Wet Deposition Processes for Radioiodines

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ABSTRACT

The results of a review of wet deposition processes for radioiodines are presented. Experimental measurements have shown that precipitation scavenging can be an important process for removal of radioiodines released to the atmosphere. Measurements of radioiodine species in routine gaseous effluents have shown that the distribution depends upon the type of nuclear facility considered. Because of differences in radioiodine species, observed scavenging rates for radioiodine in fallout are not generally applicable to routine releases from nuclear facilities. Adequate models are available to estimate the air to surface transport by wet processes. At the present time, the need is for more information on the rate constants used in the models. Reactor accident consequence calculations that assume no behavior differences among iodine species indicate that about half the predicted early deaths and illnesses are attributable to radioiodines. The distribution of species released under accident conditions will depend upon the actual sequence of events. However, for many scenarios the wet deposition of radioiodines will have an important bearing on the consequences.

SUMMARY

The purpose of this report is to review existing information on wet deposition of radioiodines. An important distinction that must be made in such a review is between measurements and models for (a) radioiodine in fallout, and (b) radioiodine released from nuclear facilities. Differences in behavior between radioiodines in categories (a) and (b) are due to differences in the chemical forms of radioiodine present. Although both categories are discussed in this report, the emphasis of this work is on the behavior of routine and accidental releases of radioiodine from nuclear fuel cycle and other installations.

After detonation of a nuclear weapon above ground, the radioiodine produced is associated mainly with particulate debris that is carried to the upper levels of the atmosphere. As the debris ages the fraction of the radioiodine associated with particulates declines and that in gaseous forms, particularly organic iodides, increases. After $\sim$10 weeks, organic iodides dominate the airborne iodine species distribution. Because ^{131}I has a half-life of about eight days, most of the observations have been made during the first few passes of a fallout plume and, therefore, principally reflect the behavior of radioiodine associated with particulate material. Measured washout ratios for fallout radioiodine are comparable to those for particulate fission products. The observed washout ratios and empirical models for wet and dry deposition of fallout both illustrate the importance of wet processes for transporting fallout from air to vegetation. The air-to-vegetation transport process is important because it is the first step in an important environmental exposure pathway for radioiodines and other fallout radionuclides.

The air-to-vegetation transport of radioiodines by both wet and dry deposition is fundamentally dependent upon the chemical forms of the released activity. Radioiodine species in nuclear facility effluents have been measured using a consistent technique since 1970. The radioiodine species sampler used separates the airborne radioiodine into four components: (a) that associated with particulate material, (b) elemental iodine (I_2), (c) hypoiodous acid (HOI), and

(d) organic iodides (CH_3I and others). Many measurements using this sampler have been made at power reactors and the data base for chemical forms of ^{131}I in effluents from these facilities is substantial. Fewer measurements have been made of the chemical forms of ^{129}I discharged from fuel reprocessing plants, but some data are available. There are also some data for radiopharmaceutical facility discharges. All the data from nuclear facilities show that the chemical forms of radioiodine in discharges differ substantially from those in fresh fallout, so observed washout ratios for fallout have limited applicability to facility effluents.

Techniques for calculating wet deposition of radioiodines are available; however, the experimental basis for parameters needed to perform the calculations is far from adequate and, in some cases, is totally lacking. The wet deposition velocity approach has been recommended for assessing the impacts of routine releases. This technique employs washout ratios, which are spatial and temporal averages of scavenging rates, and can provide reasonable average deposition rates during the growing season when transfer of airborne radioiodine to the human food chain is most important. In this approach, wet deposition rates are computed in a way analagous to that employed for dry deposition. For each radioiodine species, indicated by the subscript i, the deposition rate can be expressed as

$$\omega_{wi}(x,y) = V_{wi}\,\chi_i(x,y) = a_i W_i P_g\,\chi_i(x,y) \qquad (S1)$$

in which

$\omega_{wi}(x,y)$ = the wet deposition rate onto vegetation ($\mu Ci/cm^2$-s) at downwind location (x,y)

V_{wi} = the wet deposition velocity (cm/s) onto vegetation

$\chi_i(x,y)$ = the average air concentration ($\mu Ci/cm^3$) at the point of interest

a_i = the dimensionless retention factor for wet deposited activity on the vegetation

W_i = the dimensionless washout ratio for the species

p_g = the average rainfall rate (cm/s) during the growing season.

The total deposition onto vegetation is the sum of the contributions from each of the four species. The computation is straightforward; however, the difficulty with this procedure is the absence of information about the four retention factors (the a_i) and the four washout ratios (the W_i). Washout ratios have not been measured for the gaseous iodine species released from nuclear facilities and the data on wet deposition retention factors are quite limited. Measurements of washout ratios and retention factors are needed to quantify wet deposition of routine radioiodine releases.

Some provisional estimates of washout ratios for gaseous radioiodine species (based on theoretical considerations) and of retention factors (based on limited data for fallout) were made (Section 10). Calculations using those preliminary values suggest that wet processes are relatively less important than dry deposition for routine releases. However, these calculations conflict with field measurements of radioiodine transport through the air-vegetation-milk pathway. The field measurements showed that wet deposition was correlated with increases in the concentration of ^{131}I in the milk of cows grazed near the plant. Resolution of the differences awaits measurements of the relevant wet deposition and retention parameters.

Predictions of the chemical forms of radioiodine that would be released under accident conditions are strongly dependent upon the accident scenario assumed. For the risk dominant accidents identified in the Reactor Safety Study, the large radioiodine releases projected to occur 2-3 hours after accident initiation are expected to be principally as I_2 or as particulates (potentially CsI). The mass concentrations of iodine in the plume during such releases

would be $\sim 10^4$ times ambient atmospheric levels of ^{127}I. For accident scenarios projected to release lower fractions of the core iodine inventory, a significant percentage of the released activity is expected to be in organic form. This is also true of releases from possible sequences in which (a) the radioiodine release from containment is delayed, or (b) the containment spray systems operate to remove inorganic forms.

The status of computations of the consequences of accidental releases is similar to that for routine releases. The computational methods are established but knowledge of the scavenging rate constants used is not adequate to justify confidence in the predicted ground contamination and subsequent consequences. In the Reactor Safety Study, the stated range of scavenging rate constants is from 10^{-5} s^{-1} to 10^{-2} s^{-1}. The rate constants used were 10^{-4} s^{-1} for frontal storms and 10^{-3} s^{-1} for convective storms. These values, based principally on measurements of scavenging of particles, are used for all radioiodine species released under accident conditions.

When both liquid and gas phase transfer resistance are considered, predicted rain scavenging rate constants for relatively high airborne concentrations of I_2 are $\sim 10^{-5}$ s^{-1}. Field experiments using > 100g of stable iodine yielded four estimates of $\Lambda(I_2)$ that ranged from 2×10^{-8} s^{-1} to 3×10^{-6} s^{-1} and did not exhibit the expected dependence on rainfall rate. Similar measurements of the snow scavenging rate constant (Σ) ranged from 9×10^{-9} s^{-1} to 2×10^{-7} s^{-1}, also for relatively high airborne concentrations of elemental iodine. Those estimates of $\Sigma(I_2)$ indicated proportionality to precipitation rate, but were only 5-10% of the values for $\Lambda(I_2)$ measured under similar conditions.

On the other hand, three measured values for inorganic ^{131}I in a high relative humidity plume from a chemical reprocessing plant were substantially higher, ranging from $\sim 3 \times 10^{-5}$ s^{-1} to 4×10^{-4} s^{-1}. A single measurement for organic ^{131}I was only 1% of the value determined for inorganic ^{131}I in the same storm. Snow scavenging rates for the processing plant effluent ranged from 4×10^{-5} s^{-1} to 1×10^{-4} s^{-1}, also lower than the comparable values for rain

scavenging. The increased scavenging rates (compared with field experiments) were attributed to condensation within the plume. However, it seems possible that other constituents of reprocessing plant off-gas may have affected these measurements.

Consequence calculations made using the assumptions of the Reactor Safety Study indicate that radioiodines account for about half of the early deaths and a comparable fraction of the early illness predicted to result from the risk dominant accidents. These projected consequences are due to external radiation exposure (a) during plume passage, and (b) from radionuclides deposited on the ground by wet and dry processes. The species of iodine released and the consequences will depend on the actual sequence of events. However, for many scenarios, wet deposition of radioiodines will contribute significantly to the predicted consequences.

RECOMMENDATIONS

It is recommended that measurements be undertaken to quantify parameters used to predict wet deposition of airborne radioiodine species released from nuclear facilities. To estimate properly the environmental consequences of routine releases, measurements are needed of (a) washout ratios and (b) the retention by vegetation of radioiodine in wet deposition. Rain and snow scavenging rate constants, used to predict the consequences of accidental releases, are also dependent on the radioiodine species released. Additional data are needed to quantify scavenging rates for gaseous species in the environment.

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WET DEPOSITION PROCESSES FOR RADIOIODINES

1. INTRODUCTION

The purpose of this report is to review existing information on wet deposition of radioiodines. Much of the available data deals with radioiodines in fallout from atmospheric nuclear explosions. That material is discussed separately. The emphasis of this work is on the behavior of radioiodines released from nuclear facilities.

Precipitation scavenging is known to be an important means of air-to-surface transport of radioiodine in weapons debris. For fresh fallout, when radioiodine is mainly associated with particulates and in-cloud scavenging processes are dominant, precipitation scavenging has been observed to be the principal transport mechanism. Observed washout ratios and empirical models for fallout deposition both illustrate the importance of wet deposition processes for air-to-surface transport of fallout particulates. The species of radioiodines in fallout debris has been observed to change with time after detonation from principally particulate to the less reactive gaseous forms. The observations of scavenging of fallout are summarized in Sections 2 and 3.

Radioiodines released to the atmosphere during normal operation of nuclear facilities are primarily in gaseous form (see Section 4), and the importance of precipitation scavenging for those effluents must be evaluated independently. A principal scavenging process would be below-cloud scavenging of the gaseous species by precipitation falling through the plume. However, in convective storms, updrafts could transport portions of the plume into the cloud layer and permit in-cloud scavenging processes to operate as well. There is also limited evidence that some locations may be affected by scavenging during fog and dew formation near reactor cooling canals or ponds.[1-3] Condensation in plant effluents plumes may also lead to greater scavenging than would be anticipated from below-cloud scavenging of plume contents.[4-6]

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For normal reactor effluent releases of short-lived radioiodines (principally ^{131}I), rain is the most important form of precipitation because transfer to edible vegetables and through the milk food chain occurs only during the growing season. Direct transport to vegetation during the growning season is also the most important process for transfer of airborne ^{129}I to the human diet. However, air-to-surface transport by all forms of precipitation in all seasons adds to the soil and runoff water inventories and thus to the long-term buildup of that isotope. Following large accidental releases of radioiodines in gaseous or particulate form, direct radiation from deposited material is a major concern for persons in the contaminated area. For these potential releases, all forms of precipitation must be considered as possible transport mechanisms.

Techniques for computing air-to-surface transport, the first step in the most important environmental radioiodine exposure pathways, are presented in Section 5. Different approaches have been recommended for accidental releases and for routine effluent discharges. The limited measurements of below-cloud scavenging rate constants used for transport calculations are summarized and discussed in Section 6. Predictions of scavenging rate constants are reviewed in Section 7 and compared with the measurements. The use of scavenging rate constants has been recommended for accident consequence calculations. Estimates of washout ratios appropriate for estimating wet deposition of routine releases are discussed in Section 8. The limited available information on retention by vegetation of radioiodine in wet deposition is discussed in Section 9. The results from Sections 4, 8 and 9 are used in Section 10 to make provisional estimates of the relative importance of wet and dry deposition processes for routine releases from four types of facilities.

Section 11 contains a brief review of the importance of radioiodine in calculation of the consequences of accidents. The observations of radioiodine behavior following the TMI-2 accident are discussed. The small radioiodine releases from TMI-2 led to a review of the methods used for calculating accident consequences and the results of that review are also considered in Section 11.

2. WASHOUT RATIOS AND CHEMICAL FORMS OF RADIOIODINE IN FALLOUT

Precipitation scavenging has been identified as the principal mechanism for air-to-surface transport of ^{131}I in debris from nuclear weapons testing. The relatively large concentrations of ^{131}I in milk in the northeast in the fall of 1976[7] brought this fact to the public's attention, but it had been observed and studied previously. In seven of eleven fallout episodes examined by Machta[8,9] precipitation scavenging of particulate bomb debris was the primary mechanism transporting the radioiodine from high altitudes to the surface.

The dimensionless washout ratio has been used to characterize the observed wet deposition of radioiodine in fallout. Introduced by Chamberlain[10] in slightly different form, this ratio relates the concentration of a nuclide in precipitation to that in air:

$$W = k/\chi_m \tag{1}$$

where

W =   the washout ratio

k =   the radionuclide concentration (Ci/kg) in rainwater collected (usually near ground level)

χ_m =   the radionuclide concentration per unit mass of air (Ci/kg) at a reference height in the atmosphere (usually measured ~ 1 m above the precipitation collector).

The washout ratio is a parameter that is easily measured and one which provides a rough measure of the importance of wet deposition processes. Unless the measured air concentration can be related to the concentration with the height in the column of air through which drops fell, relating a measured washout ratio to specific scavenging processes is difficult.

Table 1 contains data from various sources on the washout ratios that have been measured for iodine isotopes. The observed washout ratios for ^{131}I during the period containing the episodes analyzed by Machta were similar to those observed at the same time for ^{140}Ba and ^{95}Zr and were also similar to washout ratios measured for gross beta radioactivity from fallout measured at other times.[11,12] Other data sumaries[13-15] indicate that the measured ^{131}I washout ratios were approximately the same as those for other fission products. Because the washout ratios for fallout are considered to reflect principally the highly efficient in-cloud scavenging process (sometimes termed rainout) of particulate debris, the data imply that the ^{131}I was associated with substantially the same distribtuion of particles as the other fallout radionuclides. Gedenov et al.[14] found that washout ratios for inorganic atmospheric dust were about three times those for fallout debris. This suggests a possible difference in particle size distributions.

TABLE 1. OBSERVED WASHOUT RATIOS FOR IODINE

Location	Isotope	Precipitation Type	Observed Washout Ratio	Reference
England	^{131}I	Rain	420	15
United States	^{131}I	Snow	100--2700[a]	13, 21
England	^{131}I	Rain	300--1500	16 and 17
	^{127}I	Rain	10--50	
Sweden	^{131}I	Rain	250--2400	20

[a] Values computed assuming all the airborne and scavenged iodine was in particulate form.

Eggleton et al.[16,17] reported measurements of washout ratios for both naturally-occurring ^{127}I and fallout ^{131}I in the fall of 1961 at Harwell, England. The washout ratios for ^{127}I ranged from 10 to 50 while those for ^{131}I (and other particulate fission products in the fallout) ranged from 300 to 1500. Analysis of air samples indicated that about 75% of the fallout ^{131}I was in particulate form, while only a small fraction of the natural ^{127}I was in particulate form.

Measurements made at Hanford yielded highly variable estimates of the species distribution of ^{131}I in ground level air. An average of 50% of the ^{131}I was associated with particulates during 1962 when weapons testing occurred frequently; however, this fraction varied from 10 to 90%. Further studies indicated that less than 10% of the gaseous iodine was I_2 or HI.[18]

It has been found that washout ratios for ^{90}Sr in particulates are inversely proportional to the precipitation rate.[19] Washout ratios for ^{131}I in the particulate bomb debris would behave similarly. That variation may account for some of the scatter in the observed values in Table 1. As discussed in the next section, wet deposition of fresh fallout is greater than for older debris. Initial values of ^{131}I washout ratios in Sweden measured soon after the resumption of weapons tested in the atmosphere were 3-5 times higher than those observed two months later.[20]

Measurements made following more recent above-ground nuclear explosions have produced comparable results. A series of measurements of the chemical form of airborne ^{131}I in ground-level air were made at four locations in New Jersey following a nuclear test in 1976. The fraction of the total ^{131}I associated with particulates was 0.4--0.7 during the first six weeks after detonation, but then declined to 0.2--0.4 during the last three weeks of the measurements. Changes were also occurring in the gaseous fraction. The organic iodide component increased from one-fourth to about three-fourths of the gaseous ^{131}I during the measurement period.[22]

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Although the iodine species measurements cited above were not coincident with measurements of washout radios at the same locations, they generally support the view that scavenging of particulates is the main transport process for radioiodine in fresh fallout. The importance of particulate savenging decreases as the particulates are removed and the gaseous fraction becomes dominant. As noted, the washout ratios for ^{127}I measured at Harwell were substantially smaller than those for fallout ^{131}I. Because the monitoring period lasted approximately nine half-lives of ^{131}I, significant decay occurred while the chemical species were changing following the bomb test in 1976. The ^{129}I in the debris would of course not decay significantly and would approach the equilibrium species distribution for ^{127}I in the atmosphere. The trend shown in Reference 22 for ^{131}I is consistent with observed distributions of stable iodine. The limited data on the behavior of ^{129}I are also consistent with this pattern. Brauer et al.[23] found that the ratio of the concentration of ^{129}I in the atmosphere to its bomb fission yield was greater than the same ratio for ^{137}Cs. This finding indicates that ^{129}I was less efficiently removed from the atmosphere by wet and dry deposition process than ^{137}Cs.

3. EMPIRICAL FALLOUT PARTICULATE DEPOSITION MODELS

An alternative means of characterizing air-to-surface transport of fallout radionuclides is to derive empirical relationships from measurements of air concentration and wet and dry deposition. Predictive equations derived from measurements in the midwestern United States[24-26] show the importance of precipitation scavenging of fallout particulates. The total radioactive particulate deposition rate (D, Ci/m^2-day) was found to be directly proportional to the volumetric air concentration (χ, Ci/m^3):

$$D = (V_d + g p) \chi \tag{2}$$

In Equation (2), p is the rainfall rate (cm/day) during the period of interest. The empirically determined constants, V_d and g, reflect the contributions of dry and wet deposition respectively. The dry deposition constant (V_d, m/day) and the wet deposition constant (g, m/cm of rain) were determined for both fresh and old fallout and are shown in Table 2.[26] The changes in the deposition parameters must reflect changes in particle size distributions with time following detonations.

TABLE 2.

EMPIRICALLY DETERMINED WET AND DRY DEPOSITION
PARAMETERS FOR FALLOUT PARTICULATES

Type of Fallout Deposition	Parameter	Fresh Fallout	Old Fallout[a]
Wet	g (m/cm of rain)	14000	4530
Dry	V_d (m/day)	1000	200

a Defined by the absence of ^{140}Ba in air and deposition samples.

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The importance of wet deposition can be seen by comparing the magnitudes of the wet and dry deposition terms in Equation (2). If the average daily rainfall rate is greater than 0.05 cm/day (about 7 inches/year), the surface deposition of older fallout radioactivity would be dominated by precipitation scavenging. Dry deposition was found to be somewhat more important for periods immediately following tests, but an average rainfall rate of only 0.07 cm/day would equalize the deposition rates for fresh fallout by wet and dry processes. In a location with an average rainfall rate of 0.25 cm/day (36 inches/year), wet processes would account for 70% and 80% of the total deposition from fresh and old fallout, respectively.[26]

4. CHEMICAL FORMS OF RADIOIODINES RELEASED
IN GASEOUS EFFLUENTS

Most of the measurements of the chemical forms of the radioiodines in the gaseous discharges from commercial nuclear facilities have been made using the species-differentiating sampler, which was developed by Keller et al.[27] Performance characteristics have more recently been reevaluated by Emel et al.[28] The sampler, shown in Figure 1, is designed to separate the airborne radioiodine into four components: (a) iodine associated with particulate material, (b) elemental iodine (I_2), (c) hypoiodous acid (HOI), and (d) organic iodine compounds. Air entering the species sampler passes, in sequence, through a high efficiency particulate filter, a bed of cadmium iodine (with excess I) adsorbed on chromosorb-P (CdI bed), a bed of 4-iodophenol adsorbed on alumina (IPh bed), and a bed of activated charcoal or silver-loaded zeolite (AgX). A second bed of activated charcoal or AgX is used as a backup bed but generally contains little radioiodine after sampling for periods of 1-2 weeks. Measurements made with this sampling system are discussed for three types of facilities in the following subsections.

4.1 Radioiodines Released from Power Reactors

The first measurements of radioiodine species at a boiling water reactor (BWR) were made at Oyster Creek in 1970.[29] Since that time the same sampling system has been used to measure radioiodine species at eight other BWRs[1,3,22,29,30] and at ten pressurized water reactors (PWRs)[31-38] in the United States. The initial radioiodine species measurements employed short sampling times. However, continuous measurements for periods of six months or more have been performed in both types of reactors, so a substantial data base has been established. These measurements have shown that the least reactive forms, HOI and organic iodides, comprise more than half of all the radioiodine released from these facilities. Table 3 contains annual composite ^{131}I species fractionations for effluents from contemporary BWRs and PWRs based on the data sources referenced above. The composites reflect many
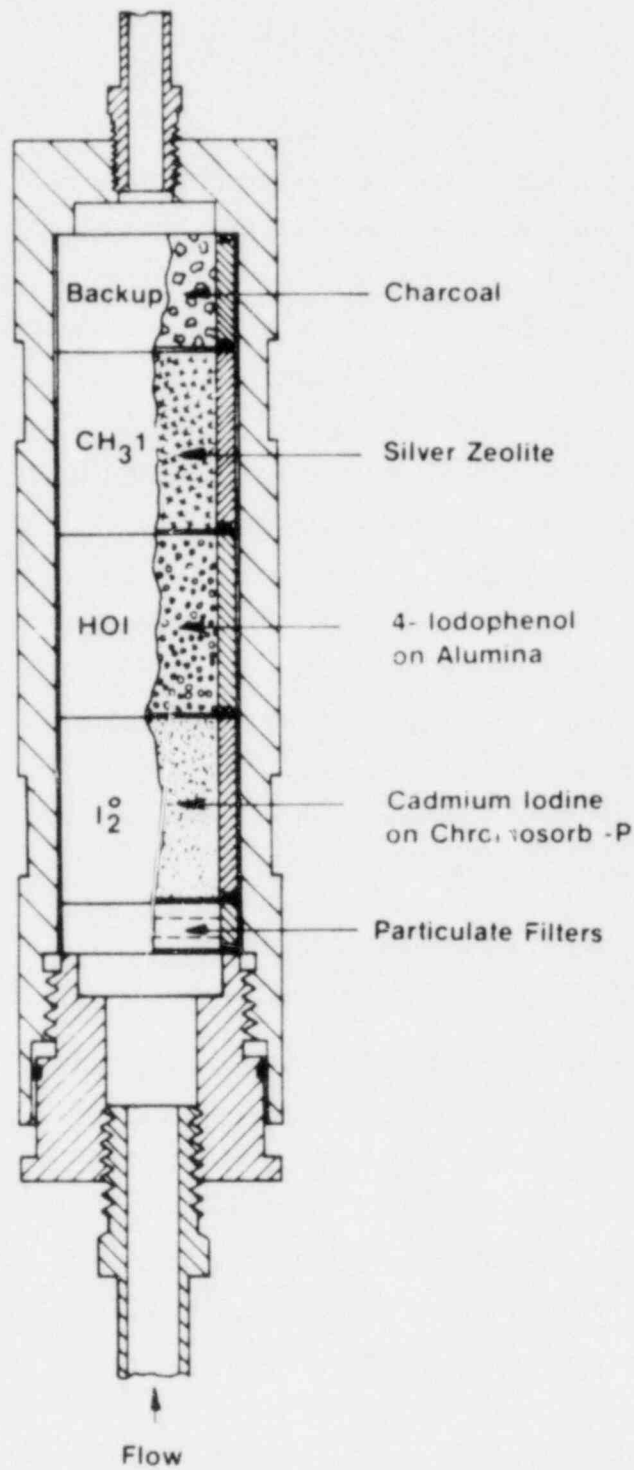
9

Figure 1. Radioiodine Species Sampler

ICPP-A-0350

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TABLE 3. AVERAGE RADIOIODINE SPECIES DISTRIBUTIONS IN
GASEOUS EFFLUENTS FROM U.S. LIGHT WATER REACTORS

| Facility Type | ^{131}I Species Distribution (percent) | | | |
	Particulates	I_2	HOI	Organic
Boiling Water Reactor[a]	7	36	26	31
Pressurized Water Reactor	2	27	40	31

[a] Distribution for a contempory BWR in which the steam jet air ejector offgas
has been treated.

individual sources within each type of facility; radioiodine species data for individual sources (and specific operating modes) are contained in the referenced reports.

A different radioiodine species sampler, developed at the Karlsruhe Nuclear Research Center, has been used to measure ^{131}I species in a PWR in Germany.[39] The Karlsruhe sampler was designed to separate the airborne ^{131}I into three components: (a) iodine associated with particulates, (b) elemental iodine, and (c) organic iodides. Hypoiodous acid (HOI) would be collected on the third bed and categorized as organic iodine in measurements made using this sampler. It was reported that a special sampling train using an IPh bed revealed that very little HOI was present in reactor containment air samples and that the use of an IPh bed to measure HOI was then discontinued.[39] The reactor containment ^{131}I species measurements in the U.S.[36] have shown that the organic iodide fraction reaches 90-96% of the total airborne ^{131}I, so the containment data are not necessarily inconsistent. However, HOI observed in the auxiliary and fuel handling buildings in the U.S. PWRs would be included in the organic fraction reported for the German PWR.

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4.2 Radioiodine Released from Fuel Reprocessing Plants

The number of measurements of radioiodine species in fuel reprocessing plant (FRP) discharges is limited. A few measurements have been made of the ^{129}I species distribution in the effluent of the Idaho Chemical Processing Plant (ICPP) using the radioiodine species sampler described above. The principal gaseous species were found to be organic iodides (60–80% of the total). The elemental iodine fractions observed ranged from 2–20% and the HOI fraction was consistently about 20%.[40]

Organic iodides have also been observed using gas chromatography at the Hanford Purex Plant and at the Savannah River Plant (SRP). Methyl, ethyl, propyl, and butyl iodides were identified. Other longer chain organic compounds may have been present but were not specifically identified. The measurements reported did not permit estimation of the fractions of the discharged activity present in various forms.[41,42]

A small amount of information has been published about radioiodine species measurements in European FRPs. A recent publication of the International Atomic Energy Agency (IAEA)[43] indicates that about 70% of the radioiodine discharged to the atmosphere at Dounreay was in organic form. This contrasts with measurements reported for the Karlsruhe plant where organic iodides comprised only about one-fourth of the ^{129}I activity released. The latter results were probably obtained using the Karlsruhe sampler described above. The measurement technique used at the Dounreay plant is not discussed in the IAEA report.

4.3 Radioiodine Released from Radiopharmaceutical and Health Care Facilities

Only a small amount of data is available on radioiodine species in gaseous effluents from radiopharmaceutical and health care facilities. The most common radioiodinated pharmaceuticals are administered in liquid form and are eliminated in excreta that are stored for decay or diluted prior to discharge. Thus the gaseous releases of radioiodine from health care facilities are normally

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a minimal fraction of the radioiodine activity that enters the facility. The principal sources of airborne activity in this category are the facilities that produce the tracer and those that manufacture iodinated pharmaceuticals.

Radioiodine species distributions were measured at a manufacturing facility for a 10-week period using the 4-component radioiodine species sampler described above. The observed distributions were variable and apparently changed with the production schedule, although a detailed correlation study was not performed to verify preliminary associations. An average of 12% of the released activity was associated with particulates and 31% was present as elemental iodine. The remainder was nearly all organic iodides (53%); only 4% of the total was released as HOI.[44]

Gas chromatographic measurements reported for a Japanese ^{131}I production facility indicate that most of the released activity was in organic form. The organic iodide fraction was found to increase as the processing proceeded.[45] No radioiodine species measurements have been reported for ^{131}I production facilities in the United States.

5. CALCULATION OF AIR TO VEGETATION TRANSPORT FOR WET PROCESSES ACTING ON EFFLUENT PLUMES

Equations (1) and (2) illustrate techniques for calculating deposition using an empirical washout factor approach and the ground-level air concentration. More sophisticated models have been developed for scavenging of contaminants from plumes by precipitation falling through the plume. In some models, precipitation scavenging is assumed to be a first-order rate controlled process and scavenging rate constants are used to describe the change of the air concentration with time. For in-cloud scavenging, the rate constant is Λ (s^{-1}) and the air concentration at time t, $\chi(t)$, is related to the initial concentration, χ_o, using

$$\chi(t) = \chi_o \exp(-\Lambda t). \tag{3}$$

For below-cloud scavenging, a similar time dependence for $\chi(t)$ is assumed with Λ (s^{-1}) used to describe scavenging by rain and Σ (s^{-1}) used to describe scavenging by snow. Like the dry deposition velocity, the scavenging rate constants Λ and Σ are parameters that largely obscure the complex microphysical processes that produce the observable results.[10,46-48]

For below-cloud scavenging of a plume from a stationary source, the expected rate of deposition of material on vegetation at the ground surface is obtained by integrating over the plume in the vertical direction.[47] For rain, the deposition rate to vegetation ω_V (Ci/m^2-s) is

$$\omega_V(x,y) = \frac{a\Lambda(x)Q}{\bar{u}\,\sigma_y(x)\,(2\pi)^{\frac{1}{2}}} \exp(-y^2/2\sigma_y^2) \exp(-\Lambda'/(x/\bar{u})) \tag{4}$$

in which

x = downwind distance (m) from the source

y =   crosswind distance (m) from plume centerline to point of interest

a =   the fraction of the radioiodine in rainwater that is retained on the vegetation

$\Lambda(x)$ =   the removal rate constant (s^{-1}) at downwind distance x

Q =   the radioiodine release rate (Ci/s)

u =   the mean wind speed (m/s)

$\sigma_y(x)$ =   the standard deviation of the crosswind concentration distribution (m)

Λ' =   an average scavenging rate constant (s^{-1}) that accounts for earlier scavenging at upwind locations (Λ' may differ from $\Lambda(x)$).

A similar equation can be written for scavenging by snow, in which the rate constants Σ and Σ' (s^{-1}) replace Λ and Λ', respectively.

Application of the washout ratio approach was recommended by Engelmann[13] as a useful predictive technique. As indicated in Section 2, a significant body of data has been collected on washout ratios for bomb debris. However, those data are not generally applicable to the scavenging of radionuclides in gaseous effluents from nuclear facilities. The radioiodine species in plant effluents differ from those in fallout. Also, the fallout debris generally entered the troposphere at a much higher elevation than plant effluents would and the concentration profiles of the radionuclides would differ markedly, so it would be fortuitous if the values for fallout debris applied.

The washout ratio approach can be made mathematically similar to that normally used to compute dry deposition rates. Slinn[49] defines a wet deposition velocity (V_w, m/s) as the ratio of the wet deposition rate onto vegetation to the

ground level air concentration. Then, assuming we know the wet deposition velocity, the deposition rate could be computed using Equation (5), which is analagous to the dry deposition equation:

$$\omega_V(x,y) = V_w \chi(x,y) \tag{5}$$

It has been suggested that the total wet deposition velocity can be written as the product Wp, where p is the precipitation rate (water equivalent values are used for snow). For gases, W is taken to be approximately equal to the partition coefficient appropriate to the gas being scavenged and the characteristics of the rainwater (e.g., the pH and concentrations of other soluble species). Thus the total wet deposition rate is

$$\omega(x,y) = Wp \, \chi(x,y) \tag{6}$$

The washout ratio approach is considered most appropriate for estimating average deposition rates because the washout ratios themselves reflect the averaging process that occurs naturally during each rain or snow storm and during the rain and snow seasons. Using field measurement data, Slinn[49] has estimated the instantaneous wet deposition velocity at several times during a storm and found it varied over three decades. Part of the variation is attributable to the rainfall rate, which varied over two orders of magnitude, but the individual washout ratios also varied by two orders of magnitude during the 7-hour storm. As noted earlier, the natural averaging process obscures such variations and their causes in most measurements of both wet and dry deposition.

For wet transport of radioiodine to vegetation, the fractional retention (a) must be included:

$$\omega_V(x,y) = aW \, p \, \chi(x,y) \tag{7}$$

To estimate the impact of wet deposition on the transport of airborne radioiodine to human foods, Equation (7) requires values of W and a for the radioiodine species that are released (see Section 4). If those values were

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known, at least as well as we know V_d for the four species (see Reference 50), then predictions of wet deposition would be comparable to those now employed for dry deposition. The status of our knowledge of these variables is discussed in Sections 8 and 9.

6. MEASUREMENTS OF SCAVENGING RATE CONSTANTS
FOR IODINE GASES

The number of field measurements made to determine the scavenging rate constants for gaseous iodine compounds is small. Engelmann and his co-workers obtained most of the available data in a series of tests in 1965-1966 at the Rimrock Lake Test Site.[5,6,11] Elemental iodine was released at an elevation of 40 feet above the ground by heating stable iodine crystals. The iodine release rates were ~ 0.3 g/s, initially for 5-minute periods and later for longer times. The iodine content of the precipitation collected at downwind locations within 100 m of the release point was measured and used to compute the scavenging rate constant. The results of these measurements, given in the referenced reports, are shown in Table 4. The values for Λ ranged from 1.9×10^{-8} to 2.5×10^{-6} s^{-1}. The expected magnitude of Λ and an expected dependence on precipitation rate (see Section 7) were not confirmed by the measurements. The measured values were from 1/2000 to 1/5 of the values predicted by Engelmann using a relatively simple model. The observed values of Σ were much lower than the measured values of Λ, a finding attributed to a slower rate of reaction of the iodine with the snow (in spite of the larger surface area of the snowflakes). The rate constants for snow of a particular type did tend to increase with precipitation rate.

Measurements made with ^{131}I discharged from two stacks of the Hanford fuel reprocessing plant yielded much larger scavenging rates. The results of these experiments are also shown in Table 4. Measured values of Λ for inorganic ^{131}I ranged from $\sim 3 \times 10^{-5}$ to 4.2×10^{-4} s^{-1}. Comparable values of Σ ranged from 4.1×10^{-5} to 1.2×10^{-4} s^{-1}. (For these measurements, inorganic ^{131}I was defined as that collected in a NaOH bubbler. The ^{131}I that passed through the bubbler and was collected on a charcoal bed was termed organic.) For both rain and columnar snow crystals, correlations with precipitation rate appeared plausible. The values for rain exceeded predicted values by about a factor of ten. It was found that the discharged air contained sufficient moisture to condense to cloud droplets and uptake of the ^{131}I by condensing water vapor was

TABLE 4. MEASURED SCAVENGING RATE CONSTANTS FOR IODINE GASES

Chemical Form	Precipitation Characteristics		Scavenging Rate Constant, Λ or Σ (s-1)
	Type	Rate (mm/hr)	
	FIELD RELEASE TESTS[a]		(Λ)
I_2	Rain	0.16	2.5×10^{-6}
I_2	Rain	0.22	1.2×10^{-6}
I_2	Rain	1.0	1.9×10^{-8}
I_2	Rain	1.8	1.6×10^{-7}
			(Σ)
I_2	Snow: Sm. Sp. Dend.[b]	0.05	3.0×10^{-8}
I_2	Snow: Pdr. & Pl. Dend.[c]	0.07	3.7×10^{-8}
I_2	Snow: Sm. Sp. Dend.[b]	0.48	1.8×10^{-7}
I_2	Snow: Unclassified	0.27	9.4×10^{-9}
I_2	Snow: Powder	0.67	5.5×10^{-8}
I_2	Snow: Powder	0.7	7.4×10^{-8}
	PROCESSING PLANT EFFLUENT TESTS		(Λ)
Inorganic[d]	Rain	0.37	$\sqrt{3} \times 10^{-5e}$
Inorganic[d]	Rain	0.52	3.2×10^{-4}
Inorganic[d]	Rain	0.87	4.2×10^{-4}
Organic[d]	Rain	0.52	3.5×10^{-6}
			(Σ)
Inorganic[d]	Snow: Columns	0.40	4.1×10^{-5}
Inorganic[d]	Snow: Columns	0.42	4.2×10^{-5}
Inorganic[d]	Snow: Columns	0.8	1.2×10^{-4}
Inorganic[d]	Snow: Agg. Sp. Dend.[f]	0.79	4.7×10^{-5}
Inorganic[d]	Snow: Agg. Sp. Dend.[f]	1.2	4.2×10^{-5}

a. Elemental iodine generated by heating iodine crystals.

b. Small spatial dendrites.

c. Powered and planar dendrites.

d. Inorganic/organic forms defined by activity trapped/not trapped by a NaOH bubbler sampling device.

e. The plume was not always over the sampling grid during this test.

f. Aggregated spatial dendrites.

postulated as the principal scavenging mechanism. Computed scavenging rates for 40-μm diameter drops were about two-thirds of the observed rates. One estimate of Λ for organic ^{131}I was given: 3.5×10^{-6} s^{-1}. The lower value, approximately 1% of that for inorganic forms, was attributed to a slower reaction rate with the water.[4-6] The possible impact on the scavenging rates of other chemical species in the processing plant plume was not discussed.

The values of Λ and Σ for processing plant effluents must be presumed to be more representative (for that type of plume) than the rate constants derived from field release tests. However, if the higher scavenging rates were due to droplet formation as hypothesized, then it is not appropriate to apply those rates to predict scavenging by rain on a summer day when there is no in-plume condensation. For the same reason, they cannot be applied to relatively dry effluent gases from other facilities.

It is possible that the difference between the Rimrock Lake data and those from the processing plant tests are partially due to differences in the airborne iodine concentration. The iodine mass concentration in the processing plant effluent is not known, but plume iodine concentrations during the field release tests were undoubtedly higher. The presence of NO$_x$ or other gases may also have affected the scavenging of iodine from the plant effluent.

Other measurements of scavenging of iodine compounds (I_2 and CH_3I) have been made in a closed vessel designed for simulations of reactor accident conditions.[51,52] The spray droplets generated were $\sim$1 mm in diameter and were composed of various solutions of boric acid or boric acid and sodium hydroxide. The results are not directly applicable to scavenging by rain in the environment; however, the experiments did provide insights into scavenging processes.

7. PREDICTIONS OF SCAVENGING RATE CONSTANTS
FOR RADIOIODINE

A thorough treatment of scavenging rates for radioiodines was first published, not surprisingly, by A. C. Chamberlain.[10] His approach, extended by Engelmann,[46,47,53] treated raindrops as a perfect sink for iodine in the gas phase. Engelmann computed the rain scavenging rate constant Λ (s^{-1}) using data on raindrop size distributions as a function of rainfall rate and both theoretical and experimental capture efficiencies. The results, shown in Figure 2, indicated that Λ should be $3\text{-}5 \times 10^{-5}$ s^{-1} for p in the range 0.5-1 mm/hr. As noted in Section 6, values of Λ measured at Rimrock Lake were substantially lower than predicted. The measured values are also shown in Figure 2 (as open circles, numbered in the sequence given in Table 4). The Rimrock Lake results appear to decrease with increasing rainfall rate, which is contrary to the prediction. Predictions made using different raindrop size spectra yielded values higher than given for drops as perfect sinks in Figure 2.

The field measurements indicated that the simple approximation of the drop as a perfect sink was not realistic. In other tests at Rimrock Lake, measured values of Λ for the more soluble gas Br_2 were in better agreement with theory, suggesting that the solubility of the gas was important and that mixing and reaction rates within the drops could be factors that determine the scavenging rate. The lower value of Λ measured for organic iodides in the field tests employing plant effluents (see Table 4) also supported this view. Postma[54] studied the effect of gas solubility and concluded that the equilibrium partition coefficient determines both the rate of gas absorption and the maximum amount of gas absorbed by stagnant drops. The detailed theoretical basis for his conclusions is given in Reference 51 and in more recent discussions of reactor safety systems.[55,56] A similar analysis, also for reactor containment spray systems, was performed by Griffiths.[57] The results of these theoretical studies can be used to estimate scavenging rate constants for radioiodines in the environment.
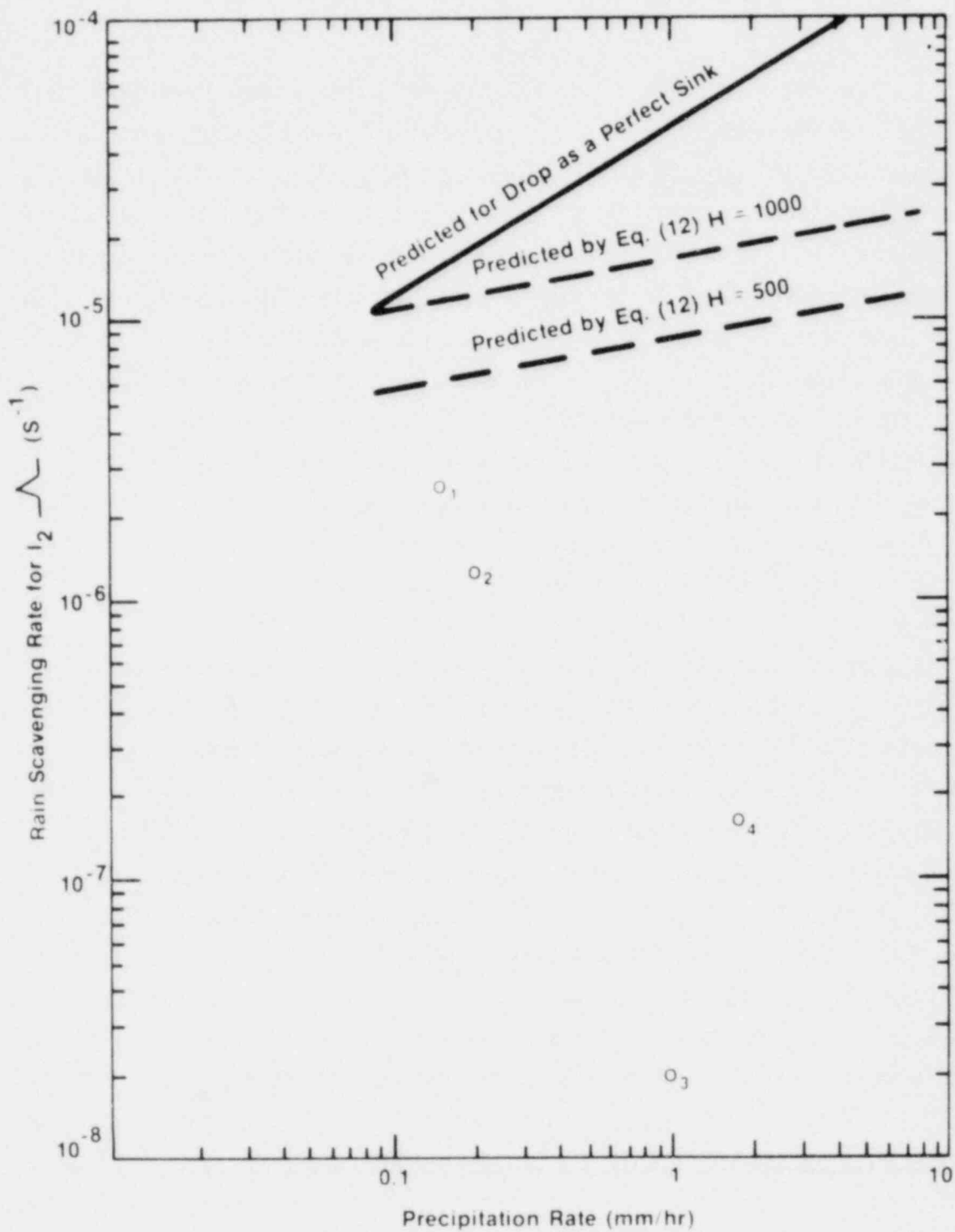
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Figure 2.   Rate Constants for Scavenging of
I_2 by Rain

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The iodine scavenging model employed for reactor containments[51,55-59] is developed in terms of spray flow rates entering a fixed volume containing airborne iodine. However, the predicted scavenging rate constant can be extracted and rewritten in environmental terms. For the case when gas phase resistance dominates, the scavenging rate constant is given by:

$$\Lambda g = \frac{6 \, k_g \, p}{d \, v_g} \tag{8}$$

in which

k_g = the gas phase transfer coefficient, essentially the deposition velocity of airborne iodine onto the drop surfaces (cm/s)

d = the drop diameter (cm)

v_g = the terminal fall velocity of the drop (cm/s).

The gas phase transfer coefficient was calculated using

$$k_g = (2 + 0.6 \, Re^{1/2} \, Sc^{1/3}) \, D/d \tag{9}$$

where

Re = $\rho d \, v_g / \mu$, the Reynolds number (dimensionless)

Sc = $\dfrac{\mu}{\rho D}$, the Schmidt number (dimensionless)

D = the diffusivity of iodine in air (cm^2/s)

ρ = the density of the gas (g/cm^3)

μ = the viscosity of the gas (g/cm-s).

The principal difference between Equations (8) and (9) and those employed by Engelmann is that Equation (8) uses the median drop size while Engelmann integrated the removal rate constant for each drop size interval over the range of raindrop sizes for rainfalls of various intensity. Nonetheless, predictions made using Equation (8) were found to be in excellent agreement with the Containment Systems Experiment measurements of washout of I_2, which implies that there was little liquid phase resistance to absorption of I_2 in their drops. As was mentioned in Section 7, the drops employed were solutions of boric acid and boric acid with NaOH.

For cases when liquid phase transport is significant, the overall resistance to transport is found using

$$\frac{1}{K} = \frac{1}{k_g} + \frac{1}{Hk_L} \tag{10}$$

in which H is the partition coefficient, K (cm/s) is the overall transfer coefficient, and the liquid phase transfer coefficient k_L (cm/s) is related the diffusivity (D_L, cm^2/s) of the gas in the liquid by

$$K_L = \frac{2 \pi^2 D_L}{3d}. \tag{11}$$

The diffusivity of I_2 in water can be calculated using the relationship given by Knudsen.[58] For a temperature of 20°C, D_L is 1.1×10^{-5} cm^2/s for elemental iodine.

A more general expression for the scavenging rate constant can be written using the relationships discussed above. Considering both gas phase and liquid phase resistance to transport, the rate constant is

$$\Lambda = \frac{4\pi^2 \alpha D H D_L p}{(\alpha D + (2/3) \pi^2 H D_L) d^2 v_g} \tag{12}$$

where the term α is

$$\alpha = \frac{k_g d}{D} = (2 + 0.6\, \text{Re}^{1/2}\, \text{Sc}^{1/3}). \tag{13}$$

Equation (12) has been used to estimate scavenging rate constants for I_2 during the Rimrock Lake tests. The results are shown in Figure 2 for two possible values of H. The dependence on precipitation rate is less important than for the perfect sink model; however, these predictions are not in any better agreement with the values of Λ measured at Rimrock Lake. In particular, the predictions do not match the observed values of Λ for higher values of p.

Hales[59] has extended the theory to consider transfer across the interface and mixing and chemical reactions in the liquid explicitly. The fact that the gas-to-liquid transfer process is reversible means the drops may release previously scavenged radioiodine when passing through air containing lower concentrations; thus the initial concentration distribution in the plume can influence the amount of precipition scavenging. The scavenging process can also modify the concentration distribution in a plume. For example, activity removed from an elevated plume in a stable atmosphere could be desorbed from the drops as they pass through clean air below the plume.[59] This downward transport of the plume has also been discussed by Slinn.[60]

Desorption of iodine from the raindrops could have influenced the Rimrock Lake test results. The sampling arcs were located close to the release point, so the plume was probably not at ground level and the drops would have fallen through air that was relatively free of I_2 prior to collection. The detailed kinetics of the possible desorption have not been examined as part of this review; however, it seems unlikely that the discrepancy between the predictions and measurements will be completely resolved by desorption of scavenged I_2.

In the calculation of accident consequences in the <u>Reactor Safety Study</u>,[61] precipitation scavenging of radioiodine and of radioactive particulates were both treated in the same manner. The common scavenging rate constants were assumed to be proportional to p with proportionality constants of 10^{-4} s^{-1} (mm/hr)$^{-1}$ for frontal storms and 10^{-3} s^{-1} (mm/hr)$^{-1}$ for convective storms. The frontal storm values are comparable to those for inorganic iodine in the condensing plume from the fuel reprocessing plant at Hanford (Table 4) and to those for below cloud scavenging of soluble dye particles.[62] For the risk dominant accidents, the released activity would almost certainly be part of a saturated plume.[63,64] The measured values for particulates did not exhibit the expected dependence on median particle size,[48,62] but Dana and Hobes[65] suggest that comparisons of experimental and calculated washout rates may not be meaningful because it is difficult to characterize adequately the actual particle size and raindrop size distributions. The calculations of particle scavenging depend strongly upon these distributions, which cannot be predicted reliably in advance. The fact that the experimentally determined values of Λ are approximately independent of particle mass median diameter is encouraging and supports the simple empirical approach used in Reference 61 for frontal storms. The basis for the convective storm predictions in Reference 61 was also empirical. Mesoscale field experimental data for a variety of particulate tracers[66-69] formed the basis for the estimate of $\Lambda = 10^{-3} \rho$ (s^{-1}). Implicit in the application of these experimental data to particles containing radioiodine is the assumption that the particle size distribution, not the chemical composition, is the principal determinant of the scavenging rate constant.

8. PREDICTION OF WASHOUT RATIOS FOR RADIOIODINE

As noted earlier, the dimensionless washout ratio (W) has been recommended for use in estimating average values of the wet deposition rate (using Equation 6). Because there are so few measurements of washout ratios that are applicable to radioiodine released from nuclear facilities, predicted values of the washout ratio are of particular interest at this time. The recent sets of estimated washout ratios are discussed below.

The washout ratio is proportional to the effective scavenging rate constant, and is obtained by integrating between ground level and the top of the plume.[49,70] Brenk and Vogt[70] employ the relationship

$$W = L \, \overline{\Lambda}/p \tag{14}$$

where L is the height of the mixed layer (m) and $\overline{\Lambda}$ is the concentration weighted average scavenging rate constant (s^{-1}). For typical German sites, they estimate a mixing height of 500 m and a long-term average rainfall rate of 0.7 mm/hr and give the approximate relationship

$$W \simeq 3 \times 10^9 \, \overline{\Lambda} \tag{15}$$

For elemental iodine, Chamberlain's curve[10] was approximated using

$$\Lambda \, (I_2) = 8 \times 10^{-5} \, p^{0.6} \tag{16}$$

and they estimate

$$W \, (I_2) \simeq 2 \times 10^5 \tag{17}$$

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for the typical German site mentioned above. Brenk and Vogt[70] employ a similar approach for aerosols. The scavenging rate constant is approximated by

$$\Lambda \text{ (aerosol)} \simeq 1.2 \times 10^{-4} \, p^{0.5} \tag{18}$$

and, using Equation (15) for their typical site, they obtain

$$W \text{ (aerosol)} \simeq 3 \times 10^{5} \tag{19}$$

These authors indicate that Equation (19) approximates the median of the log-normal distribution of three sets of observed washout ratios. However, variations within a set of measurements and between data sets are substantial.[70] No estimates of W (HOI) or W (CH_3I) are provided in Reference 70.

It is interesting to compare the values recommended in Reference[70] with those of the German Regulatory Guide (GRG), given in the same reference. For the typical precipitation rate of 7 mm/hr, Equation (17) gives a value of Λ (I_2) that is $\sim$ 1/3 that of the GRG. On the other hand, Λ (aerosol) from Equation (19) is 1.5--2 times treater than the GRG value.

Slinn[49] has estimated long term average washout rates for elemental iodine and for methyl iodide that are based on their equilibrium partition coefficients. Eggleton[71] performed an evaluation of the partition coefficient for I_2 in water. In Reference 49 W (I_2) is estimated to be 10^3, from Postma's more recent work,[54] for a concentration of 1 µg of I per liter of pH 5 rainwater. Under those conditions, the concentration in the gas phase would be 10^3 ng/m^3, roughly 100 times the natural airborne inorganic iodine concentration. The concentration of fission product iodine in air discharged from boiling water reactors[22] is about 5--10 ng/m^3. From Table 3 we see that $\sim$ 1/3 of the iodine would be I_2. Thus, the plume concentration of I_2 at the point of discharge would be slightly greater than the natural concentration, but would again approach the ambient level as the plume dispersed. Thus, the expected airborne I_2 concentration of $\sim$ 10 ng/m^3 suggests a partition coefficient of $\sim 10^4$ for pH 5 raindrops if Postma's curve[54] is used. For this estimation technique, the

appropriate long term value of W (I_2) in reactor effluents is therefore $\sim 10^4$ for the assumed conditions. The assumed value of the pH of rainfall has an important bearing on the estimated washout ratio.

The washout ratio estimated using the partition coefficient is only 1/20 of that estimated for the typical German reactor site (see above). The difference is due to the assumptions employed. Chamberlain[10] assumed that raindrops were a perfect sink (very large H) and the estimate of Reference 70 uses Chamberlain's predicted value of Λ. For normal rainwater, at a slightly less acid pH, the partition coefficient estimate of W (I_2) would be a little nearer to the value in Reference.[70]

For methyl iodide, Slinn[49] estimates a long term washout ratio of 4.2. The source of the estimate and the assumed conditions are not given, but are presumably similar to those he used for estimating W (I_2) (discussed above). Postma[55] gives an equilibrium partition coefficient of H=5 for CH_3I in water at $20^\circ C$, but no information on the concentration or pH dependence of $H(CH_3I)$, so the origin of Slinn's value is not clear.

Long term average washout ratios for particulates released near the ground are also summarized in Reference 49. Measured values for lead vary from 6.3×10^4 to 2.4×10^5. The former ratio is the average for the METROMEX (St. Louis) data[72] and represents mainly convective storms. The larger value is an annual average value from measurements at Harwell and reflects mostly precipitation from frontal storms. However, the measured washout ratios for iron are the same (2.1×10^5 and 2.3×10^5) at the two locations, so an effect of storm type is not shown by the data. The mean of these values is $\sim 2 \times 10^5$, which is comparable to the estimate of 3×10^5 for the typical German reactor site.

A washout ratio for HOI was not given in Reference 49. However, the same approach can be used; that is, the equilibrium partition coefficient of HOI can be used to estimate W (HOI). The few existing measurements[73-75] have yielded estimates of H between 300 and 900 for conditions approximating those expected in the environment around a nuclear facility. A provisional value of W (HOI) = 600 is consistent with the available experimental data on partitioning of HOI.

9. RETENTION BY VEGETATION OF RADIOIODINE IN PRECIPITATION

The initial retention by vegetation of radioiodine contained in precipitation has an important bearing on the importance of wet deposition on the air to human food transport process. Equations (4) and (7) both show the importance of the retention fraction (a). Retention of radioiodine in rainwater is the principal concern because the growing (and grazing) season is the most important for transport of radioiodine into human foods.

The initial retention on vegetation of radioiodine in rainwater should be related to the interception and retention of rain by vegetation. Horton[76] suggested that the fraction of the rainwater retained by vegetation was proportional to the height of vegetation and calculated that between 5 and 8 percent of the rainfall was retained by vegetation that covers the ground completely. An alternative approach was used in Reference 26; the deposition rate was assumed to be proportional to the areal density of the vegetation.

Nearly all of the measurements of the retention of wet deposition of radioiodine have been made for ^{131}I in fallout from nuclear weapons testing. As noted in Section 1, such results principally represent transport of particulate debris containing ^{131}I and other fission products. The fractional retention by vegetation of fallout ^{131}I carried to the ground by rain was found to be 0.5 by both Chamberlain and Chadwick in England[77] and Hull at Brookhaven National Laboratory.[78] Bergstrom and Gyllander[79] found that in light rain 30% of fallout ^{131}I was retained by vegetation, while in heavy rains the retained activity was 10-20% of the total wet deposition. Fractional retention of fallout ^{131}I in wet deposition was found to range from 0.2 to 0.7 at three midwestern locations in the United States.[1,3] The variability of these retention factors may be partially due to unquantified variations in simultaneous dry deposition on the vegetation.

Pelletier and Voilleque assumed that the wet deposition rate (onto vegetation) of old fallout particulates was proportional to areal grass density and determined the fraction of particulate fallout in rain that was intercepted

and retained by the vegetation. The values of a $(=k_r Y)$ ranged from 0.4 to 1.2 m^2/kg. The average value, 0.95 m^2/kg, corresponded to a mean retention fraction (k_r) of 0.086 for the average grass density of $Y = 0.09 kg/m^2$. An alternative calculation showed that a retention fraction of 0.059 was consistent with the data.[26] Both values are similar to Horton's estimates for retention of rainfall and support the view that the initial retention of particulates in wet deposition reflects the retention of the rainfall by vegetation. The differences between these results and the higher retention fractions given above are probably partly due to changes in the particle size distribution between fresh and old fallout. Iodine-131 was not present in the old fallout debris, so the retention factors in Reference 26 do not include any effect of iodine solubility in the rainwater or iodine binding on plant surfaces.

Specific retention factors for I_2, HOI, or organic iodides scavenged by precipitation have not been measured. Field monitoring data[3] have yielded circumstantial evidence that ^{131}I in organic form was retained only briefly following wet deposition on vegetation. It was hypothesized that the deposition occurred during dew formation at night and that volatilization of the activity occurred when the dew evaporated in late morning. Further study of both the deposition and retention aspects of the transfer of radioiodine to vegetation by wet processes is needed.

10. IMPORTANCE OF WET DEPOSITION OF RADIOIODINE RELEASED DURING NORMAL OPERATIONS

The relative importance of wet deposition processes acting on radioiodines in gaseous effluents during normal operations can be assessed by comparing the contributions of wet and dry deposition processes to the doses received by the nearby population. The most significant exposure route is the air-vegetation-milk pathway and the initial air-to-vegetation transport rates serve as the basis for the comparison.

The contributions of each effluent species to dry deposition can be estimated using deposition velocities from Reference.[50] It is recognized that there is substantial variation in the deposition velocities that have been measured in individual tests, as well as variations due to different experimental field conditions.[80] As was mentioned earlier, the use of a single mean or median transport parameter masks these variations and the complex physical processes that produce the observable relationships. However, until we are capable of modeling the complexities, the rate of dry deposition of activity on vegetation will be estimated using:

$$D(x,y) = f_1 Q \left(\frac{X}{Q}\right) V_{d1} + f_2 Q \left(\frac{X}{Q}\right) V_{d2} + f_3 Q \left(\frac{X}{Q}\right) V_{d3} + f_4 Q \left(\frac{X}{Q}\right) V_{d4} \tag{20}$$

$D(x,y)$ = the deposition rate ($\mu Ci/m^2$-s) at location (x,y)

f_i = the fraction of the total release that is species i

Q = the total release rate ($\mu Ci/s$) of radioiodine, either ^{131}I of ^{129}I

(X/Q) = the average atmospheric dispersion factor (s/m^3) for location (x,y) during the growing season

V_{di} = the dry deposition velocity for the ith species (m/s)

i   =   the subscript used to distinguish among the radioiodine species; the correspondence between subscript and species is: i=1, radioiodine associated with particulates; i=2, elemental iodine (I_2); i=3, hypoiodous acid (HOI); i=4, organic iodides (CH_3I and others).

For dairy farm pastures near (within ~ 2 km of) the facility, the differences in dispersion factors for the four species are not significant[81], so the single value of (χ/Q) in Equation (20) is an adequate approximation. The values of f_i for reactors were given in Table 3. Nominal values of measured dry deposition velocities for radioiodine species to vegetation are 1×10^{-2}, 1×10^{-23}, 5×10^{-4}, and 1×10^{-5} m/s for i=1, 2, 3, and 4, respectively.[50] We rewrite Equation (20) as

$$D (x,y) = (\Sigma f_i V_{di}) \, Q \, (\frac{\chi}{Q}) = k_d \, Q \, (\chi/Q) \qquad (21)$$

and find, by evaluating the sum, that the species weighted deposition velocity (k_d) is 3.8×10^{-3} m/s for a typical BWR and 2.9×10^{-3} ms for a typical PWR. For a fuel reprocessing plant, it is estimated to be 2.1×10^{-3} m/s. The best value for a radiopharmaceutical facility is more uncertain, but available data suggest 3.2×10^{-3} m/s.

The total wet deposition of the radioiodine in effluents carried to the same location is given by

$$\Omega (x,y) = Q \, (\frac{\chi}{Q}) \, \sum f_i a_i V_{wi} \qquad (22)$$

in which the V_{wi} (m/s) are the wet deposition velocities for the four species and the a_i are the corresponding (dimensionless) factors that reflect the initial retention of the wet deposited species by vegetation. If the wet deposition velocity is approximated by the product of the dimensionless washout ratio and the mean rainfall rate, then

$$\Omega (x,y) = Q \, (\frac{\chi}{Q}) \, p_g \, \sum f_i a_i w_i \qquad (23)$$

where p_g is the average rainfall rate (m/s) during the growing season. (Both Equations (22) and (23) assume that the mean dispersion factor during periods of rainfall does not differ from the mean for the entire growing season.)

It was pointed out in Sections 8 and 9 that measured values of W_i and a_i are not available for all the radioiodine species of interest. However, to make the desired comparison, some preliminary estimates of a_i and W_i have been presented in Table 5.

TABLE 5. PROVISIONAL ESTIMATES OF WASHOUT RATIOS AND
RETENTION FACTORS FOR RADIOIODINE SPECIES

Chemical Form	Subscript (i)	Provisional Estimates	
		Washout Ratio (W_i)	Initial Retention Factor (a_i)
Particulates	1	2×10^5	0.1
I_2	2	10^4	0.5
HOI	3	6×10^2	0.3
Organic Iodides	4	5	0.1

Using the data in Table 3 and the provisional values in Table 5, a provisional species weighted wet deposition velocity for the growing season (kw, m/s) can be estimated using

$$k_w = p_g \ \Sigma \ f_i a_i W_i \tag{24}$$

Figure 3 shows the provisional values of k_w as a function of the average growing season rainfall rate. These may be compared with the values of k_d given above. Figure 3 suggests that the rainfall rates at which wet deposition becomes an important fraction of the total deposition onto vegetation substantially exceed those expected at any U.S. reactor site. This conclusion can
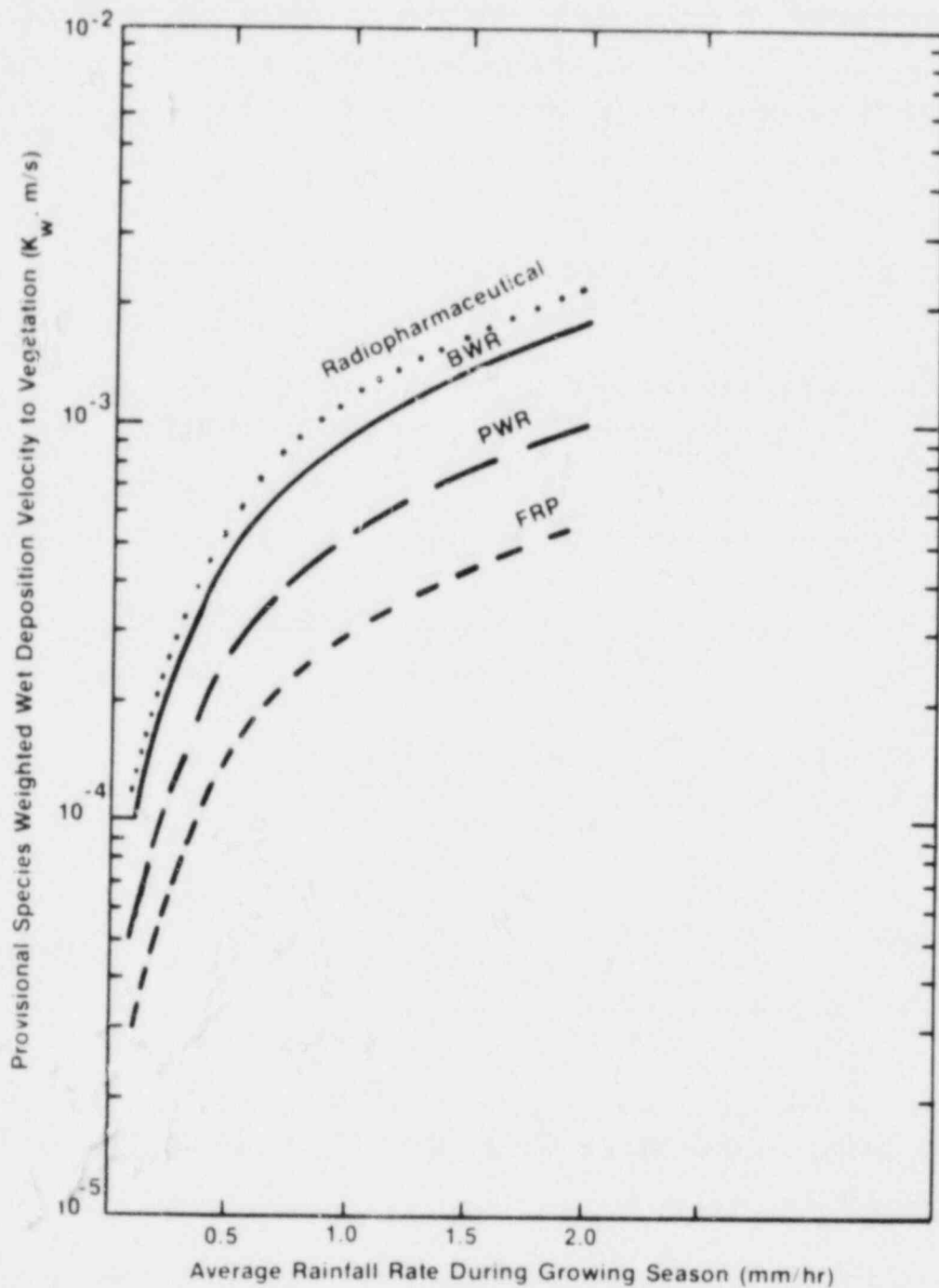
Figure 3. Preliminary Estimate of the Dependence of Species-Weighted
Wet Deposition Velocities on Rainfall Rate

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only be tentative because of (a) possible differences between dispersion factors for rainy periods and the entire growing season, and (b) the very provisional nature of the values of W_i and a_i used to derive the wet deposition curves.

The provisional estimates that suggest wet transport processes are insignificant are not consistent with observations[1,3] that significant increases in the concentration of ^{131}I in milk were correlated with precipitation. In some cases these increases may be discounted as resulting from scavenging of ^{131}I from fallout; however, that explanation does not apply uniformly. Thus, we must seriously question the applicability of the provisional estimates of W_i and a_i given in Table 5. Resolution of this question requires additional measurements.

11. IMPORTANCE OF WET DEPOSITION OF RADIOIODINE
RELEASED DURING REACTOR ACCIDENTS

Wet deposition of radioiodines and other radionuclides is an important process affecting the calculated consequences of reactor accidents. The chemical forms of the radioiodine released during such accidents have an important bearing on the estimated ground contamination and plume depletion. Information about the magnitudes of accidental releases and the species distributions expected for accidents is discussed below.

11.1 Calculated Releases Following Reactor Accidents

The Nuclear Regulatory Commission has performed a detailed evaluation of the likelihood and probable consequences of a large spectrum of reactor accidents.[63] An important facet of the consequence analysis is the behavior of radioiodines released from the fuel into the containment and other buildings and subsequently into the environment. The estimated release fractions for noble gases, radioiodines and radiocesiums for a wide spectrum of accidents considered in the Reactor Safety Study[63] are shown in Table 6. The postulated fractions of the core inventory released to the environment are quite large for some release categories (a category is representative of several possible accident sequences). The probabilities of such accidents are very small (see column two of the table) but the consequences are very large, and these accidents dominate the overall risk from nuclear power plant operation.[63]

The importance of radioiodines to the calculated consequences of such accidents has been evaluated by sensitivity analysis techniques[82]. It was found that 50% of the projected early radiation fatalities, 55% of the projected early radiation illness, and about 5% of the latent cancer deaths predicted were due to exposure to radioiodines. The predicted early injuires and fatalaties result from whole body exposure to the plume and to activity deposited on the ground by wet and dry processes in the vicinity of the plant.

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TABLE 6. ESTIMATED RELEASE FRACTIONS FOR NOBLE GASES, RADIOIODINES, AND RADIOCESIUMS FOR REACTOR ACCIDENTS[a]

Release Category for Core Accidents	Probability of Occurrence (per reactor-year)	Fraction of Core Inventory Released to Environment			
		Xe-Kr	Organic Iodine	Inorganic Iodine	Cs-Rb
PWR 1	9×10^{-7}	0.9	6×10^{-3}	0.7	0.4
PWR 2	8×10^{-6}	0.9	7×10^{-3}	0.7	0.5
PWR 3	4×10^{-6}	0.8	6×10^{-3}	0.2	0.2
PWR 4	5×10^{-7}	0.6	2×10^{-3}	0.09	0.04
PWR 5	7×10^{-7}	0.3	2×10^{-3}	0.03	9×10^{-3}
PWR 6	6×10^{-6}	0.3	2×10^{-3}	8×10^{-4}	8×10^{-4}
PWR 7	4×10^{-5}	6×10^{-3}	2×10^{-5}	2×10^{-5}	1×10^{-5}
PWR 8	4×10^{-5}	2×10^{-3}	5×10^{-6}	1×10^{-4}	5×10^{-4}
PWR 9	4×10^{-4}	3×10^{-6}	7×10^{-9}	1×10^{-7}	6×10^{-7}
BWR 1	1×10^{-6}	1.0	7×10^{-3}	0.4	0.4
BWR 2	6×10^{-6}	1.0	7×10^{-3}	0.9	0.5
BWR 3	2×10^{-5}	1.0	7×10^{-3}	0.1	0.1
BWR 4	2×10^{-6}	0.6	7×10^{-4}	8×10^{-4}	5×10^{-3}
BWR 5	1×10^{-4}	5×10^{-4}	2×10^{-9}	6×10^{-11}	4×10^{-9}

Other Accidents

Loss of Cooling in SFSP	$<10^{-6}$	10^{-1}	7×10^{-4}	7×10^{-4}	10^{-3}
Dropped Shipped Cask	6×10^{-7}	3×10^{-7}	10^{-11}	10^{-11}	1×10^{-7}
Refueling Accident	10^{-3}	9×10^{-5}	4×10^{-7}	4×10^{-7}	6×10^{-9}
Waste Gas Storage Tank Rupture	10^{-2}	2×10^{-4}	10^{-9}	10^{-9}	~ 0
Liquid Waste Storage Tank Rupture	10^{-2}	~ 0	8×10^{-8}	8×10^{-8}	6×10^{-8}
Earthquake-Induced Loss of Cooling in SFSP With Loss of Air Cooling System	3×10^{-8}	10^{-1}	7×10^{-2}	7×10^{-2}	10^{-1}

[a] From Reference 63.

The fourth and fifth columns of Table 6 show predicted organic/inorganic species distributions for radioiodines for each of the release categories and for the accidents involving spent fuel outside of the reactor core. The radioiodine species distribution is very important to the assessment of consequences because it affects (a) the expected wet and dry deposition of released radioiodine, (b) the effectiveness of containment spray systems for reducing airborne activity prior to a release, (c) the amount of radioiodine deposited on containment surfaces, and (d) the effectiveness of charcoal filtration systems used to cleanse air prior to release from the facility. The predicted species distributions in Table 6 are based on the work of Postma and Zavodoski[83]; the procedure used to estimate the fraction of the airborne iodine converted to organic form is discussed in Appendix VII of Reference 63. The total conversion of iodine to organic forms for large releases ranges from 0.4 to 0.7%. In Reference 83, the non-radiolytic conversion to organic iodides was related to airborne iodine concentration by

$$f = a \, C^b \tag{25}$$

in which

f $=$ the fraction of airborne iodine converted to organic form by non-radiolytic processes

C $=$ the initial concentration of airborne iodine (mg/m^3)

a,b $=$ constants derived from a least squares fit to data from 69 experiments with iodine concentrations ranging from 10^{-5} to $\sim 200 \, mg/m^3$. The best fit constants were $a=1.88 \times 10^{-3}$ and $b=-0.26$.

Equation (25) predicts conversion fractions (f) greater than 0.02 only for total iodine airborne concentrations of $< 10^{-4} \, mg/m^3$ ($<100 \, ng/m^3$); that is, only for those accidents that involve small release of radioiodine. For large releases, the initial airborne concentration is expected to be $\sim 100 \, mg/m^3$ and Equation (25) yeilds $f=5.7 \times 10^{-4}$, or $\sim 0.06\%$.

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Following the accident at Three Mile Island, Unit 2 (TMI-2), which resulted in low doses to individuals and to the local population, questions were raised by several groups about the validity of the models employed to predict the consequences of reactor accidents.[84-87] One of the notable features of the TMI-2 accident was the small release of radioiodine (discussed in Section 11.2). This led to the proposal[89] that the predominant iodine species release from damaged fuel would be cesium iodide (CsI). Because CsI is very soluble in water and is less volatile than I_2 (the presumed species), radioiodine releases would be far lower than those predicted in Table 6. Levenson and Rahn[85] also argued that the predictive models failed to reflect natural removal processes that would reduce the predicted release fractions. The questions raised in References 84-87 lead to a reevaluation of the methods used by the NRC to predict consequences of accidents.[64,82]

11.2 Observed Releases Following the Accident at TMI-2

As was mentioned above, the generally low release fractions of radioiodines that were observed in various reactor accidents and experimental tests were cited as evidence that the assumptions used in accident analyses were incorrect. Measurements made during and after the TMI-2 accident are discussed in some detail below because they are a principal source of information on radioiodine behavior under accident conditions at an operating power reactor. Even though the available informaton is incomplete, the data are unique and important.

During the accident at TMI-2 the amount of ^{131}I released to the environment was ∼20 Ci,[88,89] approximately 3×10^{-5}% of the core inventory at shutdown (6.9×10^7 Ci). Approximately 6.5% of the ^{133}Xe inventory was released to the environment during the same time period. Nearly all the noble gas inventory was released from the fuel, while only about 40% of the ^{131}I inventory could be accounted for using measurements of ^{131}I in the sump, in various tanks, and in other fluids.[90,91] The largest amount of ^{131}I measured in the containment atmosphere was ∼6000 Ci, based on the air concentration of 0.11 μ Ci/cm^3 measured on the sixth day. It must be noted that (a) the April 3rd sample was the first containment air sample taken, (b) the containment sprays

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had operated prior to the sample (about 10 hours after the initiating event), and (c) there was ample time for deposition of ^{131}I on containment surfaces prior to the sample, so the maximum amount of airborne iodine was definitely greater than 6000 Ci. As indicated, the overall radioiodine activity balance for TMI-2 is not yet complete. The ⁓60% of the activity that was not accounted for may be (a) in the fuel, (b) deposited on surfaces in the reactor vessel, (c) in solids in the basement of the containment building, or (d) deposited on containment building surfaces. The actual distribution of radioiodine in these locations will not be known until additional samples collected during the recovery phase have been analyzed for ^{129}I.

An important factor that kept releases from TMI-2 lower than those predicted in various accident analyses was the pressure inside the containment building. Following the hydrogen deflagration on the first day, the pressure within the containment vessel was less than atmospheric, so there was no driving force for fission product releases from the containment. As noted earlier, many of the low probability, high consequence accident scenarios postulate containment breach and radioiodine release fractions of 70% of the core inventory. That was certainly not the case at TMI-2.

The radioiodine releases that did occur were via the ventilation air discharged through filters from the Fuel Handling and Auxiliary Buildings. The principal release mechanisms were volatilization and resuspension of activity in areas that were severely contaminated early in the accident. The building ventilation systems had been turned off for extended periods of time during the first day.[38]

Airborne ^{131}I species samples were first obtained from the TMI-2 containment in late June 1979.[92] The results of two samples on 26 June and 9 July were very similar. The average species fractionation was 0.04% associated with particulates, 8.5% I_2, 9.4% HOI, and 82% organic iodides. Airborne ^{129}I concentrations were measured prior to the start of the purge of the TMI-2 containment in July 1980, but the species distribution was uncertain because of the small amounts of ^{129}I collected.[93] Shortly after the start of the purge, longer sampling times permitted better definition of the gaseous species distribution.[94]

At that time the distribution fractions were 2.1% I_2, 19% HOI, and 79% organic iodides. Following the purge, the remaining gaseous ^{129}I reached an equilibrium distribution of 2.4% I_2, 7.6% HOI, and 90% organic iodides.[94] The observed equilibrium ^{131}I and ^{129}I species distributions at TMI-2 are similar to one another and to those observed in the seale containments of PWRs during normal operation.[34-36]

11.3 Reconsideration of Accident Consequence Calculational Techniques

The U.S. Nuclear Regulatory Commission has published the results of their inquiry into the questions raised about methods of calculating consequences of reactor accidents.[64,82] It was concluded that (a) the current data base supports the view that CsI may be the predominant iodine from under accident conditions, (b) the presence of radioiodine as CsI (rather than as I_2) did not significantly change the predicted consequences for the low probability accidents that dominate the overall risk, but (c) the predicted consequences of less severe accidents would be lower if the radioiodine were CsI rather than I_2. It was also concluded that the conversion fraction from I_2 to organic iodides given in Reference 83 (see Equation (25)) was too high and would not exceed 0.1% under accident conditions.[64]

Thus, the conclusion of the Reactor Safety Study[63] and the recent reevaluation of calculation models[64] would be that radioiodines released from the improbable but severe accidents would be released as I_2 or would be associated with particulate material. The relatively large contribution of radioiodines to the predicted consequences of these risk dominant accidents would be comparable in either case. For other (more probable) accident sequences, a larger fraction of the released activity would be in organic form. The type and magnitude of the accident will therefore affect the amount of precipitation scavenging of the released activity. For risk dominant accidents that occur during storms, precipitation scavenging will be an important contributor to the early consequences of the accidents.

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16. ABSTRACT (200 words or less)

The results of a review of wet deposition processes for radioiodines are presented. Experimental measurements have shown that precipitation scavenging can be an important process for removal of radioiodines released to the atmosphere. Measurements of radioiodine species in routine gaseous effluents have shown that the distribution depends upon the type of nuclear facility considered. Because of differences in radioiodine species, observed scavenging rates for radioiodine in fallout are not generally applicable to routine releases from nuclear facilities. Adequate models are available to estimate the air to surface transport by wet processes. At the present time, the need is for more information on the rate constants used in the models. Reactor accident consequence calculations that assume no behavior differences among iodine species indicate that about half the predicted early deaths and illnesses are attributable to radioiodines. The distribution of species released under accident conditions will depend upon the actual sequence of events. However, for many scenarios the wet deposition of radioiodines will have an important bearing on the consequences.

17. KEY WORDS AND DOCUMENT ANALYSIS   17a. DESCRIPTORS

1. Radioiodine
2. Deposition
3. Precipitation

17b. IDENTIFIERS/OPEN-ENDED TERMS

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