

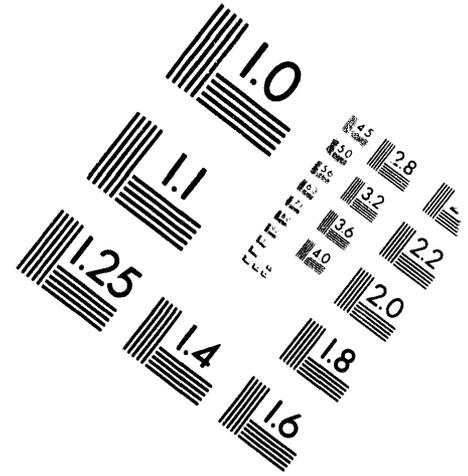
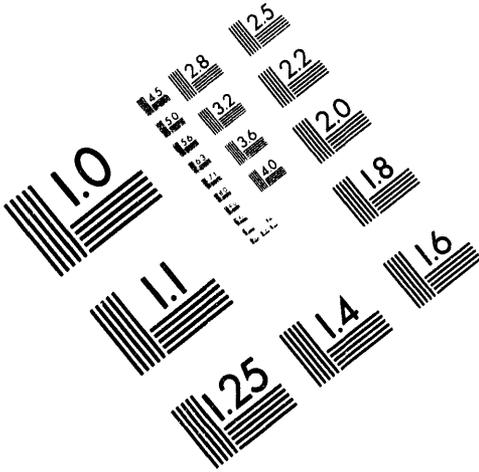


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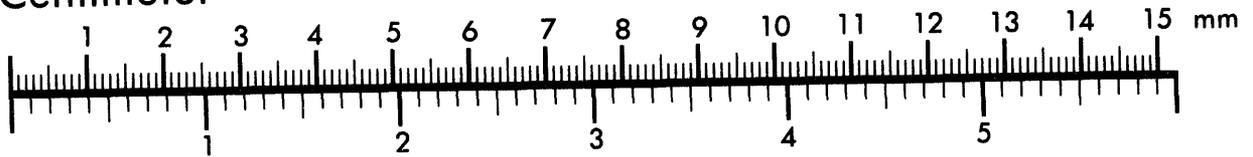
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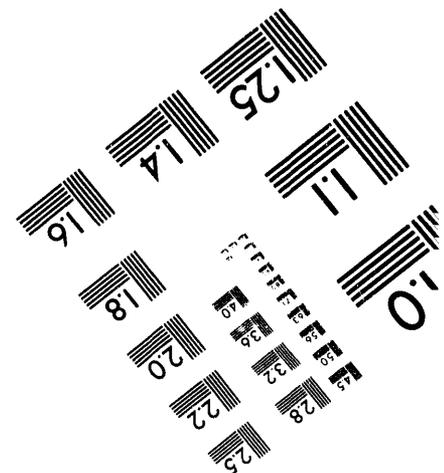
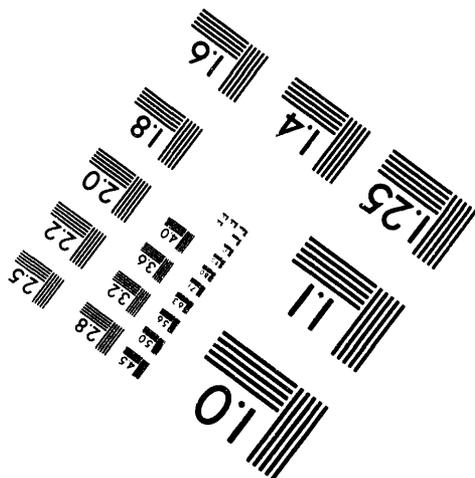
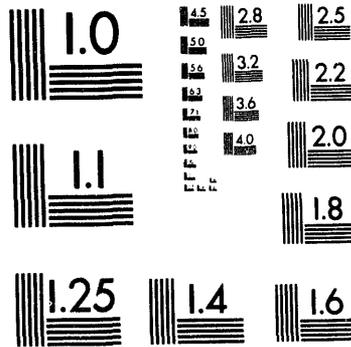
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ATMOSPHERIC DISPERSION AND DEPOSITION OF IODINE-131
RELEASED FROM THE HANFORD SITE

J. V. Ramsdell
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**ATMOSPHERIC DISPERSION AND DEPOSITION OF IODINE-131
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 curies) of iodine-131 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 (HEDR)[†] 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 (nominally, 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 iodine-131 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

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iodine-131 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.

Key words: atmospheric dispersion, deposition, iodine-131, Hanford, dose reconstruction, model uncertainty

INTRODUCTION

Early studies in the Hanford Environmental Dose Reconstruction (HEDR) Project (Ramsdell and Burk 1991a, 1991b) showed that iodine-131 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°40'N, 118°45'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.

The study area is sufficiently large that systematic variations in meteorological and climatological conditions are found across the area. The wind roses in Fig. 2 show the effects of the Rattlesnake Mountain along the west and southwest border of the Hanford Site, 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 yr⁻¹ near Hanford

to more than 250 cm yr⁻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 yr⁻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 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 iodine-131. 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, 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 represent the conditions surrounding the node. This assumption is reasonable because the averaging time is relatively long and the points of interest are far enough from the release point that the air concentration and surface contamination gradients are generally small.

Transport, diffusion, and deposition calculations are made using wind, atmospheric stability, precipitation, and mixing-layer depth fields that described the spatial and temporal variations of meteorological conditions across the study area. These fields are prepared by RATCHET as needed using hourly meteorological data from observation sites in and adjacent to the study area. Wind, atmospheric stability, mixing-layer depth, and precipitation vary in time and space throughout 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 it is a general-purpose atmospheric dispersion code, 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. These forms have significantly different deposition characteristics. For example, Voilleque and Keller (1981) give typical deposition velocities for CH_3I , I_2 , and particles as 0.00001, 0.01, and 0.001 m s^{-1} , respectively. In RATCHET, iodine-131 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 iodine-131 in the elemental form. However, it is unlikely that the iodine remained in the elemental state for long. Experimental data on 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. 1977; 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. 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 wind, atmospheric stability, and surface roughness. The surface resistance is a function of wind 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 was not included in the total resistance, dry deposition velocities in neutral conditions increase from $\sim 0.006 \text{ m s}^{-1}$ 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 for most reactive gases, and the entire range of deposition velocities is above that measured for fine particles ($\sim 1 \mu\text{m}$) 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 fall through the puffs. Default precipitation rates of 0.1, 3, and 5 mm h⁻¹ for light, moderate, and heavy rain, respectively, are included in RATCHET. 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.

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 I₂. This assumption results in the wet deposition velocities shown in Table 2 for the default precipitation rates in RATCHET. Scavenging of CH₃I by precipitation is extremely limited and is neglected as a practical matter. Wet deposition velocities for CH₃I computed in RATCHET are about three orders of magnitude lower than those for I₂. Scavenging of both I₂ and CH₃I by snow when the temperature is less than -3°C is low because the snow surface is frozen rather than wet and was 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 3 shows particle washout coefficients for the default rainfall rates in RATCHET. During periods of snow, this washout coefficient is directly related to the precipitation rate.

Given the 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 of 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 is referred to as the "source depletion" model.

In reality, deposition 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.

RACHET also accounts for radioactive decay of iodine-131 during transit and following deposition on the ground. The iodine-131 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 code includes provisions to modify the hourly meteorological data randomly to account for imprecision and, to some extent, inaccuracies in the recorded data. The model equations in RATCHET (Ramsdell et al. 1994) provide for consistent representation of the effects of these modifications on the transport, diffusion, and deposition of iodine-131. 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 the uncertainty.

Uncertainty is treated in two ways in RATCHET. Uncertainties in wind direction, wind speed, atmospheric stability, precipitation rate, and mixing-layer height are treated explicitly within the code. The explicit treatment of uncertainty in the variables and parameters listed above 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.

Source term uncertainty was treated using a set of 100 realizations of release rates and times generated by the HEDR source term model (Heeb 1994). Typical variations of the monthly and hourly source term are shown in Fig. 3. Each realization of the complete source term time series is based on and consistent with available reactor and fuel processing plant records. By using a different realization of source term time sequence in each model run, the variability in atmospheric model output reflects the uncertainty in both the source term and atmospheric models.

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 weights is constrained to be 100% and that the weight for the particulate component is the only uniformly distributed weight. The weights for I_2 and CH_3I are more likely to be near the center of the ranges than near the ends.

RESULTS

The footprints for the time-integrated iodine-131 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. Farther 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 is a function of position within the study area. In the 100 model runs, 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, Washington, and The Dalles, Oregon (which are on the upwind edge of the footprint) can be compared with the ranges at Richland, Ritzville, and Spokane, Washington, and Bonner's Ferry, Idaho (which are in the main portion of the footprint).

Analysis of the RATCHET results indicates that iodine partitioning is a primary source of variability in the time-integrated air concentrations and total deposition. Of the three components, the organic iodine fraction is 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 Hanford, the correlation between the organic fraction and time-integrated air concentration is low and positive. As the distance from Hanford increases, the correlation becomes larger. Far downwind from Hanford, 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 the total deposition is relatively high and negative near Hanford and decreases as a result of competing processes as distance increases. A large organic fraction reduces the effective deposition velocity as iodine-131 is transported downwind. Thus, a large fraction of the released iodine is available for deposition at long distances. These changes in correlation are listed in Table 5.

Fig. 5 shows the detailed variations in time-integrated air concentrations and total deposition for Richland and Bonner's Ferry.

Yakima is included in Table 5 to show the effect of intermittency on these correlations. Yakima 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 is shown in Fig. 6. For most of the study area, the maximum surface contamination is ~10% of the total deposition, which is somewhat surprising because the total deposition accumulated over a 5-yr period and iodine-131 has an approximate 8-day half-life. However, this result is consistent with the pattern of monthly releases shown in Fig. 3. 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 iodine-131 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 iodine-131 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 uncertainty in these fractions is 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 iodine-131 mass balance. Fig. 7 shows the relationship between the fraction of the iodine-131 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 iodine-131 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 iodine-131 that decayed in or departed from the study area.

CONCLUSIONS

Early HEDR Project studies determined that existing information was sufficient to reconstruct the iodine-131 releases to the atmosphere from early Hanford Site operations and to track the iodine-131 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.

Using hourly meteorological data and estimates of hourly release rates, RATCHET calculated daily time-integrated air concentrations and deposition over an $\sim 2 \times 10^5$ km² study area. 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 the large uncertainties in hourly release rates, the imprecise and sparse meteorological data, and the limited information on iodine partitioning in the atmosphere among species having

widely varying deposition properties, the variability in time-integrated air concentrations and surface contamination across the 100 realizations is smaller than might be expected, confirming that the central limit theorem works. Analysis of the atmospheric transport, diffusion, and deposition estimates indicates that the primary source of variability in the time-integrated air concentration and surface contamination estimates is associated with the uncertainty in the partitioning of iodine-131. Thus, 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 iodine-131 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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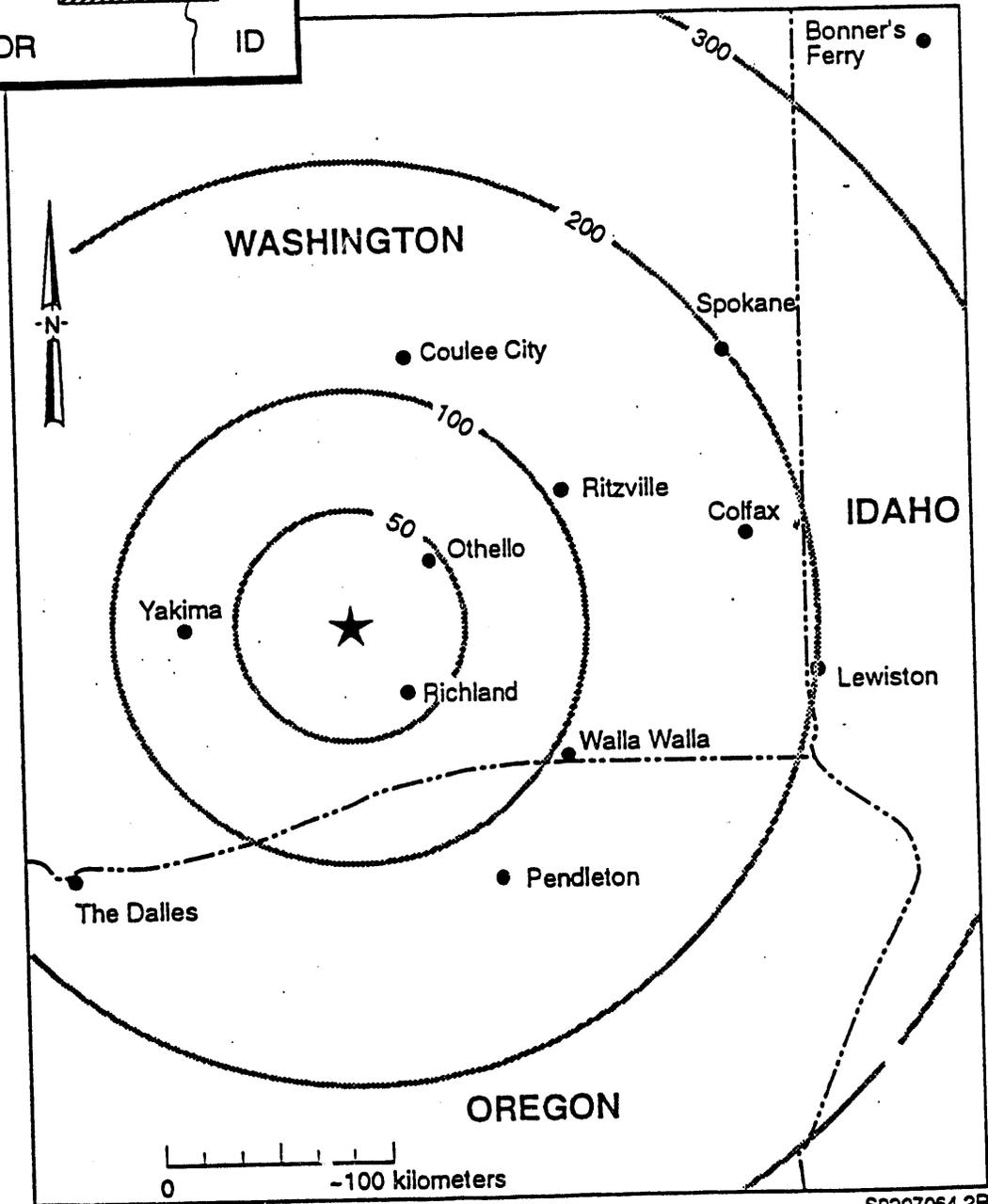
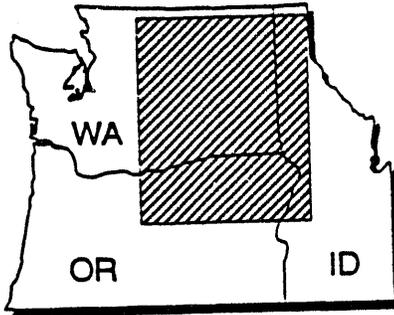
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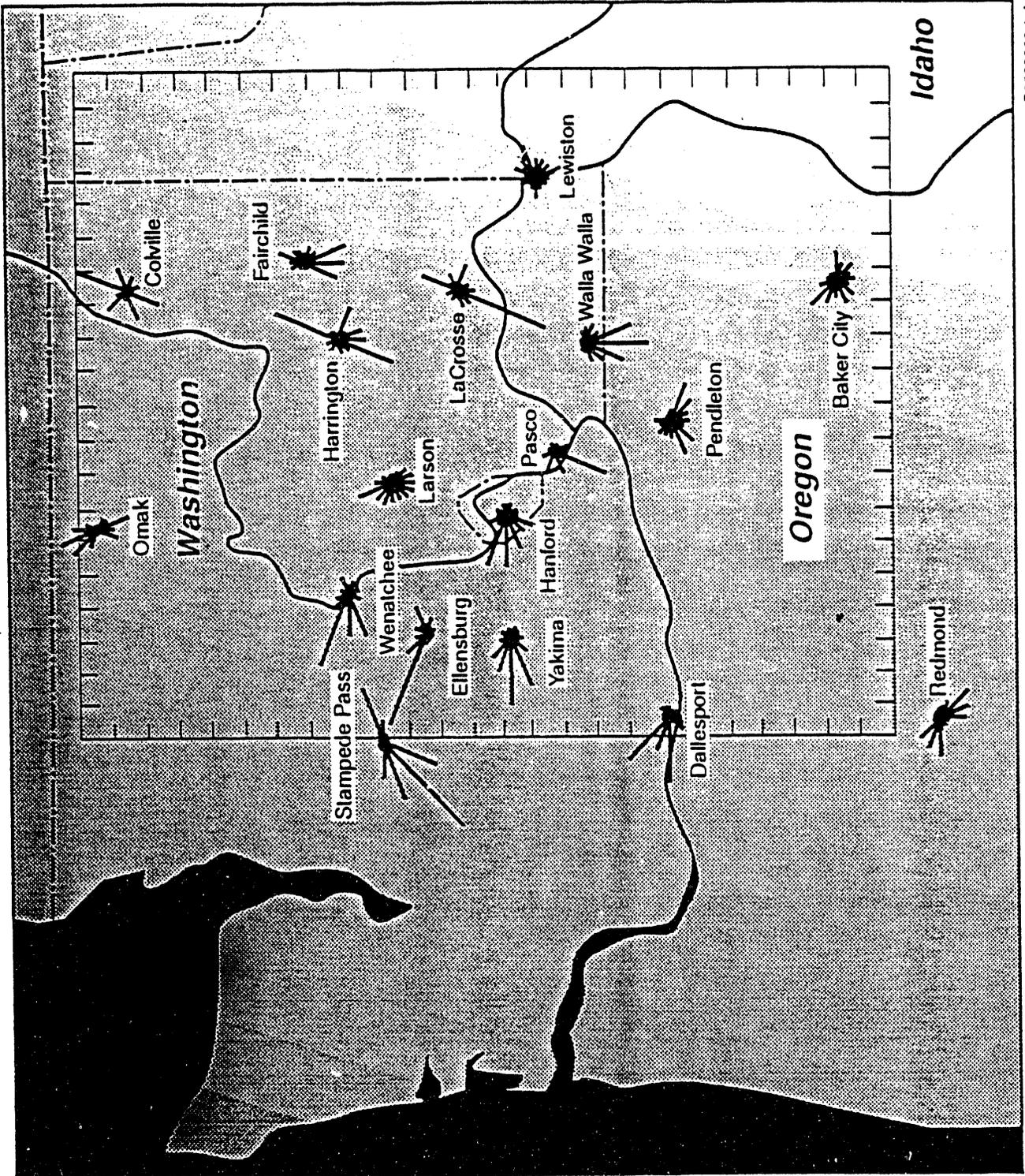
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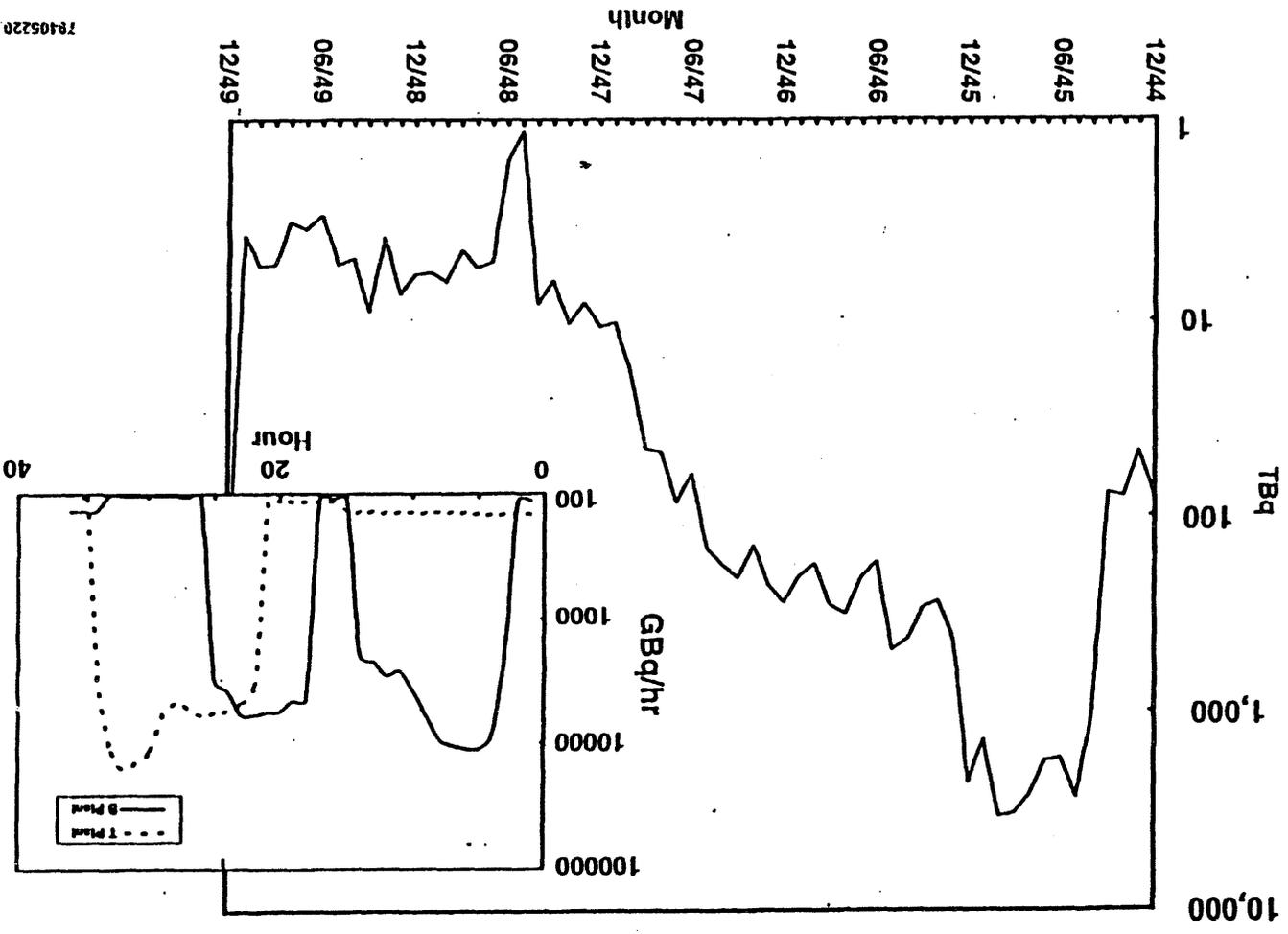


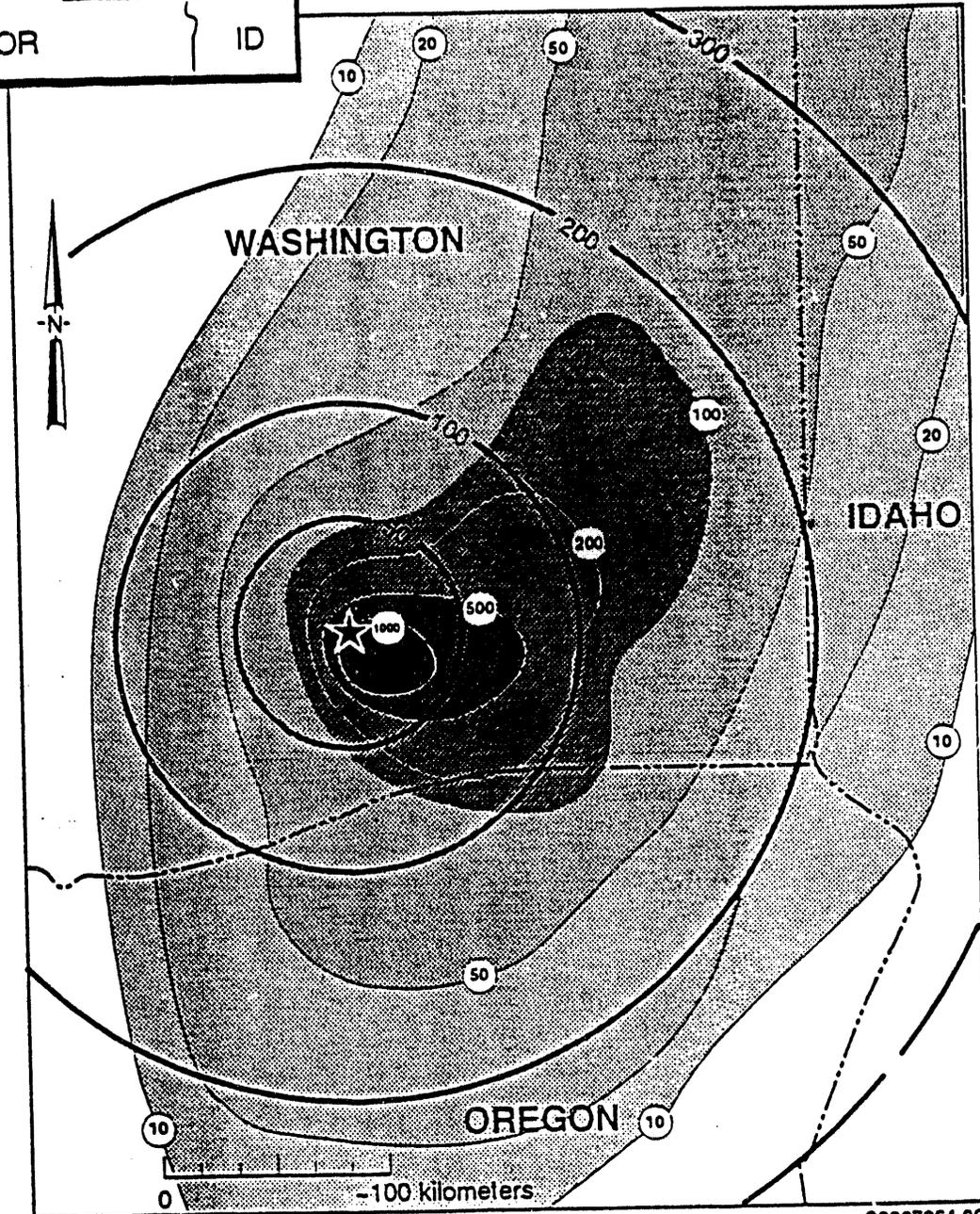
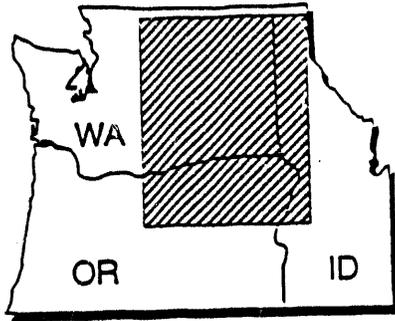
S9207064.2R

★ B & T Plant Location



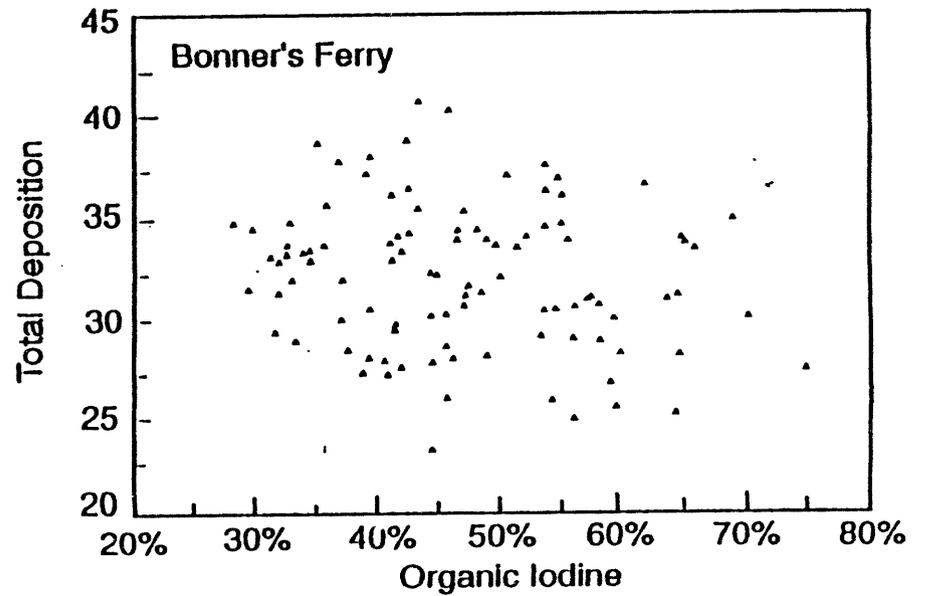
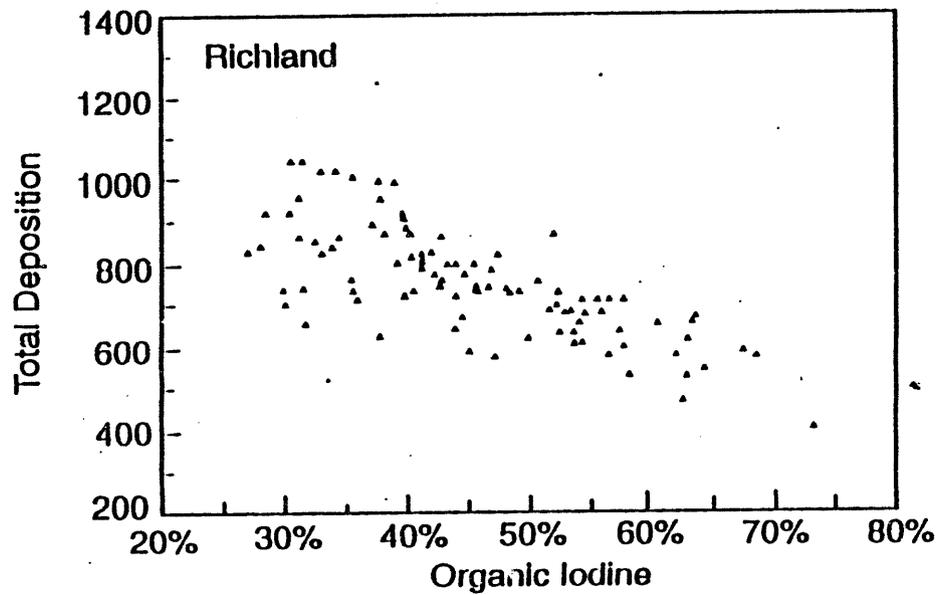
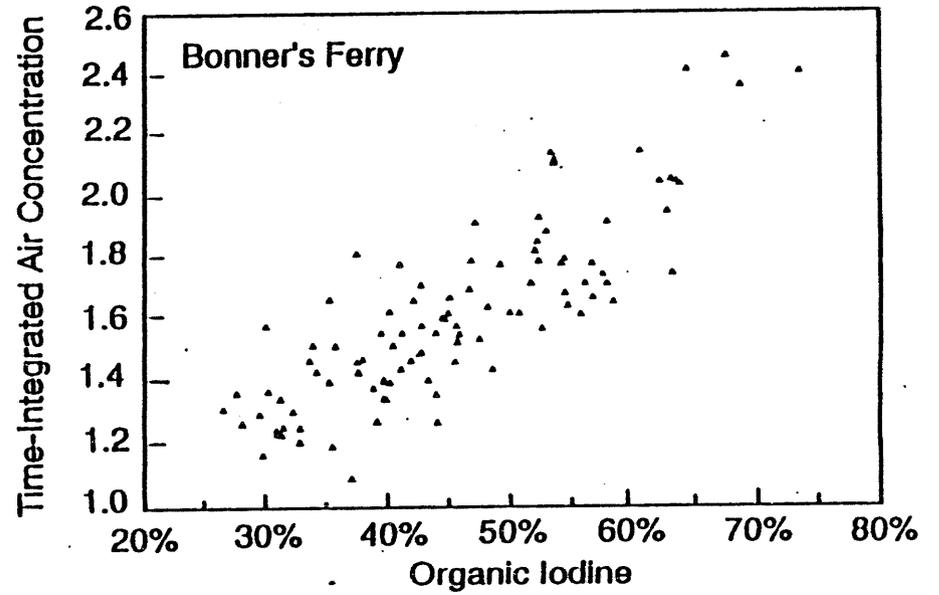
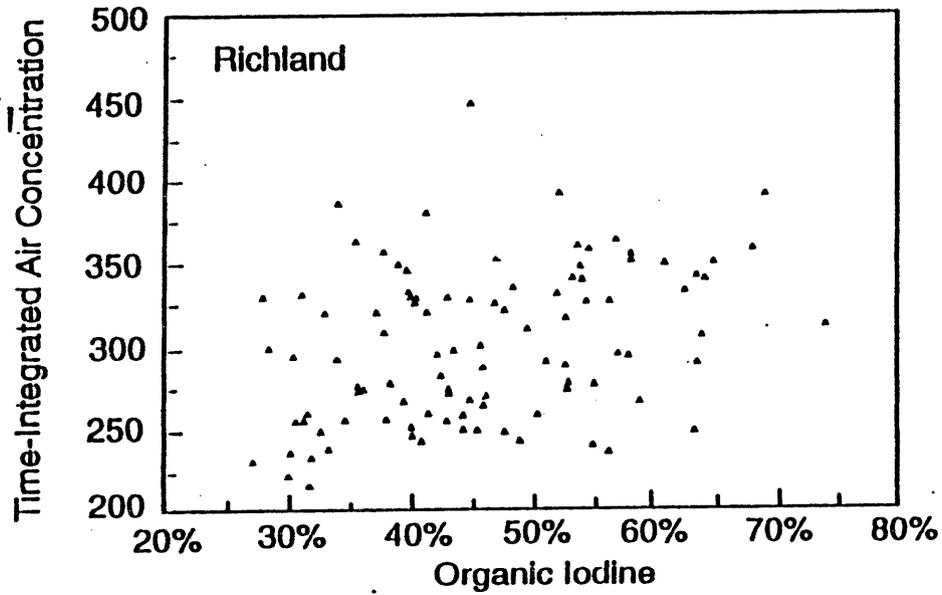
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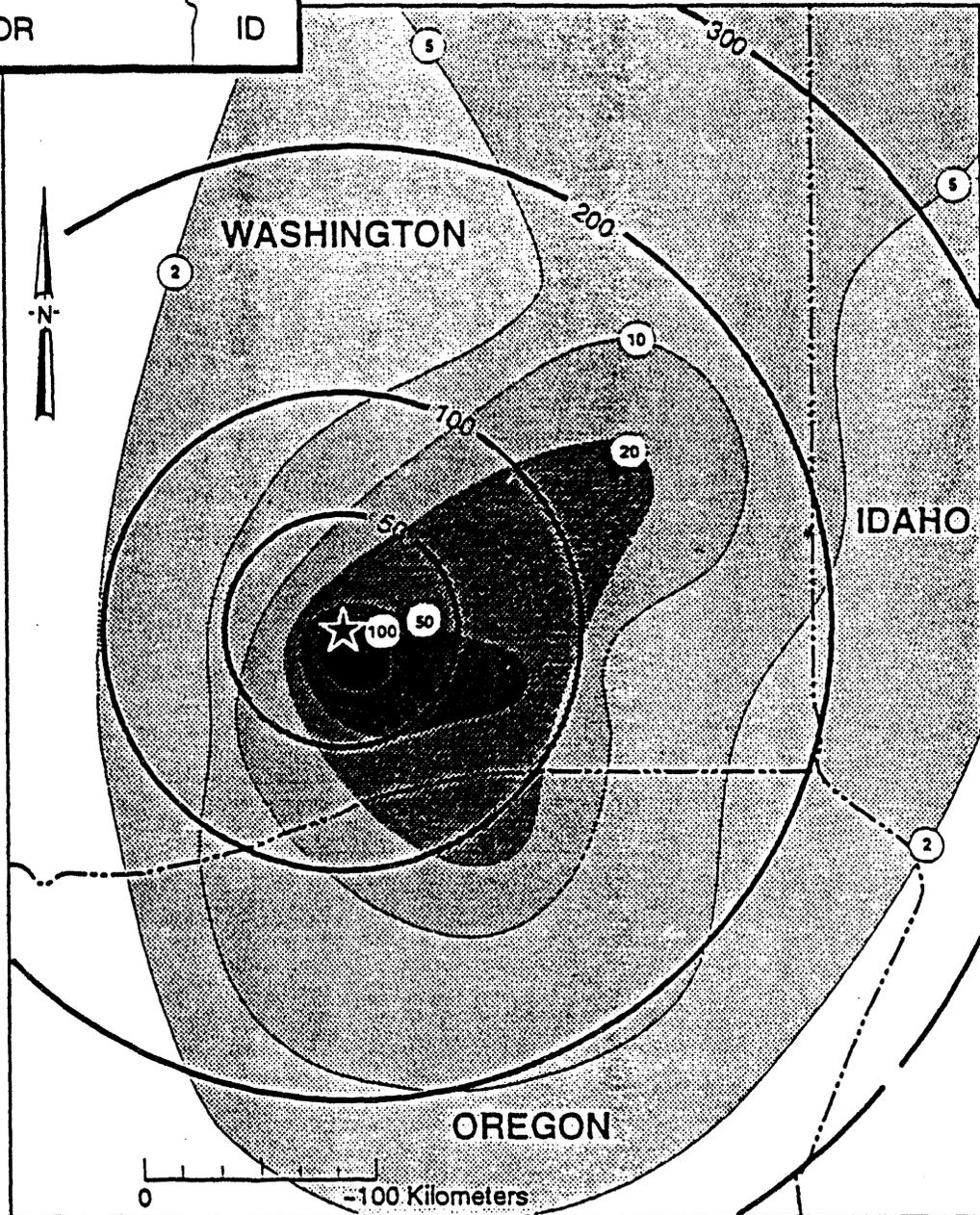
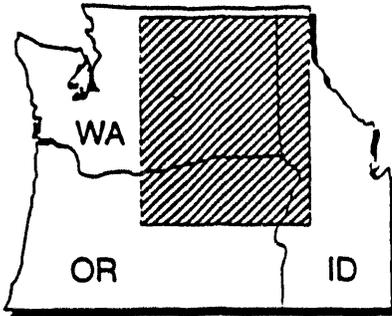




★ B & T Plant Location

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★ B & T Plant Location

S9207064.2Z

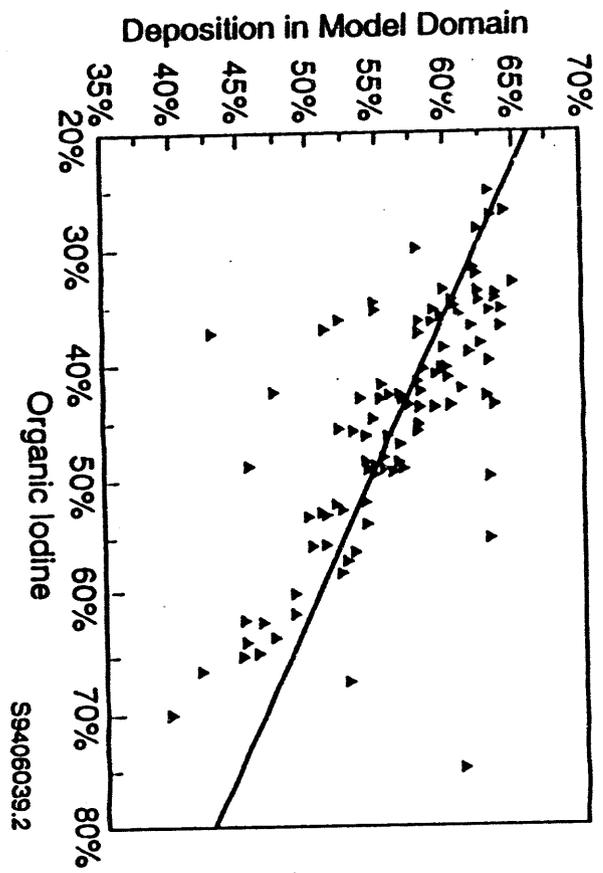


Table Captions

1. Features of the RATCHET computer code.
2. Typical wet deposition velocities for gases and particle washout coefficients.
3. Range of weight associated with iodine components.
4. Variation in total deposition within 100 realizations of RATCHET.
5. Variation of the correlation of time-integrated air concentrations and total deposition with distance.
6. Variability in the fate of iodine-131 in 100 realizations, December 1944 through 1949.

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 (L 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

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

	Deposition Velocity (m s ⁻¹)		Particle Washout
	Reactive	Nonreactive	Coefficient (h ⁻¹)
Light rain	1.4 x 10 ⁻⁵	1.4 x 10 ⁻⁸	0.254
Moderate rain	4.2 x 10 ⁻⁴	4.2 x 10 ⁻⁷	3.26
Heavy rain	6.9 x 10 ⁻⁴	6.9 x 10 ⁻⁷	4.78

Table 3. Range of weights associated with iodine components.

Component	Component Fraction	
	Minimum	Maximum
Particles	5%	45%
I ₂	11%	57%
CH ₃ I	22%	76%

Table 4. Variation in total deposition within 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	12.3
Othello	261	59.0	434	174
Pendleton	79.5	12.2	124	54.5
Richland	766	152	1400	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 5. Variation of the correlations of time-integrated air concentrations and total deposition with distance.

Location	Time-Integrated Air Concentration		Total Deposition	
	r ²	Correlation	r ²	Correlation
Richland	0.101	positive	0.465	negative
Spokane	0.702	positive	0.268	negative
Bonner's Ferry	0.705	positive	0.047	negative
Yakima	0.186	positive	0.000	not significant

Table 6. Variability in the fate of iodine-131 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

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