peak from left to right within marker area.
- E, Error number indicating failure to perform peak fit. error numbers and their definitions are:

  1, slopes on either side of peak do not meet test criteria, either because FWHM value entered is smaller than the FWHM of the extracted peak, the peak represents a Compton edge, or the extracted peak represents two (doublet) or more peaks. Note: If a doublet is detected and a fit is attempted, printout in the E column for the next peak indicates the number of iterations attempted. A zero indicates 10 iterations. 2, left portion of extracted peak falls beyond the first channel of current display group. 3, right portion of extracted peak falls beyond the last channel of current display group. 4, no convergence was accomplished during peak fit attempt. Values printed are weighted moment values rather than fitted values.
- LEFT, Channel location of left limit of peak.
- MW, Marker width-difference (in channels between left and right markers.
- CENTROID, Channel location of peak centroid.
- FWHM, Full-width half-maximum of peak.
- BKGND, Number of background counts within peak area.
- AREA, Net area (total counts minus background counts) of peak.
- %ER, Calculated fractional error.
- CTS/SEC, Counts per second calculated from net area and analyzer time.
- ENERGY, Energy in KeV of peak centroid.
- ISOTOPE, Isotope identified based on energy lines found during peak search/fit. Obtained from isotope ID library.
- HLIFE, Half-life of isotope obtained from library; listed in minutes (M), hours (H), days (D), or years (Y).
- DECAY, Calculated value proportioned to the amount of decay in seconds (counts per disintegration). Calculation is based on sample time, measurement time, clock time, live time, and half-life.
- P, Number of peaks found of that isotope.
- ENERGY, Energy line in KeV of isotope obtained from library.
- %ABN, Percent abundance for that line of the isotope in relation to total decay events.
- EFF, Efficiency of detector at energy line indicated.
- UC/US, Activity in microcuries per unit volume.
- CONF-INT, Confidence factor in microcuries per unit volume.

- Energy Tolerance Command: the energy tolerance command permits entry of a value of KeV which represents an acceptable tolerance (±) between the calculated energy and the actual energy listed in the isotope library. The command is initiated by entering CTRL T.

The program responds by printing *ENERGY TOLERANCE and awaiting an entry. The entry is terminated by SPACE. (If CTRL T is not used, ENERGY TOLERANCE defaults to 1 KeV.)

- Append Command: entries may be added to the isotope peak library via the Append Command. Each added entry is automatically added to the end of the library and assigned the next sequential line number in the library. Also, properly formatted ASCII tapes of the isotope library may be read in during execution of the Append Command by loading the tape in the teletype reader and setting teletype START/FREE/STOP switch to START. The Append Command is initiated by entering CTRL A. After CTRL A is entered, the program responds by performing a carriage return/linefeed and typing a left backet ([). An isotope peak entry may then be entered using the following format. If a tape is being read in, entries are automatically typed as the tape is read in. Example: *[Isotope name (SPACE)Half-life(SPACE)Abundance(SPACE)Energy(RETURN)

  The isotope name is entered as the one or two letter symbol of the element followed by a dash and the atomic weight of the isotope. An "M" following the atomic weight indicates metastable state. Half-life is entered in time units of minutes, hours, days, or years. The entry for half-life is a decimal number followed by the unit of time (M, H, D, or Y, respectively). Abundance is entered as a percentage and indicates the per-cent abundance of the representative isotope peak being entered. (The representative peak is the peak for that isotope that the system will detect.) Energy is entered in KeV and is the energy of the representative isotope peak. (Multiple energy entries (up to seven) may be entered, but only the first one entered is considered to be representative. Only if that peak is found are calculations made for that isotope.) Library entry is terminated by depressing RETURN key. When entries are being made in the library, RUBOUT key may be used to delete only a current entry: the isotope entry, half-life, abundance, or one of the energies. Back arrow key (←) may be used to delete the entire line. The program then automatically performs a carriage return/linefeed, repeats the left bracket, and awaits a new entry. If no entry had been started on the line when ← is depressed, the program exits append mode and returns to command mode (types asterisk).

- Delete Command: entries may be deleted from the isotope peak library via the Delete Command. An entry to be deleted is specified by its position number in the library. For example, to delete the sixth entry from the library supplied with the program, entry 6 is specified in the delete command. The next library entry then automatically becomes entry 6; all subsequent existing entries are likewise decremented and reassigned. The Delete Command is initiated by entering CTRL D. The program responds and the indicated entry number must be entered and terminated by SPACE. A sample printout for deletion of entry 6 is *DELETE:6(SPACE).

- Kill Command: the entire isotope peak library may be deleted via the Kill Command. The Kill Command is initiated by entering CTRL K. The program responds by typing KILL? and awaits an entry of Y (yes) to verify deletion of all isotopes in the library. Any other response results in return to command mode (asterisk) without any deletion.

- Detector Efficiency Command: the Detector Efficiency Command permits entry of the slope and offset values for the detector efficiency correction curve. The slope and offset of the logarithmic equation for efficiency are used to compensate for detector efficiency. The Detector Efficiency Command is initiated by entering CTRL E. The program responds by printing the following, requesting, in sequence, entries for the negative inverse of the slope and for the negative of the offset. (If CTRL E is not used -1/SLOPE defaults to 753.296 and -OFFSET defaults to 2.708.)

```
*LOG EFFICIENCY CURVE
-1/SLOPE = _______________(SPACE)
-OFFSET  = _______________(SPACE)
```

APPENDIX H

PROCEDURE FOR PROCESSING PRECIPITATION SAMPLES

This procedure was adapted from the procedure developed by the Cincinnati Laboratory of the Environmental Protection Agency.

Rain samples are collected in a polyethylene bottle containing 200 ml of 1M mercaptoacetic acid to assure collection of bound iodine scavenged by the precipitation. The sample container was sealed, labeled, and returned to the laboratory for analysis according to the following procedure.

Reagents:

The following reagents are used in this procedure.

Anion exchange resin - Dowex-1x8
Hydrochloric acid, HCl 12N (conc.), 6N
Hydroxylamine hydrochloride, $NH_2OH \cdot HCl$: 1M
Iodate carrier: 10 mg/ml as IO_3^-
Mercaptoacetic acid, HS_2COOH: 1M
Nitric acid, HNO_3: 16N (conc.)
Palladium chloride, $PdCl_2$: 0.2M
Sodium chloride, NaCl: 2M
Sodium hypochlorite, NaOCl: 5%
Sodium sulfite, Na_2SO_3: 1M
Toluene, $C_6H_5CH_3$: reagent grade

Procedure:

1. Transfer the precipitation sample to a 15-liter bucket, add 1 ml of iodate (IO_3^-) carrier (10 mg/ml). Stir for 5 minutes, add 12N HCl to pH 3 (use pH meter). Stir for 15 minutes.
2. Prepare an ion exchange column using 100 cm^3 of Dowex-1 anion resin to fill a plexiglass tube 4 cm in diameter by 8 cm high. Pass the solution through a Dowex-1 ion exchange column at a flow rate of 100 ml/minute. After the sample has passed through column, rinse column with 400 ml of 2M NaCl solution at a flow rate of 20 ml/minute.

3. Disconnect ion exchange column and transfer the resin to a 500-ml beaker. After the resin has settled, pour off any excess liquid.
4. Add 50 ml of 5% NaOCl solution. Stir the mixture with a stirring motor and teflon stirring bar for 5-10 minutes.
5. Filter solution through a sintered glass funnel retaining the liquid for analysis.
6. Repeat steps 4 and 5 twice. Discard residue.
7. Acidify the aqueous solution with 16$\underline{N}$ HNO_3, to pH <1. The HNO_3 is to be added carefully and should be done in the hood because of the vigorous evolution of Cl_2.
8. Transfer the solution to a 500-ml separatory funnel, add 50 ml toluene, 10 ml of 1$\underline{M}$ $NH_2OH{\cdot}HCl$, and shake for two minutes.
9. Drain aqueous phase into a 500-ml separatory funnel containing 20 ml of toluene, add 5 ml to 1$\underline{M}$ $NH_2OH{\cdot}HCl$, and shake for two minutes.
10. Combine toluene from steps 8 and 9 in one separatory funnel, wash with 20 ml water, and discard the aqueous rinse.
11. Add 10 ml water, and 5 drops of 1$\underline{M}$ Na_2SO_3 to the separatory funnel and shake until the toluene is clear. (If toluene is still pink, add a few additional drops of NA_2SO_3 to completely reduce I_2.) Drain aqueous phase into a 40 ml centrifuge tube.
12. Add 10 ml water to separatory funnel and 5 drops of 1$\underline{M}$ Na_2SO_3, and shake for two minutes. Add the 10 ml of aqueous solution to the centrifuge tube, acidify with 6$\underline{N}$ HCl to pH 1, heat in a water bath for 5 minutes.
13. Add 1 ml of 0.2$\underline{M}$ $PdCl_2$ to precipitate PdI_2, heat for 5 minutes in hot water bath.
14. Filter PdI_2 onto a tared 0.8-μm membrane filter. Gelman Metricel filters GA-4 have been satisfactory for this operation. Wash twice with 5-ml portions of water. Dry at 70°C in a partially covered petri dish for 30 minutes and weigh for yield determination.
15. Mount for counting.

APPENDIX I

PROCEDURE FOR PROCESSING GRASS SAMPLES

This procedure was adapted from the procedure developed by the Cincinnati Laboratory of the Environmental Protection Agency. A sample of fresh grass to which iodide carrier has been added is digested to a dried pulp in a caustic medium to prevent loss of iodine. After ashing until uniform at 600°C, the sample is acidified and iodine is distilled into caustic solution. The distillate is acidified and the iodine is extracted into CCl_4. After back extraction, the iodine is precipitated as PdI_2 for counting.

Reagents:

The following reagents are required for processing of the vegetation samples.

Carbon tetrachloride, CCl_4
Hydrazine sulfate, $(NH_2)_2 \cdot H_2SO_4$: saturated
Hydrogen peroxide, H_2O_2: 30%
Iodide carrier: 10 mg/ml
Nitric acid, HNO_3: $6\underline{N}$
Palladium chloride, $PdCl_2$: $0.2\underline{M}$
Sodium bisulfite, $NaHSO_3$: $1\underline{M}$
Sodium hydroxide, NaOH: $4\underline{N}$, 10%
Sodium hydroxide, NaOH: $0.5\underline{N}$
Sodium hypochlorite, NaOCl: 5%
Sodium nitrate, $NaNO_3$: solid
Sodium nitrite, $NaNO_2$: $1\underline{M}$
Sulfuric acid, H_2SO_4: $36\underline{N}$ (conc.), $12\underline{N}$

Procedure:

1. Divide the freshly cut grass sample (up to 1000 g wet weight) into as many large stainless steel evaporating pans (12" x 6" x 4") as needed. Dissolve 200 g NaOH in 2000 ml water. Add 1.0 ml iodide carrier into the solution and mix. Pour the solution uniformly

over the grass in each pan. Digest on hot plate until all the grass can be combined in a single pan. (Note 1).

2. Evaporate until the sample is completely dry. (Note 2).
3. Ash in a muffle furnace for 1 hour at 600°C. Remove from furnace and cool.
4. Grind the ashed lumps in a grinder, or pour into a large mortar and grind the lumps with a pestle until uniform. Return ash to the pan and mix thoroughly with 10-15 g $NaNO_3$. Ash in muffle furnace for an additional 15 minutes at 600°C. Remove from furnace and cool.
5. Repeat step 4 as often as needed until the ash is a homogeneously light gray in color, free of carbon.
6. Let cool to room temperature. Gently break up the ash. Transfer with spatula and powder funnel to a 3-l single-neck, ground glass distillation flask, containing ~700 ml distilled water. (Note 3).
7. Assemble a condenser, separatory funnel and 250-ml erlenmeyer trap containing 20 ml $4\underline{M}$ NaOH. Connect to the flask and circulate water through condenser.
8. To the contents of the flask add 50 ml 30% hydrogen peroxide from the separatory funnel. Mix well using magnetic stirrer, then add 350 ml $18\underline{N}$ sulfuric acid dropwise (~10 ml/min.) stirring continuously while acid is being added to prevent boil over. (Note 4). Remove stirrer and replace with burner.
9. Distill iodine at a full rolling boil until reaction is complete. (Note 5). Remove heat, then add 10 ml $36\underline{N}$ H_2SO_4, 10 ml saturated hydrazine sulfate, and 10 ml 50% H_2O_2 in sequence from the dropping funnel. (Note 6). Distill for another 5-10 minutes to rinse the condenser. Discard solution in flask.
10. Transfer distillate to a beaker and add 5 ml NaOCl. Boil for 5 minutes, then cool to room temperature.
11. Using a pH meter, adjust the distillate to pH 2 with $6\underline{N}$ HNO_3 then add 2-5 ml $1\underline{M}$ $NaHSO_3$ a drop at a time to reduce IO_3^- to I^-. Transfer to a 250-ml separatory funnel.
12. Add 3-5 ml $1\underline{M}$ $NaNO_2$ to oxidize the iodide to free iodine. Mix well. Add 5 ml CCl_4 and shake for 1-2 minutes. Draw off organic layer into a clean 60-ml separatory funnel containing 10 drops $1\underline{M}$ $NaHSO_3$ and 5 ml water.
13. Add 5 ml CCl_4 and 1 ml $1\underline{M}$ $NaNO_2$ to original separatory funnel containing the aqueous layer and shake for 1-2 minutes. Combine organic layer with that in separatory funnel in step 12.

14. Repeat step 13 and discard the aqueous layer.
15. Shake separatory funnel until CCl_4 layer is decolorized, let phases separate and transfer aqueous layer to a centrifuge tube.
16. Add 5 drops 1M $NaHSO_3$ and 5 ml water to separatory funnel and shake several minutes. Allow phases to separate and add aqueous layer to centrifuge tube from step 15.
17. Add 1 ml water to separatory funnel and shake several minutes. When phases separate add this aqueous layer to centrifuge tube. Discard the organic layer.
18. Add 1 ml of 6N HCl, then heat in a water bath several minutes to expel CCl_4 and SO_3.
19. Add 2 ml 0.2M $PdCl_2$, heat gently in a water bath, stirring to aid coagulation. Cool.
20. Filter through a tared 0.8-μm pore size membrane filter and wash with 20 ml water.
21. Dry at 70°C for 30 minutes, cool, weigh, mount, and count.

Notes:

1. After solution starts to boil, push the grass down until it is covered. Add water if necessary. Careful observation is necessary at this point, because sample will boil over if a channel is allowed to form. It is advisable to keep poking the grass with a heavy glass rod to prevent this. The initial digestion is performed at a medium high setting on the hot plate. After combining sample into one pan, reduce heat to medium to prevent sample loss due to spattering.
2. This drying process can also be completed by overnight drying in an oven set at 175°C.
3. Add sample to the water in the flask with stirring. Wash tray with additional water and add rinsing to the flask.
4. Add enough sulfuric acid to neutralize the sample, then add 50 ml in excess. Boil rapidly for 10 minutes after all acid has been added.
5. Usually, distillation is complete when no more brown iodine vapors are evident, but even so, the reaction may not be complete because other valence states of iodine could still be present which are not distilled.
6. Acidifying the sample and adding hydrazine sulfate will reduce iodine to iodide. The hydrogen peroxide oxidizes it to I_2 which is now distilled over as the condenser is rinsed.

Reagent Preparation:(1)

1. Hydrazine sulfate, saturated. Dissolve 4g $(NH_2)_2 \cdot H_2SO_4$ in 100 ml boiling water.
2. Palladium chloride, 0.2M. Dilute 71 ml of analytical grade $PdCl_2$ (5% solution) to 100 ml with water.
3. Sodium nitrate, $NaNO_3$ solid.
4. Zinc, powdered, analytical grade.

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(1) Krieger, H. L. and S. Gold, Procedures for Radiochemical Analysis of Nuclear Reactor Aqueous Solutions, Environmental Monitoring Series, EPA-R4-73-014, NERC-Cincinnati, May, 1973.

APPENDIX J

TABULATIONS OF RADIOIODINE SPECIES AND RELEASE RATES MEASURED AT QUAD CITIES

TABLE J-1

QUAD CITIES RADIOIODINE SPECIES BY SAMPLING PERIOD

Sampling Period	1977 Dates		Fraction (%) of Total Release Rate in Each Form							
			Main Chimney				Combined Reactor Building Vents			
	Start	End	Partic.	I_2	HOI	Organic	Partic.	I_2	HOI	Organic
1	6-15	6-25	8.3	51.2	21.3	19.2	5.6	10.8	58.2	25.4
2	6-25	7-7	11.3	55.2	25.0	8.5	1.7	9.1	38.8	50.3
3	7-7	7-15	17.2	50.9	23.9	8.1	2.7	8.3	37.6	51.4
4	7-15	7-26	13.3	46.7	28.6	11.4	8.6	23.7	23.7	44.1
5	7-26	8-4	4.3	41.2	29.5	25.1	0.3	10.0	54.8	34.9
6	8-4	8-16	6.8	44.9	27.8	20.5	7.1	22.2	53.2	17.5
7	8-16	8-26	20.8	60.8	14.5	3.9	2.3	32.6	27.3	37.7
8	8-26	9-5	4.0	58.5	18.4	19.2	6.2	14.1	53.7	26.1
9	9-5	9-14	6.3	35.7	41.4	16.5	< 0.1	4.8	58.2	36.9
10	9-14	9-23	3.4	41.3	30.8	24.5	2.2	24.2	29.0	44.6
11	9-23	9-30	< 1.3	< 3.0	20.3-21.1	75.5-78.9	3.7	23.9	32.8	39.6
12	9-30	10-12	3.6	24.8	14.1	57.5	4.7	19.7	20.8	54.9
13	10-12	10-21	5.3	14.0	19.4	61.3	6.7	25.8	11.8	55.7
14	10-21	10-28	2.0	27.6	14.6	55.8	16.5	19.4	8.1	56.0

TABLE J-2

AVERAGE DAILY I-131 RELEASE RATES FROM THE QUAD CITIES REACTOR BUILDING VENT

	I-131 Release Rates (nCi/sec)				
Day	June	July	August	September	October
1		4.28	2.67	2.26-2.33	3.21
2		3.41	2.34	1.66-1.71	3.54-3.58
3		4.02	2.11	3.10-3.17	3.02-3.06
4		5.37	3.86	3.38-3.45	3.88-3.92
5		3.71	1.87	2.62-2.69	3.87-3.91
6		2.94	1.67	4.14	4.01-4.05
7		16.6	2.00	5.75	4.26-4.30
8		2.99	1.54	3.44-3.51	4.10-4.14
9		3.42	2.56	3.10-3.17	3.65
10		3.01	2.71	2.93-3.00	4.24
11		3.32	1.97	2.52-2.59	5.29
12		2.92	1.78	2.09	9.97
13		2.87	2.40	1.77	5.92
14		3.58	1.62	1.97-2.01	3.14
15	12.8	2.31	1.89	1.61-1.65	3.84
16	12.4	3.13	2.03	0.835-0.869	3.36
17	1.41	3.33	1.35-1.42	2.25-2.29	3.96
18	0.976	3.11	1.02-1.09	1.69-1.72	3.45
19	0.996	3.26	1.01	1.81-1.85	2.55
20	0.993	3.45	1.21-1.27	2.03-2.06	2.95
21	1.74	1.90	1.13-1.18	2.36	12.26-12.30
22	3.84	2.59	1.31-1.38	4.04	7.52-7.56
23	5.20	3.24	1.42	4.77	4.78-4.82
24	3.65	2.75	1.18-1.23	1.47	8.19-8.23
25	3.43	8.41	1.52-1.57	1.48	17.86-17.90
26	3.18	13.1	2.09-2.17	9.10	18.10-18.14
27	2.66	12.5	2.95	13.7	24.0
28	2.64	9.23	2.57-2.64	7.18	9.42

TABLE J-2, (cont.)

AVERAGE DAILY I-131 RELEASE RATES FROM THE QUAD CITIES REACTOR BUILDING VENT

	I-131 Release Rates (nCi/sec)				
Day	June	July	August	September	October
29	3.53	21.2	1.68-1.75	4.57	7.19
30	4.02	4.14	1.20-1.27	3.35-3.39	5.29
31	-	3.37	1.78-1.85	-	6.81

TABLE J-3

AVERAGE DAILY I-131 RELEASE RATES FROM THE QUAD CITIES MAIN CHIMNEY

Day	I-131 Release Rates (nCi/sec) June	July	August	September	October
1		28.7	6.13	6.25	8.12
2		22.9	6.15	7.65	8.74
3		24.2	8.08	7.37	7.21
4		31.1	9.25	4.70	11.7
5		17.9	7.85	3.66	12.1
6		20.2	7.61	3.22	9.69
7		45.8	7.13	19.6	10.9
8		18.6	8.33	3.88	14.2
9		14.8	8.31	9.61	15.1
10		24.5	5.75	7.74	14.6
11		25.6	10.4	5.94	12.1
12		13.3	6.17	7.25	99.5
13		35.3	26.1	10.5	19.6
14		35.5	12.1	9.92	20.3
15	9.73	34.1	14.3	7.76	16.7
16	65.2	18.0	12.6	11.0	13.4
17	7.58	39.9	11.5	14.1	17.7
18	6.84	36.1	18.3	9.55	14.5
19	7.67	48.6	51.8	9.76	16.0
20	6.91	32.8	38.2	12.6	55.8
21	22.1	27.5	54.4	26.7	161
22	50.1	14.2	27.0	22.3	57.7
23	42.8	34.2	37.1	9.26	34.1
24	58.3	19.0	22.4	7.09	19.2
25	25.7	22.9	79.3	4.86	30.7
26	22.0	13.9	102.3	50.6	13.5
27	26.9	9.75	41.6	123.1	57.4
28	31.5	10.4	22.3	17.8	17.4

TABLE J-3, (cont.)

AVERAGE DAILY I-131 RELEASE RATES FROM THE QUAD CITIES MAIN CHIMNEY

Day	I-131 Release Rates (nCi/sec)				
	June	July	August	September	October
29	55.2	7.55	16.2	11.1	13.6
30	51.6	9.62	5.54	9.27	9.98
31	-	5.72	10.1	-	11.0

APPENDIX K

PREPARATION OF STABLE IODINE SAMPLING MEDIA

INTRODUCTION

Careful preparation of each type of collection substrate for use in atmospheric sampling is required for the purpose of ensuring maximum collection efficiency and minimum blank values. Each collection medium is unique and requires a different method of preparation and handling for this work.

PREPARATION OF PARTICULATE FILTERS

Nucleopore polycarbonate filters with a pore size of 0.4 micrometers were used to collect iodine associated with particulates. This filter shows minimum interaction with the common gaseous iodine species. The preparation of these filters includes a clean up procedure designed to remove any surface contamination that may have occurred through previous handling. The filters are washed in boiling distilled-deionized water and allowed to air dry in an iodine free area. Filters are stored in batches of ten in plastic filter holders with paper disc spacers between filters. The filter packs are sealed in polyethylene bags for storage or shipment.

PREPARATION OF TBAH COATED FILTERS

Tetrabutyl ammonium hydroxide (TBAH) coated filters were used to collect inorganic iodine gases in the environment. Preparation of the inorganic collection media involves the impregnation of 47-mm Whatman 41 cellulose paper filters with a solution of tetrabutyl ammonium hydroxide (TBAH) and glycerol in water. The method of impregnation consists of placing a filter on the surface of an aqueous solution containing 40% TBAH and 10% glycerol for approximately 1 minute. After this time, the impregnated filters are allowed to air dry under an infrared lamp in an iodine free room for a period of approximately 30 minutes. Batches of ten filters with spacers are placed in a plastic filter holder and sealed in a polyethylene bag.

PREPARATION OF ACTIVATED CHARCOAL

Activated coconut shell charcoal (Barnabey-Cheney Type AC, 8-12 mesh) was used to collect the gaseous organic iodine fraction. The charcoal requires a clean up step for removal of the 50 to 300 parts per billion of iodine typically present.

Approximately 100 grams of charcoal are placed in a quartz tube and attached to a vacuum system. The pressure is lowered to approximately 10^{-5} Torr. The temperature of the tube is slowly raised to $750^{\circ}C$ and maintained at these conditions for a period of 10-14 days. After this time, the temperature is lowered to ambient at a rate of $25^{\circ}/hr$. An activated charcoal trap in the vacuum system venting line filters the air that enters when the system is returned to atmospheric pressure after cooling.

Charcoal columns are made by packing 0.5 gram of the cleaned activated charcoal in 3/8" o.d. thin walled flourocarbon tubing measuring approximately 3" in length. Both ends of the tubing are plugged with quartz wool which had been heated to $500^{\circ}C$ for 12 hours and stored in a dessicator. After this procedure is completed, the columns are capped with 3/8" o.d. aluminum tube fittings and stored in sealed plastic bags until use.

APPENDIX L

PROCEDURE FOR LOADING, EXCHANGE, AND UNLOADING STABLE IODINE SAMPLING TRAIN COMPONENTS

INTRODUCTION

The stable iodine sampler consists of a sequence of filters held in a 47-mm open faced sampling head followed by three charcoal beds. The filters are arranged in the sampling head as follows: (1) Nucleopore particulate filter, 0.4-µm pore size, (2) teflon support filter, 5-µm pore size, (3) tetrabutyl ammonium hydroxide (TBAH)-coated filter, (4) teflon support filter, 5-µm pore size, (5) TBAH-coated filter, and (6) teflon support filter, 5-µm pore size. The sampled air passes first through the Nucleopore filter, then through the other filters in sequence, and on to the charcoal beds.

The following procedures are used for loading the sampling train in the laboratory, exchanging samplers in the field, and unloading the exposed sampling train in the laboratory. When the entire sequence is performed in the field, sample unloading is performed first followed by loading of new sampling media in the same filter holder. Precautions against stable iodine contamination are taken.

SAMPLE LOADING

Sample loading is to be performed in a room remote from species sampler media preparation and other laboratory work involving significant quantities of stable iodine. Prior to sample loading, the person must wash hands and arms. All tools are to be used only in the preparation area and for stable iodine sampler work only. Load the filters in the proper sequence into the filter holder. Label the filter head with a tag indicating the date on and the location. Place the filter head in a new plastic bag. Remove plugs and connect three charcoal beds in series using two 3/8" tube to 3/8" tube connectors and the tools reserved for stable iodine sampler work. The two ends of the charcoal bed series should be kept sealed with plugs. Label each charcoal bed with a sequence number, the date on, and the location. Place the charcoal bed array in a new plastic bag. Place the bagged filter head, the bagged charcoal beds and the tools in a third new plastic bag for transport to the field locations. The bag should then be placed in a box used only for the stable iodine sampler materials and kept away from boxes containing IPh or in-plant sampling apparatus or sampling media.

SAMPLE EXCHANGE

Sample exchange is performed in the field using tools which are only used for stable iodine sampler work. Perform the exchange before changing the media in the environmental radioiodine sampler if there is one located nearby. Turn off the pump and record the off time and data. Disconnect the tubing from the third charcoal bed and insert the plug in the end of the charcoal beds. Separate the charcoal bed train from the filter head and plug the other end. Record the date off on each charcoal bed label and on the filter head label. Place the filter head and the charcoal beds in separate new plastic bags. Remove the end plug and connect the new charcoal bed sequence to the filter head; check to verify that the bed labeled #1 is closest to the filter head. Remove the plug and connect the other end of the charcoal bed train to the tubing from the pump. Place the tools and the bagged components in a bag and then in the box for return to the laboratory. Start the pump and record the date and time in the log book. (If there is an environmental radioiodine sampler nearby, do not restart the stable iodine sampler until the environmental radioiodine sampler media have been exchanged.)

SAMPLE UNLOADING

Sample unloading is to be performed in the same location used for sample loading and using the same single-purpose tools. Open the filter head and separate the filters. Place filters (1), (3), and (5), each with its teflon support filter, in separate new plastic filter holders. Label each bag or filter holder with the type of filter and the on and off dates. Separate the charcoal beds and plug each one at both ends. Place each bed in a new plastic bag or vial and label it. Place all six bagged components in a large new plastic bag labeled with the date on, date off, and sampling location.

APPENDIX M

ANALYTICAL PROCEDURES FOR STABLE IODINE

After collection, each stable iodine sample component was analyzed quantitatively for iodine using neutron activation analysis (NAA). The analytical procedure involves sample irradiation, chemical separation, and gamma ray spectrometry for the quantitative determination. The procedure is outlined in the flow diagram.

PACKAGING AND IRRADIATION

Each sample filter and charcoal segment was individually packed into a 1-dram polyethylene vial using precleaned stainless steel forceps. The vials were previously washed in hot nitric acid and rinsed with distilled, deionized water. The backup spacer filter is carefully separated and discarded before packing the collecting filter. The activated charcoal samples were uncapped, the quartz wool removed, and the contents poured into a 1-dram polyethylene vial. This packing step produces six individual samples to be irradiated for each collection event. Flux monitors were made by placing a known concentration of iodine on a 25-mm diameter paper filter which was sealed in precleaned polyethylene.

Three sample vials and one flux monitor were irradiated for 10 minutes with thermal neutrons at a flux of $\sim 4 \times 10^{12}$ n cm^{-2} sec^{-1}.

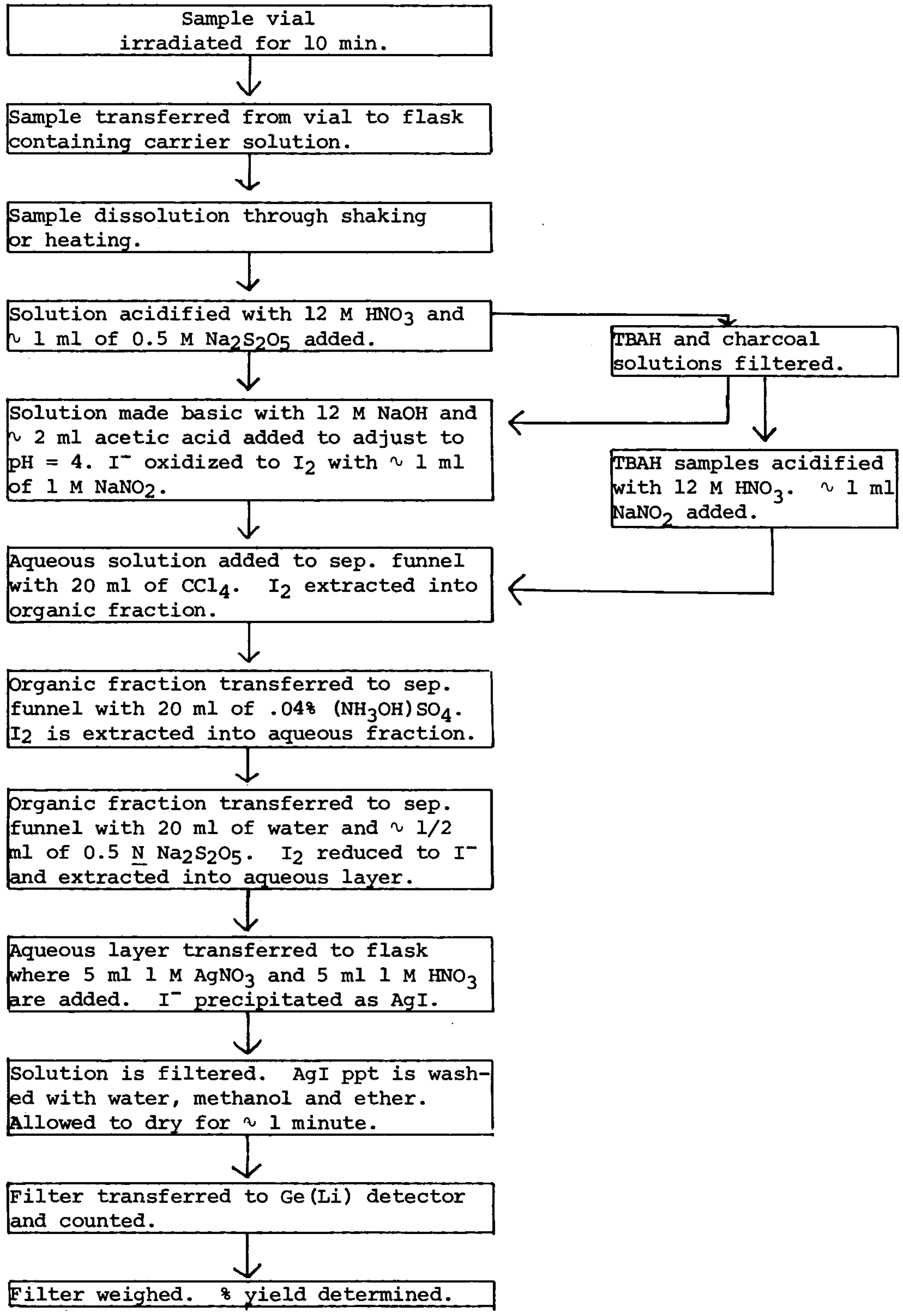
CHEMICAL SEPARATIONS

Immediately after irradiation the vial was opened and the contents quantitatively transferred to an Erlenmeyer flask containing 5 ml of carrier solution, which has an equivalent weight of iodine equal to 50 mg of AgI. The carrier solution was prepared in 1 M NaOH. The separation procedures for the 3 types of samples are described below.

Particulate Filters

After the filter was added to the carrier solution it was oxidized by the dropwise addition of a 5% ClO^- solution. The solution was acidified to a phenopthalein end point with 16 M HNO_3 and 0.5 M $Na_2S_2O_5$ solution is added dropwise to reduce the

FLOWCHART FOR IODINE ANALYSES



Sample vial irradiated for 10 min.

Sample transferred from vial to flask containing carrier solution.

Sample dissolution through shaking or heating.

Solution acidified with 12 M HNO_3 and ∿ 1 ml of 0.5 M $Na_2S_2O_5$ added.

TBAH and charcoal solutions filtered.

Solution made basic with 12 M NaOH and ∿ 2 ml acetic acid added to adjust to pH = 4. I^- oxidized to I_2 with ∿ 1 ml of 1 M $NaNO_2$.

TBAH samples acidified with 12 M HNO_3. ∿ 1 ml $NaNO_2$ added.

Aqueous solution added to sep. funnel with 20 ml of CCl_4. I_2 extracted into organic fraction.

Organic fraction transferred to sep. funnel with 20 ml of .04% $(NH_3OH)SO_4$. I_2 is extracted into aqueous fraction.

Organic fraction transferred to sep. funnel with 20 ml of water and ∿ 1/2 ml of 0.5 N $Na_2S_2O_5$. I_2 reduced to I^- and extracted into aqueous layer.

Aqueous layer transferred to flask where 5 ml 1 M $AgNO_3$ and 5 ml 1 M HNO_3 are added. I^- precipitated as AgI.

Solution is filtered. AgI ppt is washed with water, methanol and ether. Allowed to dry for ∿ 1 minute.

Filter transferred to Ge(Li) detector and counted.

Filter weighed. % yield determined.

periodate to the iodide. The solution is made basic to phenolpthalein with 12 M NaOH and adjusted to a pH of 4 through the addition of approximately 2 ml of glacial acetic acid. The aqueous solution is quantitatively transferred to a 125-ml separatory funnel containing 20 ml of CCl_4 and 1 ml of 1.0 M $NaNO_2$ where the iodide is oxidized and the I_2 extracted into the organic layer after vigorous shaking. The organic fraction was transferred to a separatory funnel containing 20 ml of 0.04% $(NH_3OH)SO_4$ and the aqueous fraction is transferred to another separatory funnel containing 20 ml of CCl_4 to reextract any remaining I_2. The organic fraction from the second separatory funnel is added to the $(NH_3OH)SO_4$ separatory funnel and washed with the aqueous phase. This washing is designed to selectively remove any bromine found in the organic layer (i.e. selective reduction of bromine to bromide). The organic fraction is transferred to a separatory funnel containing 1/2 ml of 0.5 M $Na_2S_2O_5$ in 20 ml of water where the iodine is reduced to the iodide and extracted into the aqueous layer. The organic layer is discarded and the aqueous fraction is transferred to an Erlenmeyer flask where 5 ml of 1 M $AgNO_3$ and 5 ml of 1 M HNO_3 are added to precipitate the iodide as AgI. The solution containing the insoluble precipitate is filtered onto a preweighed glass fiber filter and washed with water and methanol and dried with ether.

Charcoal Beds

The charcoal samples are transferred to an Erlenmeyer flask containing 5 ml of iodate carrier solution which is made up in 1 M NaOH. The flask is placed on a hot plate for approximately 5 minutes and small portions of distilled deionized water are added to prevent vigorous boiling. After heating the solution is allowed to cool whereupon it is acidified to the phenopthalein endpoint as previously described in the previous section. After the addition of the sulfite the mixture is filtered to remove the washed charcoal. This solution is made basic with 12 M NaOH and buffered to a pH of 4 with acetic acid. The remainder of the chemical separation follows that which is used in particulate filter procedure.

TBAH Filters

The TBAH filters are transferred to an iodate carrier solution in 1 M NaOH and subjected to vigorous boiling for a period of approximately 7 to 10 minutes. Distilled deionized water is added to the flask to prevent the solution from boiling to dryness. This allows the partial thermal decomposition of TBAH to occur alleviating most of the interference of the tetrabutylammonium cation that is present. The flask is allowed to cool and approximately 5 ml of distilled water is added at which time the mixture is acidified to the phenopthalein end point with 16 M

HNO_3 and filtered to remove the remaining portions of the leached filter. At this point, the filtered solution is further acidified by the addition of ∿ 1 ml of 12 M HNO_3 and ∿ 1 ml of 1 M $NaNO_2$ is added to selectively oxidize the iodide to iodine. Glacial acetic acid is replaced with 12 M HNO_3 to create the appropriate oxidizing medium for the remaining chemical separation steps. This solution is quantitatively transferred to a separatory funnel containing 20 ml of CCl_4 where the solution is subjected to the same separation procedure described for the particulate filter.

The filter containing the AgI precipitate was transferred to a calibrated Ge(Li) detector and counted. The data were recorded on magnetic tape for computer analysis. After counting, the filter was weighed and the chemical yield determined. All recorded spectra were analyzed by the URI IBM-360-75 computer using the programs GAMNL and PIDAQF. The measured activities were corrected for chemical separation yield and the total weight of iodine on the filter was determined.