

Line-Loss Determination for Air Sampler Systems

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Pacific Northwest Laboratory Operated by Battelle Memorial Institute

Prepared for U.S. Nuclear Regulatory Commission

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and suggestions were made for incorporating variable resuspension	rates.			
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<u>ABSTRACT</u>

Iodine deposition can potentially bias the results of radioiodine air sampling systems. To develop guidance and acceptance criteria for determinations of line-loss correction factors, the data on laboratory sampler simulations, field tests on samplers, and experimentally measured iodine deposition rates were reviewed. Sampling system design features and operating conditions at several power reactors are discussed.

Measurements of iodine deposition rates on various air sampler construction materials were reviewed, and predicted air sampler performance based on the data was presented. Three examples of field tests of air sampler performance for radioiodine were examined. A model of iodine deposition and resuspension was extensively reviewed, and suggestions were made for incorporating variable resuspension rates.

Three principal methods for determining radioiodine line-loss factors were defined and compared: in-place field tests, laboratory mock-up with modelled extrapolations to various release rate modes, and modelling based on laboratory data on similar materials. Guidelines for applying these methods were given. Research was recommended to determine whether the three methods were comparable so the less-expensive method could be substituted for the preferred field tests.

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SUMMARY

Air sampling systems in nuclear power reactors are used to measure the concentration of several airborne contaminants in reactor containments and ventilation effluent. Samples are aspirated at one point and transported to the collection point through long lengths of tubing in which significant fractions of the contaminants can be deposited and later resuspended. This deposition and delayed resuspension can introduce a significant bias in calculations of concentration. The U.S. Nuclear Regulatory Commission (NRC) is concerned with the methods used to account for the sample biases introduced by air sampler line losses of radioiodine in particular, because the problem seems to be most severe for that contaminant. This study reviews the iodine line-loss determination methods and recommends a regulatory strategy, measurement alternatives and necessary research.

The report first reviews ventilation and containment air sampling system construction. Descriptions for 28 systems at eight reactors are given to illustrate the range of characteristics and operating parameters. The range of characteristics of the stack sampling systems is summarized. Also summarized are the available data on the distribution of radioiodine forms typical of the operating modes of boiling-water reactors (BWRs) and pressurizedwater reactors (PWRs). The deposition and resuspension of iodine on materials differs, depending on the chemical form of the iodine.

The report next reviews the published work on the deposition of iodine on air sampler construction materials. The deposition velocities experimentally determined are summarized. The performance of typical sampling systems was extrapolated from the data. If deposition was the only process occurring in sampling tubes, the resulting bias in collected samples could be severe and some construction materials, notably Teflon® and polyethylene, may actually be superior to the typical metals. The relative effects of humidity and iodine chemical forms are illustrated. The elemental and hypoiodous acid (HOI) forms are many times more likely to deposit than the methyl iodide. The effect of humidity does not seem to be consistent for all the materials for which deposition data were found.

Thirdly, a review of the reported field tests on two sampling systems indicated that estimating sample bias, based on deposition alone, is too pessimistic; also, that the samplers tested performed well for iodine. The tests were conducted by injecting iodine into the ventilation stream and comparing the results from the permanent sampler with those of a sampler with in-stack or short-coupled collectors.

The report also reviews some of the tests conducted by Science Applications International Corporation (SAIC) for utilities in response to NRC requests for performance data. Sampler performance was estimated by interpreting experimental data using a model. Test data is included in the report. Again, these tests show that the iodine line loss is not as bad as would be predicted when only deposition is considered. The laboratory tests show that deposition and resuspension are both occurring and that the rate

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of resuspension is not constant, and that resuspension can be delayed significantly following the initial deposition. EG&G Idaho, Inc., for the NRC, has attempted to conduct a limited duplication of the experimental data in nearly identical tests, but it is unclear whether the SAIC results were verified.

The SAIC researchers developed a model to incorporate the deposition and resuspension rates determined from the experiments. This was to extrapolate what the observed sampler performance (or line-loss correction factor) might be under certain iodine release scenarios. The model deficiencies are examined and a method is developed to incorporate time-dependent iodine resuspension rates.

The report summarizes three approaches to estimating the transmission of iodine through air sampling lines--in-place testing, laboratory simulation, and modeling, based on deposition and resuspension data determined for a set of air sampler materials. The advantages and disadvantages of each approach were discussed. The report recommended in-place testing using existing iodine release levels or injected nonradioactive elemental iodine. If inplace testing is impossible to apply on a wide scale, then the other approaches may be advised after an initial demonstration that the results are comparable to an in-place test. Laboratory testing should continue with a broader range of operating conditions and materials to develop a generic sampling system design with optimal performance.

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1.0 INTRODUCTION

Air sampling systems are used to measure the concentration of airborne radioactive contamination in reactor containments and ventilation effluents. Samples travel from the point of aspiration to the collection point through long lengths of tubing, in which significant fractions of sampled contaminants can be deposited that may later be reentrained in the air. This can introduce a significant bias in calculations of source concentrations. The U.S. Nuclear Regulatory Commission (NRC) is concerned with methods used to account for sample biases introduced by sampler line losses. The immediate concern is for radioiodine sampling because the line-loss problem appears to be more severe for that contaminant than for other radioactive contaminants. Pacific Northwest Laboratory (PNL) has developed guidance and acceptance criteria for procedures to determine radioiodine line loss.

Under this project, PNL has completed the following tasks: 1) review of available data on line-loss measurements and identification of ranges of parameters and results of the measurements, 2) review of line-loss measurement methods, 3) review of a radioiodine line-loss model, 4) discussions with NRC inspectors and visits to five nuclear plants to view radioiodine sampling systems and identify sampling problems, 5) a summary of key parameters and problems in iodine air sampling, and 6) preparation of draft guidance for performing line-loss measurements and acceptance criteria for data. This final report summarizes the completed work.

Section 2.0 presents an overview of the radioiodine air sampling problem. Section 3.0 reviews measurement methods for determining iodine deposition on air sampler construction materials. Section 4.0 describes three in-place testing methods performed on air samplers, and Section 5.0 is a review of laboratory testing on simulated air sampling systems. Application of the model reviewed in Section 5.0 is discussed in Section 6.0. Suggested improvements in models for calculating dry deposition velocities and deposition rates and an alternate modeling approach are given in Section 7.0. Section 8.0 is a discussion of the data reviewed, and PNL's recommended interim guidelines are presented.

2.0 OVERVIEW OF THE RADIOIODINE AIR SAMPLING PROBLEM

The radio air sampling problem is discussed in the following sections.

2.1 <u>IODINE IN REACTOR EFFLUENTS</u>

Extensive studies have been made of the quantities of radioiodine species in light-water reactor (LWR) effluents. The iodine species distribution and quantities differ widely among various plants, plant areas, and plant operating modes. The distribution of the iodine species in gaseous effluents is of interest because iodine transport through tubes is highly dependent on the iodine form. For three pressurized-water reactors (PWRs) and three boiling-water reactors (BWRs), the average distribution of iodine species in the total plant effluent was estimated by Pelletier et al. (1978a,b). Table 2.1 shows distributions estimated for a hypothetical year's operation, including the various operating modes of the plants. For PWRs, the primary coolant system in the auxiliary building and the containment purges were the most important sources of airborne radioiodine. For BWRs, the import source was the condenser area. For both plant types, the emission rates fluctuate considerably, depending on the power level and maintenance operations.

2.2 AIR SAMPLER CHARACTERISTICS

The air sampling systems used to sample the radioiodine from reactor stacks and ducts have various characteristics that depend on the plant design. Design parameters are affected by the physical layout between the sample extraction and collection locations, safety restrictions in the area, interpretation of regulations and standards, and the detection capability of the analytical or monitoring equipment. Table 2.2 lists characteristics of iodine sampling systems at eight plants (data taken in 1986). In some cases, the system consists of sections with different characteristics in series, as denoted by Series Part A, B, etc. Besides stack systems, some information is included for containment samplers where available.

<u>Table 2.1</u> .	Average Percent Radioiodine (¹³¹ I) Spe	cies
	Distributions for Reactor Effluents	

Radioiodine	PWR	BWR
Species	<u>(Pelletier et al. 1978b)</u>	<u>Pelletier et al. (1978a)</u>
Elemental Iodine	27	28
Hypoiodous Acid	40	20
Organic Iodides	31	40
Particles	2	12

System	Series Part	<u>Tube Size, in</u> 00 ID	- Angle	Bends Number	Total	Equiv Horizontal	alent Lengt Vertical	<u>h, ft</u> Totał	Flow, scfm	Heat Traced?	Operating Temperature, •F	Actual Flow, cfm	Iodine Monitor
	 					REACT	DR A		I				· L · · · · · · · · · ·
Unit 1 Main Low Range		0.75				[191.3	1.67	Yes	?	?	No
Unit 1 High Range	A B	0.75 0.25						10.9 182.4	0.06 0.06	Yes Yes	? ?	? ?	No No
Unit 2 Main Low Range	1	0.75				1		197.3	1.67	Yes	?	?	No
Unit 2 High Range	A B	0.75 0.25						9.17 187.5	0.06 0.06	Yes Yes	?	? ?	No No
Containment		1.0											1
						REACT	DR B						
Main Stack Low Range		1.0			-			?	2.12	No	RT	2.12	Yes
High Range	A	1.0						185	0.21	Yes	130	0.23	No
Standby Gas Treatment Stack	В	0.375	}					ζ2	0.21	Tes	130	0.23	NO
Low Range		1.0						?	2.12	No	RT	2.12	Yes
High Range	A B	1.0 0.375						33 <2	0.21 0.21	Yes Yes	180 <180	0.25 0.25	No Yes
Containment Drywell		0.375, 0.5, 0	.75 vario	us lengtl	hs in ser	ies		240	0.2	Yes	<300	0.29	No
Containment Atmosphere		0.375, 0.5, 0	.75 vario	us lengtl	hs in ser	ies		120	0.2	Yes	<185	0.24	No
/	·					REACTO	DR C		,				·,
Líne 1 Line 2		0.75 0.252	90 • 90 •	16 16	1,440 1,440	85.9 85.0	55 55	140.9 140.0	2 0.06		RT 30		
						REACTO	DR D						
Main Stack Low Range	A	1.25	90.	3	270	0 -	230	230	2.5	Yes	90	2.59	No
High Range Subsample	в	0.75 0.375	90 • 90 •	3 1	270 90	70 0	15 3.5	85 3.5	2.3 0.06	Yes Yes	90 90	2.39 0.1	No No
Standby Gas Treatment Stack						1							
Low Range High Range		0.75	90.	6	540	90	71	161	2	Yes	90	2.08	No
Existing Proposed Subsample		0.375 0.375	90.	6	540	90		156 <4	0.06 0.06	No No	90 RT	0.06 0.06	No No

TABLE 2.2. Summary of Reactor Sampling Systems

<u>TABLE 2.2</u>. (contd)

System	Series Part	<u>Tube</u>	<u>Size, in.</u> ID	Angle	<u>Bends</u> Number	Total	<u>Equiva</u> Horizontal	<u>lent Length</u> Vertical	<u>n. ft</u> Total	Flow, scfm	Heat Traced?	Operating Temperature, •F	Actual Flow, cfm	Iodine Monito
	-		-	·			REACTO	R E				·	· · · · · ·	1
MY1		1.13							157	10		RT		
L		-					REACTO	RF					<u>.</u>	
Main Stack Low Range High Pango		1		90	4	360	150	100	250	1.4 to 2.1	Yes	70	1.4 to 2.1	No
Existing		1							250	0.21	Yes	70	0.21	No
Grab Monitor		? ?							<10 <4	0.21 0.0035	No No	RT RT	0.21 0.0035	No Yes
							REACTO	RG				-		
	A B C		1.370 1.370 0.618						88.0 198.0 198.0	12.83 14.00 4.00		RT RT RT		
L	1			.I.			WNP-	2						
Reactor Bldg. Low Range	A B	1.25 0.75	1.12 0.62	90 - 90 - 50 - 45 - 40 -	4 6 1 1 1	360 675	10.5 6.8	1 25.4	11.5 31	5 1.4	No No	RT RT	5 1.4	No No
High Range		0.5		90 - 42 - 38 - 8 -	6 2 2 2	716	92.5	- 24.25	116.75	0.05	Yes	>32		No
Turbine Bldg.	A	1.25	1.1Ż	90 · 50 · 45 ·	2 2 2 2	370	11.7	13.7	23.8	7.5	Yes	112	8.09	No
	В	0.75	0.62	90.	2	180	0.5	5.5	6	2.3	Yes	112	2.48	No
Radwaste Bldg. A & B		?		90 ·	1	141	6.7	11.7	16.5	?	No	RT	? ,	No
Radwaste Bldg. C.		?		45.	2	90	3.8	12.2	14.4	?	No	RŤ	?	No
							Unrein (1985)						
1 2 5 6		L	0.874 0.874 0.870 0.835						100.0 50.0 256.0 246.0	3.00 3.00 2.00 2.80		25 to 30 25 to 30 25 to 30 25 to 30 25 to 30		

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The stack systems are divided into low- or high-range (post-accident) systems. The range of characteristics is shown in Table 2.3.

Many systems have "real-time" iodine monitors; others employ collection cartridges for subsequent analysis. Sampling lines have as few as two bends in the line; others as many as sixteen 90° bends. Most high-range systems have long sampling lines with low flow rates compared to the low-range systems. Where high-range systems have short lines, they are subsampling lines from the low-range system, which help minimize iodine line losses and response time following iodine transients in the sampled air flow.

No information was available on expected humidities in accident conditions. The operating temperatures for most heat-traced systems are apparently set to exceed the maximum expected dewpoint. Some heat-traced systems merely provide freeze protection in the event of condensation.

There are many parameters or characteristics of the air sampling system that could affect how much of the originally sampled iodine makes it through the tubing to where it is to be collected or monitored. These include

- temperàture
- humidity
- iodine concentration

 -level
 -variability
 -duration
- iodine species distribution
- particle size
- tube length
- tube diameter
- sample flow rate
- Reynolds number
- tube material
- tube surface condition

 roughness
 contamination.

TABLE 2.3. Range of Stack System Characteristics

<u>Range</u>	Tube Inside <u>Diameter, in.</u>	Flow Rate, scfm	Total <u>length, ft</u>	Operating <u>Temperature, °F</u>
High	0.25 to 0.87	0.05 to 0.21	2 to 250	>32 to 180
Low	0.67 to 1.38	1.4 to 12.8	30 to 250	Ambient to 112

The effects of some of these parameters are unknown, and some will be discussed in the following sections.

However, even when these parameters are taken into account and overcome, problems are still encountered when ventilation exhaust is sampled for radioiodine content. Some of these problems include

- To what extent and under what conditions sampling data are biased.
- Some forms of iodine deposit more readily than others.
- The various iodine forms are present in normal and abnormal exhausts to differing degrees and are highly variable.
- Required range of measurability is large.
- All the operating conditions are not known, especially accident conditions.
- The NRC is unsure how the utilities are to demonstrate sampling bias.
- Remedial measures forced on the utilities now may later have to be retracted.
- Utilities distrust enforcement actions required in an atmosphere of unclear scientific justification.
- Licensees are unsure how useful source term measurements are compared to measurements in the environment.
- Radiation exposure around the high-level sampler is difficult to control.

Clearly, some of these problems are regulatory and political. It is hoped that this report will provide some basis for guidelines to improve the quality of iodine air sampling.

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3.0 MEASUREMENTS OF IODINE DEPOSITION ON AIR SAMPLER CONSTRUCTION MATERIALS

Before providing guidance on methods for measurements of elemental iodine line losses in reactor effluent air samplers, it is useful to review the measurement methods that have been used. This section summarizes work performed to characterize the deposition of radioiodine onto materials used in air sampling and other power reactor systems.

The first work discussed (Hemphill and Pelletier 1978) was performed for the Electric Power Research Institute (EPRI) to explain why airborne iodine compounds seem to differ in various parts of a power reactor and during different operations. The second work discussed (Kabat 1983) was performed specifically to address material problems in iodine air sampling. Iodine deposition rates were measured, and we have applied them to estimate the performance of example air sample delivery systems where deposition is the only iodine/surface interaction considered.

3.1 <u>HEMPHILL AND PELLETIER</u>

Hemphill and Pelletier (1978), of Science Applications International Corporation (SAIC), reported deposition velocities and resuspension rates for elemental iodine on coupons of various materials, including aluminum (type 5052, which contains 2.5% magnesium and 0.25% chromium), galvanized steel, concrete, and two paint formulations. The test chamber was aluminum with an applied Teflon*(a) coating. Deposition and resuspension measurements were also made on the test chamber to account for those effects.

In Hemphill and Pelletier's (1978) test procedure, elemental gaseous iodine is generated by the reaction

$$IO_3^- + 5I^- + 6H^+ = 3I_2 + 3H_2O$$
(3.1)

In a two-neck boiling flask, 20% H₂SO₄ and potassium iodate are heated and stirred to produce the above reaction. The gaseous iodine is continuously purged with a flow of helium. Radioactive sodium iodide solution is added to the reaction mixture with a peristaltic pump so the generation rate of $^{131}I_2$ is essentially constant. The helium-iodine mixture is diluted with filtered air in the mixing chamber. The dilution air is not usually humidity controlled, but can be if required. Iodine concentrations were measured with an early version of the species sampler, consisting of two particle filters in series (Flanders F-700 paper)(b), an elemental iodine adsorber (10% by weight

- (a) Teflon is a registered trademark of E. I. du Pont de Nemours & Co., Wilmington, Delaware.
- (b) Flanders Filters, Inc., Washington, North Carolina.

cadmium iodide on Chromosorb $P_{\bullet}(a)$, an hypoiodous acid (HOI) adsorber (4-iodophenol on activated alumina), and an organic iodide adsorber [tri-ethylenediamine-(TEDA-) impregnated charcoal].

In the samplers, a significant amount of the iodine was collected on both of the particle filters. The authors reasoned that particles should be collected mostly on the first filter and not penetrate to the second; therefore, it was suspected that much of the iodine on the filters was really adsorbed vapors. This suspicion was investigated with two sets of the test data. It was assumed that the amount collected on the second filter was adsorbed vapor, and that the first filter would have adsorbed the same amount. The difference between the measured radioiodine and the amount assumed to be adsorbed on the first filter was taken to be the true particleborne amount. As shown in Table 3.1, those estimated amounts of particles were only a few percent of the sum of the iodine assumed to be adsorbed on the filters plus the iodine measured on the adsorber stage of the sampler; therefore, the iodine measured on the filters was combined with that of the elemental iodine adsorber to give the total elemental iodine fraction.

Iodine concentrations were measured upstream and downstream of the test achamber during the iodine injection part of the experiment, which lasted 2 h. After iodine injection was completed, iodine sampling at the outlet of the chamber was continued for up to 30 to 330 h to measure the resuspension rate.

Teet 12	Measured	<u>Activity</u>	<u>True % Activity</u>		
<u>Samples</u>	<u>Filters</u>	<u>CdI2 Bed</u>	<u>Particles</u>	<u> </u>	
1 2 3 4 5 Mean	33.7 52.1 23.0 31.9 24.7 33.1	66.3 47.9 77.0 68.1 75.3 67.0	12.9 14.3 4.9 9.6 0.5 8.4	87.1 85.7 95.1 90.4 99.5 91.6	
Test 17 <u>Samples</u>					
1 2 3 4 Mean	25.2 12.0 14.8 5.3 14.3	74.8 88.0 85.2 94.7 85.7	2.9 3.4 2.6 2.1 2.8	97.1 96.6 97.4 97.9 97.2	

<u>TABLE 3.1</u>. Effect of Subtracting Adsorbed Iodine from Particulate Stage (Hemphill and Pelletier 1978)

(a) Chromosorb P is a registered trademark of Johns-Manville Products Corp., Manville, New Jersey.

After the injection of iodine, a portion of the specimen was removed to measure the surface activity by direct counting. The surface activity count was also verified by mass balance on the iodine generator, samplers, etc.

Tests were conducted at humidities in the 20% to 90% relative humidity range and temperatures in the 64° to 85°F range. Air flows in the test chamber were both laminar and turbulent. Turbulence was created by a small Teflon fan inside the exposure chamber. The authors pointed out that the deposition velocity under laminar conditions would be applicable only for similar conditions; therefore, the turbulent data would be of most use in sampling systems.

Table 3.2 lists the deposition data for the aluminum specimens and the Teflon coating on the exposure chamber. Note that for aluminum the deposition rates were higher for the high-humidity tests than for the low-humidity tests. For Teflon coating, there was no correlation with humidity; however, all the Teflon tests were under laminar flow, which would be diffusion controlled. The authors did observe that the resuspension rate seemed to decline with time for aluminum, which may have been due to some irreversible deposition or an artifact of the experimental and data reduction method. It was also observed that the species distribution measured at the chamber inlet was more than 90% to 98% elemental iodine. The distribution shifted during the resuspension experiment to the less-reactive HOI and organic iodine, so that the elemental fraction was 62% to 95%.

<u>TABLE 3.2</u> .	Deposition	Velocities and	Resuspension	Rates for Aluminum
	and Teflon	Coating (Hemph	ill and Pellet	ier 1978)

<u>Coating</u>	Deposition Velocity, cm/s	Resuspension <u>Rate, s⁻¹</u>	% Relative Humidity	Flow <u>Profile</u>
Teflon #3	0.014		42 - 44 30 - 37	Laminar
#5	0.015		46 - 50	Laminar
#6	0.010		42 - 43	Laminar
#7	0.026	7.5 (-6)	36 - 39	Laminar
#12	0.014	7.5 0.1 (-6)	54 - 61	Laminar
#14	0.0076	8.5 1.8 (-6)	>90	Laminar
#16	0.0064		<26	Laminar
Aluminum #8	0.038		<26	Laminar
#9	0.051	1.1 0.6 (-5)	46 - 52	Laminar
#13	0.063	3.5 0.5 (-6)	>90	Laminar
#17	0.51	3.3 (-5)	>88	Laminar
#18	0.43	1.5 0.7 (-5)	>92	Turbulent
#28	0.075	<1.8 (-6)	<26	Turbulent

3.2 <u>KABAT</u>

Kabat (1983) reported deposition velocities of different iodine species onto coupons of various materials with laminar flow conditions. The materials tested included copper, carbon steel, stainless steel, aluminum, polyvinylchloride (PVC), polyethylene, Teflon, and Buna-N.

Some metal specimens were tested as received, and others were tested after cleaning. The cleaning process began with an ethanol rinse. Aluminum was cleaned with 1% NaOH; the other metals with 1% HCl. They were then rinsed with distilled water and methanol. The plastic specimens were tested as received with no cleaning. The specimens were 560 to 1600 mm² in size.

The specimens were placed in glass chambers through which the test gas flowed. The flow rate of the gas was sufficiently high that iodine depletion was negligible. This permitted the use of a single iodine species sampler at the outlet of the exposure chamber for the determination of exposure concentration. The species sampler contained copper screens, HOI adsorbent, and TEDA-impregnated charcoal, as described by Kabat (1976).

The individual iodine species were generated and mixed with air of the desired humidity level. Elemental iodine vapor was generated from a chilled solution of elemental iodine in distilled water. The HOI was generated by purging the carrier air through a 5×10^{-8} <u>M</u> solution of elemental iodine in distilled water. The methyl iodide was released from a cylinder. In each test, several specimens were exposed to one of the iodine forms. The amount of iodine deposited on the specimens was determined by direct counting. The concentrations of each iodine species in the test gas were

Elemental	Iodine	10 μg/L ± 30%
HOI		$0.01 - 0.05 \mu g/L$
CH3I	,	$1 \mu g/L \pm 20\%$

The experimental parameters were

Temperature	20° - 24°C
Pressure	l atm`
Relative Humidity	5% and 97% ± 3%
Carrier Gas	air
Reynolds Number	16

The measured deposition velocities for metals and plastics are given in Tables 3.3 and 3.4, respectively.

Kabat used the deposition data to estimate the performance of sampling tubes using as an example a 30-m-long tube with a 12.5-mm inside diameter and 100-lpm flow rate. The fraction of each iodine form that would deposit while passing through the tube was calculated by Equation (3.1).

Fraction deposited =
$$1 - Exp(4v_dL/v_D)$$
 (3.1)

Iodine <u>Form</u>	% Relative Humidity	<u>Copper</u>	Carbon <u>Steel</u>	Stainless <u>Steel</u>	<u>Aluminum</u>
I2	5 Uncleaned	2.0E-01	3.4E-02	1.8E-02	8.4E-03
I_2	5 Cleaned	2.3E-01	1.1E-01	8.7E-02	1.7E-02
I ₂	97 Uncleaned	2.1E-01	1.3E-01	1.6E-01	8.0E-02
I ₂	97 Cleaned	2.3E-01	2.5E-01	2.0E-01	1.8E-01
нот	5 Uncleaned	1.0E-03	1.4E-03	4.0E-04	1.9E-04
HOI	5 Cleaned	1.4E-02	8.0E-04	3.3E-03	2.5E-03
HOI	97 Uncleaned	2.2E-02	2.0E-03	1.8E-03	1.2E-03
HOI	97 Cleaned	2.7E-02	3.8E-03	4.4E-03	5.6E-03
CH3I	5 Uncleaned	2.0E-06	8.0E-06	1.0E-05	8.0E-06
CH ₃ I	5 Cleaned	1.0E-05	8.0E-06	7.0E-06	1.0E-05
CH ₃ I	97 Uncleaned	7.0E-06	4.0E-06	8.0E-06	4.0E-06
CH ₃ I	97 Cleaned	7.0E-06	4.0E-06	8.0E-06	1.0E-04

TABLE 3.3. Deposition Velocities for Metals, cm/s

TABLE 3.4. Deposition Velocities for Plastics, cm/s

Iodine <u>Form</u>	% Relative <u>Humidity</u>	PVC	Poly- <u>ethylene</u>	<u>Teflon</u>	<u>Buna-N</u>
I ₂	5	1.0E-01	4.0E-03	3.5E-02	2.0E-01
I ₂	97	2.7E-01	1.5E-02	2.0E-02	2.7E-01
HOI	5	1.2E-01	3.3E-02	2.5E-04	1.5E-01
HOI	97	1.3E-01	2.2E-02	9.5E-04	1.6E-01
CH3I	5	3.4E-04	1.0E-04	3.4E-05	1.9E-04
CH ₃ I	97	6.0E-04	3.2E-05	2.5E-05	4.5E-04

where v_d = deposition velocity, cm/s L = tube length, cm

 v_a = average gas velocity in tube, cm/s D = tube inside diameter, cm.

3.3 DISCUSSION

The estimated fractional deposition for metal specimens are shown in Figures 3.1 to 3.3. The cleaned metal specimens generally had greater elemental iodine deposition than those tested as received from the supplier. This was usually, but not always, the case for the other iodine species as



FIGURE 3.1. Estimated Fraction of Elemental Iodine Deposited in a 30-m Metal Tube, 12.5-mm Inside Diameter, 100-1pm Flow Rate

well. This observation may not necessarily apply to tubing because the surface treatments that tubes undergo in fabrication are probably different than those for sheet metal.

Figure 3.4 shows the fraction of each form of iodine that would deposit in the example sampling tube made of each of the material types tested without any cleaning. The figure shows that more elemental iodine would deposit than HOI, and more HOI than methyl iodide for all the materials except polyethylene and PVC. In those two cases, HOI was more depositing than elemental iodine; however, methyl iodine was still the least depositing. Generally, more iodine deposited at 97% relative humidity than at 5% relative humidity. The high humidity significantly enhanced deposition over that of the lowhumidity tests of elemental iodine and HOI, except in the cases of HOI on polyethylene and elemental iodine on Teflon. Accounting for deposition only, the performance of the various materials tested without the cleaning process and at 97% relative humidity is estimated in Figures 3.5 and 3.6. The line loss for each form of iodine in the example sampling tube was calculated as before, except the losses were weighted by the fraction of each gaseous iodine form estimated to be present in the total effluent of BWRs and PWRs,





as previously shown in Table 2.1. Thus the sum of the losses (shown by the stacked bars in Figures 3.5 and 3.6) for each form represents the total estimated gaseous iodine losses in the sampler when sampling from the total effluent from that type of plant. The figures suggest that the ranking of preferred gaseous iodine sampling line materials may be (best to worst) Teflon, polyethylene, aluminum, carbon steel, stainless steel, copper, PVC, and Buna-N.







FIGURE 3.4.

Estimated Percentage of Iodine Deposited in Example Tube $\space{1.5}$ as a Function of Materials and Humidity



<u>FIGURE 3.5</u>. Estimated Gaseous Iodine Line Loss for Sampling Total Boiling-Water Reactor Effluent at 97% Relative Humidity



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Estimated Gaseous Iodine Line Loss for Sampling Total Pressurized-Water Reactor Effluent at 97% Relative Humidity
4.0 IN-PLACE DETERMINATIONS OF AIR SAMPLER PERFORMANCE

In-place determinations of air samples performance are discussed in the following sections.

4.1 EPRI TESTS

While obtaining data for EPRI on the sources of radioiodine in BWRs, Pelletier et al. (1978a) made iodine transmission tests on simulated sampling tubes. Three stainless steel and three aluminum tubes, each 0.635 cm (1/4 in.) inside diameter, were installed at a reactor site. Radioiodine from the plant ventilation system was injected into each tube. Iodine species samplers were used to measure the inlet and outlet activity of 131I, 133I, and 135I. The tubes varied in length from 183 to 732 cm (6 to 24 ft), and the flow velocities were 300 to 540 cm/s (5.7 to 10.26 lpm). The resulting mean residence times were between 0.4 and 2.4 s. Before testing began, ventilation air flow was maintained through the tubes for a month so that the measured transmission would be for equilibrium conditions. Inlet and outlet sampling was then done for a 40.9-h period.

The model used in Pelletier's study to interpret the results was an earlier form of a model to be described in Section 5.0. The resuspension rate is for total iodine. Deposition velocities and resuspension rates were calculated assuming equilibrium in the surface activity and equal deposition velocity and resuspension rates for the different iodine isotopes. The decontamination factor equation for each isotope includes the deposition velocity and resuspension rate as parameters. Calculating the ratio of the decontamination factors for the isotopes through the tubes eliminated deposition velocity and allowed solution for the resuspension rate. Once the resuspension rate was calculated, the deposition velocity was found from the decontamination factor equation for one isotope. The authors did not give detailed data, but reported the mean and median deposition velocities and resuspension rates from the tests as shown in Table 4.1.

<u>TABLE 4.1</u>. Deposition Velocity and Resuspension Rate for Equilibrated Tube Experiment (Pelletier et al. 1978a)

	<u>Mean Value (</u>	<u>Std. Dev.)</u>	<u> Median Values </u>			
	Resuspension Rate	Deposition Velocity	Resuspension Rate	Deposition Velocity		
<u>Tubing</u>	<u>x E-6/s</u>	<u>x E-3 cm/s</u>	<u>x E-6/s</u>	<u>x E-3 cm/s</u>		
Stainless Steel	11.2 (4.0)	78 (12)	7.9	76		
Aluminum	6.6 (3.4)	30 (9)	3.5	25		

During the tests it was observed that some of the iodine collected on the particle filters could be removed by purging and collected on the down stream CdI_2 bed. This was assumed to be gaseous iodine adsorbed and then desorbed from the filter paper. For this reason, the iodine collected on the particle filter was combined with that collected on the CdI_2 adsorber and counted as I_2 .

4.2 STROM AND HESBOL

Another example of in-place performance testing was conducted at Swedish BWRs by Ström and Hesböl (1977), who described a typical sampling system as having two circuits. The primary circuit isokinetically extracts the sample from several points in a cross section of the stack, transports the sample to the collection point, and returns the excess sample to the stack. The sample collector draws a subsample from the primary circuit. The dimensions of the primary circuit tube are 10 m (32.8 ft) long, 10 cm (3.94 in.) inside diameter, and 60 lps (3600 lpm or 127 cfm) flow rate.(a) The main iodine species observed in the effluents has been methyl iodide. Particle sizes are usually smaller than 4 μ m.

The stack effluent monitoring systems at Swedish BWRs are tested before reactor startup. The testing series includes a test of sampling representativeness(b) and of iodine and particle line losses. The representativeness is checked by injecting 131I-tagged methyl iodide into the ventilation stream and sampling in the stack at the elevation of the sampler nozzles and at the regular sample collection point. Methyl iodide is used because it is largely nondepositing. The tracer for the particle line-loss test is a fluorescent dye-tagged dioctyl-phthalate aerosol that has a mass median diameter of about 20 μ m. The concentration is measured in the stack and compared with the results from the regular particle samples. The tracer for the iodine line-loss test is 131I₂, and the concentration is measured at the sampler intake and at the regular sample collection point.

The $^{131}I_2$ tracer is produced by the reaction of sodium iodine and potassium dichromate solution in a portable hood and is injected into the ventilation air with a small air stream. The total amount of injected radioiodine is about 1 to 2 mCi ^{131}I . The quantity of stable iodine injected in the

- (a) Private conservations with Hesböl revealed a technique developed to eliminate chemisorption of iodine on surfaces. The stainless steel tubing is polished inside and heat treated with steam to obtain a very thin layer of Fe_2O_3 on the inside walls. The same is done with glass sampling tubes to obtain a thin layer of SiO_2 . This technique has only been applied to experimental sampling systems associated with tests at the Malviken reactor. It is unknown how long such a treatment of the tubing would be effective.
- (b) Representativeness in this case means that the fraction of injected methyl iodide observed by the samplers is in proportion to the ratio of sampler to stack flow rates.

stacks varied from 10 to 100 mg and contained 3 to 5 mCi (24 to 40 ng) 131 I. The duration of injection is about 1/2 h.

The in-stack sampling rig is suspended by a rope from a pulley at the top of the stack. Samplers are attached to the rig in the same plane as the regular sampler nozzles. The flow rate through each sampler is measured individually. The samplers include

- eight filter holders behind isokinetic nozzles
- four centripeters(a) for particle-size measurement
- four iodine samplers consisting of a filter followed by a TEDAimpregnated charcoal cartridge. The filters are directly exposed to the flow. No nozzles are used, so the samples are not isokinetic.

The results shown in Table 4.2 show agreement within a factor of two, which was judged as being acceptable. A significant part (30% to 50%) of the 131 collected by the in-stack sampler was on the particle filter. (There was one extreme case of 90% collection on the filter.) With two filters in series, 11% of the 131 penetrating the first filter was collected on the second so some of the iodine was absorbed by the filter medium. Therefore, the iodine on the first filter was probably both particle-borne and absorbed gas. Comparison of the particle data is invalid, because the particle-borne iodine samples in the stack were not collected isokinetically.

4.3 ONTARIO HYDRO

Curtis and Guest (1986) reported in-place tests of iodine line losses in a stack sampler at Unit 4 of the Ontario Hydro Bruce A Power Station. The

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<u>TABLE_4.2</u> .	Average Particle-Borne and Gaseous Igdine
	Concentrations in Four Stacks, pCi/m ³
,	(Ström and Hesböl 1977)

	Stack Identification						
	011	RI	RII	<u>B2</u>			
Gaseous Iodine							
In-Stack Sampler	1270	10150	1220	14830			
Regular Sampler	1430	8400	2490	13000			
Particle-Borne Iodine							
In-Stack Sampler	1317	4920	11000	9170			
Regular Sampler	232	4526	1020	4100			

(a) A device that continuously separates airborne particles into size fractions.

sampler consisted of a horizontal multinozzle intake probe and a vertical delivery line 20 m long, with a 2.54-cm outside diameter and a 72-lpm flow rate. At the sampling elevation, the stack had a 1.32-m² cross section. The sampler tubing was 316 stainless steel and had been in service for several years. The air sample was collected on a glass fiber filter followed by a TEDA-impregnated carbon cartridge.

To accommodate the in-place test, the air sampler was modified with the addition of a Y-shaped stream splitter at the point where the sampling line exits the stack. A TEDA-impregnated carbon sample cartridge was short-coupled to one leg of the splitter. The other leg of the splitter was connected to the existing sampler line. The Airflow through both legs was kept equal, presumably 72 lpm. (If the flow rate was 72 lpm, the multinozzle probe would have been operating at twice its normal flow rate.)

To conduct a velocity traverse and further corroborative sampling, the stack was penetrated in five locations approximately 1 m above the sampler probe. Through each penetration, five samples were collected at equal time intervals at different locations (a total of 25) concurrently with the sampling experiments being run on the split stack sampler. Iodine was collected on a TEDA-impregnated carbon cartridge.

Initial tests using the three iodine samplers simultaneously were conducted using the iodine normally released from the plant, without the addition of iodine to the ventilation system. The iodine levels were far too low, so any bias between the samplers was masked by the counting statistics. Consequently, stable methyl iodide and elemental iodine were injected into the stack flow upstream of the fan. Collected iodine samples were analyzed using neutron activation. (A submicron particle tracer, sodium fluorescein, was also injected for particle line-loss studies.) The methyl iodide was made airborne by bubbling air through the liquid. Elemental iodine was generated by heating solid iodine in flowing air. The test durations were 3 to 8 h.

The test results are shown in Table 4.3. From these tests, it appears that the existing sampling system yielded results comparable with the splitter and the traversing samples. No significant bias caused by iodine line loss was discernible, and the repeatability of the test was good. These results do not show whether there was some resuspension of iodine after the injection experiment was concluded. In some cases, fresh collection cartridges were installed after the iodine injection experiment and were operated for 24 to 72 h. No residual iodine was collected, showing that for this sampling system, there was no significant retention and subsequent resuspension of iodine.

<u>TABLE 4.3</u>. Recovery of Injected Species (Curtis and Guest 1986)

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			Percent Recovery of <u>Injected Compound</u>				
		Injection		Splitter	Station		
Test	Injected	Concentration,	Traverse	Outlet	Monitor		
<u>No.</u>	<u>Species</u>	<u>ug/m³</u>	<u>Sample</u>	<u>Sample</u>	<u>Sample</u>		
1	CH3I	43 ± 3	98	95	99		
2	CHAI	89 ± 7	-	103	98		
3	CHAI	51 ± 4	-	96	104		
4	I ₂	22 ± 2	100	93	97		
5	12	41 ± 3	-	100	93		
6	15	34 ± 3	-	97	100		
`7	Uranine	1.1 ± 0.1	99	100	97		
	Particles	4					
8	Uranine	1.1 ± 0.1	115	106	112		
	Particles						
9	Uranine	1.1 ± 0.1	109	105	111		
	Particles						

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5.0 LABORATORY SIMULATIONS OF AIR SAMPLERS

Previous sections reviewed iodine deposition rate determinations on specimens of air sampler construction materials and in-place tests of iodine line loss in existing air samplers. In this section, we review work done to test simulated air sampling systems in the laboratory to determine values for the parameters affecting iodine/surface interaction. Because the experimental method and the interpretation of results are closely tied to a theoretical model of iodine behavior in an air sample, the model will be described first.

5.1 <u>REVIEW OF A MODEL FOR RADIOIODINE TRANSMISSION THROUGH</u> <u>SAMPLE DELIVERY LINES</u>

5.1.1 <u>Deposition and Resuspension</u>

Airborne concentrations of radioiodines released from nuclear operations are measured by sampling the contaminated atmosphere and withdrawing the sample through a sample delivery line. However, concentrations for both particles and gases change in the sampling line. The sampled concentrations can be modified by the processes of dry deposition and resuspension within the sample line. Dry deposition describes the processes by which radioiodines are removed to the internal surfaces of the sample delivery line, and resuspension describes the processes by which previously deposited radioiodines are again made airborne. Dry deposition and resuspension are a function of the radioiodine composition, surface properties of the sample delivery line, and the sampling flow rates. Dry deposition is usually greatest for turbulent air flow, and is increased significantly by discontinuities, such as elbows in the sampling line. Resuspension is greater for turbulent flow than for laminar flow. Neither dry deposition nor resuspension can be predicted accurately; measurements are required.

Dry deposition occurs for both particle and gaseous radioiodines. The removal rates of airborne radioiodines to the internal surfaces of a sample delivery line are described by dry deposition velocities. The dry deposition velocity, v_d , is defined as the deposition flux, N, (amount deposited per unit area per unit time) divided by the average airborne concentration, C, above the deposition surface. Thus, the dry deposition velocity is

 $v_d = - N/C$

(5.1)

(The minus sign is required because the flux to the surface is a negative quantity, while the deposition velocity is defined as positive.) The deposition velocity is usually reported in centimeters per second (Sehmel 1980).

Resuspension also occurs for both particle and gaseous radioiodines. The removal rates of radioiodines from the internal surfaces of the sample delivery line are described by resuspension rates. The resuspension rate is defined as the fraction of radionuclide resuspended per unit area per unit time; that is, in s^{-1} . The resuspension rate, r, is

$$r (units of s^{-1}) = \frac{N, flux away from internal surface (m^{-2} time)^{-1}}{G, radionuclide concentration on surface (m^{-2})} (5.2)$$

5.1.2 Iodine Mass Balance Equations

The model described here comes largely from Unrein et al. (1985) at Science Applications International Corporation (SAIC). The material has been supplemented by information in SAIC reports to various power reactor operators. The model has evolved and different forms have been used to account for differences in experimental methods and data.

The mechanisms described in the model are dry deposition onto the internal surfaces of the sample delivery line, chemical species transformation on the surfaces, and resuspension of deposited radioiodines. It is emphasized that although dry deposition velocities and resuspension rates cannot be predicted accurately as a function of controlling parameters, even less is known about species transformation rates on surfaces of sample delivery lines.

This model describes the sample delivery line as a sequence of shorter line segments composed of four hypothesized compartments in each segment. For each segment, dry deposition, chemical transformations on the surfaces, and resuspension rates are considered. Radioactive decay of 131 I is considered in the model because some of the sampling perjods for resuspension are long when compared to the 8.04-day half-life of 131 I. The model approach for calculating resuspension rates is an iterative data analysis technique to predict dry deposition and calculate resuspension rates. Details of this technique are not available.

Each segment of the model sample delivery line is shown in Figure 5.1. There are four compartments in each line segment: one compartment for radioiodines on the interior surfaces, and three compartments for airborne particles (iodine associated with particles) and elemental iodine, hypoiodous acid (HOI), and organic iodides. A single compartment is used for iodine associated with particles and elemental iodine because they are measured jointly in the experimental procedure. Although it is assumed that there is a single form of radioiodine on the surface, there are different reaction rates on the surface to form the different radioiodine species that are resuspended.





The differential equations used to describe the mass balance in any line segment are

$$\frac{dq_{12}}{dt} = I_{12} + r_{12}q_s - (\lambda + \lambda_v + d_{12}) q_{12}$$
(5.3)

$$\frac{dq_3}{dt} = I_3 + r_3 q_s - (\lambda + \lambda_v + d_3) q_3 \qquad (5.4)$$

$$\frac{dq_4}{dt} = I_4 + r_4 q_s - (\lambda + \lambda_v + d_4) q_4 \qquad (5.5)$$

$$\frac{dq_s}{dt} = d_{12}q_{12} + d_3q_3 + d_4q_4 - (\lambda + r)q_s$$
(5.6)

- where q_{12} = instantaneous airborne radioiodine activity, I_2 ; both particulate and gaseous (stages 1 and 2 of the species sampler), μ Ci
 - q_3 = instantaneous airborne activity of HOI (species sampler) stage 3), μ Ci
 - q_4 = instantaneous airborne activity of organic iodides (species sampler stage 4), μ Ci

q_s = instantaneous total radioiodine surface activity in the sampling line, μCi

 I_{12} = mean injection rate of particulate and gaseous I_2 into the line, $\mu Ci/s$

 I_2 = mean injection rate of gaseous HOI into the line, μ Ci/s

- I_4 = mean injection rate of gaseous organic iodide into the line, $\mu Ci/s$
- r_{12} = average resuspension rate constant for particulate and gaseous I_2 , s^{-1}

 r_3 = average resuspension rate constant for HOI, s^{-1}

 $r_4 = average resuspension rate constant for organic radioiodine, s^{-1}$

 $\lambda = radioactive decay constant for ¹³¹I, 9.97 \times 10⁻⁷ s⁻¹$ $\lambda_v = rate of removal of radioiodine from the tube, Q/V s⁻¹$ $d_{12} = deposition rate constant for particulate and gaseous I₂$ $d_3 = deposition rate constant for HOI$

 d_A = deposition rate constant for organic iodides

r = total resuspension rate of $r_{12} + r_3 + r_4$ (s⁻¹).

The rate constants d_{12} , d_3 , d_4 , and λ_V can be expressed in terms of other parameters. The deposition rate constants, d_i , for the different chemical species are the following

$$d_{12} = \left(v_d \text{ particles} \right) (A/V) \tag{5.7}$$

$$d_3 = (v_d \text{ HOI})(A/V)$$
(5.8)

$$d_4 = (v_d \text{ organic})(A/V)$$
 (5.9)

where A = the internal surface area of the sampling line segment, cm²

 v_d = area average dry deposition velocity for the noted species, cm/s

V = the volume of the sampling line segment, cm^3 .

The rate constant for removal of radioiodines in the air exiting the sample delivery line segment is

$$\lambda_{V} = Q/V \tag{5.10}$$

where Q =the air sampling flow rate, cm³/s.

5.1.3 Calculation of Dry Deposition Velocities

In an earlier version of the model, dry deposition velocities were calculated as

$$v_{d} = \frac{\left(C_{in} - C_{out}\right)}{A \bar{C}} Q \qquad (5.11)$$

where $(C_{in} - C_{out})$ = the difference in concentration between the inlet and outlet of the sample delivery line, $\mu C_i/cm^3$

 \bar{C} = the average radioiodine concentration in the sampling line during the injection period, $(C_{in} + C_{out})/2$, $\mu Ci/cm^3$.

This equation is an approximation and is useful where deposition is low. For high rates of deposition, it yields abnormally low values for v_d (see further discussion in Section 7.1).

In a modified modeling approach, the entire sample delivery line length is treated as a sequence of shorter line segments. Dry deposition velocities are calculated from transmission factors, TF_i , for each line segment, and the transmission factor, TF, for the total sample delivery line. The transmission factor is the ratio of outlet to inlet concentrations for the respective line lengths. If the number of line segments is n, and the line segments are of equal length, the transmission factor for each segment is $(TF)^{1/n}$. The average dry deposition velocity for the total sample delivery line is calculated from the equation

$$v_{d} = \frac{Q\left(1 - TF_{i}\right)}{A_{i} TF_{i}}$$
(5.12)

where A_j is the surface area of the i-th line segment of the sample delivery line. As the number of segments (n) becomes larger, v_d approaches an asymptotic value.

5.1.4 <u>Calculation of Resuspension Rates</u>

In keeping with the approach to consider the sampling tube as divided into short segments, the resuspension rate constants and surface activities are calculated for each segment of the sampling line, and then the average resuspension rate constant is determined. An iterative technique is used.

In this technique, the surface activity, q_{SO} , of radioiodines residing on the internal surfaces of the sampling line is assumed uniform over the length of the sample delivery line. The initial surface activity, q_{SO} , is calculated from the following mass balance

$$q_{so} = (C_{in} - C_{out}) Q T$$
(5.13)

where T is the elapsed time of sampling line exposure to fresh airborne radioiodines. The surface activity as a function of time is

$$q_{s}(t) = \frac{d_{12} q_{12}}{a_{s}} \left(1 - e^{-a_{s}t} \right) + \rho_{s0}e^{-a_{s}t}$$
(5.14)

where a_s is the $r_{12} + r_3 + r_4 + \lambda$ is the total removal rate constant describing the loss of radioiodine from the surface. The average surface activity during a sampling period is

$$\bar{q}_{s} = \frac{d_{12}q_{12}}{a_{s}} + \frac{d_{12}q_{12}\left(1 - e^{-a_{s}t_{s}}\right)}{a_{s}^{2}t_{s}} + \frac{q_{s0}\left(1 - e^{-a_{s}t_{s}}\right)}{a_{s}t_{s}}$$
(5.15)

It is assumed that time derivatives are equal to zero in Equations (5.3), (5.4), and (5.5); thus, resuspension rate constants are calculated after assuming that airborne concentrations are constant during each sampling period. After substituting the respective products of measured concentrations for each sampling period and the line volume, $C_j \times V$, into Equations (5.3), (5.4), and (5.5), the equations for calculating the resuspension rate constants for each sampling period follow

$$r_{12} = C_{12} V (d_{12} + \lambda_v + \lambda) / \bar{q}_s$$
 (5.16)

$$r_{3} = c_{3} V (\lambda_{v} + \lambda) / \bar{q}_{s}$$
 (5.17)

$$r_4 = c_4 V \left(\lambda_v + \lambda\right) / \bar{q}_s$$
 (5.18)

After q_{SO} is calculated, the analysis approach is to calculate selfconsistent values of the average surface activity and resuspension rate constants for the first measurement period. Then, surface activities are calculated for the next sampling period. The process is repeated for each of the remaining sampling periods.

With the experimental data, calculated resuspension rate constants often vary considerably over the sampling periods, as much as two orders of magnitude; however, average values are used in subsequent modeling of equilibrium and transient performance.

5.1.5 Equilibrium Transmission Factors

It is usually assumed that when the I_2 input is constant, the surface and airborne activity will eventually be at equilibrium levels. At equilibrium, the total airborne activity, qCi, for all radioiodine species arising from the constant input of the depositing species, I_v , is

$$q_{e} = q_{12e} + q_{3e} + q_{4e}$$
(5.19)

where the subscript e refers to the equilibrium condition. Equations (5.3) through (5.6) are solved for q_{12} , q_3 , and q_4 assuming $I_3 = I_4 = d_3 = d_4 = 0$ $dq_i/dt = 0$. Adding the resulting three equations and grouping constants yields

$$q_{e} = \frac{a_{s}I}{b} + \frac{d_{12}I(r_{3} + r_{4})}{a_{3}b}$$
(5.20)

where

$$b = a_{s}a_{12} - r_{12}d_{12} \tag{5.21}$$

$$a_{12} = d_{12} + \lambda_v + \lambda \tag{5.22}$$

$$a_3 = a_4 = \lambda_V + \lambda \tag{5.23}$$

At equilibrium, the output rate of radioiodine from the sample delivery line is $q_e\lambda_V$ (μ Ci/s). Because the input rate of I₂ is I₁₂, the equilibrium I₂ transmission factor, TF_e, is $q_e\lambda_V/I_{12}$. The deposition rate constant and the averaged resuspension constants are used to calculate TF_e for the depositing species of radioiodine.

To estimate a transmission factor for an inlet mixture of radioiodine containing HOI and organic iodides (which are considered nondepositing) a transmission factor of 1 is assumed for the HOI and organic species. The mixture TF_e is then computed using the weighted average, where the transmission factor for each species is weighted by its relative abundance in the effluent. It is unusual to have data on the actual species distribution, so SAIC calculates an example overall transmission factor based on species distributions reported in the literature (e.g., Pelletier et al. 1978a,b; Mandler et al. 1981). SAIC also estimates the time to reach equilibrium: from one to several days.

5.1.6 <u>Transient Transmission Factors</u>

The foregoing procedure is used by SAIC to estimate the transmission factor following step changes in radioiodine concentration; however, it is not exactly clear how. For a step increase in iodine concentration, the estimated transmission factor is initially low and gradually increases to the TF_e . The transmission factor is also often estimated for the cases of 1) an instantaneous cessation of iodine emissions, and 2) pulse emissions above an equilibrium value followed by a cessation in emissions. In both cases, the total cessation would be observed initially as a sudden concentration decrease, followed by a more gradual decrease to the new level.

5.2 <u>SAIC TESTS</u>

SAIC has conducted laboratory simulations of iodine transmission through sample delivery lines for some utilities. The customer's sample delivery lines are simulated by using tubing of equal lengths, size, and type that is then coiled to occupy a small space. Radioiodine is injected into the sample delivery lines, and samples of airborne activity are taken. Based on sample analyses, deposition velocity and resuspension rate constants are estimated, and equilibrium and transient elemental iodine transmission factors are calculated. The experimental procedures are described in greater detail below. For some utilities, tests had been performed previously in which sample delivery line physical and operational characteristics were similar; in these cases, deposition and resuspension rates from the earlier tests are used, and the performance of the sample delivery lines in question is predicted using the model described in Section 4.0.

5.2.1 Experimental Method

The experimental method is divided into two main parts: deposition and resuspension. The resuspension portion of the experiment follows the deposition portion as soon as sample collectors are changed and the flow system is altered so radioiodine is no longer injected into the line.

<u>Deposition</u>

Figure 5.2 is a schematic of the experimental apparatus. The elemental gaseous iodine was generated as described in Section 3.1 [Equation (3.1)]. Up to two sample delivery lines can be exposed to the airborne radioiodine at a time. Tubing of the same type, size, and length as that being simulated is coiled in a 7-ft diameter and coupled in the system.

Sample delivery line flow rates and enclosure temperature are controlled to simulate the conditions of interest. Usually, these conditions are those normally expected in the plant; however, other conditions are sometimes used to test performance under abnormal operations or to find operating parameters that will result in improved performance. The duration of the exposure to airborne radioiodine is usually less than 3 h. The humidity is the recorded value and has not usually been a controlled parameter.

The test apparatus includes a single inlet radioiodine sampler and an outlet sampler for each line. Samples are usually collected with SAIC's radioiodine species samplers (Pelletier et al. 1978a), but total iodine



FIGURE 5.2. Iodine Line-Loss Test Apparatus (Unrein et al. 1985)

collection cartridges are used if the concentration is too low for good analyses by the species samplers. The species sampler consists of the following components, or stages, in series

- filter paper or cartridge for particle collection
- cadmium iodide cartridge for the I² collection
- 4-iodophenol on alumina for hypoiodous acid (HOI) retention
- TEDA-treated charcoal cartridge for total collection of remaining iodine, taken to be organic iodides when following the above stages
- backup TEDA-treated charcoal cartridge.

Sampling cartridges are counted for 1^{31} I using a Ge(Li) spectrometer whose calibration is traceable to the National Bureau of Standards. Because some sampling intervals are long compared to the half-life of 1^{31} I (8.04 days), the results are corrected for decay occurring during sampling and between sampling and analysis. This correction factor assumes that the 1^{31} I concentration was constant during the sampling period. Counting uncertainties of 2 σ are generally less than a few percent for the deposition experiment.

The inlet species sampler analysis typically shows that 98% of the radioiodine is found on the I_2 stage, 1% each on the HOI and organic iodide stages, and the balance on the particulate stage. Total inlet radioiodine concentrations are on the order of 10^{-6} to 10^{-7} mCi/cm³. The total iodine concentration (radioactive and nonradioactive species) is not reported, but it is presumed that >90% of the total is not $13^{11}I$.

The sample delivery line outlet samplers show that total radioiodine concentrations range from a fraction of a percent to nearly 100% of the inlet concentration. The outlet species distribution is different from the inlet distribution if significant deposition and chemical reaction occurred. If the deposition has been severe, it would not be unusual to find nearly equal amounts (nearly 1/3 of the total amount) of radioiodine on each of the I₂, HOI, and organic species stages and a few percent on the particle filter. For slight deposition cases, the outlet species distribution is nearly equal to the inlet distribution.

<u>Resuspension</u>

After the samples for estimating deposition have been collected, the iodine generator is shut down and the sample cartridges replaced. To estimate the radioiodine resuspension rates, the air supply is switched to filtered laboratory air. Flow rates are held constant. Inlet samples are no longer taken, but outlet sampling continues over long periods of time with periodic changeout of cartridges. Resuspension experiments may continue for several weeks. Intervals between sampler cartridge changeout are nonuniform, and become longer with time. Sampling durations range from a few hours at the start to several days at the end of sampling. The test conditions are those normally expected in the plant; however, other conditions are sometimes used to test performance under abnormal operations or to find operating parameters that will result in improved performance.

Sample cartridges are counted in the manner already described. Counting uncertainties of 2 σ range from a few percent at the start of the experiment to slightly under 100% by the end of the experiment. Concentrations of the radioiodine species are considerably lower during this part of the experiment. If the iodine concentration becomes too low, analysis of one or more of the stages of the species sampler becomes impractical. The experimenter will then switch to a sampler consisting of a particle filter and a pair of TEDA-treated charcoal cartridges in series. This sampler is not species selective. Sometimes the species distribution is inferred from earlier resuspension samples.

Radioiodine concentrations generally decrease with time; however, occasional order-of-magnitude increases in concentration will be observed followed by a resumption in decreasing concentration. Species distributions during the resuspension experiment are either similar to that observed during the deposition experiment or with a slight decrease in collection on the gaseous I_2 stage and small increases on the other stages, suggesting that some reaction takes place over time.

5.2.2 <u>Test Results</u>

SAIC laboratory simulation test results were reported by Unrein et al. (1985) to simulate common effluent air samplers. The published results are supplemented here with data provided by staff from three of the reactor sites. The tests at least partly cover the range of air sampler operating characteristics commonly observed at reactor sites during normal operation. The range of characteristics include

- tube inside diameters -- 1/4 to 1-3/8 in.
- flow rates -- 0.06 to 14 cfm
- tube lengths -- 50 to 256 ft
- temperature -- 25° to 30°C
- relative humidity -- 25% to 75% (laboratory conditions; an uncontrolled parameter).

All tubes were either 304 or 316 stainless steel as clean as received from the distributor. This range of characteristics may adequately cover effluent samplers under normal conditions, but there is some question whether accident conditions or containment sampler conditions are satisfactorily covered.

Table 5.1 is a summary of the simulated sampling line characteristics, test conditions, and results at four of the reactor sites listed in

											<u>Resuspensi</u>	on Experiment
	Tube			Surface Area-to-				Injectio	n Experime	nt		Average 131 _I
Reactor <u>System</u>	Inside Diameter, in.	Flow Rate, cfm	Tube Length, <u>ft</u>	Volume Ratio, <u>1/ft</u>	Residence Time, s	Approx. Reynold's <u>No.</u>	Duration, min	Inlet, μCi/mL _ <u>E-7</u>	Observed C/Co	Deposition Velocity, cm/s	Duration,	Resuspension Rate, E-6/s
14a	0.252	0.06	140	191	48	376	30		0.0013	0.027	742	1
C2a	0.252	0.06	140	191	48	376	30	19.7	0.00133	0.023	521	0.075
14b	0.252	0.06	140	191	48	376	126		0.0045	0.021	904	1
С2Ь	0.252	0.06	140	191	48	376	128	4.24	0.0045	0.019	904	0.38
13a	0.752	2.00	141	64	13	4192	30		0.23	0.054	742	7
C1a	0.752	2.00	141	64	´ 13	4192	30	19.7	0.235	0.054	521	0.95
GC	0.618	<i>4</i> .00	198	78	6	10209	?	2.77	0.397	0.06	232	0.28
15	0.870	2.00	256	55	32	3623	258		0.58	0.0095	282	7
С1Ь	0.752	2.00	141	64	13	4192	128	4.24	0.62	0.018	904	2.2
13b	0.752	2.00	141	64	13	* 4192	126		0.62	0.018	904	10
E1	1.126	10.00	157	43	7	14009	285	2.18	0.68	0.043	692	1.08
Gb	1.370	14.00	198	35	9	16119	?	0.805	0.687	0.038	234	0.91
Ga	1.370	12.83	88	35	4	14773	?	6.15	0.693	0.076	237	0.071
16	0.835	2.80	246	58	20	5292	258		0.72	0.0088	282	10
11	0.874	3.00	100	55	8	5417	72		0.75	0.02	455	6
12	0.874	3.00	50	55	4	5417	72		0.78	0.032	455	5

TABLE 5.1. Summary of Air Samplers Simulated in Tests

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Table 2.2. The first several columns in the table describe the sample line simulated -- inside diameter, length, flow rate, surface-to-volume ratio, residence time, and Reynold's number. The next part of the table summarizes the iodine injection part of the experiment, listing the experiment duration, the injected radioiodine concentration, the total 131 I transmission factor observed during the injection experiment, and the reported elemental iodine deposition velocity. The rest of the table lists the duration of the resuspension experiment and the average resuspension rate for all 131 I as calculated using the model. To aid in comparisons, the tests shown in Table 5.1 are listed in ascending order of observed total 131 I transmission factor (TF or C/Co).

Note that Tests Clb and C2b are repeats of Tests Cla and C2a, respectively. These two tests were repeated to collect improved data for the resuspension experiment. The results for the repeat tests differ greatly from each other. It should also be observed that Tests 3 and 4 reported by Unrein et al. (1985) are equivalent to Tests Cl and C2.

The deposition velocities reported range from 0.0088 to 0.07 cm/s. This is comparable to Kabat's data for I_2 on uncleaned metal at low humidity. While the deposition velocity varies by only a factor of 10, the transmission factors observed cover a wide range because of the differences in sample characteristics. The reported average resuspension rates range from 7.1 x 10^{-8} s⁻¹ to 1.0 x 10^{-5} s⁻¹.

Part of the large difference in the reported resuspension rates in Table 5.1 is because Unrein et al. (1985) used a preliminary form of the model described earlier to estimate the resuspension rates. For the other tests, the average resuspension rates varied from 7.1 x 10^{-8} s⁻¹ to 2.2 x 10^{-6} s⁻¹. This is comparable to the variability in resuspension rates determined for each sampling period in the resuspension part of the tests. The data for the resuspension tests are given in Appendix A.

Table 5.2 lists more detailed data for the iodine injection part of some of the tests. The inlet and outlet concentrations for total 131 I and the species distributions are given. Note that for the Reactor C2 sampler, the outlet species distribution differs greatly from that of the inlet. This was interpreted as a result of the reaction of I₂ with the tube surface, producing the other less-depositing iodine compounds. In this case, the reaction occurred where deposition was high, perhaps because of the high surface area/tube volume ratio and long residence time. In the other tests, the I₂ is the dominant iodine form in the inlet and outlet ends of the tube as generated in the injection apparatus.

Detailed data on iodine species distributions, concentrations, and resuspension rates are given in Appendix A. The shift in iodine species distribution where deposition was high was also observed in the long-term resuspension experiments as shown in Table A.3. The 131 I concentrations measured during weeks of the resuspension experiment can fluctuate from 2 to 3 orders of magnitude, sometimes even increasing after many days, as shown in

ç		¹³¹ I	Species Distribution, %						
Reactor <u>System</u>		µCi/cm ³ _ <u>x_E-7_</u>	Parti- <u>culate</u>	12	HOI	<u>Organic</u>			
Cla	Inlet	19.7	1.5	98.4	0.05	0.01			
	Outlet	4.62	1.4	97.7	0.9	0.1			
C1b	Inlet	4.24	0.24	99.2	0.45	0.07			
	Outlet	2.63	0.57	98.8	0.41	0.2			
C2a	Inlet	19.7	1.5	98.4	0.05	0.01			
	Outlet	0.0262	7.9	46.8	33.3	12.1			
C2b	Inlet	4. 24	0.24	99.2	0.45	0.07			
	Outlet	0.0191	3.1	36.2	36.7	24.0			
Ε	Inlet Outlet	2.18 1.48	2.2 2.1	96.9 95.7	0.8 2.1	0.1			
Ga	Inlet	6.15	_{NA} (a)	NA	NA	NA			
	Outlet	4.26	NA	NA	NA	NA			
Gb	Inlet	0.805	NA	NA	NA	NA			
	Outlet	0.553	NA	NA	NA	NA			
Gc	Inlet	2.77	NA	NA	NA	NA			
	Outlet	1.1	NA	NA	NA	NA			

<u>TABLE 5.2</u>. Inlet and Outlet Concentrations and Iodine Species Distributions (from reactors noted in Table 2.2)

(a) NA = Not Available

Table A.4. Consequently, the resuspension rates calculated using the method described in Section 5.1 can fluctuate greatly, as shown in Table A.5. Resuspension rates for each species are then inferred from whatever species distribution data were available, as was done to generate data for Reactor C Test 2b in Table A.6.

5.2.3 Predicted Transmission Factors

A comparison of the tabulated values of transmission factors measured during the injection experiment and the predicted equilibrium values points out the difference that accounting for resuspension can make. The predicted equilibrium values suggest that the eventual performance of a sampling line can be better than what might be determined from a short test; however, to our knowledge, there has been no direct experimental verification of the predicted transmission factors based on these laboratory tests. Test C2b in Table 5.3 shows what may be typical of post-accident samplers (PAS). The observed transmission performance during the injection experiment was exceedingly poor. The predicted equilibrium transmission performance is much better, because resuspension is accounted for; however, it is still not as good as is desirable, and the time to reach the improved performance is long, perhaps too long compared to the length of a significant release event of short duration. This result has prompted some utilities to modify the PAS to extract the high-level samples as a subsample from the routine sampling system operated in the normal manner, thus taking advantage of the better performance of the normal sampler to transport the sample to the subsampling point.

SAIC's reports present predicted transmission factors for depositing iodine species during transient events. The purpose is to assist the customer in interpreting sample data during and following such events. Examples are given for performance following a step increase and decrease in concentration. Because the injection part of the experiment simulates a step increase of short duration, it seems reasonable to assume that the predicted performance immediately following a step increase should be comparable to the performance observed during the experiment. As shown in Table 5.4, this was generally true except for the Reactor C Tests 1b and 2b. The model predicts that if the increased concentration persists, the performance improves until the equilibrium value is reached in the time given in Table 5.3.

Example transient performance is also given for a step decrease if the sampling system had been operating at equilibrium and only sampling depositing species of iodine. Table 5.5 shows the predicted measured airborne concentration of previously deposited iodine that would be measured by 24-h samples terminated on the day shown following a decrease to zero iodine in the stack. The quantity shown is that relative to the concentration previous to the event. The iodine collected is previously deposited iodine that has become resuspended. If depositing iodine was only a portion of the total

<u>TABLE 5.3</u> .	Predicted Equilibrium and Measured Transmission
	Factor Following a Step Increase in Depositing
	Iodine Species

	Measured in		Approximate
Reactor	Injection	Predicted at	. Time to Reach
<u>System</u>	<u>Experiment</u>	<u>Equilibrium</u>	<u> Equilibrium</u>
C1b	0.62	0.86	2 wk
Ć2b	0.0045	0.19	2 wk
E	0.68	0.88	>200 h
Ga	0.693	0.71	24 h
Gb	0.687	0.82	>160 h
Gc	0.397	0.48	>160 h

<u>TABLE 5.4</u>. Predicted Short-Term and Measured Transmission Factor Following a Step Increase in Depositing Iodine Species

Reactor <u>System</u>	Measured in Injection <u>Experiment</u>	Short-Term Performance <u>Predicted</u>
C1b	0.62	0.67
C2b	0.0045	0.15
E	0.68	0.68
Ga	0.693	0.69
Gb	0.697	0.69
Gc	0.397	0.40

<u>TABLE 5.5</u>. Measured 24-h Concentration Relative to Starting Concentration for a Step Decrease to Zero Iodine in Stack

	Reactor System											
<u>Day</u>	Ga	Gb	GC	<u> </u>	<u>C1b</u>	C2b						
1	0.023	0.15	0.17	0.20	0.18	0.22						
2	0.021	0.13	0.15	0.16	0.15	0.20						
3	0.018	0.11	0.14	0.12	0.12	0.18						
4	0.016	0.10	0.13	0.097								
5	0.015	0.08	0.11	0.076	0.08	0.15						
6	0.013	0.07	0.10	0.060								
7				0.047	0.06	0.13						
8	0.011	0.05	0.08									
10					0.03	0.10						
15					0.01	0.06						

iodine before the event, the total measured iodine after the event would be proportionately lower. Unfortunately for comparison and verification purposes, the situation modeled here does not correspond to the conditions under which the resuspension data are taken. It would be of interest to see if the model could reproduce both the deposition and resuspension data.

5.3 EG&G TESTS

Edson et al. (1987) of EG&G Idaho conducted tests on simulated air sampler tubing. The experimental method was similar to that of SAIC,

described in Section 5.2.1. The tests apparatus was similar to that shown in Figure 5.2, except the relative humidity was controlled at 50%, and iodine was generated by the subliming radiolabeled I_2 crystals in air flowing at 5 cm³/min. The generation rate was about 0.024 mg/min. The airborne iodine was transported to a chamber where it was mixed with additional room air. Iodine sampling initially used a species sampler similar to that described earlier, except a silver zeolite cartridge was used instead of the TEDA-impregnated charcoal as the fourth stage of the sampler.

The test parameters were as follows

- temperature -- 30°C
- relative humidity -- 30%
- simulated sampling line:
 - 304 stainless steel
 - 150-ft length made from 20-ft sections welded together and coiled into a 5-ft diameter
 - 0.86-in. inside diameter
 - 27.3 ft/s air velocity inside tube stated airflow rate of 5.5 cfm (inconsistent with the above two parameters, which indicate an airflow of 6.6 cfm) 55.8 ft⁻¹ surface-to-volume ratio.

Four tests were conducted on the same tubing. The problems with the experimental method are as interesting as the results. Each tests consisted of a 4-h injection experiment followed by a 100-h resuspension experiment. During the injection experiment, six continuous and consecutive samples were collected at the inlet to the tube (via a subsample from the mixing chamber) and six samples from the outlet of the tube. Inlet and outlet samples covered the same time periods. For the resuspension experiment, iodine generation was stopped, and six continuous consecutive samples were collected from the tube outlet.

After the first test, the inlet samples showed an erratic iodine generation rate. For the second test, the apparatus was modified with the installation of a stainless steel union in the 1/4-in. Teflon tube from the iodine generator to the mixing chamber. This allowed the experimenter to temporarily disconnect the injection tube form the mixing chamber, operate the generator for 1 to 2 h until the rate was stable (exhausting into a charcoal filter), and reconnect the tube to the mixing chamber. Significant quantities of particulate were observed in the outlet samples of the first test during the resuspension experiment, so a particulate sample was collected at the tube inlet mixer during part of the second test's resuspension phase. The second test showed that the particulate material was also present at the inlet mixer samples, so the mixing chamber air was filtered during the remaining tests. (The particles were identified as coming from the brushes of the fan motor in the test chamber.) The species samplers used in the first two tests showed that no HOI or organic iodides were collected, so only a particle filter followed by a charcoal cartridge were used to collect samples for the remaining tests. Also, inlet mixer samples were collected during the first 6 h of the resuspension experiment of Tests 3 and 4 to determine whether iodine injection had really stopped, because a large increase in outlet concentration was observed several hours into the resuspension part of Test 2.

Test 3 showed that the stainless steel union in the 1/4-in. Teflon injection line was a significant site of iodine deposition an later resuspension. The union was modified so that the ends of the Teflon tubing butted up to each other, effectively removing the ion as a deposition site for Test 4. Also, the test tubing was cleaned with solvent before Test 4 to remove particles that had collected from the first two tests and to investigate the effect of cleaning. These test changes demonstrate that some refinement of the experimental method is necessary before reliable data will be collected.

6.0 MODEL-BASED PREDICTIONS OF IODINE AIR SAMPLER PERFORMANCE

Science Applications International Corporation (SAIC) has used the previously described model techniques to analyze the performance of several iodine air sampler systems for reactor sites. Two sites have made these reports available to us. The characteristics of the analyzed systems are listed in Table 6.1. The model used has been generally described earlier; however, SAIC sometimes modifies the model as appropriate to fit various conditions.

To apply the model to predicting sampler line loss, a deposition velocity and resuspension rate must be selected. In one case, only the total radioiodine resuspension rate was given. Presumably, values from experimental data taken for a tube size and flow rate that best fits the situation would be chosen. Care must be taken to choose values derived with the same model to be used to predict sampler performance. This is particularly true for choosing resuspension rates. This is illustrated in the first six lines of Table 5.1 where three of the tests reported by Unrein et al. (1985) were the same as tests made for utilities. The resuspension rates in the report furnished to the utility were significantly lower than reported by Unrein because an earlier form of the iodine line-loss model was used to derive the values in his report. The resuspension rates reported by Unrein cannot be used in the model described in Section 5.1 nor usefully compared with values obtained by other methods. Deposition velocities seem to be less sensitive to the version of the model used. This will be discussed in Section 7.0.

To determine if sampler characteristics could be used as an indicator of appropriate line-loss parameters, some preliminary analysis was performed using scatterplots of sampler characteristics (tube diameter, length, flow rate, Reynolds number, residence time, and surface-to-volume ratio) versus the listed deposition velocities and resuspension rates. The analysis revealed no significant correlation. This was disappointing; however, it was probably due to differences in the methods used to calculate the resuspension rate and deposition velocity from test to test. A lack of correlation would add to the uncertainty in choosing, from existing data, the model parameters for input to a purely theoretical prediction of sampler performance. This would lend credence to the argument that each system should be tested. The apparent lack of repeat tests to demonstrate the precision of the results / casts some doubt on whether the results can be reliably applied to the real systems in question. Also, no sensitivity analysis has been performed to show the effects of the assumptions on the results.

The SBGT B and the MAIN C lines of Table 6.1 are both PAS systems. We presume that the deposition and resuspension values were selected from earlier tests with similar systems. The MAIN C line is for a PAS system modified to use a subsample from the normal sampling system line, MAIN A. The elemental iodine transmission through MAIN A and MAIN C combined results in about 77% transmission. Compared with the similar line, SBGT B, the modified system should perform much better. (It is unfortunate that many reactor PAS

Reactor (from <u>Table 2.2)</u>	<u>Line</u>	Inside Diameter 	Line Length, ft	Flow Rate, <u>lpm</u>	Surface- to-Volume _Ratio, L/cm	Residence Time, s	Approximate Reynolds Number	Operating Temperature, °C	<u>Ass</u> Deposition Velocity, cm/s	umed Resuspension Rate, E-6/s	Predic Species <u>Transm</u> _ <u>1</u> 2	ted Indi Equili ission I HOI	ividual ibrium <u>Factor</u> <u>CHzI</u>
D	SBGT B	0.95	156	1.70	4.20	120	252	32	?	?	0.025	0.85	0.99
F	1	2.54	2250	40.00	1.57	58	2228	21	0.01	1	0.54	0.93	0.99
F	2	2.54	250	99.12	1.57	23	5521	21	0.01	1	0.77	0.97	1
D	MAIN A	3.18	230	70.80	1.26	47	3155	32	?	?	0.85	1	1
D	MAIN A	1.91	85	65.14	2.10	7	4837	32	combined	combined end-to-end with above			
D	SBGT A	1.91	161	42.48	2.10	20	3155	32	?	?	0.86	1	1
D	MAIN C	0.95	3.5	1,70	4.20	3	252	32	?	?	0.9	1	1

<u>TABLE 6.1</u>. Sampling Systems Analyzed Theoretically

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6.2

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systems are of the same type as SBGT B.) Line 2 in Table 6.1 is a hypothetical illustration of how the performance of line 1 in the table could be improved by increasing the flow rate from 40 to 99 lpm.

ł

7.0 SUGGESTED MODELING IMPROVEMENTS

Improvements are suggested here for calculating dry deposition velocities and resuspension rates. The improvements for dry deposition are the use of one equation for high removal rates, and a series expansion of that equation for analysis of low removal rates. The improvements for resuspension include the estimation of the quantities of each radioiodine species that have not resuspended at the conclusion of each experiment. The analysis of the resuspension data suggests that for each radioiodine species there may be two main types of species resuspended, a readily resuspended form during the early portion of the resuspension experiment and, subsequently, a less readily resuspended form.

7.1 DRY DEPOSITION VELOCITIES

Dry deposition velocities can be calculated by using the inlet and outlet concentrations for the sample delivery line. The descriptive equations are derived using a mass balance for flow in a tube. Airborne concentrations and air velocities are assumed uniform for any cross section of the tube. The differential equation (Sehmel 1968) describing the change in the quantity of airborne radioiodine and the deposition flux to the surface is

$$\frac{\pi D^2}{4} u dC = -(v_d C) (\pi D dx)$$
(7.1)

where u is the average air velocity, and x is the distance along the tube axis. After rearranging and integrating from an inlet concentration, C_0 , to an outlet concentration, C, the change in airborne concentration can be expressed as

$$\ln \left(\frac{C}{Co}\right) = -4 \left(\frac{V_d}{u}\right) \left(\frac{L}{D}\right)$$
(7.2)

where L equals the total tube length. After rearranging, the dry deposition velocity is calculated from the expression

$$v_d = -\left[\begin{pmatrix} \underline{u} \\ 4 \end{pmatrix} \begin{pmatrix} \underline{D} \\ L \end{pmatrix} \right] \ln \left(\frac{\underline{C}}{\underline{C}_0} \right)$$
 (7.3)

Equation (7.3) can be used to calculate dry deposition velocities when there is a large change in airborne concentrations. However, if dry deposition is small, the ratio C/C_0 is nearly unity, and dry deposition velocities cannot

be calculated accurately using Equation (7.3). For small changes in airborne concentration, the first term of a power series expansion for the logarithm is used to evaluate the dry deposition velocity. Thus for small deposition, the dry deposition velocity is calculated from either of the following expressions:

$$v_{d} = -\frac{\left(C_{0} - C\right)}{\bar{C}} \left(\frac{Q}{A}\right)$$
(7.5)

where A is the internal surface area of the sample delivery line (A = π DL). This is the same equation [see Equation (5.11)] used in the SAIC approach.

Equation (7.4) is needed only to evaluate dry deposition velocities for small changes in concentrations. An indication of the range of small concentration changes will be illustrated by assuming the product (u/4)(D/L) is equal to unity. Dry deposition velocities calculated from Equations (7.4) and (7.5) are shown in Table 7.1 for comparison.

The ratio of dry deposition velocities calculated from Equation (7.4) divided by the dry deposition velocity calculated from the series expansion of Equation (7.5) is shown in the last column of Table 7.1. Either equation

	i i	' <u>Dry Deposition Velocity, cm/s</u>					
Percent		from	Series Expansion	(Eq. 7.4)/			
<u>Deposition</u>	<u> </u>	<u>Equation (7.4)</u>	from Equation (7.5)	<u>(Eq. 7.5)</u>			
30	0.70	0.3567	0.3529	1.011			
20	0.80	0.2231	0.2222	1.004			
10	0.90	0.1054	0.1053	1.001			
5	0.95	0.0513	0.0513	1.000			
1	0.99	0.010050	0.010050	1.000			
0.5	0.995	0.005013	0.005013	1.000			
0.4	0.996	0.004008	0.004008	1.000			
0.3	0.997	0.003005	0.003005	1.000			
0.2	0.998	0.002002	0.002002	1.000			
0.1	0.999	1.00E-03	1.00E-03	1.000			
1.E-0Ź	0.9999	1.00E-05	1.00E-05	1.000			
1.E-03	0.99999	1.00E-05	1.00E-05	1.000			
1.E-04	0.999999	1.00E-06	1.00E-06	1.000			
1.E-05	0.9999999	1.00E-07	1.00E-07	1.000			

TABLE 7.1.	Comparison	of	Calcul	′ated [Dry De	position	Velocities

can be used between 10^{-5} to 5% deposition. For deposition greater than about 5%, the series expansion becomes less exact. The series expansion [either Equation (5.11) or (7.5)] is needed only if dry deposition is even less than 10^{-5} %.

The SAIC approach using small line segments in Equation (5.12) is an unnecessary procedure that attempts to approximate Equation (7.4). We recommend that when deposition is low (even less than 10^{-9} %), Equation (7.5) can be used. However, when deposition is high (all cases of interest for this review), Equation (7.4) should be used.

Deposition velocities calculated by Equation (7.2) are compared with those reported by SAIC in Table 7.2. Ratios of dry deposition velocities are shown in the last column. The ratio is the literature value for the dry deposition velocity divided by the dry deposition velocity value calculated for this report. The average ratio is near unity, with a maximum ratio of 1.011.

7.2 <u>RESUSPENSION_ANALYSIS</u>

The analysis of the SAIC model has been limited for three reasons. First, the assumption that the derivative with respect to time is zero in Equations (5.3) to (5.5) may be too great a simplification. The time rate of change is experimentally observed, is quantified, and should be included in the analysis. Second, effects of error propagation resulting from inaccuracies in calculating dry deposition velocity may result in uncertainties in resuspension rates calculated with the SAIC iterative model. Third, SAIC's and the following analyses of resuspension data for each radioiodine species suggest that resuspension rates decrease as a function of time for each radioiodine species. Only an average of resuspension rates (which vary over three orders of magnitude) for each radioiodine species is included in the SAIC model.

The following subsections describe an alternate approach to modeling resuspension. This approach describes the amounts of radioiodine on the internal surfaces of sample tubes, not the airborne concentrations. The results of this approach suggest that resuspension rates decrease with time. In addition, the analysis suggests that resuspendable quantities of each radioiodine species remaining on the internal surface of the sample delivery line at the conclusion of each resuspension experiment can be estimated. Chemical reaction rates occurring on surfaces have not been addressed, but organic iodines are believed to be formed. Assumptions used to develop this model include the following:

- Only the radioiodine that is resuspendable is considered. Permanently fixed radioiodine does not enter into the analysis.
- Only resuspension and radioactive decay are assumed to change the amount of each radioiodine species on surfaces.

TABLE 7.2. Comparison of SAIC's Reported Deposition Velocities to Values Calculated by Preferred Method

						Dry Deposition Velocity,							
					D / / / /)	cm/s							
	0	л	1	(Cin)/	· UU/(4L)=		Calc.	<u>Ratio</u>					
line	Q3/c	U,	L,	(C, .)	$q/(\pi UL),$	Penarted	Poport	Reported					
<u>Erne</u>			<u></u>	Zout	5	Reported	Report	Calculated					
Prom SAIL LUSLOMER REPUBLS													
C _{in} /C	put = (4.6)	2E-7)/(1.97E-6)									
. .	944	1.91	4293	0.235	0.0367	0.054	0.0531	1.02					
Reactor U, $2A = (2.625.0)/(1.075.6)$													
$C_{in}/C_{out} = (2.52E-9)/(1.9/E-6)$													
. .	28.3	0.64	4267	0.00133	0.00330	0.023	0.0219	1.05					
Reactor C, 1B													
$C_{in}/C_{out} = (2.63E-7)/(4.24E-7)$													
	944	1.91	4293	0.620	0.0367	0.018	0.0175	1.03					
Reacto	or C. 2B		·										
C _{in} /C	put = (1.9)	1E-9)/(4.24E-7)		_							
	28.3	0.64	4267	0.00450	0.00330	0.019	0.0178						
•													
From (Inrein et	al: (19	85)										
1	1416	2.22	3048	0.75	0.0666	0.020	0.0192	1.04					
2	1416	2.22	1524	0.78	0.133	0.032	0.0330	0.97					
3a	.944	1.91	4293	0.23	0.0366	0.054	0.0538	1.00					
3b	944	1.91	4293	0.62	0.0366	0.018	0.0175	1.03					
4a	28.3	0.64	4267	0.0013	0.00330	0.027	0.0219	1.23					
4b	28.3	0.64	4267	0.0045	0.00330	0.021	0.0178	1.18					
5	944	2.21	7803	0.58	0.0174	0.0095	0.0095	1.00					
6	1321	2.12	7498	0.72	0.0265	0.0088	0,0087	1.01					
Reacto	or G, A		-										
Q = 12	2 . 83 cfm												
L ≈ 88	3 ft												
C_{in}/C_{c}	$_{out} = (4.2)$	6E-7)/(6.15E-7)									
	6056	3,48	2682	0.693	0.207	0.076	0.0759	1.00					
Reacto	or G, B												
Q = 14	4 cfm			4									
L = 19	98 ft					•							
C_{in}/C_{c}	$C_{in}/C_{out} = (5.53E-8)/(8.05E-8)$												
	6608	3.48	6035	0.687	0.100	0.038	0.0375	1.01					
Reacto	or G, C												
$Q = 4 \mathrm{cfm}$													
L = 19	98 ft		1										
$C_{in}/C_{out} = (1.10E-7)/(2.77E-7)$													
	1888	1.57	6035	0.397	0.0634	0.060	0 0586	1 024					
	1000	1.0,	0000	5.551	V.00JT	0.000	0.0000	1.024					

- The iodine resuspended at the first part of a sampling tube does not redeposit and resuspended several times before being collected at the end of the tube.
- Chemical reactions on surfaces of the sample delivery line are neglected.
- The amount of each radioiodine species still available for resuspension at the completion of each experiment can be estimated from the data.

The proposed model describes the time rate of change of amounts of each radioiodine species, Ni, on internal surfaces of the sample delivery lines as modified by resuspension rates, r, and radioactive decay. The time rate of change is described by

$$\frac{dN}{dt} = -(r + \lambda) N$$

where λ = the radioactive decay constant for ^{131}I

- N = the amount of the resuspendable iodine species on the tube surface
- r = the resuspension rate, 1/time
- t = the time since the start of the resuspension experiment.

Equation (7.6) integrates to

$$\ln \left(\frac{N}{N_0}\right) = -(r + \lambda) t \qquad (7.7)$$

(7.6)

where N is the resuspendable amount of the iodine species on the tube surface at the beginning of the experiment. When the left-hand side of Equation (7.7) is plotted versus time, the slope of the line at any instant is the sum of the resuspension rate plus the decay rate. The resuspension rate within an experimental time interval is evaluated from the data obtained at the limits of the time interval. The challenge in Equation (7.7) is to estimate N₀. The method we used to estimate N₀ is presented in the next subsection.

7.2.1 <u>Calculation_of_No</u>

 N_0 is estimated from the total mass of each species resuspended during each experimental time interval with appropriate decay correction back to the start of the experiment. The procedure is illustrated with SAIC resuspension

data (provided by one of SAIC's customers) from a simulated sampling tube. The simulated tube was from Reactor C, described in Table 5.7, and the resuspension data are detailed in Table 7.3. The amounts of resuspended ¹³¹ I were determined for 15 successive sampling intervals of duration shown in Column A of the table. Column B shows the time elapsed from the start of the resuspension experiment to the end of each time increment. The reported airborne ¹³¹ I concentrations measured during each sampling interval are listed in Column C of the table and converted into mass units in Column G using Equation (7.8),

where the air flow rate during resuspension is $944 \text{ cm}^3/\text{s}$, and the time duration is given in Column A for the collection of each sample.

Species distribution data were obtained for the first 14 samples. The majority of radioiodines resuspended were in the particle and elemental iodine fractions. The data for particles and elemental iodine are given in Table 7.3, and the data for HOI, organic, and total iodines are given in Appendix B in Tables B.1 through B.3. The percentages of the 131I measured as particles and elemental iodine are listed in Columns D and E (Table 7.3), and their sum, the total percentage of the 131I accounted for as particles + elemental iodine, is reported in Column F. The product of the particle + elemental fraction and the total amount of resuspended 131I in Column G is shown in Column H, the estimated mass of the particle + elemental iodine were stable rather than radioactive, N₀ would be the sum of the amounts in Column H; however, the iodine is radioactive, so the amounts resuspended during each sampling interval are corrected for decay, using Equation (7.9), to estimate the corresponding amounts at the start of the resuspension experiment.

$$R_{ot} = R_t e^{\lambda t}$$
(7.9)

where R_{+} = mass resuspended during an interval

 R_{ot} = original mass on the tube surface corresponding to the mass resuspended, R_t , during the sampling interval.

These corrected amounts resuspended, Ro_t , are termed "undecayed" and are listed in Column I of Table 7.3. The desired value of N_0 is the sum of these decay corrected masses, as shown in Equation (7.10) and at the bottom of Column I.

$$N_{o} = \Sigma \quad (R_{ot}) \tag{7.10}$$
TABLE 7.3. Calculation of Resuspension Rates Versus Time for Particles Plus Elemental Iodine

		-	131				Resusp	ended nCi/l	nlerval	nCi	12 + Partic	le on Surfac	e	Normalized	ln(N/N_)	Resuspensi	on Rates,	Section-1
	Sample	lest	Concen-	Speci	65	Percent			Under ayed									Ratio
Period	Duration	Cumulstive Minimum	tration,	Perce		Particles	1311	Particles +1-	Particles +1.5	Undecayed	Ad susted	UnadiusLed	Decayed Adjusted	Unadiusted	Adjusted	Uncayed Unadjusted	Decayed Adjusted	Adjusted/ Unadjusted
2FR0	0	0	0	0	0	0	0	0	0	1093.97	1243.97	1093.97	1243.97	0 -	0			
1	890	890	7.072-09	2.3	95.1	97.4	356.40	347.13	366.10	727.87	87/.87	690.15	832.3/	-0.4607	-0,4018	7.6E-06	6.5E-06	0.86
2	1440	2330	2.93E-09	0.7	95.2	95.9	238.98	229.18	263.44	464.43	614.43	404.03	534.52	-0.9961	-0.8447	5.20-06	4.1E-06	0,79
3	1440	3770	7.90E-10	1.7	90.7	92.4	64.43	59.54	74.59	389.84	539.84	311.16	430.89	-1.2573	-1.0602	2.0E-06	1.5E-06	0,74
4	1470	5240	7.11E-10	1.6	88.6	90.2	59.20	53.40	73.05	316.79	466.79	231.58	341.23	-1.5526	-1.2935	2.4E-06	1.6L-06	0.70
5	1994	8124	4.68E-10	1.1	87.6	88.7	49.94	44.30	67.82	248.97	398.97	162.61	260.58	-1.9062	-1.5632	Z.1E-06	1.4E-06	0.65
6	2436	9560	1.85E-10	1.8	85.1	86.9	25.53	22.18	39.29	209.68	359.68	118.39	203.08	-2.2236	-1.8125	1.22-06	7.16-07	0.60
)	3066	12620	7.218-11	3.8	81.2	85.0	12.50	10.62	22.59	187.09	337.09	87.97	158.50	-2.5206	-2.0603	6.2E-07	3.5E-07	0.57
8	3120	15740	6.23E-11	8.6	84.1	92.7	11.01	10.21	26.16	160.93	310.93	62.79	121.32	-2.8577	-2.3277	8,06-07	4.3E-0/	0.54
. 9	4030	19770	5.98E-11	0.9	90.9	91.8	13.65	12.53	40.87	120.07	270.07	36.81	82.81	-3.3917	-2,7095	1.2E-06	5.8E-07	0.48
10	4715	24485	3,268-11	5.2	68.6	73.8	8.71	6.43	27.78	92.29	247.29	21.35	56.04	-3.9367	-3.1000	9.3E-07	3.8E-07	0.41
11	5280	29765	2.00E-11	1.0	79.1	80.1	5,98	4.79	28.40	63.88	213.68	10.78	36.06	-4.6203	-3.5404	1.2E-06	3.9E-07	0.34
12	5/15	35480	1.248-11	0.7	83.7	84.4	4.01	3.39	28.27	35.62	185.62	4.27	22.25	-5.5463	-4.0239	1.7E-06	4.1E-07	0.24
13	5760	41240	8.89E-12	0.8	87.6	68.4	2.90	2.56	30,19	5.43	155.43	0.46	13.20	-7.7719	-4,5458	5.4E-06	5. JE -07	0.09
14	5820	47060	1:17E-12	5.4	79.0	84,4	0.39	0.33	5.43	0	150.00		8.99		-4.9294		1.0E-07	
15	7200	54260	1.90E-13				0.0/7											

Total Resuspended = 1093.97 150 <---Assumed Remaining

I

J

7.7

1

COLUNNS---> A

c

D

E

F

G

ы

7.2.2 <u>Calculation of Surface Activity as a Function of Time</u>

The resuspension rates are calculated from the surface activity as a function of time. The mass of resuspendable iodine species on the surface of the tube as a function of time is calculated by a two-step process. First, the mass on the surface at the end of each sampling interval, decay-corrected back to the start of the experiment, $N_{\rm ot}$, is calculated by

$$N_{ot} = N_{ot-1} - R_{ot}$$
(7.11)

where N_{ot-1} = mass on surface after the previous interval

Rot = decay-corrected mass of species resuspended during sampling
interval ending at time t.

The N_{ot} is calculated by Equation (7.11) for each sampling interval and the results are reported in Column J of Table 7.3. The undecayed surface mass, N_{ot}, as a function of time (Columns J versus B) is shown as the lower curve in Figure 7.1. The estimated mass, N_t, on the surface at time t is calculated by decay correcting N_{ot} to t by

$$N_{t} = N_{ot} e^{-\lambda t}$$
(7.12)





7.8

The calculated N_+ values are shown in Column L.

7.2.3 <u>Calculation of Resuspension Rate</u>

Resuspension rates are estimated from the slope of ln (N_{+}/N_{0}) plotted versus time [(i.e., Equation (7.7)]. The values of ln (N_{+}/N_{0}) are listed in Table 7.3, Column N, and are plotted versus time as the lower curve in Figure 7.2.

Resuspension rates, r, for each sampling interval are calculated from the expression



<u>FIGURE 7.2</u>. Particle Plus Elemental Iodine Normalized Surface Activity Versus Time

and are shown in Column P.(a) Both Column P and Figure 7.2 indicate that resuspension rates are a function of time. Additionally, from an examination of Figure 7.2, there may be more than one form of resuspendable particle + elemental iodine. A rapidly resuspended form controls resuspension for about the first 10,000 min. A slower resuspending form controls resuspension for times greater than about 10,000 min. Appendix Tables B.1 through B.3 show similar calculations for the other iodine species. Results for the other iodine species comparable to those in Figures 7.1 and 7.2 are plotted in Figures B.1 through B.6.

Figure 7.2 suggests that for a portion of the experiment, resuspension rate (slope) is roughly constant with curvature at the beginning and end of the experiment. The following subsection illustrates a method for eliminating the curvature at the end of the experiment by estimating the amount of resuspendable iodine remaining on the surface at the end of the experiment. When this estimated remaining amount is included in the calculation of resuspension rates, the rates become more constant for that time period.

7.2.4 <u>Remaining Resuspendable Mass</u>

A nearly constant final resuspension rate may be found if one assumes that a quantity of resuspendable iodine was still on the surface at the end of the experiment. The method to estimate the remaining mass is illustrated using the case of the lower curve in Figure 7.1. A straight line is projected from the data between 10,000 and 35,000 min out to the final data point at 42,000 min. This projected line yields an initial estimate of the amount remaining on the surface, $N_{\rm Of}$, undecayed back to the start of the experiment. This estimated amount is added to the original estimate of resuspendable material from Equation (7.10). The adjusted $N_{\rm O}$ is

$$N_{o}$$
 adjusted = $N_{o}(eq. 4.27) + N_{of}$ (7.14)

The initial estimate of N_{of} is tested by calculating adjusted values for N_{ot} using Equation (7.11) and replotting the adjusted values. The final results are shown in Table 7.3, Column K, and adjusted N_{ot} versus time is shown by the upper curve in Figure 7.1.

(a) It can be shown that an equivalent short-cut method for calculating resuspension rates uses the undecayed surface masses as shown below.

$$r = \frac{\ln \frac{N_{ot}}{N_{ot-1}}}{\Delta t}$$

7.10

If the final part of the curve is not as linear as desired, new values can be selected for N_{of} as estimated from the plot of the adjusted data. It is important not to choose an N_{of} so large as to change significantly the curvature of the plot for the initial part of the experiment. Choosing too large an N_{of} could be avoided by establishing a linearity criterion. One possible criterion would be to set a maximum limit on the standard error of a linear regression of the $ln(N/N_o)$ versus time data covering the interval in question.

This estimation approach seems successful for the particle + elemental specie for linearizing the data beyond 10,000 elapsed minutes as shown in the upper curve in Figure 7.1. The value assumed for N_0 for particles and elemental iodine was 150 nCi. Resuspension rates using both unadjusted (for N_0f) and adjusted surface activities are shown in Columns P and Q of Table 7.3. The resuspension rates in both cases decrease with time and range from 1 x 10⁻⁷ to 7.6 x 10⁻⁶ s⁻¹. (Tables B.1 through B.3 and Figures B.1 through B.6 show similar calculations and plots for the other species.)

There are uncertainties in calculated resuspension rates. Table 7.3, Column R, shows the ratio of resuspension rates calculated from data adjusted for N_{of} to those from the unadjusted data. Resuspension rates based on surface activity adjusted for N_{of} are always less than those based on the unadjusted surface activity. The assumption of an N_{of} made little difference for the initial resuspension rates, but reduced the variability of the resuspension rates for times greater than about 10,000 min. Appendix C addresses the effect of estimating different amounts of remaining resuspendable surface activity on resuspension rates during the final three-fourths of the experiment.

7.3 COMPARISON OF RESUSPENSION RATES

Table 7.4 is a comparison of resuspension rates calculated by the SAIC method as reported in their report with the resuspension rates calculated as described above accounting for N_{Of} . Listed after Line A in the table are the resuspension rates for each species for each sampling interval per the SAIC method. Following that are listed (after Line B) the initial resuspension rates calculated by this report's method and the ratio (after Line C) of the initial SAIC rate and this report's rate. The most substantial difference was for the HOI and organic species where SAIC's values are more than an order of magnitude less than calculated using adjusted surface activities.

Table 7.4 next lists (after Line D) the resuspension rates used in SAIC's model for estimating iodine transmission factors. These rates are the

	Sampling Period	Cumulative		Resusnensi	on Rate, s-1	
Sampling <u>Period</u>	Duration,	Time, min	I ₂ + Particles	<u>HOI</u>	<u>Organic</u>	<u>Total</u> 131 ₁
A: Repor	ted by SAIC		-			
1	890	890	1.0×10^{-5}	9.9 x 10^{-8}	9.2 x 10^{-8}	
2	1440	2330	$6.5 \times 10^{-0}_{-6}$	1.3×10^{-7}	5.5×10^{-0}	
3	1440	3770	2.4×10^{-6}	9.5×10^{-0}	4.1×10^{-8}	
4	1470	5240	2.7×10^{-6}	1.6×10^{-7}	4.3 x 10_8	
5	1884	/124	2.4 x 10	1.6×10^{-7}	4.6 x 10	
. 0	2430	9500	1.3 X 10-7	1.0 X 10 4.0 X 10-8	2.5×10^{-8}	
2	3120	12020	7 0 v 10-7	4.9 X 10-8	2.7×10^{-8}	
9	4030	19770	1 1 2 10-6	5.0×10^{-8}	1.5×10^{-8}	
10	4715	24485	75×10^{-7}	5.2×10^{-8}	1.3×10^{-7}	
11	5280	29765	8.3×10^{-7}	1.2×10^{-7}	2.3×10^{-8}	
12	5715	35480	9.6 x 10 c	7.6×10^{-8}	4.4×10^{-8}	
13	5760	41240	1.4×10^{-0}	9.3×10^{-8}	3.1×10^{-8}	
14	5820	47060	3.1×10^{-7}	2.9×10^{-6}	1.0×10^{-6}	
15	7200	54260	8.3×10^{-6}	5.1×10^{-3}	2.5×10^{-5}	
B: Calcu inter	lated by met val:	thod develop	ed in this re	port for the	first time	
1	890	890	6.5×10^{-6}	1.3×10^{-6}	1.2×10^{-6}	6.4×10^{-6}
C: Ratio time	of rates pe interval:	er SAIC meth	od to this re	port's metho	d for the fi	rst
1	890	890	1.54	0.076	0.077	
D: Averag	ge rate used	d by SAIC to	model all tim	me intervals	:	
				2.1×10^{-6}	8.4×10^{-8}	4.0×10^{-8}
E: Rates for final part of experiment for the interval from 10,000 to 42,000 min, based on adjusted surface activities:						
Avera	ge:		4.4×10^{-7}	3.6×10^{-7}	1.3×10^{-7}	5.4×10^{-7}
Standa	ard Deviatio	on:	0.8×10^{-7}	1.3×10^{-7}	1.4×10^{-7}	0.9×10^{-7}
Range	:		4.4×10^{-7}	3.6×10^{-7}	1.3×10^{-7}	5.4 x 10 ⁻⁷
			$to to 5.8 \times 10^{-7}$	to 6.2 x 10 ⁻⁷	$to 4.3 \times 10^{-7}$	to 6.8 x 10 ⁻⁷

TABLE 7.4. Comparison of Resuspension Rates for Reactor C Sampling Line 1b

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averages of the rates for each of the 15 sampling periods. Finally reported in the table (after Line E) are the average, standard deviation, and range of resuspension rates for the 10,000- to 42,000-min interval calculated as described in this report.^(a)

The resuspension rates used by SAIC to characterize resuspension for HOI and organic radioiodine are still significantly lower than the average rates for the final part of the experiment calculated per this report's method. These comparisons suggest that an iodine line-loss model should address resuspension rates as a function of time. If these data are representative of sampling lines, then it may be reasonable to select two resuspension rates for modeling the elemental + particle species: 1) the initial rate from the first sampling interval, and 2) an average of the rates from the sampling intervals from 10,000 min to the end of the experiment.

⁽a) A resuspension rate perhaps more representative of the time interval could be calculated from a linear regression of the appropriate ln (N/N_0) versus time data. For example, in the case of organic iodine, the resulting resuspension rate would be 1.45 x 10^{-7} s⁻¹. Performing a linear regression would also facilitate the use of a linearity criterion for choosing N_{0f} .

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8.0 <u>DISCUSSION AND GUIDELINES</u>

This chapter provides a discussion of test data, research needs and interim guidelines included in this report.

8.1 DISCUSSION OF TEST DATA

Test data are discussed in the following subsections.

8.1.1 <u>Transmission Factors</u>

The objective of the research reviewed in this report has been to find a way to correct the measured radioiodine concentration in ventilation exhaust streams for the loss of iodine in the tubing that transports the sample from the extraction point to the collection/monitoring device. The simplest way to correct the measured concentration is the use of a line loss or transmission factor in the denominator of the emission calculation, thereby increasing the emission rate to account for the line loss. The laboratory tests on simulated sampling lines seem to indicate that transmission factors may change curing transients in the real emission rate, mostly as a result of the resuspension and surface reaction of deposited iodine. This effect seems to be the most severe where iodine deposition is high.

Based on the laboratory simulation described here, the performance of a sampling system during a transient release would be variable; however, possibly predictable. During a release of a high concentration, if it were of short duration (<1 day), then the performance of the sample delivery line would be more like that of the deposition phase of the laboratory simulation than the so-called predicted equilibrium performance. If the sample delivery line had a very low-equilibrium transmission factor, the release would be unnoticeable in the sampler data for some period of time. Following cessation of a high release, resuspension may dominate and the apparent performance (transmission factor) would be much greater than the equilibrium value. If the release rate were to continue for many weeks, then the performance may approach the predicted equilibrium value.

However, a sustained constant release rate seems unlikely in light of the observations of Pelletier et al. (1978a,b), possibly diminishing the value of an equilibrium transmission factor.

To our knowledge, there has been no experimental verification of predicted transmission factors under either equilibrium or transient conditions. Such verification would be desirable if, on a wide scale, utilities were to rely on the Science Applications International Corporation (SAIC) approach of modeling transmission factors based on laboratory measurements of deposition and resuspension rate constants. Concern for the apparent poor repeatability of the laboratory measurements also dictates a need for demonstrable repeatability. We are uncertain about the interpretation of the in-place data, using the methods tried to date, relative to the concept of transient transmission factor. Certainly where a sampler is found to have a high transmission factor in an in-place test, the possible higher equilibrium transmission factor is probably not very different, and using the observed transmission factor would probably be acceptable. If the observed transmission factor were very low, then one would have to consider that under some conditions, the actual transmission factor may be different depending on the trending of the emission rate.

Another concern is that the measurements of deposition and resuspension rate constants are done only (and probably to facilitate experiments of practical duration and detection limits) using very high concentrations of radioactive and stable iodine. There has as yet been no work demonstrating that these rate constants are not functions of either concentration, time, surface characteristics, temperature, humidity, or isotopic exchange. The present laboratory simulations and modeling at least are very helpful in defining performance problems and testing their solutions. However, it is not yet known if this approach is ready to be adopted on either an interim or final basis for quantitative use in measurements of radioiodine releases.

8.1.2 Discussion of Model

The model described by Unrein et al. (1985) has been discussed, and certain improvements to the approach were suggested. An analysis procedure was developed for evaluating dry deposition velocities and resuspension rates from measurements of airborne iodine concentrations entering and exiting sample delivery lines. The procedure was used to reanalyze data reported by SAIC for a simulated sample delivery line. Although simplifying assumptions are made in both modeling approaches, a conclusion is that dry deposition velocities should be calculated with the model presented here [see Equations (7.3) and (7.5)]. The use of hypothesized short segments of the sample delivery line as assumed by SAIC for the purpose of estimating the deposition velocity is unwarranted, and may introduce calculational errors.

Resuspension rates decrease with time, a fact that was not emphasized in SAIC's method. Therefore, the use of resuspension rates averaged over the entire experimental time interval is of questionable value, because resuspension rates changed significantly.

Resuspension rates were calculated based on amounts of each radioiodine species on walls of a sample delivery line. Resuspension rates are a function of time and chemical species. For the initial resuspension time period, the resuspension rate for particle + elemental iodine is $6.5 \times 10^{-6} \text{ s}^{-1}$, $1.3 \times 10^{-6} \text{ s}^{-1}$ for hypoiodous acid (HOI), $1.2 \times 10^{-6} \text{ s}^{-1}$ for organic iodides, and $6.4 \times 10^{-6} \text{ s}^{-1}$ for total radioiodine. Resuspension rates decreased with time to about an order of magnitude lower after 10,000 min elapsed time.

Essentially all of the initially deposited radioiodine was resuspended during the experiment. About 1170 nCi of radioiodine was deposited on the tube surface during the deposition part of the experiment. From Table B.3, about 1182 nCi (decay corrected to the start of the resuspension experiment) were collected and accounted for during the resuspension experiment. The resuspension rate calculation method described herein adds a few percent to reduce the variation in the resuspension rate for the final part of the data, but the total quantity is within the probable error of the calculation of total deposit.

8.1.3 <u>Summary of Three Available Methods</u>

The literature reviewed here suggests three approaches to estimating transmission (or line-loss) factors to use in the calculation of radioiodine emissions. Included are

- in-place testing
- laboratory simulations
- performance modeling based on a data base of deposition and resuspension rates determined in the laboratory.

The approaches could be adapted from the methods reviewed earlier in the report. To assist in selecting a method, the advantages and disadvantages of each are summarized below.

1. In situ tests by injection of iodine or use of iodine levels already present in the ventilation.

Advantages

- a. The result would be specific for the tested system.
- b. A repeatable result would be hard to dispute.
- c. Expensive generic lab testing could be avoided that would otherwise be needed before adopting the other two methods.
- d. The test is conducted with the actual tubing with its own operating history and surface contaminants (oils, carbon dust, etc.), which may produce results significantly different from tests with other tubes.

Disadvantages

- e. In situ tests are probably more difficult to conduct than lab tests.
- f. Tests may disrupt site operations.
- g. The injection of radioiodine may be prohibited; however, stable iodine may be usable.

- h. Tests may require some facility modifications at the stack and sampling skids.
- i. A short test does not address transient response, but transmission factors may be conservative during a step increase and equilibrium conditions, and about right during the critical initial phase of a step decrease.
- j. In situ tests are expensive for the industry as a whole.
- 2. Measurement of deposition velocity and resuspension rate in the laboratory for tubing simulating the actual system followed by model extrapolation to equilibrium and transient conditions.

Advantages

a. The results should be applicable to the system tested.

Disadvantages

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- b. The predicted equilibrium and transient responses have not been verified.
- c. The model used to predict performance is unpublished and unavailable for detailed peer review.
- d. To date, most customers are unsure about the results to the extent that they have gone through the motions for the NRC's benefit and are unwilling to apply the results.
- e. There is insufficient evidence of precision in the data.
- f. There has been no verification that the results can be confidently transferred to the real system.
- g. The tubing used in the testing may not (perhaps cannot) have the same surface contamination or operating history as the simulated tubing.
- 3. Model equilibrium and transient performance based on measurements of deposition velocity and resuspension rates for similar systems.

Advantages

a. Potentially the least expensive, fastest method for the utilities.

Disadvantages

- b. A more extensive data base would have to be developed but may be less expensive for the industry as a whole than tests for each site by methods 1 or 3.
- c. There is a lack of correlation in the basic data between the important deposition/resuspension parameters and the sampler characteristics making the selection of the basic inputs uncertain.
- d. A sensitivity analysis of the effects of the selection of the input deposition/resuspension parameters on the predicted performance is needed.
- e. Disadvantages b, c, d, e, and f from above also apply.

8.2 RESEARCH NEEDS

The NRC currently requires utilities to demonstrate the effectiveness of air sampling systems. This requires that the methods used to do so have an acceptable level of demonstrable precision. Such is not the case at present, either because the work has not been performed or not reported. Also, there is little evidence to show that the line-loss determination methods described in this report yield results that can be used to quantify iodine release values with much confidence under all conditions that may be important. Also, little is known about the details of the line-loss mechanisms involved.

There are several lines of inquiry that would assist in evaluating the differences between the line-loss methods that should be addressed, including the following:

- Compare laboratory determined transmission factors against in-place data in some blind tests.
- Compare a model-determined transmission factor against laboratory and in-place determinations in blind tests.
- Investigate the typical range of variability in effluent radioiodine concentrations and the rate of concentration fluctuations. The useful assumption of equilibrium between deposition and resuspension may not be realistic if the sampling tubes are continually challenged by transients.
- Address the effects of accident conditions by including them in the range of tested parameters.

8.5

- Consider the effects of resuspension in the ranking of materials. Kabat's work definitely showed a ranking in the deposition performance of materials. It may be that an iodine sampling system should be made of different materials than one designed for particles.
- Address the need for source term data in conjunction with field data of deposition and uptake in food products.

8.3 INTERIM GUIDELINES

Until research has been performed in the areas outlined above, the NRC has three options:

- 1. Cease enforcing the requirement for defensible source term measurements for radioiodine in ventilation exhausts.
- 2. Accept the use of transmission factors determined by one of the available imperfect methods.
- 3. Require the use of a blanket transmission factor by groups of similar sampler types or configurations.

The first option satisfies the argument that the only useful radioiodine measurements come from field sampling of deposition, air concentration, and uptake in food products. The conclusion of that argument is that source sampling is 1) not needed, 2) useless, or at best, 3) corroborative evidence. The second option satisfies the desire to use the best available technology for source term measurements, which in turn, can then be used in support of field sampling programs and dispersion modeling. Carefully chosen transmission factors should yield conservative emission rates, but not to the point of being unrealistic. Perhaps using the transmission factor for elemental iodine, the most depositing iodine form, is more than sufficiently conservative. The third option is not recommended because it does not use the best available technology, is prone to yield, at best, upper-bound source terms, and at worst, is a pretense for real data.

Our recommendation is to pursue the second option and to use the transmission factor performance determination methods in the following order of preference, with the final selection based on practicality and quality of results:

- 1. in-place tests using existing levels of radioiodine or injected nonradioactive elemental iodine
- 2. laboratory simulation of the system that shows repeatability after some previous demonstration of adequate equivalence to in-place tests
- 3. modeling based on an adequate data base from lab tests of similar systems where repeatability is demonstrated.

We prefer the in-place test because of the advantages cited in Section 8.1.3, but recommend some modifications to overcome some of the disadvantages. If a tested system shows such poor performance where resuspension is an important factor, it would then be preferable to replace the system with one that has little line loss rather than trying to estimate the transmission factor response to transient effects that may only be observed belatedly or guessed at by other methods. Proper quality assurance must be observed with either of the methods, including traceable calibration of experimental equipment, peer review of procedure and results, and benchmarking of calculations and models.

8.7

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A-i

SAIC PRIVATE CUSTOMER TEST DATA FOR RESUSPENSION EXPERIMENTS

TABLE A.1.

Resuspension Data from Reactor C (from Table 2.2) Test la

	,	Average	Species Di	stribu	tion,	%
<u>Period</u>	Duration, min	Concentration (a) $(\mu Ci/cm^3)$ of $131I$	¹³¹ I Associated <u>with Particulate</u>	0rg _ <u>I</u> 2_	anic <u>HOI</u>	<u>Iodides</u>
1	345	2.05×10^{-9} (1.4%)	33.6	58.6	6.9	1.0
2	840	2.90×10^{-10} (2.8%)	11.7	75.6	8.7	4.0
3	1,251	3.62 x 10 ⁻⁹ (1.9%)	1.2	92.8	4.4	1.5
4	5,694	1.44 x 10 ⁻¹⁰ (1.0%)	6.3	74.3	14.2	5.2
5	5,830	8.71 x 10 ⁻¹¹ (1.6%)	4.3	79.5	12.2	4.0
6	10,080	5.20 x 10 ⁻¹¹ (1.4%)	11.3	82.2	3.6	3.0
7	7,200	3.86 x 10 ⁻¹¹ (1.8%)	4.1	72.5	7.8	15.6
Σ =	31,240					
		<u>Abnormal C</u>	<u>onditions</u>			
8 ^(b)	7,020	1.62 x 10 ⁻¹¹ (1.3%)	8.6	87.2	1.7	2.5
9 ^(c)	6,240	1.58 x 10 ⁻¹¹ (2.5%)	2.0	91.4	3.3	3.3

(a) 2 σ counting uncertainties are given in parentheses.
(b) Sampling flow rate increased from 944 to 1888 cm³/s during this period.
(c) Sampling line temperature raised from 30° to 50°C during this period; normal flow rate used.

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		Average	Species Dis	<u>tribut</u>	<u>ion, %</u>	
<u>Period</u>	Duration,	Concentration(a) (μ Ci/cm ³) of 131I	131 _I Associated <u>with Particulate</u>	<u> </u>	<u>H0 I</u>	Organic <u>Iodides</u>
1	890	7.07×10^{-9} (0.6%)	2.3	95.1	1.4	1.3
2	1,440	2.93 x 10 ⁻⁹ (1.3%)	0.7	95.2	2.9	1.2
3	1,440	7.90×10^{-10} (2.6%)	1.7	90.7	5.3	2.3
4	1,470	7.11 x 10 ⁻¹⁰ (0.9%)	1.6	88.6	7.7	2.1
5	1,884	4.68×10^{-10} (1.1%)	1.1	87.6	8.8	2.5
6	2,436	1.85 x 10 ⁻¹⁰ (1.4%)	1.8	85.1	10.6	2.5
7	3,060	7.21 x 10 ⁻¹¹ (2.0%)	3.8	81.2	9.7	5.4
8	3,120	6.23×10^{-11} (1.3%)	8.6	84.1	5.2	2.2
9	4,030	5.98 x 10 ⁻¹¹ (1.5%)	0.9	90.9	6.4	1.8
10	4,715	3.26×10^{-11} (1.6%)	5.2	68.6	7.7	18.5
11	5,280	2.00 x 10 ⁻¹¹ (1.6%)	1.0	79.1	16.6	3.3
12	5,715	1.24×10^{-11} (1.8%)	0.7	83.7	9.9	5.7
13	5,760	8.89 x 10 ⁻¹² (2.6%)	0.8	87.6	8.8	2.9
14	5,820	1.17×10^{-12} (4.7%)	5.4	79.0	11.6	4.0
15	7,200	1.90×10^{-13} (4.2%)		(b)		
Σ =	54,260					

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TABLE A.2. Resuspension Data from Reactor C (from Table 2.2) Test lb

(a) 2 σ counting uncertainties are given in parentheses. (b) Radioiodine species distribution not measured.

TABLE A.3.

Resuspension Data from Reactor C (from Table 2.2) Test 2a

		Average	Species Dist	<u>ributi</u>	<u>on, %</u> (a)
<u>Period</u>	Duration, min	Concentration(b) (<u>µCi/cm³) of 131</u>	¹³¹ I Associated <u>with Particulate</u>	<u> I_2 </u>	HOI	Organic <u>Iodides</u>
1	345	1.92×10^{-10} (18%)	19.4	55.3	22.8	<2.5
2	840	4.38 x 10 ⁻¹¹ (9%)	<2.7	54.2	31.7	<11.4
3	1,251	3.19×10^{-11} (13%)	<6.6	71.4	18.8	3.2
4	5,694	1.01×10^{-11} (21%)	<5.6	43.5	18.6	32.3
5	5,530	1.92×10^{-12} (44%)	38.3	30.5	<11.6	<19.6
6	10,080	$<5.2 \times 10^{-1.4}$		(c)		
7	7,200	5.84 x 10 ⁻¹² (4%)		(c)		
Σ =	31,240					
		<u>Abnormal</u>	<u>Conditions</u>			
8(q)	7,020	8.90×10^{-13} (2.9%))	(c)		
g(e)	6,240	9.23 x 10^{-13} (21%)		(c)		
Σ =	44,500			١		

(a) The distributions shown assume that the undetected species were present at levels just below the detection limit. Assuming that the undetected species were completely absent would give the other extreme values for the distributions.

(b) 2 σ counting uncertainties are given in parentheses.

(c) Radioiodine species distribution not measured.

Sampling flow rate increased from 28.3 to 198 cm 3 /s for this period. (d)

(e) Sampling line temperature raised from 30° to 50°C during this period; normal flow rate used.

		Average	Species Distribution, %(a)				
<u>Period</u>	Duration,	Concentration(b) (μ Ci/cm ³) of 131I	131 ₁ Associated <u>with Particulate</u>	Organic <u>I2 HOI Iodides</u>			
1	890	4.33×10^{-10} (3.0%)	<0.34	38.4 35.9 25.4			
2	1,440	4.39×10^{-10} (1.8%)	,	(b)			
3	1,440	3.37×10^{-10} (1.6%)		(b)			
. 4	1,470	6.69 x 10 ⁻¹⁰ (1.9%)		(b)			
2 5	1,884	1.36×10^{-10} (5.0%)		(b)			
6	2,436	5.45×10^{-11} (2.9%)		(b)			
7	3,060	7.36 x 10 ⁻¹² (2.7%)		(b)			
8	3,120	2.78×10^{-12} (4.5%)		(b)			
9	4,030	6.49×10^{-11} (1.3%)		(b)			
. 10	4,715	3.66×10^{-11} (1.4%)		(b)			
11	5,280	5.21 x 10 ⁻¹¹ (1.0%)		(b)			
12	5,715	1.75×10^{-11} (3.6%)		(b)			
13	5,760	1.74×10^{-11} (4.1%)		(b)			
14	5,820	2:05 x 10 ⁻¹² (21%)		(b)			
15	7,200	2.10×10^{-13} (71%)		(b)			
Σ =	54,260						

TABLE A.4. Resuspension Data from Reactor C (from Table 2.2) Test 2b

(a) 2 σ counting uncertainties are given in parentheses. (b) Radioiodine species distribution not measured.

<u>TABLE A.5</u>. SAIC-Estimated Resuspension Rate Constants for Reactor C (from Table 2.2) Tests 1a and 2a

	Average Resuspension Rate Constants (s ⁻¹) ⁽⁸⁾							
		1a			2a			
Measurement Period	12+ Particles	H OI	Organic	I2+ Particles	HOI	Organic		
1	1.7 x 10 ⁻⁶	5.3 x 10 ⁻⁸	7.7 x 10 ⁻⁹	2.6 x 10 ⁻⁷	1.3 x 10 ⁻⁸	6.6 x 10 ⁻⁹		
2	2.5×10^{-7}	9.9 x 10 ⁻⁹	4.6 x 10 ⁻⁹	6.2 x 10 ⁻⁸	3.0×10^{-9}	1.6 x 10 ⁻⁹		
3	3.8×10^{-6}	7.1 x 10 ⁻⁸	2.4 x 10 ⁻⁸	4.8 x 10 ⁻⁸	2.3 x 10 ⁻⁹	1.2 x 10 ⁻⁹		
4	1.7 x 10 ⁻⁷	1.2 x 10 ⁻⁸	4.4 x 10 ⁻⁹	1.9 x 10 ⁻⁸	9.0 x 10 ⁻¹⁰	4.7 x 10 ⁻¹⁰		
5	1.5×10^{-7}	9.0 x 10 ⁻⁹	2.9 x 10 ⁻⁹	5.0 x 10 ⁻⁹	2.4×10^{-10}	1.3 x 10 ⁻¹⁰		
6	1.7×10^{-7}	2.6×10^{-9}	2.2×10^{-9}			10		
7	1.8×10^{-7}	7.3 x 10 ⁻⁹	1.5 x 10 ⁻⁸	2.9 x 10 °	1.4 x 10 ⁻	7.2 x 10 '		
Mean ^(b)	9.2 x 10 ⁻⁷	2.4 x 10 ⁻⁸	8.7 x 10 ⁻⁹	7.0 x 10 ⁻⁸	3.4 x 10 ⁻⁹	1.8 × 10 ⁻⁹		
			<u>High Flow I</u>	Rate(c)				
8	2.1 x 10 ⁻⁷	2.1 x 10 ⁻⁹	3.2 x 10 ⁻⁹	1.7 x 10 ⁻⁸	3.2 x 10 ⁻⁹	1.7 x 10 ⁻⁹		
			<u>High Temperat</u>	ture(d)				
9 ·	2.2 x 10 ⁻⁷	3.2 x 10 ⁻⁹	3.2 x 10 ⁻⁹	1.5 x 10 ⁻⁸	7.1 x 10 ⁻¹⁰	3.7 x 10 ⁻¹⁰		

(a) Average species distribution fractions used for all periods.
(b) Unweighted average values for measurement periods above.
(c) Sampling flow rate increased from 944 to 1888 cm²/s for Line 1 and from 28.3 to 198 cm²/s for Line 2 during Period 8.

(d) Sampling line temperature raised from 30° to 50°C during Period 9.

		Average I	Resuspension	Rate Constant:	s (s ⁻¹) ^(a)	
		<u>1b</u>			2b	
Measurement Period	1 ₂ + Particles_	I2+ HOI	Organic	<u>Particles</u>	HOI	<u>Organic</u>
1	1.0 x 10 ⁻⁵	9.9 x 10 ⁻⁸	9.2 x 10 ⁻⁸	6.5 x 10 ⁻⁷	3.2 x 10 ⁻⁸	1.7 x 10 ⁻⁸
2	6.5 x 10 ⁻⁶	1.3 x 10 ⁻⁷	5.5 x 10 ⁻⁸	7.2 x 10 ⁻⁷	3.5 x 10 ⁻⁸	1.8 x 10 ⁻⁸
3	2.4 x 10 ⁻⁶	9.5 x 10 ⁻⁸	4.1 x 10 ⁻⁸	6.1 x 10 ⁻⁷	2.9 x 10 ⁻⁸	1.5 x 10 ⁻⁸
4	2.7×10^{-6}	1.6 x 10 ⁻⁷	4.3 x 10 ⁻⁸	1.3 x 10 ⁻⁶	6.5 x 10 ⁻⁸	3.4×10^{-8}
5	2.4×10^{-6}	1.6×10^{-7}	4.6 x 10 ⁻⁸	3.1 x 10 ⁻⁷	1.5 x 10 ⁻⁸	7.7 x 10^{-9}
6	1.3 x 10 ⁻⁶	1.0×10^{-7}	2.5 x 10 ⁻⁸	1.4 x 10 ⁻⁷	6.8 x 10 ⁻⁹	3.6×10^{-9}
7	6.3×10^{-7}	4.9×10^{-8}	2.7 x 10 ⁻⁸	2.2 x 10 ⁻⁸	1.1 x 10 ⁻⁹	5.7 x 10^{-10}
8	7.9 x 10 ⁻⁷	3.0×10^{-8}	1.3×10^{-8}	1.0×10^{-8}	4.9×10^{-10}	2.6×10^{-10}
9	1.1×10^{-6}	5.2 \times 10 ⁻⁸	1.5 x 10 ⁻⁸	3.0×10^{-7}	1.4 x 10 ⁻⁸	7.5 x 10 ⁻⁹
10	7.5 x 10 ⁻⁷	5.3 x 10^{-8}	1.3×10^{-7}	2.2×10^{-7}	1.1×10^{-8}	5.6 x 10^{-9}
11	8.3×10^{-7}	1.2×10^{-7}	2.3×10^{-8}	4.3×10^{-7}	2.1×10^{-8}	1.1×10^{-8}
. 12	9.6 x 10^{-7}	7.6×10^{-8}	4.4×10^{-8}	2.1 x 10^{-7}	1.0×10^{-8}	5.2×10^{-9}
13	1.4×10^{-6}	9.3 x 10^{-8}	3.1×10^{-8}	2.9×10^{-7}	1.4×10^{-8}	7.5×10^{-9}
14	3.1×10^{-7}	2.9×10^{-8}	1.0×10^{-8}	5.0 x 10^{-8}	2.4×10^{-9}	1.3×10^{-9}
15	8.3×10^{-8}	5.1 x 10 ⁻⁹	2.5 x 10 ⁻⁹	7.5 x 10 ⁻⁹	3.7 x 10 ⁻¹⁰	1.9 x 10 ⁻¹⁰
Mean ^(c)	2.1 x 10 ⁻⁶	8.4 x 10 ⁻⁸	4.0 x 10 ⁻⁸	3.5 x 10 ⁻⁷	1.7 x 10 ⁻⁸	9.0 x 10 ⁻⁹

(a) Average species distributions for Periods 1--14 used for Period 15.
(b) Average species distributions from Test A used for all periods.
(c) Unweighted average values for measurement periods above.

A.6

<u>TABLE A.7</u>. Resuspension Data for Reactor G (from Table 2.2) Test A

Duration,	Average ¹³¹ I <u>Concentration, <i>µ</i>Ci/cc(a)</u>
1185	2.82E-10 (5.4%)
1540	1.82E-10 (4.1%)
1420	1.46E-10 (3.1%)
1355	6.92E-11 (5.7%)
1455	6.51E-11 (4.6%)
1500	3.63E-11 (1.6%)
1440	3.92E-11 (0.6%)
1440	6.40E-11 (2.6%)
1440	2.07E-11 (4.6%)
1440	1.80E-11 (4.0%)
14215	· · ·
	Duration, min 1185 1540 1420 1355 1455 1500 1440 1440 1440 1440 14215

(a) 2 σ fractional counting uncertainties are given in parentheses.

<u>TABLE A.8</u> .	Resuspension	ı Data f	for	Reactor	G
-	(from Table	2.2) Te	est	В	

Period <u>Number</u>	Duration,	Average ¹³¹ I <u>Concentration, <i>µ</i>Ci/cc(a)</u>
1 2 3 4 5 6 7 8 9 10 Σ =	1200 1395 1470 1455 1400 1530 1440 1290 1380 <u>1500</u> 14060	7.04E-10 (1.7%) 4.31E-10 (0.6%) 9.86E-11 (1.7%) 5.92E-11 (4.5%) 3.15E-11 (6.1%) 3.13E-11 (2.3%) 3.11E-11 (2.1%) 9.56E-12 (5.3%) 1.90E-11 (1.4%) 8.11E-12 (2.4%)

(a) 2 σ fractional counting uncertainties are given in parentheses.

A.7

Period <u>Number</u>	Duration,	Average ¹³¹ I <u>Concentration, <i>µ</i>Ci/cc</u> (a)
1	1275	2.24E-09 (2.3%)
2	1440	8.40E-11 (1.0%)
3	1230	5.61E-11 (3.2%)
4	1470	4.39E-11 (5.0%)
5	1980	2.12E-11 (4.0%)
6	1080	1.37E-11 (4.0%)
7	1440	8.07E-12 (1.9%)
8	1440	6.57E-12 (1.9%)
9	1440	5.97E-12 (3.3%)
10	_1110	9.30E-12 (3.3%)
Σ =	13905	

TABLE A.9. Resuspension for Reactor G (from Table 2.2) Test C

(a) 2 σ fractional counting uncertainties are given in parentheses.

		(from lable 2.2) lest				
<u>Period</u>	Duration,	Average Concentration(a) (µCi/cm ³) ¹³¹ I	Average Resuspension Rate Constant, s ⁻¹			
1 2 3 4 5 6 7 8 9 10 11 12 13 14	1005 1440 1620 1925 735 2880 2880 4290 1695 5595 5700 5820 4500	$\begin{array}{c} 2.14 \times 10^{-9} (1.9\%) \\ 3.39 \times 10^{-10} (1.4\%) \\ 9.05 \times 10^{-10} (1.8\%) \\ 4.95 \times 10^{-10} (1.1\%) \\ 1.55 \times 10^{-10} (2.4\%) \\ 1.15 \times 10^{-11} (2.8\%) \\ 7.77 \times 10^{-11} (2.8\%) \\ 7.88 \times 10^{-11} (1.7\%) \\ 3.56 \times 10^{-11} (1.9\%) \\ 2.08 \times 10^{-11} (2.8\%) \\ 1.01 \times 10^{-12} (1.6\%) \\ 9.03 \times 10^{-12} (3.4\%) \\ 4.80 \times 10^{-12} (1.9\%) \\ 1.50 \times 10^{-12} (2.5\%) \end{array}$	2.7×10^{-6} 5.9×10^{-6} 2.2×10^{-6} 1.5×10^{-7} 5.6×10^{-7} 3.6×10^{-7} 4.6×10^{-7} 4.6×10^{-7} 4.6×10^{-7} 1.9×10^{-7} 1.2×10^{-7} 1.5×10^{-7}			
Σ =	41,525		x = 10.75 E-7			

<u>TABLE A.10</u>. Resuspension Data for Reactor E (from Table 2.2) Test

(a) 2 σ fractional counting uncertainties are given in parentheses.

TABLE A.11. Measured Distributions of Resuspended Radioiodine Species (%) for Tests Reported by Unrein et al. (1985)

<u>Line</u>	Particulate(a)	<u> </u>	HOI	Organic <u>Iodides</u>
1(þ)	5.1	81.5	1.0	2.4
2(b)	10.4	85.8	1.3	2.6
3	9.2	79.3	7.0	4.5
3	2.5	85.5	8.0	4.0
4	-12.0(c)	~53.0	~23.0	~12.0
4	<0.4(d)	38.0	36.0	25.0
5	6.8	83.9	5.6	3.7
6	4.9	85.9	5.5	3.8

(a) Radioiodine associated with particles.

(b) Laboratory air drawn through the line was not filtered.

(c) Species concentrations variable; one or more components were below detectable levels during sampling periods.

(d) Radioiodine species measurements made during first resuspension period; total activity measured during the other 14 periods.

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APPENDIX B

TABLES AND FIGURES FOR CALCULATION OF HOI, ORGANIC, AND TOTAL IODINE RESUSPENSION

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FIGURE B.4. HOI Normalized Surface Activity Versus Time









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			131 ₁						nC1 HOI on	Surface		Normal1zed	1n(N/N_)	Resuspens	on Rates,	Section-1
	sample	Test	Concen-	. .	Resuspended nCl/Interval							Ratio				
Period	Duration Minimum	Minimum	pCi/cc	HOI	1311	H01	Undecayed HO1	Undecayed Unadjusted	Undecayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Unadjusted	Vecayed Adjusted	Uecayed Unadjusted	Decayed Adjusted	Adjusted/ Unadjusted
ZERO	0	0	0	0	0	0	0	58.01	78.01	58.01	78.01	0	0			
L	890	890	7.07E-09	1.4	356,40	4.99	5,26	52.75	72.75	50.01	68,98	-0.1483	-0.1231	1.8E-06	1.3E-06	0.73
2	1440	2330	2.93E-09	2.9	238,98	6.93	7.97	44.78	64.78	38.96	56.36	-0.3981	-0.3251	1,92-06	1.3E-06	0.71
3	1440	3770	7.90E-10	5.3	64.43	3.41	4.28	40.50	60.50	32.33	48.29	-0.5847	-0.4796	1.2E-06	7.92-07	0.68
4	1470	5240	7,112-10	1.1	59.20	4.56	6.24	34.27	54.27	25.05	39.67	-0.8398	-0.6762	1.9E-06	1.22-06	0.65
5.	1884	7124	4.68E-10	8.8	49,94	4.39	6.73	27.54	47.54	17.99	31,05	-1.1710	-0.9213	1.98-06	1.2E-06	0.61
6	2436	9560	1.85E-10	10.6	25.53	2.71	4.79	22.75	42.75	12.84	24.13	-1,5079	-1.1732	1.3E-06	7.3E-07	0.56
1	3060	12620	7.21E-11	9.7	12.50	1.21	2.58	20,17	40.17	9.48	18.89	-1.8111	-1.4184	6.6E-07	3.4E-07	0.52
8	3120	15740	6.23E-11	5.2	11,01	0.57	1.47	18.70	38.70	7.30	15.10	-2.0732	-1.6421	4.0E-07	2.0E-07	0.49
9	40 30	19770	5.98E-11	6.4	13.65	0.87	2.85	15.85	35.85	4,86	10.99	-2.4795	-1.;9596	6.8E-07	3.2E-07	0,45
10	4715	24485	3.26E-11	7.7	8.71	0.67	2.90	12,95	32.95	3.00	7.62	-2.9633	-2.3258	7.1E-07	3.0E-07	0.42
11	5280	29765	2.008-11	16.6	5,98	0.99	5.89	7.07	27.07	1.19	4.57	-3.8850	-2,8383	1,9E-06	6.2E-07	0.32
12	5715	35480	1.24E-11	9.9	4.01	0.40	3.32	3.75	23.75	0.45	2.85	-4.8600	-3,3107	1.8E-06	3.8E-07	0.21
13	5760	41240	8.89E-12	.8.8	2.90	0.26	3.01	0.75	20.75	0.06	1,76	-6.8195	-3.7904	4.72-06	3.92-074	0,08
14	5820	47060	1.17E-12	11.6	0.39	0.045	0.75	0	20.00	0	1.20		-4.1751		1.0E-07	
15	7200	64260	1.90E-13		0.077											
Total Resus;			pended	58,01	20 <as< td=""><td>sumed Remat</td><td>ning</td><td></td><td></td><td></td><td></td><td></td><td></td></as<>	sumed Remat	ning									

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$\underline{\text{TABLE B.1}}$. Calculation of Resuspension Rates Versus Time for HOI

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TABLE B.2.	Calculation	of	Resuspension	Rates	Versus	Time	for	Organic	Iodine
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			131						C1_Organtc	on Surface		Normalized	$\ln(N/N_{o})$	_Resuspenst	on Rates,	Section-1
Period	Sample Duration Minimum	Test Cumulative Minimum	Concen- tration, Ci/cc	Percent Organic	Resusp 1311	<u>Organic</u>	/Interval Undecayed Organic	Undecayed Unadjusted	Undecayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Ratio Adjusted/ Unadjusted
ZERO	0	0	0	0	- 0	0	0	28.93	78,93	28.93	78.93	0	0			
1	890	890	7.07E-09	1,3	356,40	4.63	4.89	24.04	74.04	22.80	70.21	-0.2382	-0.1171	3.5E-06	1,2E-06	0.35
2	1440	2330	2.932-09	1.2	238.98	2.87	3,30	20.75	70.75	18.05	61,55	-0.4718	-0,2488	1.7E-06	5,3E-07	0.31
3	1440	3770	7.90E-10	2.3	64,43	1,48	1.86	18,89	68.89	15,08	54,99	-0.6517	-0,3615	1.1E-06	3.1E-07	0,28
4	1470	5240	7.11E-10	2.1	59.20	1.24	1,70	17.19	67,19	12.57	49.12	-0.8339	-0.4744	1.12-06	2.8E-07	0.26
5	1884	7124	.4.68E-10	2,5	49.94	1,25	1.91	15.28	65,28	9,98	42.63	-1.0644	-0,6159	1.0E-06	2.6E-07	0.24
6	2436	9560	1,85E-10	2.5	25,53	0.64	1.13	14.15	64.15	7.99	36,22	-1.2870	-0.7790	5.3E-07	1.28-07	0,23
7	3060	12620	7.21E-11	5.4	12.50	0.67	1.44	12.71	62.71	5.98	29,49	-1.5769	-0.9846	5,8E-07	1.2E-07	0,21
8	3120	15740	6.23E-11	5.5	11.01	0.24	0,62	12.09	62,09	4.72	24.23	-1,8135	-1.1811	2,7E-07	5.3E-08	0,20
9	4030	19770	5,98E-11	1.8	13.65	0.25	0.80	11.29	61.29	3,46	18.79	-2.1231	-1.4351	2.8E-07	5.4E-08	0.19
10	4715	24485	3.26E-11	18,5	8.71	1.61	6,96	4.33	54.33	1.00	12.57	-3.3641	-1.8375	3.4E-06	4.3E-07	0.13
n	5280	29765	2.00E-11	3,3	5,98	0,20	1.17	3.16	53,16	0.53	8,97	-3.9952	-2.1751	1.0E-06	6.9E-08	0.07
12	5715	,35480	1.24E-11	5,7	4.01	0.23	1,91	1.25	51.25	0.15	6,14	-5.2652	-2,5534	2.7E-06	1.1E-07	0.04
13	5760	41240	8.89E-12	2,9	2. 90	0.08	0,99	0.26	50,26	0.02	4.27	-7.1885	-2.9174	4.6E-06	5.6E-08	0.01
14	5820	47060	1.17E-12	4.0	0,39	0.02	0.26	0	50,00	0	3.00		-3.2705		1.5E-08	
15	7200	54260	1.902-13		60.08											

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Total Resuspended = 28,93

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			131 ₁	Resu	spended		nC1 131 on	Surface		Normalized	$\ln(N/N_0)$	Resuspenst	on Rates,	Section-1
Period	Sample Duration Minimum	Test Cumulative Minimum	concen- tration, <u>µCi/cc</u>	131 131	nterval Undecayed	Undecayed Unadjusted	Undecayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Decayed Unadjusted	Decayed Adjusted	Ratio Adjusted/ Unadjusted
ZERO	0	0	0	0	0	1182.43	1302.43	1182,43	1302.43	0	0			
1	890	890	7.07E-09	356.40	375.88	806.56	926,56	764.76	878.54	-0.4358	-0.3937	7.2E-06	6.4E-06	0.89
2	1440	2330	2.93E-09	238.98	274.70	531, 86	651.86	462.69	567.08	-0.9383	-0.8315	4.8E-06	4.1E-06	0.84
3	1440	3770	7.90E-10	64.43	80.73	451.13	571.13	360.08	455.86	-1.1890	-1.0498	1.9E-06	1.5E-06	0.80
4	1470	5240	7.11E-10	59.20	80.98	370.15	490.15	270,58	358.30	-1.4748	-1.2906	2.2E0-6	1.7E-06	0.77
5	1884	7124	4.68E-10	49.94	76.46	293.69	413.69	191.81	270.19	-1.8188	-1.5729	2.0E-06	1,5E0-6	0.73
6	2436	9560	1.85E-10	25.53	45.21	248.48	368.48	140.29	208.04	-2.1316	-1.8343	1.1E0-6	7.9E-07	0.69
7	3060	12620	7.21E-11	12.50	26,58	221.90	341.90	104.33	160.76	-2.4277	-2.0921	6.2E-07	4.1E-07	0.66
8	3120	15740	6.23E-11	11.01	28.22	193.68	313.68	75.57	122.39	-2.7503	-2.3648	7.3E-07	4,6E-07	0.63
9	4030	19770	5.98E-11	13.65	44.52	149.16	269.16	45.74	82.53	-3.2524	-2.7588	1.1E-06	6.3E-07	0.59
10	4715	24485	3,26E-11	8.71	37.64	111.52	231.52	25.79	53.55	-3.8252	-3.1914	1.0E0-6	5.3E-07	0,52
11	5280	29765	2.00E-11	5.98	35.46	76.06	196.06	12.83	33.07	04.5236	-3.6734	1.2E0-6	5.2E-07	0.43
12	5715	35480	1.24E-11	4.01	33.49	42.57	162.57	5.10	19.48	-5.4457	-4,2024	1.7E-06	5.5E-07	0.32
13	5760	41240	8.89E-12	2.90	34,15	8.42	128.42	0.72	10.91	-7.4108	-4,7826	4.7E0-6	6.8E-0/	0.15
14	5820	47060	1.17E-12	0.39	6.43	1.99	121,99	0.12	7.32	-9.2025	-5.1820	4.1E-06	1.5E-07	
15	7 200	54260	1.90E-13	0.077	1.99	0	120.00	0	4.68		-5.6290		3.8E-08	

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TABLE B.3. Calculation of Resuspension Rates Versus Time for Total Iodine

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Total Resuspended = 1182.43

D

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B C

120 <---Assumed Remaining

E F G

COLUMNS--> A

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В.б

APPENDIX C

SELECTING REMAINING RESUSPENDABLE MASS

The approach for selecting a value for the remaining resuspendable surface radioiodine presented in Section 7.2.4, "Remaining Resuspendable Mass," can be difficult to apply in some cases. This is illustrated for the organic radioiodine where the data after about 24,000 min show some inflection in the lower curves in Figures C.2 and C.5. To reduce the inflection in the curvature and the variability in resulting resuspension rates, a value of 50 nCi was selected for N_{of}. This value seems high because only 29 nCi of organic radioiodine was measured during the entire resuspension experiment and also the organic is the least depositing of the various species. It may be reasonable to expect; however, that such a relatively large amount could be resuspendable after the end of the experiment because the resuspendable organic could be generated by reaction of the particle + elemental iodine with surface contaminants.

Table C.1 and Figures C.1 and C.2 show the result of making a lower estimate of N_{of} for the organic specie, 15 nCi instead of 50 nCi. Table C.1 is in the same format as Table B.2, except some repetitive columns are omitted. As expected, the difference between the adjusted and unadjusted resuspension rates is less in Table C.1 than when 50 nCi was used for N_{of}. Figures C.1 and C.2 show that the adjusted plots of ln(N) and ln(N/No) versus time are less linear than those shown in Figures C.2 and C.5.

The average and standard deviation of resuspension rates from 10,000 to 42,000 min are shown in Table C.2 for comparison. The change in resuspension rate is approximately inversely proportional to the change in assumed N_{of} . Given the assumptions made in calculating and using resuspension rates, either value may be just as useful.

A value based on a linear regression of the appropriate data may be the most useful approach, making the standard error of the vertical deviations a convenient basis for a linearity criterion. Table C.3 illustrates the results of calculating linear regressions of the ln (N/N_0) versus time data between 10,000 and 42,000 min. The results are shown for when 0, 15, and 50 nCi organic iodine are assumed remaining on the surface after 42,000 elapsed minutes. The resulting slopes and resuspension rates are significantly different at the 95% confidence limit.

C.1

	nCi Org	anic	Resuspension_Rates, s ⁻¹					
Test Cumulative, min	<u>On Surf</u> Undecayed <u>Unadjusted</u>	ace Undecayed <u>Adjusted</u>	Decayed <u>Unadjusted</u>	Decayed <u>Adjusted</u>	Ratio Adjusted/ <u>Unadjusted</u>			
0	28.93	43.93						
890	24.04	39.04	3.5E-06	2.2E-06	0.64			
2330	20.75	35.75	1.7E-06	1.0E-06	0.60			
3770	18.89	33.89	1.1E-06	6.2E-07	0.57			
5240	17.19	32.19	1.1E-06	5.8E-07	0.55			
7124	15.28	30.28	1.0E-06	5.4E-07	0.52			
9560	14.15	29.15	5.3E-07	2.6E-07	0.49			
12620	12.71	27.71	5.8E-07	2.8E-07	0.47			
15740	12.09	27.09	2.7E-07	1.2E-07	0.45			
19770	11.29	26.29	2.8E-07	1.2E-07	0.44			
24485	4.33	19.33	3.4E-06	1.1E-06	0.32			
29765	3.16	18.16	1.0E-06	2.0E-07	0.20			
35480	1.25	16.25	2.7E-06	3.2E-07	0.12			
41240	0.26	15.26	4 .6E-06	1.8E-07	0.04			
	15 <ass< td=""><td>umed Remainin</td><td>g</td><td></td><td></td></ass<>	umed Remainin	g					
	CORRI	ESPONDING COLU	JMNS IN TABLE I	8-2				
В	Н	I	Ν	0	Р			

<u>TABLE C.1</u> .	Calculation of Resuspension Rates for Organic I	odine
	Assuming a Reduced Amount Remaining	

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C.2





C.3

TABLE C.2. Comparison of Resuspension Rates

	<u> </u>	<u>d_N</u> of
	<u>15_nCi_</u>	<u>50 nCi</u>
Average	3.3E-07	1.3E-07
Std. Dev.	3.5E-07	1.4E-07

<u>TABLE C.3</u>. Linear Regressions of Organic Iodine Normalized Surface Activity Versus Time

	0	15	50
Slope	-1.9×10^{-4}	-8.3 x 10 ⁻⁵	-6.9 x 10 ⁻⁵
Intercept	1.3	-0.15	-0.12
Standard Error of Vertical Deviations	0.43	0.072	0.030
Resuspension rate, s $^{-1}$	2.9 x 10 ⁻⁶	3.9×10^{-7}	1.45 x 10 ⁻⁷