



Department of Energy

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OCT 26 1998

Frank J. Congel, Director
Incident Response Division
Office for Analysis and Evaluation
of Operational Data
U.S. Nuclear Regulatory Commission
11545 Rockville Pike
Rockville, Maryland 20852-2738

REVIEW OF RADIOIODINES

In your August letter (enclosed), you requested that the Federal Radiological Monitoring and Assessment Center (FRMAC) Monitoring Working Group address the issue of potential underestimation of radioiodines released by a nuclear power plant accident, and impacts this might have on public health protection and worker turn-back guidance. Please find enclosed a letter from the Radiation and Indoor Environments National Laboratory of the U.S. Environmental Protection Agency (EPA), summarizing the review of this topic by FRMAC scientific staff. This review includes input from Dr. Harvey Clark, of Bechtel Nevada, FRMAC Assessment Working Group Chairman, and Mike Smith and Jim Benetti of EPA, of the Monitoring Working Group. Input was also received from Dr. Robert Bores of Nuclear Regulatory Commission Region I. The reviewers concur that there is some potential for underestimating total radioiodine utilizing current sampling methods, and the FRMAC monitoring and assessment manuals will be revised to compensate for this potential for underestimation. This will also be an agenda topic for the next Monitoring and/or joint Monitoring/Assessment Working Groups Meeting, and the outcome of subsequent discussions will be forwarded to you.

Should you have any questions regarding this topic, please contact me at (702) 295-1299, or by FAX at (702) 295-2383.

Donald R. Elle, Director
Emergency Management Division

EMD:BWH-99092
EMT 4-5

Enclosure:
As stated

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UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

RADIATION AND INDOOR ENVIRONMENTS NATIONAL LABORATORY

P.O. BOX 98517

LAS VEGAS, NEVADA 89193-8517

October 22, 1998

Bruce Hurley
Federal Radiological Monitoring
and Assessment Center
U.S. Department of Energy
P.O. Box 98518
Las Vegas, Nevada 89193

Dear Dr. Hurley,

This letter addresses your recent request of Jerry Martin, FRMAC Monitoring and Analysis Working Group chair to respond to issues raised by Mr. David Minnaar, Chief of the Radiological Protection Section of the State of Michigan, and amplified by Frank J. Congel of the U.S. Nuclear Regulatory Commission.

The paper which Mr. Minnaar has attached suggests that the fraction of radioiodine which may change form during transport in the event of an accident involving a radioiodine release may be significant. The primary issue raised by Mr. Minnaar, is that there is a potential for underestimation of radioiodine concentration in air, if it is assumed that the majority of radioiodine is released and transported in the elemental state. The underestimation results from analyzing only the silver zeolite cartridge, presuming that all iodine activity is found there.

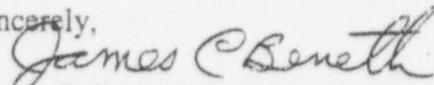
To address the monitoring aspects of this issue, I have requested a change to the FRMAC Monitoring and Assessment Manual, which clarifies that the laboratory analysis of air samples for radioiodine and other isotopes should be conducted on both the silver zeolite (or charcoal) cartridge and its associated particulate prefilter and that laboratory results should be displayed so as to enable a clear identification of the total iodine present in the complete air sample. This change merely clarifies the FRMAC procedure which is currently followed. This approach is in widespread use within the nuclear power industry in in-plant and environmental monitoring for iodine (both routine and unplanned releases). Please note that this is not currently a field measurement technique, as it requires laboratory gamma spectroscopy to be conducted. To improve the timeliness of such measurements, I am recommending that field teams monitor the activity of both the filter and cartridge and report above background values of either, as part of their sampling procedure. Assessment may then prioritize the collection and analysis of the samples if desirable (sample courier, priority analysis, etc.).

In addition, I am reviewing the literature and obtaining information on radioiodine species sampling media and cartridge holders for use with our existing air sampling equipment. Although it is not in common use, there appears to be a method to discriminate between elemental and organic iodine, which employs a cascade of several cartridges using carbon impregnated with various compounds, and having differing retention efficiencies for iodine compounds. From initial discussions with individuals having experience in this area, I believe that we should be able to add this capability to our current field monitoring methodology at nominal cost, if it is desirable. However, the Assessment Group needs to deliberate on this, since this additional data may not be usable in predictive models, and since current predictive methods may be adequately conservative to obviate the need of species discrimination. Harvey Clark informs me that sufficient conservatism could be incorporated into ARAC predictions if speciation were suspected by simply turning off the feature which depletes the plume due to deposition, thereby bounding the iodine population dose.

To summarize: the current air sampling methodology used by FRMAC, the States, and the utilities, are capable of collecting better than 95% of airborne radioiodines on either the particulate filter or iodine cartridge. If both the filter and cartridge are analyzed and considered in the assessment of individual or population dose, radioiodine will not be underestimated, regardless of its chemical form. Gamma spectral analysis is necessary to accurately quantify radioiodine present on the sampling media in the presence of interferences, so the emphasis needs to be placed on timeliness of getting media with significant activity to the laboratory.

I will plan to add this subject to our next Monitoring Working Group and/or joint Monitoring/Assessment Group meeting, and will report to you or to your successor. Please contact me if I can further assist you.

Sincerely,



James C. Benetti, Health Physicist
U. S. Environmental Protection Agency



UNITED STATES
NUCLEAR REGULATORY COMMISSION

WASHINGTON, D.C. 20555-0001

Dr. Donald Elle, Director
Federal Radiological Monitoring
and Assessment Center
U.S. Department of Energy
P.O. Box 98518
Las Vegas, Nevada 89193

Dear Dr. Elle:

This is to request your assistance in addressing the measurement of radioiodines in the environment following an accidental release of radioactive material from a nuclear power plant. Radioiodines released from a reactor accident may undergo a chemical transformation while airborne and after deposition. The chemical transformation of radioiodines may affect the accuracy of field measurements.

In his letter to the U.S. Nuclear Regulatory Commission dated July 7, 1998, Mr. David W. Minnaar, Chief of Radiological Protection Section in the State of Michigan, has expressed his concern regarding the potential underestimation of radioiodines using the current method of counting the activity contained in a silver-zeolite cartridge (copy enclosed). The States and licensees routinely use this method in emergency planning drills and exercises to demonstrate that they meet the FEMA Radiological Emergency Preparedness Exercise Manual (REP-14), Objective 8, Criterion 4, which states that "field teams should demonstrate as a minimum the capability to sample and measure airborne radioiodine as low as 10E-7 uCi/cc."

Ultimately the objectives are to determine if the emergency worker turn-back guidance (FRMAC Assessment Manual, Method M.2.2) should be revised or if additional protective actions are needed for the public (Method M.3.0). Therefore, the environmental monitoring methods must allow the iodine airborne concentration to be measured promptly, in the presence of noble gases and other fission products, and be related to the gamma dose rate. We believe that consistency between the Federal and State responders in environmental monitoring procedures is important. I am, therefore, requesting that the Federal Radiological Monitoring and Assessment Center (FRMAC) Monitoring working group address this issue and provide your response to us by October 30, 1998. We intend to provide our response to Mr. Minnaar using your assessment of the issue. We also intend to provide the results of your assessment to the Federal Radiological Preparedness Coordinating Committee (FRPCC) for distribution to other States.

If you have any questions, please contact Mr. Aby Mohseni of my staff at (301) 415-6409.

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| INFO | | _____ |
| MGR | <input checked="" type="checkbox"/> | _____ |
| AMBFS | | _____ |
| AMTS | | _____ |
| AMNS | <input checked="" type="checkbox"/> | _____ |
| AMEM | | _____ |

Sincerely,
Frank J. Congel
Frank J. Congel, Director
Incident Response Division
Office for Analysis and Evaluation
of Operational Data

Enclosure: As stated

cc w/encl: See next page

98-08260244 2 pp

D. Elle

- 2 -

cc w/encl:

David W. Minnaar, Chief
Radiological Protection Section
Drinking Water and Radiological
Protection Division
Department of Environmental Quality
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Lansing, MI 48909

Russell Salter
Federal Emergency Management Agency
500 C Street, S.W.
Washington, D.C. 20472



Sent 7/7/98

JOHN ENGLER, Governor
DEPARTMENT OF ENVIRONMENTAL QUALITY

"Better Service for a Better Environment"
HOLLISTER BUILDING, PO BOX 30473, LANSING MI 48909-7873

INTERNET: www.dec.state.mi.us
RUSSELL J. HARDING, Director

REPLY TO

DRINKING WATER & RADIOLOGICAL
PROTECTION DIVISION
3423 N MARTIN L KING JR BLVD
PO BOX 30630
LANSING MI 48909-8130

July 7, 1998

Mr. Timothy Martin, Director
Office of Analysis and Evaluation
of Operational Data
U.S. Nuclear Regulatory Commission
11545 Rockville Pike
Rockville, Maryland 20852-2738

Dear Mr. Martin:

The purpose of this letter is to request technical assistance from your office for an improved understanding of the behavior of airborne radioiodines potentially released from U.S. commercial nuclear power reactors during a severe reactor accident, including the associated dose consequences to impacted members of the public. After a preliminary discussion of this issue with Mr. Roland Lickus, State Liaison Officer, U.S. Nuclear Regulatory Commission (NRC), Region III, Mr. Lickus recommended we direct our concerns to your office.

Existing guidance for off-site field measurement of airborne radioiodines (FEMA REP-2, REV 2/June 1990) suggests the use of an air sampler equipped with a particulate prefilter and an efficient radioiodine absorber, such as a silver-zeolite cartridge. The guidance presumes that airborne radioiodine concentrations can be characterized by presuming essentially total representation as inorganic iodine vapors, which would then be collected by and measured from the silver-zeolite cartridge.

We are aware, however, that evidence exists that radioiodines (once released into an airborne plume) may undergo chemical form changes during atmospheric transport such that radioiodines will likely be found in the form of particulates and as iodine vapor (both organic and inorganic forms) after some nominal transport time, regardless of the chemical form at the point of release (see enclosure). Obviously, this would lead to a potential for only partial detection of total inhalable radioiodines when field measurements for radioiodines are confined to a silver-zeolite cartridge reading only. Radioiodine dose assessments based on field measurements could then be significantly underestimated.

In order to be prudently conservative in promptly estimating off-site radioiodine dose consequences from field measurements during a nuclear reactor accident, we believe that the potential for chemical form partitioning needs to be resolved so that public thyroid doses are not significantly underestimated as may occur when following the existing FEMA guidance. If NRC can provide an assessment of this issue and recommend a technically sound field airborne radioiodine sampling and analysis procedure, it would greatly benefit our emergency response program for off-site nuclear plant accident dose assessment.

Your prompt attention to this request will be greatly appreciated.

Sincerely,

David W. Minnaar, Chief
Radiological Protection Section

DWM:RT
Enclosure

cc: Mr. Roland Lickus, NRC, Region III
Mr. Daniel Sibo, MDSP/EMD
cc/enc: Mr. Dennis Hahn, MDEQ/RPS

ENCLOSURE

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ATMOSPHERIC DISPERSION AND DEPOSITION OF ^{131}I RELEASED FROM THE HANFORD SITE

J. V. Ramsdell, Jr., C. A. Simonen, K. W. Burk, and S. A. Stage*

Abstract—Approximately 2.6×10^4 TBq (700,000 Ci) of ^{131}I were released to the air from reactor fuel processing plants on the Hanford Site in southcentral Washington State from December 1944 through December 1949. The Hanford Environmental Dose Reconstruction Project developed a suite of codes to estimate the doses that might have resulted from these releases. The Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET) computer code is part of this suite. The RATCHET code implements a Lagrangian-trajectory, Gaussian-puff dispersion model that uses hourly meteorological and release rate data to estimate daily time-integrated air concentrations and surface contamination for use in dose estimates. In this model, iodine is treated as a mixture of three species (inorganic gases, organic gases, and particles). Model deposition parameters are functions of the mixture and meteorological conditions. A resistance model is used to calculate dry deposition velocities. Equilibrium between concentrations in the precipitation and the air near the ground is assumed in calculating wet deposition of gases, and irreversible washout of the particles is assumed. RATCHET explicitly treats the uncertainties in model parameters and meteorological conditions. Uncertainties in ^{131}I release rates and partitioning among the nominal species are treated by varying model input. The results of 100 model runs for December 1944 through December 1949 indicate that monthly average air concentrations and deposition have uncertainties ranging from a factor of two near the center of the time-integrated plume to more than an order of magnitude near the edge. These results indicate that ~10% of the ^{131}I released to the atmosphere decayed during transit in the study area, ~56% was deposited within the study area, and the remaining 34% was transported out of the study area while still in the air. *Health Phys.* 71(4):568–577; 1996

Key words: ^{131}I ; dose assessment; radioactivity, airborne; emissions, atmospheric

INTRODUCTION

EARLY STUDIES in the Hanford Environmental Dose Reconstruction (HEDR) Project (Ramsdell and Burk 1991a, b) showed that ^{131}I releases from the B and T fuel processing plants at the Hanford Site were of sufficient

magnitude that there was a large area in the vicinity of Hanford where doses could be of concern. As a result, the study area shown in Fig. 1 was selected for atmospheric dispersion and deposition modeling. This rectangular area is centered at $46^{\circ}40'\text{N}$, $118^{\circ}45'\text{W}$ and extends ~500 km from north to south and 400 km from east to west. Geographically, the study area extends from central Oregon to northern Washington and from the crest of the Cascade Mountains to the eastern edge of northern Idaho. The study area is bounded by the major topographic features of the region and is situated with more of the area on the downwind side of the releases, given the prevailing wind direction.

Rattlesnake Mountain is the most notable topographic feature in the immediate vicinity of the Hanford Site. It is a treeless ridge that runs along the southwest boundary of the Hanford Site and has an elevation that exceeds 1,050 m. The southwestern slope of Rattlesnake Mountain is gentle, but the northeast face of Rattlesnake Mountain is extremely steep. The elevation of the Hanford Site in the vicinity of the reactor fuel processing plants is about 225 m. There is a broad valley between Rattlesnake Mountain and the processing plants. To the north and east of the processing plants, the terrain slopes toward the Columbia River. Light nighttime drainage winds may cause plumes to drift either to the west toward Rattlesnake Mountain or to the east toward the Columbia River.

The study area is sufficiently large that systematic variations in meteorological and climatological conditions are found across the area. Wind roses are a graphical means of showing the climatological distribution of wind directions at a location. The wind roses in Fig. 2 show the effects of Rattlesnake Mountain, the channeling of flow by the mountain valleys along the western edge of the study area, and the prevailing southwest winds over most of the Mid-Columbia Basin (see Pasco, LaCrosse, and Harrington in Fig. 2) and the Spokane area (see Fairchild in Fig. 2). Annual precipitation varies from a low of ~16 cm y^{-1} near Hanford to more than 250 cm y^{-1} in the Cascade Mountains on the western edge of the study area. The annual precipitation along the eastern edge of the study area is generally more than 50 cm y^{-1} .

The size of the study area, the variations in meteorological conditions, the length of time being modeled, and the desire to model uncertainty led to the selection of

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(Manuscript received 17 August 1994; revised manuscript received 6 December 1995, accepted 5 June 1996)

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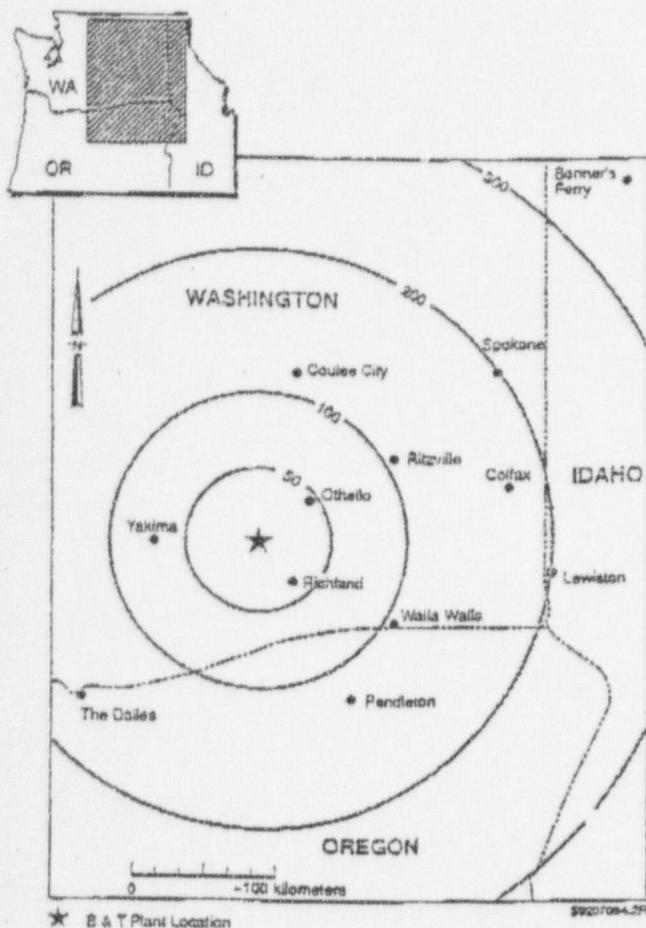


Fig. 1. Hanford Environmental Dose Reconstruction Project study area.

a Lagrangian-trajectory, Gaussian-puff dispersion model as the appropriate approach for the HEDR Project (Ramsdell 1991, 1992). Consequently, the Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET) (Ramsdell et al. 1994) computer code was developed to estimate atmospheric dispersion and deposition of ^{131}I . This paper describes RATCHET and some of the results from the modeling activities.

TECHNICAL APPROACH

Features of the RATCHET computer code are shown in Table 1. Sequences of Gaussian puffs represent plumes released from ground-level and elevated sources. As the puffs move, 24-h time-integrated air concentrations and surface contamination are calculated at nodes by summing the contributions from puffs as they move past the nodes. Subsequent HEDR computational codes (Ikenberry et al. 1992) use the time-integrated air concentration and surface contamination at each node to calculate monthly, annual, and cumulative doses in the area surrounding the node. This use of point values as averages for small areas is reasonable because the integration times are relatively long and the points of interest

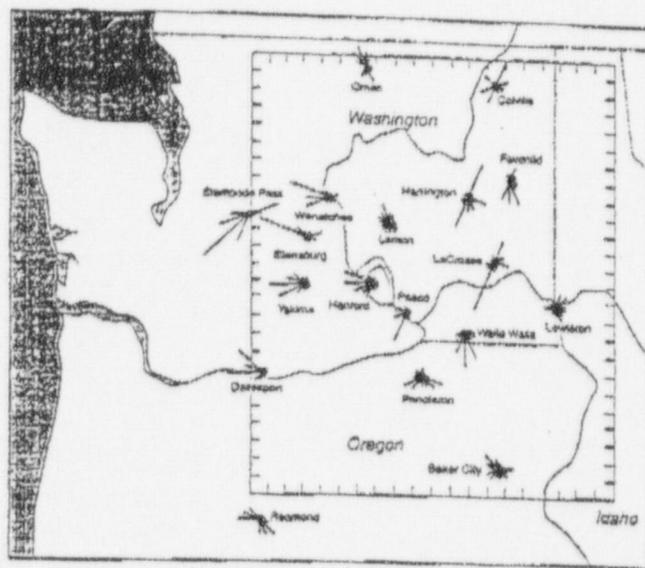


Fig. 2. Wind roses for the HEDR study area, December 1944 through December 1949. By convention, bars of a wind rose point in the directions from which the wind blows, and the lengths of the bars are proportional to the direction frequency.

are far enough from the release point that the spatial gradients of air concentration and surface are generally small.

Transport, diffusion, and deposition calculations are made using wind, atmospheric stability, precipitation, and mixing-layer depth fields that describe the spatial and temporal variations of meteorological conditions across the study area. These fields are prepared by RATCHET using hourly meteorological data from observation sites in and adjacent to the study area. The temperature at the Hanford Meteorological Station is used in the model, but only as a function of time. The spatial variation of temperature is not modeled because temperature is primarily used to calculate plume rise, which takes place in the immediate vicinity of the Hanford Meteorological Station. Stage et al. (1993) describe the meteorological data set used by RATCHET.

Although RATCHET is a general-purpose atmospheric dispersion code, on the basis of results of an analysis by Napier (1992), RATCHET was developed specifically to estimate the transport, diffusion, and deposition of iodine. Iodine is a special material that exists in three forms in the atmosphere: it is found in organic gases (e.g., CH_3I), in inorganic gases (e.g., I_2), and attached to aerosol particles. In RATCHET, ^{131}I is treated as a mixture of these three species with deposition characteristics that are a weighted average of the characteristics for each of the species. The weights assigned to each component are equal to the fraction of the total iodine in the component.

Burger (1991) states that the fuel processing operations at Hanford should have released ^{131}I in the elemental form. However, it is unlikely that the iodine remained in the elemental state for long. Experimental data on

Table 1. Features of the RATCHET computer code.

| Features | RATCHET |
|------------------------|--|
| Study area | ~200,000 km ² covering eastern Washington, northeastern Oregon, and northern Idaho |
| Node spacing | 9.65 km (6 mi) |
| Source term | 100 sequences of hourly release rates for the period from December 1944 through December 1949 |
| Meteorological data | Hourly data for 25 locations in and adjacent to the study area; data were not available for all stations for all times |
| Wind fields | 1/r ² interpolation of measured winds |
| Topographic effects | Major effects of topography are implicit in measured weather data; regional variation of surface roughness is treated explicitly |
| Wind profile | Diabatic wind profiles based on similarity theory |
| Stability | Spatially varying based on wind, cloudiness, and time of day—not discrete classes |
| Precipitation | 7 precipitation types, spatially varying precipitation fields, 3 precipitation regimes with different precipitation rate distributions for each regime |
| Mixing layer depth | Spatially varying based on meteorological conditions at the meteorological stations |
| Plume rise | Briggs' equations |
| Diffusion coefficients | Based on travel time and turbulence levels; computed in the code based on meteorological data and surface roughness |
| Dry deposition | Calculated using resistance model to incorporate effects of wind speed, surface roughness, atmospheric stability, and iodine species on dry deposition |
| Wet deposition | Reversible scavenging of gases and irreversible washout of particles determined as a function of iodine species and precipitation rate |
| Iodine representation | Partitioned between highly reactive gases (I ₂), slightly reactive gases (CH ₃ I), and particles |
| Uncertainty | Uncertainty in meteorological data treated in RATCHET calculations; uncertainty in source term and iodine partitioning treated as part of code input |
| Model time step | 15-s minimum, 15-min maximum |
| Output frequency | Daily |

iodine releases reported by Ludwick (1964) indicate that about two-thirds of the iodine changed form in the time required to travel 3.2 km (2 mi), about one-third was in organic species, and the remaining third was associated with particulate material. This partitioning of iodine is consistent with other results; e.g., in plumes from stacks at the Hanford Site (Ludwick 1967; Perkins 1963, 1964), in the plume following the Chernobyl reactor accident (Aoyama et al. 1986; BIOMOVs 1990; Bondiotti and Brantley 1986; Cambray et al. 1987; Mueck 1988), and in natural atmospheric iodine (Voilleque 1979). Consequently, the partitioning of iodine in RATCHET is independent of travel time.

Deposition rates are proportional to the concentration in the air near the surface. The proportionality constant is referred to as the deposition velocity. The three ¹³¹I forms have significantly different deposition characteristics. For example, Voilleque and Keller (1981) give typical deposition velocities for CH₃I, I₂, and particles as 0.00001, 0.01, and 0.001 m s⁻¹, respectively. Current-generation applied atmospheric dispersion models estimate the deposition velocity using an electrical systems analogy. In this analogy, described by Seinfeld (1986), the deposition process is assumed to be controlled by a network of resistances, and the deposition velocity is the inverse of the total resistance of the

network. Resistances are associated with 1) atmospheric conditions; 2) physical and chemical characteristics of the material; and 3) physical, chemical, and biological properties of the surface.

Following the resistance analogy, dry deposition velocities in RATCHET are equal to the reciprocal of the sum of the three component resistances: an aerodynamic resistance, a surface resistance, and a transfer resistance. The aerodynamic resistance is a function of windspeed, atmospheric stability, and surface roughness. The surface resistance is a function of wind speed and surface roughness. Transfer resistances are usually associated with the characteristics of the depositing material and surface type [e.g., Wesley and Hicks (1977) associated transfer resistance with stomatal openings in plants].

In RATCHET, the transfer resistance is used as a mathematical means of placing a lower limit on the total resistance (or equivalently an upper limit on deposition velocity). A limiting resistance is needed because both the aerodynamic and surface resistances decrease as wind speed increases. If the transfer resistance is not included in the total resistance, dry deposition velocities in neutral conditions increase from ~0.006 m s⁻¹ at a wind speed of 1 m s⁻¹ to greater than 0.06 m s⁻¹ at a wind speed of 10 m s⁻¹. Deposition velocities at the upper end of this range are higher than normally assumed

most reactive gases, and the entire range of deposition velocities is above that measured for fine particles (~ 1 m s $^{-1}$) and nonreactive gases (McMahon and Denison 1979; Sehmel 1980). Assuming transfer resistances of 10 s m $^{-1}$ for reactive gases and 100 s m $^{-1}$ for fine particles yields dry deposition velocities that are more consistent with reported values.

RATCHET treats wet deposition of gases and particles separately. Wet deposition of gases is modeled assuming equilibrium between gas concentrations in the air and precipitation. Wet deposition of particles is modeled using a washout coefficient, assuming irreversible collection of particles as the precipitation falls through the puffs.

Scavenging rates for gases are based on solubility, assuming equilibrium conditions between the gas concentration in the air near the ground and in the precipitation. With this assumption, the scavenging rate for gases is expressed as a wet deposition velocity (Slinn 1984). The solubility coefficients used to calculate the scavenging rate are inversely related to the Henry's Law constant for the gas being scavenged. Slinn (1984) provides guidance in their selection. A solubility coefficient of 500 is assumed for I_2 . This assumption results in the wet deposition velocities shown in Table 2 for the default precipitation rates in RATCHET. Scavenging of H_3I by precipitation is extremely limited and may be neglected as a practical matter. Wet deposition velocities for CH_3I computed in RATCHET are about three orders of magnitude lower than those for I_2 . Scavenging of both H_3I and CH_3I by snow when the temperature is less than 0°C is low because the snow surface is frozen rather than wet and is ignored by RATCHET.

The wet deposition model for particles assumes that precipitation falls through the full vertical extent of the puffs and collects particles by collision. The scavenging rate for particles is expressed as a washout coefficient (the fraction of airborne material removed by precipitation each hour). RATCHET uses an expression in which the washout coefficient is proportional to the precipitation rate to the three-fourth's power to estimate the wet deposition of particles by rain (Slinn 1984). Table 2 shows particle washout coefficients for the default rainfall rates in RATCHET. During periods of snow, the washout coefficient is directly related to the precipitation rate.

Given dry and wet deposition velocities, the surface contamination that accumulates at any point during a short period is equal to the product of a transfer coefficient

(deposition velocity), the concentration in the air, and the time period. To this contamination, RATCHET adds the contamination resulting from the washout of particles. The total surface contamination at a point during any period is the sum of the contributions from all puffs.

RATCHET maintains a mass balance. Material is removed from puffs as it deposits on the surface by dry and wet deposition. The mass to be removed from each puff is determined by multiplying the model time step by the deposition flux over the area covered by the puff. This mass is subtracted from the total mass in the puff. Material is not selectively removed from the bottom of the puff. This method of removal of mass from the plume is referred to as a "source depletion" model.

In reality, dry deposition and wet deposition of gases results in a mass deficit in the layer of air next to the surface. Source depletion models instantaneously propagate this deficit through the full vertical extent of the puff. This propagation is unrealistic, particularly in stable atmospheric conditions. Using the resistance analogy to estimate deposition velocities does not deal with this problem explicitly. However, using the resistance analogy results in lower deposition velocities during stable conditions, which reduces the magnitude of the error associated with the source depletion model.

RATCHET also accounts for radioactive decay of ^{131}I during transit and following deposition on the ground. The ^{131}I activity in the atmosphere is decreased hourly, and deposited activity is decreased to account for decay until midnight on the day of deposition.

RATCHET is a deterministic model. No single model run provides information on uncertainty in the time-integrated concentrations or surface contamination estimated by the code. However, RATCHET was designed to permit evaluation of uncertainty using a Monte Carlo approach. The Monte Carlo approach, which is described in detail in the RATCHET code documentation (Ramsdell et al. 1994), is consistent with and complements the approach found in other codes used in the HEDR Project.

Meteorological uncertainty is treated in two ways in RATCHET. Uncertainties in wind direction, wind speed, atmospheric stability, precipitation rate, and mixing-layer thickness are treated explicitly within the code. The explicit treatment of uncertainty in these variables and parameters leads to the implicit treatment of uncertainty in all model calculations using these variables and parameters. Uncertainties in other model parameters, such as release rates and iodine partitioning, are treated in model input by changing values from realization to realization.

Hourly data for each meteorological station are modified by addition of random components prior to use of the data in preparation of the meteorological data fields used in atmospheric dispersion and deposition calculations. This modification of the hourly meteorological data accounts for imprecision and, to some extent, inaccuracies in the recorded data. The model equations in

Table 2. Typical wet deposition velocities for gases and particle washout coefficients.

| | Deposition velocity (m s $^{-1}$) | | Particle washout coefficient (h $^{-1}$) |
|---------------|------------------------------------|----------------------|---|
| | Reactive | Noareactive | |
| Light rain | 1.4×10^{-3} | 1.4×10^{-6} | 0.254 |
| Moderate rain | 4.2×10^{-4} | 4.2×10^{-7} | 3.26 |
| Heavy rain | 6.9×10^{-4} | 6.9×10^{-7} | 4.78 |

RATCHET (Ramsdell et al. 1994) provide for consistent representation of the effects of the modifications on the transport, diffusion, and deposition of ^{131}I . Consequently, the code produces sets of time-integrated air concentrations and surface contamination that, while consistent with all available data, are yet different. The results from a set of model runs must be analyzed to evaluate uncertainty.

For wind speeds and wind directions, the random components were drawn from uniform distributions. The range of random values for wind direction for the HEDR calculations was $\pm 11.25^\circ$ because prior to 1965 wind directions were reported in compass points rather than degrees. For wind speeds the range of values was two reporting units for reported winds greater than zero. When calm winds were reported, directions were selected randomly from the range 000° to 360° . The range of wind speeds used during calm conditions depended on the wind speed reporting units. For speeds reported in miles per hour or knots, a range of 0 to 2 was used; for speeds reported in meters per second, the range was 0 to 1.

Within RATCHET, atmospheric stability is represented by the reciprocal of the Monin-Obukhov length (Monin and Obukhov 1954), which is a scaling length for vertical motions in the atmospheric boundary layer. Golder (1972) provides a method of estimating the reciprocal of Monin-Obukhov length, which is a continuous variable, from the discrete stability classes used in many dispersion models and surface roughness.

Random sampling was used to account for uncertainty in determination of stability classes and in estimating the reciprocal of the Monin-Obukhov length from the stability class and surface roughness. Uncertainty in stability class was estimated on the basis of a comparison of stability classes estimated by various methods using the Hanford Site meteorological data. This uncertainty was represented as a conditional probability distribution. The stability class used to determine the reciprocal of the Monin-Obukhov length was drawn randomly from this conditional probability distribution given the stability class determined using the method developed by Pasquill (1961), Gifford (1961), and Turner (1964). The uncertainty in conversion of stability class to reciprocal of the Monin-Obukhov length was treated assuming a uniform distribution of these values within the range shown by Golder given the stability class and surface roughness.

Precipitation rates generally are not included in hourly meteorological observations, but the current weather is reported. When the current weather at a meteorological station indicated that there was precipitation, RATCHET used an empirical conditional precipitation rate distribution to select a precipitation rate for use in wet deposition calculations. Empirical precipitation distributions for use in the HEDR Project were prepared using data available for stations in and near the atmospheric model domain. Separate sets of distributions were prepared for regions having annual precipitation less than $\sim 25 \text{ cm y}^{-1}$, 25 to 50 cm y^{-1} , and greater than

50 cm y^{-1} . Within each set of distributions there is a distribution for use when the reported weather is light rain, another for use with moderate rain, and a third for use with heavy rain. Similarly, there are distributions for light, moderate, and heavy snow.

When the conditional distributions are not available, default precipitation rates of 0.1, 3, and 5 mm h^{-1} are used for light, moderate, and heavy rain, respectively. The corresponding default precipitation rates for light, moderate, and heavy snow are 0.03, 1.5, and 3.3 mm h^{-1} , respectively. These rates are consistent with hourly precipitation rates observed at the Hanford Site.

RATCHET determines the mixing-layer thicknesses used to limit vertical dispersion directly from the meteorological data, including wind speed and stability. Ranges of values have been reported for several of the coefficients in the equations used to calculate the mixing-layer thickness. Each time these equations are used, coefficients are randomly selected from uniform distributions defined by the ranges of reported values. Thus, RATCHET varies the mixing-layer thickness to account for both the uncertainty in meteorological conditions and the uncertainty in mixing-layer thickness model parameters.

Source-term uncertainty is treated by varying the release sequences in 100 source-term realizations. These sequences of release rates and times were generated by the HEDR source-term model (Heeb 1994). Each realization of the complete source-term time series was based on and is consistent with available reactor and fuel processing plant records. By using a different realization of the source-term time sequence in each RATCHET model run, the variability in atmospheric model output reflects the uncertainty in both the source term and atmospheric models.

Typical variations of the monthly and hourly source term are shown in Fig. 3. The variations among the 100 source-term time sequences account for uncertainties in the times that batches of fuel were processed, the amount of ^{131}I released during the processing, and changes in the

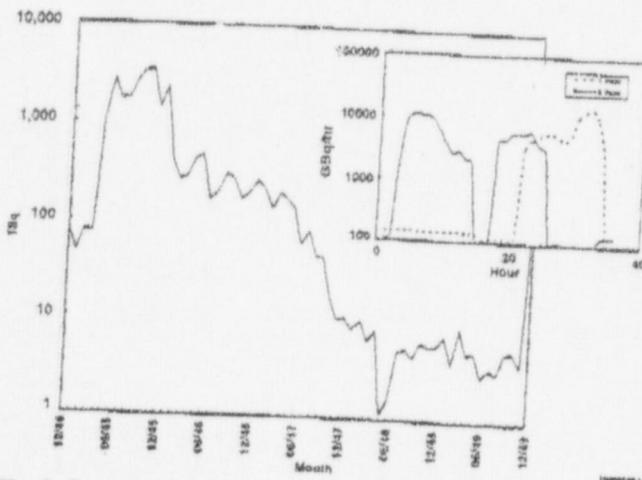


Fig. 3. Typical monthly and hourly variations in source term.

rate of evolution of the iodine during fuel processing. According to Heeb (1993), the uncertainty in timing of releases ranges from a few hours to a day or more. As a result, the uncertainty in release rate at a specific time is large. The release rate may range from 0 to several terrabecquerels per hour. In contrast, the uncertainty in total release over long periods, for example months or years, is much smaller. Coefficients of variation of the monthly release estimates are typically 0.1 or lower (Heeb 1994).

For HEDR model runs, the iodine associated with particles was assumed to be uniformly distributed between 5% and 45%, and the I_2 was assumed to constitute 20% to 60% of the gaseous iodine. The remainder of the iodine was assumed to be CH_3I . The range of values for each iodine fraction, based on these assumptions, is shown in Table 3. Note that the sum of the fractions is constrained to be 100% and that the fraction for the particulate component is the only uniformly-distributed fraction. The fractions for I_2 and CH_3I are more likely to be near the center of the ranges than near the ends. The distribution of iodine among the three species was changed from realization to realization but not within a realization.

RESULTS

The footprints for the time-integrated ^{131}I air concentrations and surface deposition are consistent with the wind roses shown in Fig. 2. Fig. 4 shows a footprint based on the median deposition at each of the nodes from 100 model runs. The highest values near the Hanford Site were found to the east and southeast of the release point, which is consistent with the prevailing winds at the Hanford Meteorology Station. Further from the Site, the highest values are found to the northeast, which is consistent with the prevailing southwest winds in the Mid-Columbia Basin. The pattern shown in Fig. 4 is somewhat broader than the footprint for a typical model run because the spatial correlations in deposition within a model realization were lost by using median values.

Variability of the total deposition from realization to realization is a function of position within the study area. In the 100 realizations, the ranges of values at nodes in the main part of the footprint are generally less than a factor of four, but the ranges for several nodes at the edge of the footprint exceed an order of magnitude. Table 4 shows statistics for 12 locations; the ranges of values at Yakima, WA, and The Dalles, OR (which are on the upwind edge of the footprint) can be compared with the

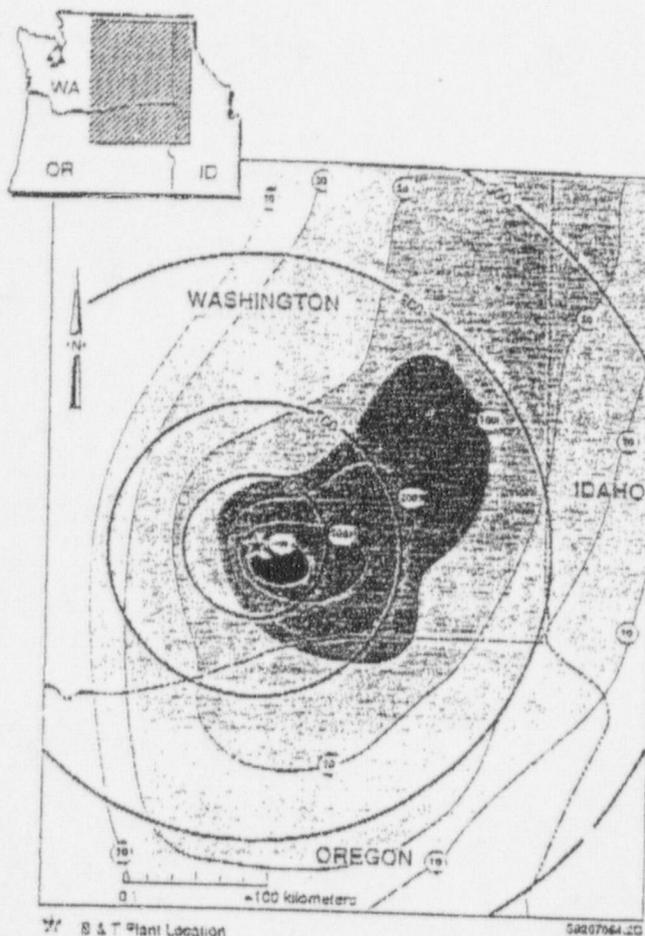


Fig. 4. Median estimates of total ^{131}I deposition (kBq m^{-2}), December 1944 through December 1949.

Table 4. Variation in total ^{131}I deposition in 100 realizations of RATCHET.

| Location | Mean (kBq m^{-2}) | Standard Deviation (kBq m^{-2}) | Maximum (kBq m^{-2}) | Minimum (kBq m^{-2}) |
|----------------|------------------------------|--|---------------------------------|---------------------------------|
| Bonner's Ferry | 31.4 | 4.1 | 41.5 | 21.7 |
| Colfax | 57.1 | 10.5 | 87.4 | 33.9 |
| Coulee City | 24.9 | 6.1 | 48.1 | 14.6 |
| Lewiston | 18.6 | 2.9 | 24.3 | 12.2 |
| Othello | 261 | 59.0 | 434 | 174 |
| Pendleton | 79.5 | 12.2 | 124 | 54.5 |
| Richland | 766 | 152 | 1,400 | 385 |
| Ritzville | 194 | 36.4 | 309 | 115 |
| Spokane | 80.1 | 11.6 | 119 | 55.5 |
| The Dalles | 3.2 | 1.4 | 7.2 | 0.9 |
| Walla Walla | 70.3 | 18.7 | 160 | 44.8 |
| Yakima | 6.7 | 2.3 | 13.2 | 2.6 |

Table 3. Ranges of iodine species fractions.

| Component | Component fraction | |
|-----------------------|--------------------|---------|
| | Minimum | Maximum |
| Particles | 5% | 45% |
| I_2 | 11% | 57% |
| CH_3I | 22% | 76% |

ranges at Richland, Ritzville, and Spokane, WA, and Bonner's Ferry, ID (which are in the main portion of the footprint).

The way in which iodine was partitioned among the three species had a significant effect on time-integrated

air concentrations and total deposition at nodes. Of the three species, the organic iodine fraction was most highly correlated with variations in time-integrated air concentrations and total deposition. However, this correlation is a function of position within the study area. Near the Hanford Site the correlation between the organic fraction and time-integrated air concentration is low and positive. As distance increases, the correlation becomes larger. Far downwind from the Hanford Site variations in the organic fraction account for more than 70% of the variability in time-integrated air concentrations. In contrast, the correlation between the organic fraction and total deposition is relatively high and negative near the Hanford Site and decreases as distance increases.

Fig. 5 illustrates these changes in the correlation between organic fraction and time-integrated air concentrations and total deposition by showing the model output for all 100 realizations for Richland and Bonner's Ferry. The changes in correlation between organic fraction and time-integrated air concentrations and total deposition with distance are shown more quantitatively in Table 5. Richland, Spokane, Bonner's Ferry lie near the long-term plume axis at distances of approximately 50 km, 200 km, and 350 km, respectively.

Table 5. Spatial variation of the correlations between the ^{131}I organic species fraction and time-integrated air concentrations (MBq s m^{-3}) and total deposition (kBq m^{-2}).

| Location | Approximate distance (km) | Time-integrated air concentration | | Total deposition | |
|----------------|---------------------------|-----------------------------------|-------------|------------------|-----------------|
| | | r^2 | Correlation | r^2 | Correlation |
| Richland | 50 | 0.101 | positive | 0.465 | negative |
| Spokane | 200 | 0.702 | positive | 0.268 | negative |
| Bonner's Ferry | 350 | 0.705 | positive | 0.047 | negative |
| Yakima | 60 | 0.186 | positive | 0.000 | not significant |

The changes in correlation are explained physically by considering the effect of deposition on air concentration as distance increases. The effective deposition velocity for iodine is negatively correlated with organic fraction. As a result, when the organic fraction is small, iodine deposition near the source will be large and concentrations far downwind will be low. At the other extreme, when the organic fraction is large, there will be relatively little deposition near the source and concentra-

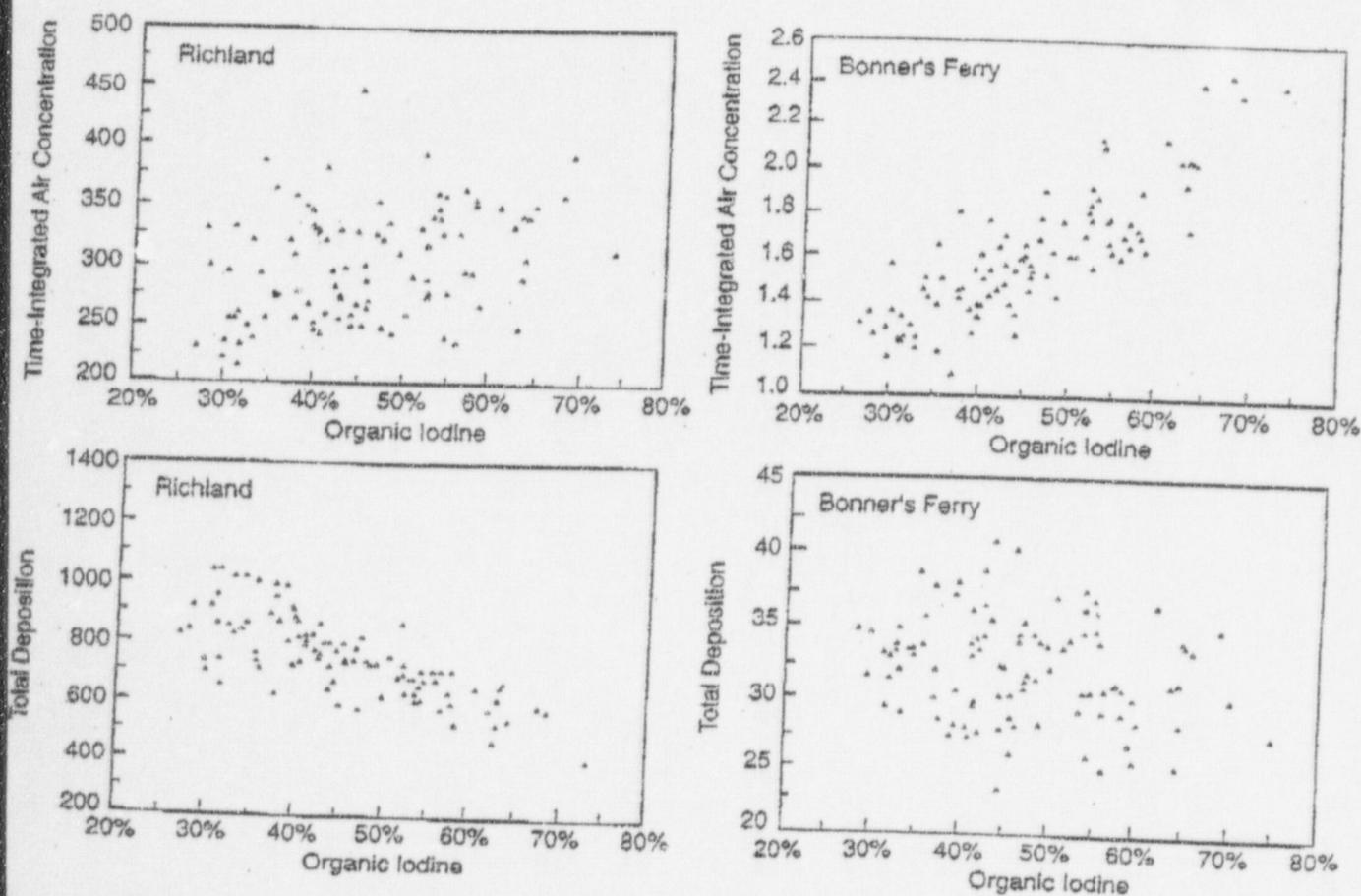


Fig. 5. Variation in estimates of total time-integrated air concentration (MBq s m^{-3}) and deposition (kBq m^{-2}) at Richland, Washington, and Bonner's Ferry, Idaho, as a function of percent organic iodine (CH_3I).

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Table 6. Variability in the fate of ^{131}I in 100 realizations, December 1944 through 1949.

| | Total release (TBq) | Deposited in study area (%) | Decayed in study area (%) | Left study area (%) |
|--------------------|---------------------|-----------------------------|---------------------------|---------------------|
| Mean | 2.57×10^4 | 56.5 | 9.6 | 33.9 |
| Standard deviation | 1.37×10^3 | 6.5 | 0.90 | 5.4 |
| Maximum | 2.93×10^4 | 71.4 | 11.6 | 45.9 |
| Minimum | 2.33×10^4 | 37.3 | 7.8 | 23.9 |

tions far downwind will be relatively high. However, these higher concentrations do not result in correspondingly high deposition at distance because the effective deposition velocity is low.

Yakima, which is included in Table 5, is slightly farther from the release point than Richland. Thus, the correlation between the organic fraction and time-integrated air concentrations at Yakima is consistent with correlations at other locations. On this basis, the correlation between total deposition at Yakima and the organic fraction might be expected to be similar to the correlation for Richland; however, Table 5 shows that there is no correlation. Although there is no proof, intermittency is suspected to be one of the reasons for the lack of correlation since RATCHET output indicates that released material reached Yakima only a few times each year.

The footprint for median estimates of the maximum surface contamination at any time during the 5-y period is shown in Fig. 6. For most of the study area, the maximum surface contamination is ~10% of the total deposition. This result is consistent with the pattern of monthly releases shown in Fig. 3. While it is highly unlikely that the maximum surface contamination occurred simultaneously over the entire area, it is likely that the maxima for most locations in the main portion of the footprint occurred during the last half of 1945.

Heeb (1994) estimates that $\sim 2.6 \times 10^4$ TBq of ^{131}I were released from Hanford during the first 5 years of operation. The ultimate fate of this iodine was estimated using the mass balance statistics generated by RATCHET (see Table 6). It is estimated that ~56% of the ^{131}I released at Hanford was deposited within the

study area, ~10% decayed while in transit in the atmosphere within the study area, and the remaining 34% left the area in the air. The uncertainties in these fractions are relatively small in comparison with the uncertainties in the values at specific nodes or for shorter time periods.

The variability in the organic iodine fraction is a major contributor to the variations in the overall ^{131}I mass balance. Fig. 7 shows the relationship between the fraction of the ^{131}I deposited in the study area and the organic fraction. The variations in the organic fraction account for almost 50% of the variations in the total deposition. Variations in the organic fraction account for similar percentages in the amount of ^{131}I that decayed during atmospheric transport within the study area and the amount that left the area. In contrast to the variation shown in Fig. 7, these last two correlations were positive. Increasing the organic fraction increases the percentage of ^{131}I that decayed in, or departed from, the study area.

CONCLUSIONS

Early HEDR Project studies determined that existing information was sufficient to reconstruct the ^{131}I releases to the atmosphere from early Hanford Site operations and to track the ^{131}I through the environment. Therefore, the HEDR Project developed a suite of computer codes to estimate the radionuclide releases, track them through the environment, and estimate doses. The RATCHET computer code is part of this suite.

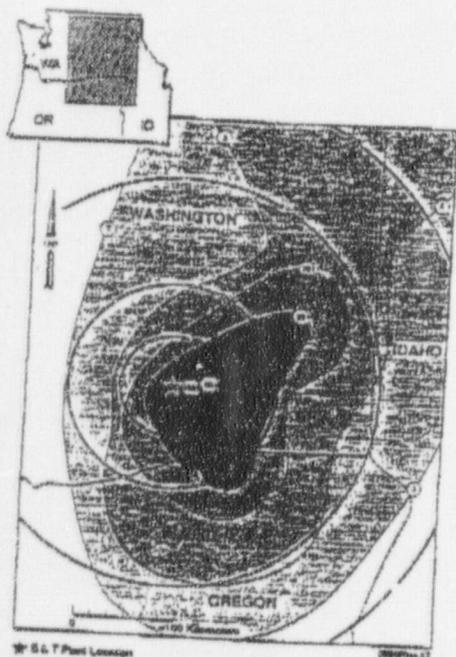


Fig. 6. Median estimates of the maximum surface contamination (kBq m^{-2}), December 1944 through December 1949.

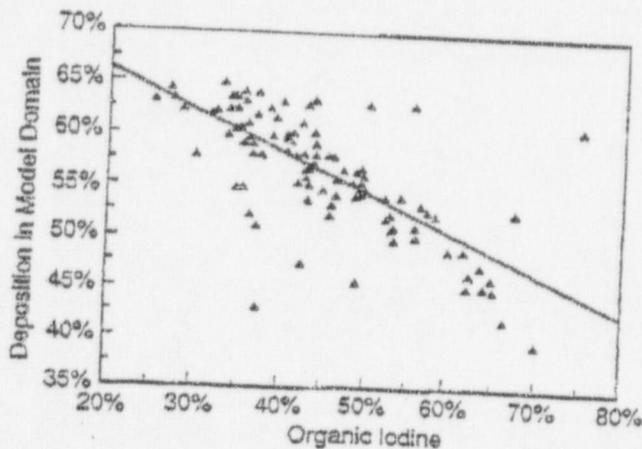


Fig. 7. Variation in percent of ^{131}I deposited as a function of the percent of organic iodine.

Using hourly meteorological data and estimates of hourly release rates, RATCHET calculated daily time-integrated air concentrations and deposition over a study area of $\sim 2 \times 10^5 \text{ km}^2$. The resulting time-integrated air concentration and deposition patterns are consistent with expectations based on regional wind data and the results of earlier studies of material transported from Hanford. Monte Carlo techniques used in RATCHET calculations, along with 100 realizations of the release rate time series, resulted in characterization of the uncertainties in the time-integrated air concentrations and surface contamination in a manner that preserved both spatial and temporal correlations.

Given large uncertainties in hourly release rates, imprecise and sparse meteorological data, and limited information on iodine partitioning in the atmosphere among species having widely varying deposition properties, large variations might be expected in the time-integrated air concentrations and surface contamination calculated by RATCHET for specific nodes. These large variations do, indeed, exist in the daily values. However, the radiation doses of interest in the HEDR Project were long-term, cumulative doses. Therefore, the results shown here are based on the results of integration over a period of more than 5 y. This integration smooths out the day-to-day variability. Consequently, the variability in time-integrated air concentrations and surface contamination across the 100 realizations is relatively small. The results in Table 4 show that, throughout the footprint of the time-integrated plume, the coefficient of variation of the total deposition is typically of the order of 0.2. This value is only a factor of four larger than the coefficient of variation of the total ^{131}I release, which is about 0.05 (see Table 6).

Analysis of the atmospheric transport, diffusion, and deposition estimates indicates that a primary source of variability in the time-integrated air concentration and surface contamination estimates is associated with the uncertainty in the partitioning of ^{131}I . This is an area where further work appears to be warranted.

The following conclusions resulted from this work:

- Sufficient data existed to reconstruct radionuclide releases and define their uncertainty.
- The available meteorological data, while sparse, were sufficient to estimate the transport, diffusion, and deposition of the ^{131}I released to the atmosphere.
- The time-integrated air concentration and surface contamination estimates are reasonable and are consistent with available data.

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