Abstract—Between 1944 and 1956, radioactive $^{131}$I was released into the atmosphere from operations at the Oak Ridge National Laboratory in Oak Ridge, TN. The releases occurred from stacks and from building vents and openings in three different chemical forms: elemental, organic, and particulate. During their transport in the atmosphere, different forms of iodine react differently with other atmospheric chemicals and moisture, and are removed from the plume at different rates by the processes of dry and wet deposition. A modified Gaussian plume model was developed to address the processes of radioiodine speciation, deposition, depletion, and dispersion in the atmosphere, and to propagate uncertainties in input parameter values through to the ground-level concentrations of $^{131}$I in air. A unique approach was used to develop an implicitly correlated set of hourly meteorological parameters for any day of a month for each month of the year from ten years of available data between 1987 and 1996. The model was validated for both annual average and short-term releases. For the annual average releases, the predictions of ground-level concentrations of $^{131}$I from the model were within a factor of 2 of measured field data. For two of the three sets of available weekly data, the measurements fell within the 95% subjective confidence interval of model predictions. Predictions of ground-level air concentrations were made on an annual average basis for the entire period of release and on a short-term, episodic basis for a 1954 accident.

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Key words: $^{131}$I; dose assessment; radiation, environmental; modeling, meteorological

INTRODUCTION

Between 1944 and 1956, radioactive $^{131}$I was released to the atmosphere from the radioactive lanthanum processing facility at the Oak Ridge National Laboratory (ORNL) in Oak Ridge, TN (Apostoaei et al. 1998). Most releases occurred from two ORNL stacks: the Chemical Processing Pilot Plant Stack (the 3020 Stack) between 1944 and 1950, and the central off-gas-processing stack (the 3039 Stack) between 1950 and 1956. Releases also occurred directly from the building housing the lanthanum processing facility. Once released into the atmosphere, $^{131}$I was dispersed and transported downwind by prevailing winds. During transport, it underwent a number of chemical transformations because of the reactions of its various chemical forms with other atmospheric chemicals and moisture. The different chemical forms of radioiodine were also removed from the atmosphere in differing amounts by the processes of wet and dry deposition.

A member of the public would have been exposed to $^{131}$I released from the radioactive lanthanum operation via inhalation of contaminated air or ingestion of contaminated food products derived from deposition of $^{131}$I from the atmosphere to vegetation. A dose reconstruction study was conducted to estimate the radiological dose to an exposed individual’s thyroid gland, the excess lifetime risk of the individual developing thyroid cancer, and the probability of causation for an already diagnosed thyroid cancer. In order to calculate these quantities, it is necessary to first calculate the ground-level concentrations of $^{131}$I in air and on pasture vegetation grazed by cows and goats during the years of release.

A dispersion model (SENES Oak Ridge Atmospheric Dispersion Model for Jodine-131; SORAMI) based on a Gaussian plume modeling approach (Hanna et al. 1982), was developed for this analysis. The primary reasons for developing a new model were to simulate the chemical transformations of iodine from one chemical form to another as a function of downwind distance, to address the calculation of the wet deposition velocities using the approach described in Apostoaei et al. (1998), and to address the depletion of radioiodine from the plume as a result of removal by the processes of wet and dry deposition. The transformation chemistry of iodine in the atmosphere is discussed first in the next section. Governing mathematical equations of the SORAMI model and model validation using field data are presented in the following section. Final simulations were conducted separately for the routine releases and for a 1954 accident in which large quantities of $^{131}$I were released in a short period of time. For both routine releases and the accident,
separate simulations were conducted for releases from the stack and from the building vents and openings. Predictions of air concentrations were made for a 38-km-radius region surrounding Stack 3039.

Uncertainties in input parameters were identified and propagated through to the predictions of air concentrations for all simulations. For the routine releases, hourly joint probability distributions were developed for the wind speed, atmospheric stability, and wind direction for each month of the year. The approach used allows for the correlation structure of the measured data to be implicitly maintained while sampling the distributions for repeated simulations using a Monte Carlo method.

ATMOSPHERIC CHEMISTRY

Background

During its transport in the atmosphere, $^{131}$I undergoes several chemical transformations depending on the chemical form in which it is released from the stack. In addition, all forms of $^{131}$I undergo radioactive decay with a half-life of 8.04 d. For travel times of less than an hour, within which most of the chemical transformations occur, removal of $^{131}$I by radioactive decay would be insignificant.

$^{131}$I was released from the RaLa processing facility in three different chemical forms: a reactive form (consisting primarily of $^{131}$I in elemental form), a nonreactive form (consisting of $^{131}$I in organic forms), and a particulate form. A detailed literature survey of the reactions of iodine in the atmosphere (Chameides and Davis 1980; Jenkins and Cox 1985; Garland and Curtis 1981; Garland 1967) was conducted. It was found that elemental iodine ($I_2$) photo-dissociates rapidly in the presence of sunlight with a half-life of 20 s. Methyl iodide photo-dissociates in the atmosphere in the presence of sunlight with a half-life of 64 h, which indicates that only the iodine released in elemental form undergoes significant photodissociation during its travel in the region of interest. Models of iodine chemistry did not contain reactions for the formation of methyl iodide in the atmosphere even though evidence exists for its formation.

An experimental study was conducted at Hanford (Ludwick 1964) in which iodine was released into the atmosphere in elemental form. Measurements of different forms of iodine in the atmosphere indicated that beyond a distance of roughly 3 km, 30% of iodine was in particulate form, 36% was in organic form, and the remaining 34% was in the elemental form. Ludwick (1967) used stack gas measurements and measurements 5 miles downwind of the stack to determine that the original iodine, released in the elemental form, partitioned into 15%, 43%, and 42% particulate, organic, and elemental forms, respectively. Ramsdell et al. (1994), after a review of several papers, concluded that the partitioning of iodine into different forms at 3,200 m in Ludwick’s experiments (Ludwick 1964) was consistent with the results of other measurements of iodine in the plumes from other stacks at the Hanford site (Ludwick 1967; Perkins 1963, 1964), with the partitioning of iodine in the plume following the Chernobyl accident (Aoyama et al. 1986; Bondietti and Brantley 1986; Cambray et al. 1987; Mueck 1988), and with the partitioning of natural iodine in the atmosphere (Voilleque 1979).

A number of references (Garland 1967; Clough et al. 1965; Chamberlain and Wiffen 1959) indicate that the majority of iodine is sorbed onto submicron-range aerosol particles. If the relative humidities at Hanford and Oak Ridge were comparable, the partitioning of elemental iodine into particulate form would occur in similar proportions at both places. Measurements of particle concentrations in the submicron range are not directly available from either Oak Ridge or Hanford for the period of interest (1944–1956). Recent measurements of PM10 (particles less than 10 μm in size) at Oak Ridge (18–24 μg m$^{-3}$ between 1992 and 1996; McElhoe 1998) and Hanford (32 μg m$^{-3}$; Neitzel et al. 1993) indicate comparable values.

Relative humidity at Oak Ridge is consistently higher than that at Hanford (Apostoaei et al. 1998). Since conversion of condensation nuclei to submicron size aqueous aerosols is enhanced by increased relative humidity in the atmosphere (Winkler 1973; Fitzgerald 1975), it is reasonable to assume that there would be increased sorption of iodine onto aerosols at Oak Ridge, resulting in a higher rate of transformation of the elemental form of iodine into the particulate form.

Moyers and Duce (1972) provide sorption mechanisms for the dissolution of gaseous I$_2$ and HI species into aqueous, submicron aerosols formed on sea-salt condensation nuclei in a marine atmosphere. They also provide speciation reactions for iodine in the aqueous phase that lead to the formation of I$^-$, IO$_3^-$, and I$_2$. Perkins (1964) had also observed the same three forms of iodine in the particulate matter. Presence of these forms of iodine in the atmosphere suggests that the mechanisms suggested by Moyers and Duce (1972) are also applicable to a nonmarine atmosphere.

Approach and input parameterization to address radioiodine chemistry

For estimating the concentrations of radioiodine in different chemical forms at ground-level at various locations (all of which at or beyond 3 km from the stack), the following approach is used, based on the background presented earlier:

- The distance to complete transformation, denoted by $x_c$, is treated as an uncertain parameter and is allowed to take a uniform distribution between 2 and 3.2 km to indicate the large uncertainty in the exact value of the distance at which the chemical transformation process is complete.
- If all of the $^{131}$I were to be released in elemental form and if the atmospheric conditions at Oak Ridge were identical to those at Hanford, it can be assumed that the proportion of $^{131}$I in each of the three chemical forms identified earlier would be
equal at all locations of concern. Because Oak Ridge has a significantly more humid climate than Hanford, the amount of particulate formed from the release of $^{131}$I in elemental form would be higher at Oak Ridge.

- It is assumed that $^{131}$I released in organic and particulate forms would not undergo any change because of its slow rate of photo-dissociation.
- Iodine chemistry is assumed to remain the same throughout the year with no seasonal patterns.
- Beyond $x_c$, the iodine in the three forms do not transform further into other forms, thus allowing the higher proportion of particulate form to be maintained beyond this distance. However, the proportions of the three forms change as a function of distance because of the different deposition (hence, removal) rates of the three forms of iodine. The term $g_{kj}$ is used to represent the fraction of iodine that is transformed from form $j$ to form $k$. Based on earlier discussions, if $k$ and $j$ take on the values 1, 2, and 3 for elemental, organic, and particulate forms, respectively, then $g_{12} = g_{13} = g_{23} = g_{32} = 0$ and $g_{22} = g_{33} = 1$. The only parameters, which then must be parameterized, are $g_{11}$, $g_{21}$, and $g_{31}$.
- For $g_{21}$, a uniform distribution was assumed with a central value of 0.33 and a large range (0.2−0.45). This assumption results directly from the literature data and the conclusions from Ramsdell et al. (1994), accounting for uncertainties. For $g_{31}$, a larger range of probable values was selected (0.2−0.8) to account for the increased humidity at Oak Ridge compared to that at Hanford, to account for the lack of confirmatory knowledge regarding the applicability of the mechanisms of Moyers and Duce (1972) to the non-marine atmosphere at Oak Ridge, and to reflect the range of measured values from the literature surveys discussed earlier. The upper-bound value was increased to 0.8 to account for generally high relative humidity at Oak Ridge and also to account for the very high relative humidity during summer nights at Oak Ridge (Holland 1953). The lower value was set at a level (0.2) above the measured values, also to reflect the generally higher relative humidity at Oak Ridge than at Hanford. Finally, $g_{11}$, the fraction that remains in elemental form, is calculated as $(1-g_{21}-g_{31})$.

**ATMOSPHERIC DISPERSION**

The SORAMI model

Part of the routine releases and a major portion of releases from the 1954 accident on April 29 occurred directly from building vents and openings. Wet and dry deposition can remove significant portions of contaminants from the plumes released at or near ground-levels (Horst 1984, 1977). Therefore, depletion by wet and dry deposition processes must be accounted for, in addition to the iodine speciation chemistry, by the model used to predict the transport of radiiodine in the atmosphere. The governing equation of a modified Gaussian plume model developed for this analysis is as follows:

$$C_{u,k} = \left[ \sum_{j=1}^{3} \left( \frac{Q_j}{Q_{0,j}} \right) \right] \cdot \frac{1}{2\pi \sigma_x \sigma_y u} \cdot e^{-\frac{2}{2\sigma_x^2}} \cdot \left( e^{-\frac{(z-h)^2}{2\sigma_z^2}} + e^{-\frac{(z+h)^2}{2\sigma_z^2}} \right),$$

where

- $C_{u,k}$ = the concentration of $^{131}$I in chemical form $k$ in air at location $(x,y,z)$ (Bq m$^{-3}$);
- $Q_{0,j}$ = the release rate of $^{131}$I in chemical form $j$ from a source located at (0,0,h) (Bq s$^{-1}$);
- $(Q/Q_{0,j})_{x\rightarrow x_{2}}$ = source depletion correction for the $j$th chemical form between downwind distances $x_1$ and $x_2$ (dimensionless);
- $g_{kj}$ = fraction of $^{131}$I released in chemical form $j$ that is transformed to chemical form $k$ within the distance $x_c$;
- $x_c$ = distance from source at which all chemical transformations are assumed to be complete (m);
- $h$ = the effective release height (= height of the physical stack + plume rise) (m);
- $\sigma_y$ = crosswind dispersion coefficient of a contaminant in air (m);
- $\sigma_z$ = vertical dispersion coefficient of a contaminant in air (m); and
- $u$ = average wind speed at the source elevation during the hour being simulated (m s$^{-1}$).

The source depletion correction terms were obtained from the formulation presented in Hanna et al. (1982) as shown below.

$$\frac{Q_j(x)}{Q_{0,j}} = \left[ \exp \int_{0}^{x} \frac{d\epsilon}{\sigma_z \exp(h^2/2\sigma_z^2)} \right]^{-\left(\frac{2}{\pi}\right)^{\frac{1}{2}}\left(\frac{V_{dep,j}}{u}\right)} ,$$

where

- $\epsilon$ = integration variable which defines the downwind distance from the source (m); and
- $V_{dep,j}$ = total deposition velocity of the $j$th chemical form of $^{131}$I (m s$^{-1}$).

Other quantities in eqn (2) have been defined earlier. It should be noted that $\sigma_z$ is a function of downwind distance in eqn (2). To derive the quantity $(Q/Q_{0,j})_{x\rightarrow x_{2}}$ in...
eqn (1), the following relationship was used:

$$\left( \frac{Q_j}{Q_{0,j}} \right)_{s, m} = \left( \frac{Q}{Q_{0,j}} \right)_{0, m}.$$

(3)

Other processes, not explicitly shown in the above equations but included in the new code, are the plume rise (based on Briggs 1969, 1975), vertical wind-speed profile for the estimation of wind speed at stack height, and distances to virtual point sources for ground-level or near-ground area sources (based on Turner 1970). The total deposition velocity in eqn (2) is estimated as the sum of the dry and wet deposition velocities. Estimation of wet deposition velocities and the values used for dry deposition velocities, for each chemical form of $^{131}$I, are presented in the next section.

**Input parameters**

Two separate analyses were conducted for the releases of $^{131}$I between 1944 and 1956. The first one was conducted for routine releases. These releases were represented by annual average estimates of source terms from the stacks and the building housing the radioactive lanthanum facility. For validation purposes, field data from 1967 to 1969 were used. The second analysis was conducted for the accidental release of 29 April 1954. Stack releases for the 1954 release lasted for just under 1 h, and direct releases from the building housing the RaLa processing facility lasted for 2.5 h. For the analysis of the accidental release, the SORAMI model was validated using a limited number of available short-term monitoring and release data.

**Routine releases**

The primary input parameters for the simulation of the SORAMI model are described below.

**Hourly wind speed, wind direction, and atmospheric stability.** The SORAMI model requires, for routine releases, the input of wind speed, wind direction, and atmospheric stability for each hour of simulation. Directly usable data in electronic form for the years 1944 to 1956 were not available even though summaries for some of these years are available (Holland 1953). Some data were available from strip-chart recordings for 1950 and later. Electronic data from the ORNL meteorological station MT2 (ORNL 1992) were available for ten years from 1987 to 1996. These data represented hourly measurements of the wind speed, wind-direction, and stability class at an elevation of 10 m above the ground, and they were statistically analyzed to generate the probabilities of obtaining specific values of wind speed ($u$), wind direction ($d$), and atmospheric stability ($s$) for a given hour of the day for a given month. The following assumptions were made for the statistical analysis:

- Significant variations of $u$, $d$, and $s$ occur on an hourly scale. Once the hourly variations in $u$, $d$, and $s$ are accounted for, seasonal variations in the values of $u$, $d$, and $s$ are handled on a monthly scale; i.e., within a month, each day is statistically the same as any other day in that month. The use of one month as the time scale to account for the seasonal variations of $u$, $d$, and $s$ is dictated primarily by the amount of available data. For example, for the month of January, the choice of the monthly time scale results in 31 data points for a given hour of the day (e.g., 8 a.m.) from one year's meteorological data set. For the 10-year period between 1987 and 1996, 310 (= 31 × 10) data points are available for that hour. Thus, 310 alternate values of $u$, $d$, and $s$ are available for the development of their probabilities of occurrence for a given hour in January. If the time scale is made finer than 1 mo, the number of data points is reduced and the uncertainties on the probabilities increase. Furthermore, there is no overwhelmingly compelling rationale for the meteorology of one week in January to be statistically different from that of another week.

- The probabilities of the occurrences of $u$, $d$, and $s$ during a given hour of the day for a given month are assumed to be mutually dependent. In other words $u$, $d$, and $s$ are treated as correlated parameters. Since, for a given hour, the meteorological data provide simultaneous measurements of these three parameters, the joint probability distribution for the three parameters for a given hour of the month in a specific month can be easily generated as follows:

$$p(u_{i,m}, s_{i,m}, d_{i,m}) = p(d_{i,m}) \times p(u_{i,m}|d_{i,m}) \times p(s_{i,m}|u_{i,m}, d_{i,m}),$$

(4)

where

- $i =$ subscript representing an hour of the day;
- $m =$ subscript representing the month;
- $p(u,s,d) =$ the joint probability of the simultaneous occurrence of events $u$, $s$, and $d$;
- $p(d) =$ probability of event $d$;
- $p(u|d) =$ probability of event $u$ given that $d$ has occurred;
- $p(s_{u,d}) =$ probability of event $s$ given that $u$ and $d$ have occurred;
- $u =$ wind speed;
- $d =$ wind direction; and
- $s =$ atmospheric stability class.

The following approach was implemented to sample $u$, $d$, and $s$ for a given hour of the day for a specific month:

- Wind speed was divided into six classes, 0–1.5 m s$^{-1}$, 1.5–3 m s$^{-1}$, 3–4.5 m s$^{-1}$, 4.5–6 m s$^{-1}$, 6–7.5 m s$^{-1}$, and 7.5–10 m s$^{-1}$.

The probabilities of obtaining specific values of wind speed ($u$), wind direction ($d$), and atmospheric stability ($s$) for a given hour of the day for a given month can be easily generated as follows:
m s\(^{-1}\), and >7.5 m s\(^{-1}\); wind direction was divided into 36 sectors of 10° each; and the atmospheric stability was divided into six discrete classes, A to F, according to the Pasquill-Gifford classification scheme.

Using the meteorological data from 1987 through 1996 (for each hour of the day for a given month), the cumulative joint probability distributions were developed for the simultaneous occurrences of a given wind speed class, wind direction class, and the atmospheric stability class using eqn (4). Specifically, the probability of the occurrence of each of the 36 wind directions was first developed for each hour of the day for each month of the year. This represents the first term in eqn (4). Next, the probability of the occurrence of each of the six wind speed classes for each wind direction class was developed. This represents the second term in eqn (4). Finally, the probability of the occurrence of each atmospheric stability class for each wind speed class and for each wind direction class was developed. This represents the last term in eqn (4).

- The probability density functions (pdfs) representing the three terms on the right-hand side of eqn (4), developed in the previous step, were then sampled using a Latin Hypercube Monte Carlo sampling scheme (Iman and Shortencarier 1984). First, the pdf for wind direction (d) was sampled for a given hour of day for a given month. For that wind direction, the pdf for the wind speed (u) was sampled for the same hour of the day for the same month. Finally, the pdf for the stability class (s) was sampled for the just-sampled wind speed and wind direction. This process is repeated 400 times for each hour of the day for the 12 calendar months.
- The 12 sets of 400 values of implicitly correlated d, s, and u for each hour of day for each month were then used to generate the meteorological input files for the SORAMI model.

**Lateral and vertical dispersion coefficients.** For the rural simulation mode, the SORAMI model uses Briggs’ approximations (Hanna et al. 1982) to the Pasquill-Gifford curves (Turner 1970) for the estimation of \(\sigma_L\) and \(\sigma_z\), the lateral and vertical dispersion coefficients, respectively. An uncertainty factor was assigned to each of these estimates. The uncertainty factor was sampled using a uniform distribution between 0.67 and 1.5 for each stability category. The uncertainty factor is used to multiply the values of \(\sigma\) and \(\sigma_z\) estimated by the Briggs’ approximation for each stability class. The upper and lower bounds of the uncertainty factor were developed such that the lower and upper bounds of one class are very close to the expected value of the adjacent classes.

**Mixing heights.** When the vertical extent of the plume becomes equal to the mixing height at a given downwind distance from the source, the contaminant is assumed to be uniformly distributed in the vertical dimension below the mixing height. Mixing heights representative of different stability classes were obtained from Holzworth (1972) for the Oak Ridge region. Afternoon conditions were assumed to represent stability classes A and B (unstable categories), morning conditions were assumed to represent stability classes E and F (stable categories), and the values for stability classes C and D (neutral category) were derived from the values for stable and unstable conditions. Sufficiently large ranges of parameter values were used to represent the uncertainty in the knowledge of mixing heights. Table 1 presents the mixing height data used in the analysis. For each of the 400 alternate realizations of u, d, and s, the mixing height is sampled from the appropriate distribution in Table 1.

**Release information such as emission rate, height and diameter of the stack, and temperature and velocity of the effluent releases.** A unit emission rate for \(^{131}\)I was prescribed for each hour to obtain the ground-level air concentration per unit release (\(y/Q\)), which can then be multiplied by the actual emission rate to estimate the hourly air concentration for each given hour at a desired location. Table 2 presents the values of other parameters used in the analysis. As mentioned earlier, Stack 3020 was used for \(^{131}\)I releases between 1944 and 1950, and Stack 3039 was used between 1950 and 1956. However, because of the close proximity and elevations of the two stacks, it was decided to treat all releases as having occurred from Stack 3020.

Ambient temperature was assumed to remain at 293°K. The temperature data are used in the model to calculate the buoyancy plume rise from the stack release as a function of the temperature difference between the ambient air and stack effluents. Because effluents were released to the stack at ambient conditions, the contribution of buoyancy to the plume rise would be small compared to the momentum contribution.

**Dry and wet deposition velocities.** Dry and wet deposition processes contribute to the depletion of radioiodine from the plume. They also contribute to the accumulation of radioiodine on the pasture grass and increase in ground-level air concentrations. Depletion of radioiodine from the plume is controlled by deposition processes that must account for the presence of all major

<table>
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<tr>
<th>Stability Class</th>
<th>Minimum mixing height (m)</th>
<th>Maximum mixing height (m)</th>
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</thead>
<tbody>
<tr>
<td>A, B (unstable)</td>
<td>1,000</td>
<td>2,000</td>
</tr>
<tr>
<td>C, D (neutral)</td>
<td>500</td>
<td>1,500</td>
</tr>
<tr>
<td>E, F (stable)</td>
<td>300</td>
<td>650</td>
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Table 1. Mixing height data used for analysis. A uniform distribution was chosen between the minimum and maximum values.
landforms and features in the plume path. Since the SORAMI model uses a Gaussian plume approach, an average value of the deposition velocity representative of the study region is required.

Information on the values of dry deposition velocities for the Oak Ridge region were obtained from the National Oceanic and Atmospheric Association’s Atmospheric Turbulence and Diffusion Division (ATDD) at Oak Ridge and from the Oak Ridge National Laboratory. Data from experimental measurements reported by Heinemann and Vogt (1980) were also used. For the dry deposition of iodine in elemental form, a uniform distribution between 1 and 6 cm s\(^{-1}\) was selected as representative of a study region consisting of forest canopies and pastureland. This range of values is consistent with measurements of other highly reactive chemical species, such as gaseous HNO\(_3\), in the Oak Ridge region. For the deposition of iodine in the particulate form, a log-uniform distribution between 0.05 and 0.5 cm s\(^{-1}\) was selected for the study region. This range of values is consistent with measurements on submicron-size particles, such as sulfate aerosols, in the Oak Ridge region. For the organic form, a log-uniform distribution between 0.001 and 0.05 cm s\(^{-1}\) was selected, as the organic form of iodine is extremely nonreactive.

The wet deposition velocity is estimated as a product of the washout ratio, \(WR\) (expressed as \(\text{m}^3\text{air L}^{-1}\text{rain}^{-1}\)), and the hourly precipitation rate, \(P\) (expressed as \(\text{mm h}^{-1}\)). Washout ratio is defined as the ratio of the concentration of iodine in rainwater to its concentration in air at ground level. The washout ratio is assumed to depend on the amount of rain during each rain event, and each rain event is assumed to last for a one-hour period. Thus, a rainfall lasting several hours would be treated as several events, each lasting for an hour. The washout ratio is calculated using the following empirical equation (Slinn 1978; NRC/CEC 1994):

\[
WR = \frac{WR_1^h \times P^{-s}}{24},
\]

where

\(WR_1^h\) = the washout ratio for a 1 mm hourly rain;

\(P\) = the precipitation rate per rain event (\(\text{mm h}^{-1}\)); and

\(s\) = a rain exponent, an empirical parameter (unitless) that accounts for the observed decrease in the washout ratio with increasing rates of precipitation.

\[WR_d = WR_1^{NCI} \times E^{-a}\] (6)

where

\(WR_d\) = the washout ratio on a daily basis;

\(WR_1^{NCI}\) = the washout ratio for a 1 mm daily rain;

\(E\) = the precipitation rate per rain event (per day) (\(\text{mm} \text{d}^{-1}\)); and

\(a = 0.7\) = a rain exponent, an empirical parameter [unitless] that accounts for the observed decrease in the washout ratio with increasing rates of precipitation.

Since parameter values for \(WR_1^{NCI}\) and \(a\) were readily available (NCI 1997), the washout ratio, \(WR_1^{NCI}\), was derived by equating the net daily deposition rate from eqn (5) with that derived from eqn (6) to obtain the following equation:

\[
WR_1^h = \frac{1}{24} \cdot \frac{(WR_1^{NCI} \times E^{-a})}{P_a},
\]

where \(P_a\) is the average rainfall for the day (=E/24).

Washout ratio is a measure of the solubility of the gas, and for gases, essentially all raindrops attain their equilibrium concentration within 10 m of fall (Slinn 1978). The distribution of \(WR_1^{NCI}\) (for elemental form of \(^{131}\)I) was assumed to be log-triangular with a minimum of 2,100, mode of 5,000, and a maximum of 10,700 \(\text{m}^3\text{air L}^{-1}\). This range is based on a review of NCRP recommendations (NCRP 1993), on an expert elicitation performed for the US Nuclear Regulatory Commission and the European Community (NRC/CEC 1994), on NCI (1997), and on Slinn (1978). The distribution of \(WR_1^{NCI}\) for the particulate form of \(^{131}\)I is assumed to be log-triangular between 1,000 and 5,400 \(\text{m}^3\text{air L}^{-1}\) with a mode of 2,500 \(\text{m}^3\text{air L}^{-1}\). This range is based on measurements made in the USA in the aftermath of the Chernobyl accident as summarized by Richmond et al. (1988) and on an expert elicitation performed for the US Nuclear Regulatory Commission and the European Community (NRC/CEC 1994). The distribution of \(WR_1^{NCI}\) for organic forms of \(^{131}\)I is assumed to be log-triangular between 4 and 18 \(\text{m}^3\text{air L}^{-1}\) with a mode of 8 \(\text{m}^3\text{air L}^{-1}\) (Slinn 1978; NRC/CEC 1994; NCI 1997).

The rain exponent \(s\) was also considered an uncertain variable. Based on the available information (Brenk and Vogt 1981; Schwarz 1985, as cited by NRC/CEC 1994; NRC/CEC 1994), a relatively large range (0.2–0.6; uniform distribution) was chosen for this parameter.

Hourly precipitation rate. The hourly precipitation rates were developed from the ten years of hourly rainfall data available from the measurements conducted at the ATDD station on Illinois Avenue in Oak Ridge between 1986 and 1996. The 1995 data were not complete and, therefore, not used. Since direct rainfall data for the years 1944 to 1956 were not available in electronic form, it was

<table>
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<th>Table 2. Stack release parameters.</th>
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<td>Stack</td>
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<tr>
<td>3020</td>
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<td>3039</td>
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\(^{1}\) Meyers, T., NOAA’s Atmospheric Turbulence and Diffusion Division, Oak Ridge, TN 37830. Personal communication; 1998.

\(^{2}\) Lindberg, S., Oak Ridge National Laboratory, Oak Ridge, TN 37830. Personal communication; 1998.
decided to generate hourly rainfall data. Using the ATDD data from 1986 to 1996, hourly rainfall estimates were generated for each hour of the day for a given month. It was not possible to generate each hour’s data for every single day of the month because ten years’ data would provide only ten measured values for each hour and day of a month. Therefore, focusing on a “representative day of a month” allowed the use of 310 (31 d × 10 y) measured values for every hour of a day in January. The next step in the process was to develop a strategy to address the uncertainty associated with the measurements. Previous methods to generate rainfall data have relied on a Markov Chain approach (Richardson and Wright 1984; Richardson 1981; Schroeder et al. 1985). The Markov Chain approach models a stochastic process using the assumption that the event at a given time step \((t+1)\) is dependent only on the previous time step \((t)\).

The 310 sets of hourly information available for the month of January were analyzed to determine the frequency with which it rained a certain amount in any given hour given the fact that it did or did not rain during the previous hour. Statistical analysis of the 10 y data provided the probabilities for each month of the occurrence of a given amount of rain given information whether or not rain occurred in the previous hour. Using an arbitrary initial condition of no rainfall during the last hour of December, 400 alternate sets of hourly rainfall data were then generated sequentially for each hour of the “representative day” for each of the 12 months from January through December. Sampling of the conditional probabilities for each hour was conducted using a Latin Hypercube-based Monte Carlo methodology described in Iman and Shortencarier (1984).

Statistics of the final rainfall data sets were compared with those of the observed rainfall for 30 y presented in Holland (1953) and those of the 10 y of data between 1986 and 1996, and are presented in Apostoaei et al. (1998). The comparisons of measurements to modeling results showed good agreement between the modeled data set and the actual measurements of precipitation at Oak Ridge during recent years. The 95% subjective confidence intervals of modeled rainfall were seen to be larger than those of the measured data.

Release geometry for releases from buildings. The building that housed the radioactive lanthanum processing facility was approximately 29 m × 30 m in area (Clinton Laboratory Drawing No. CL-706D-9 dated 18 November 1944). The building was 12 m tall at its highest point (Clinton Laboratory Drawing No. CL-706-D-54 dated 8 January 1945). Since releases from the building occurred from vents and openings at various levels within the building, an average, constant release height of 10 m was chosen. The entire release is treated as a single area source with a diameter that marginally exceeds the longest diagonal of the building. The diameter was treated as an uncertain parameter with a uniform distribution varying from 29 m to 44 m. The area source is converted into a virtual point source at an elevation of 10 m and upwind of the building. The distance from the building to the virtual point source is estimated on the basis of the sampled values of source diameter at the building and the atmospheric stability, both expressed as uncertain variables. The distance from the virtual source as well as the vertical extent of the plume at the location of the building (downwind from the virtual point source) then automatically emerge as uncertain variables because of the treatment of the prescribed release height of 10 m as the height of the virtual source.

Ramsdell and Fosmire (1998) show that the building wake effects are important for predictions in the first few hundred meters from the source (typically 200–300 m). They also show that most building wake models in use, such as Fuquay model (Gifford 1968), NRC Regulatory Guide 1.145 (U.S. NRC 1982), Ramsdell’s model (Ramsdell 1990), and Huber’s model (Huber 1984), converge to open-terrain solutions obtained from Gaussian plume models beyond 300–400 m from the source. Since the receptor locations of interest for this study are beyond 3 km from the source, it is not necessary to model either the building wake effects from the Building 706-D (housing the RaLa processing facility) or the effects from the presence of other, near-source, buildings.

1954 accident

The accident on 29 April 1954 started sometime before 5:00 p.m. and resulted in releases of \(^{131}I\) until 7:00 p.m. Releases from the stack ceased at about 5:30 p.m., while direct releases from the building continued to occur until 7:00 p.m. Meteorological parameters were obtained from an inter-company (Oak Ridge National Laboratory) correspondence from P. R. Guinn to J. C. Hart (Guinn 1954). These parameters represent data at half-hour intervals from the ORNL Health Physics Group Meteorological Station 012, which is a mid-valley location within Bethel Valley where the building is located (Holland 1953; Hillsmeier 1963). Table 3 presents the data from Guinn (1954).

Wind direction. Each wind direction from Table 3 was interpreted as representing a 22.5°C sector from which the wind blew towards the monitoring station. Each reading would, therefore, represent a full sector of 22.5°C, and the maximum uncertainty associated with each reading would be ± 11.25°C on either side of the designated direction. For each of the five half-hour periods of the accident, a maximum uncertainty of ± 11.25°C was accordingly assigned about the readings recorded in Table 3, and the parameter was sampled using a uniform distribution.

Wind speed. To address the uncertainty associated with a given integer value of the wind speed in Table 3, the lower limit of wind speed was chosen as 0.5 subtracted from the integer value, and the upper limit of wind speed was chosen as 0.5 added to the integer value, and the range was sampled using a uniform distribution.
atmospheric stabilities were estimated from the stability classification procedure promulgated by the Nuclear Regulatory Commission and presented in Brenk et al. (1983). Vertical and horizontal dispersion coefficients (1983). Vertical and horizontal dispersion coefficients

Atmospheric stability and dispersion coefficients. The temperature differences presented in Table 3 were used to evaluate the atmospheric stability during a given hour. The temperature difference was interpreted as representing the difference in measured air temperatures at two elevations; atmospheric stabilities were estimated from the stability classification procedure promulgated by the Nuclear Regulatory Commission and presented in Brenk et al. (1983). Vertical and horizontal dispersion coefficients ($\sigma_z$ and $\sigma_y$, respectively) are directly affected by the stability class. Uncertainties in $\sigma_z$ and $\sigma_y$ were assigned in the same manner as presented earlier for routine releases.

Other parameters. Other parameters are treated in the same manner as described earlier for routine releases. Rainfall data, obtained from strip-chart records for 29 April 1954 from the ORNL Health Physics Group Meteorological Station 012 in the Bethel Valley, indicated that there was only one period, between 6:00 and 7:00 p.m., during which any precipitation (0.67 in.) occurred on April 29, 1954. The amounts of rainfall in the two half-hour periods between 6:00 and 7:00 p.m., however, are unknown. However, Guinn (1954) states that the rain started at 6:15 p.m. This means that there was a 15-min period in the first half-hour period and the full 30-min period during the second half-hour period during which the net precipitation occurred. Assuming that the rainfall intensity was fairly uniform during the 45-min period, the first 15 min, between 6:15 and 6:30 p.m. would account for one-third (0.22 inches) of the entire rainfall (0.67 inches). It was decided to use this value as the most likely value of the rainfall in the period between 6:00 and 6:30 p.m. A triangular distribution with a large range spanning 0.11 to 0.44 inches was assigned to account for the uncertainty in the knowledge of the rainfall intensity during this period. Rainfall for the second period between 6:30 and 7:00 p.m. was then obtained as the difference between the total rainfall and the rainfall between 6:00 and 6:30 p.m. An uncertainty of $\pm 10\%$ was assigned, along with a triangular distribution with a most likely value of 0%, to the total rainfall for the entire period to account for any errors in measurements. Estimation of the wet deposition velocities of individual chemical forms for the accidental release is conducted in the same manner as described for routine releases using half-hour duration for a rain event (instead of the 1-h duration used for routine releases).

Model validation for routine releases

Annual average release rates from stacks (Bethel Valley Stacks 3039, 3020, and 2026 and Melton Valley Stacks 7911 and 7512) in two adjacent valleys in the Oak Ridge Reservation, separated by a distance of about 1.8 miles, were available for the 1967–1969 period. Measured ground-level air concentrations from 9 monitoring locations (Fig. 1) surrounding the ORNL facility were also available for this period. Using the hourly meteorological data, generated according to the discussions presented earlier and constant release rates matching the reported releases in 1967, 1968, and 1969 (Table 4), the annual average concentrations were estimated for the 9 monitoring locations using the SORAMI model.


Table 3. Weather conditions between 4 and 8 p.m. on 29 April 1954 (Guinn 1954), from the ORNL Health Physics Group Meteorological Station 012.

<table>
<thead>
<tr>
<th>Time (p.m.)</th>
<th>Temperature (°F)</th>
<th>Relative humidity (%)</th>
<th>Temperature difference (°F)</th>
<th>Direction (from)</th>
<th>Speed (miles/hour)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4:00</td>
<td>78</td>
<td>57</td>
<td>−2</td>
<td>W</td>
<td>9</td>
</tr>
<tr>
<td>4:30</td>
<td>73</td>
<td>95</td>
<td>−2</td>
<td>S</td>
<td>12</td>
</tr>
<tr>
<td>5:00</td>
<td>74</td>
<td>70</td>
<td>−1</td>
<td>SE</td>
<td>8</td>
</tr>
<tr>
<td>5:30</td>
<td>75</td>
<td>66</td>
<td>−1</td>
<td>S</td>
<td>8</td>
</tr>
<tr>
<td>6:00</td>
<td>73</td>
<td>69</td>
<td>−1</td>
<td>S</td>
<td>6</td>
</tr>
<tr>
<td>6:30</td>
<td>65</td>
<td>90</td>
<td>−1</td>
<td>S</td>
<td>15</td>
</tr>
<tr>
<td>7:00</td>
<td>65</td>
<td>95</td>
<td>−1</td>
<td>SP</td>
<td>12</td>
</tr>
<tr>
<td>7:30</td>
<td>64</td>
<td>100</td>
<td>−1</td>
<td>DI</td>
<td>3</td>
</tr>
<tr>
<td>8:00</td>
<td>65</td>
<td>100</td>
<td>−1</td>
<td>DI</td>
<td>1</td>
</tr>
</tbody>
</table>

a The temperature difference was interpreted as representing the difference in measured air temperatures at two elevations. The two elevations were presented as 4 ft and 183 ft in Holland (1953), and as 5 ft and 145 ft in Hillsmeier (1963).

b Direction indeterminate.
Measurements at the monitoring stations involved the adsorption of gaseous iodine in an activated charcoal canister located downstream from a Vose LB-5211 particulate filter (Oakes et al. 1981). The measured concentrations were converted to annual average ground-level concentrations of $^{131}$I in air and reported by Binford et al. (1970). Table 5 presents the annual average concentrations of $^{131}$I at the nine monitoring stations. Table 5 also presents the distance and direction of each monitoring station from the RaLa processing facility.

It should be noted that all releases from the Bethel Valley Stacks were treated as having occurred from Stack 3039, while that from the Melton Valley was dominated by Stack 7911. Detailed rationale is provided in Apostoaei et al. (1998) for the use of Stack 3020 as surrogate for Stack 3039; the most compelling reason was that the average absolute percent difference in the predictions from the two stacks were within 5% of each other (Apostoaei et al. 1998). Stack 7911 is located 1,300 m east and 1,070 m south of Stack 3039.

As discussed earlier, the hourly meteorological input for a given month is representative of any day of the month. Therefore, 400 alternate realizations of the ratio of air concentration at a receptor point to the source term ($\chi/Q$) were used. These realizations were developed using eqns (1)-(4). From these, 400 alternate 1-d average values of the $\chi/Q$ ratios for each hour of the day for each of the twelve months were developed using eqns (1)-(4). From these, 400 alternate 1-d average values of the $\chi/Q$ ratios

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**Table 4.** Amounts of $^{131}$I released from Bethel and Melton Valley Stacks between 1967 and 1969 (Binford et al. 1970).

<table>
<thead>
<tr>
<th>Year</th>
<th>Releases from Bethel Valley Stacks (10$^{10}$ Bq)</th>
<th>Releases from Melton Valley Stacks (10$^{10}$ Bq)</th>
<th>Total releases (10$^{10}$ Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Stack 2026</td>
<td>Stack 3039</td>
<td>Stack 3020</td>
</tr>
<tr>
<td>1967</td>
<td>11 (13%)</td>
<td>64 (78%)</td>
<td>0.6 (1%)</td>
</tr>
<tr>
<td>1968</td>
<td>2.2 (6%)</td>
<td>27 (72%)</td>
<td>0.2 (1%)</td>
</tr>
<tr>
<td>1969</td>
<td>23 (39%)</td>
<td>30 (51%)</td>
<td>0.1 (0%)</td>
</tr>
</tbody>
</table>
Table 5. Measured annual average ground-level concentrations of 131 I.

<table>
<thead>
<tr>
<th>Ambient 131I monitoring station; distance and direction from the RaLa Stack</th>
<th>Measured concentrations (10^-4 Bq m^-3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kerr Hollow (HP-31); 11.4 km, 49°</td>
<td>1967 1968 1969</td>
</tr>
<tr>
<td>Midway Gate (HP-32); 10.6 km, 37°</td>
<td>10.4 5.18 8.51</td>
</tr>
<tr>
<td>Gallaher Gate (HP-33); 7.4 km, 274°</td>
<td>7.03 3.33 7.03</td>
</tr>
<tr>
<td>White Oak Dam (HP-34); 3.1 km, 189°</td>
<td>8.51 4.44 8.88</td>
</tr>
<tr>
<td>Blair Gate (HP-35); 7.0 km, 292°</td>
<td>9.62 8.88 7.40</td>
</tr>
<tr>
<td>Turnpike Gate (HP-36); 8.8 km, 14°</td>
<td>3.70 3.70 5.18</td>
</tr>
<tr>
<td>Hickory Creek Bend (HP-37); 7.6 km, 119°</td>
<td>3.33 3.33 3.70</td>
</tr>
<tr>
<td>Gallaher Bend (HP-38); 5.6 km, 31°</td>
<td>2.96 3.70 4.07</td>
</tr>
<tr>
<td>Townsite (HP-39); 13.8 km, 78°</td>
<td>8.51 6.66 9.25</td>
</tr>
<tr>
<td>Midway Gate (HP-32); 10.6 km, 37°</td>
<td>6.29 5.18 4.44</td>
</tr>
</tbody>
</table>

* Direction is measured clockwise as the angle between the line connecting the monitoring station and the RaLa Stack and the line going north from the RaLa Stack. North from the RaLa Stack 302° is treated as 0°.

were developed for each month by averaging over the 24 h. These 400 values were then randomly sampled with replacement, 31 times, and the 31 values were summed to represent one alternate realization of the 31-d time-integrated χ/Q ratio for the month of January. The same process was repeated 400 times to represent 400 alternate realizations of the 31-d time-integrated concentrations for January. The process was repeated for each calendar month. For each month, the lower bound, central estimate, and upper bound of the 95% subjective confidence interval and arithmetic mean values were developed from the 400 alternate realizations. The lower bound, central estimate, and upper bound values of the 95% subjective confidence interval and the arithmetic means of the χ/Q ratios for all the 12 months were summed up and divided by 365 d to obtain, respectively, the lower bound, central estimate, and upper bound values of the 95% subjective confidence interval and the arithmetic means of annual average χ/Q ratios. Concentrations at each location of interest were then estimated using eqn (1).

Implementation of the source depletion formulation within the SORAMI model, based on eqns (1)-(3), was first independently verified using the example problems provided in Horst (1984, 1977). Since amounts of iodine released in individual chemical forms during the 1967–1969 period are not known, it was decided to estimate the concentrations of iodine at the 9 receptor locations on the basis of three different assumptions: (1) all of the iodine was released in elemental form; (2) all of the iodine was released in the organic form; and (3) all of the iodine was released in the particulate form.

Concentrations were predicted directly at the 9 locations of interest using the SORAMI model. Concentrations were also predicted at the nodes of a polar grid along each of the equally spaced 16 compass directions starting with N and going clockwise 22.5° to NNE, NE, and so on until NNW. Along each direction, 25 equally spaced nodes, 1.5 km apart, were selected. Concentrations at each of the 9 monitoring stations were then estimated from the predicted concentrations at the four nodes surrounding the location of the monitoring station using an interpolation scheme. The interpolation scheme used a weighted approach, with weights calculated as the inverse of the square of distance from the monitoring station (where the prediction was needed) to each of the four surrounding nodes. The concentration at a monitoring station was then estimated as the weighted sum of the product of the normalized weight and predicted concentration at each of the four surrounding nodes.

Predicted estimates of the central value (median) of the annual average ground-level concentrations of 131I in air at each of the nine monitoring stations were then divided by the measured annual average concentrations presented in Table 5 to obtain the P (predicted)/O (observed) ratio for each of the nine monitoring stations. The geometric means of the P/O ratios for each of the monitoring stations using estimates for all three years are presented in Table 6. It should be noted that the directly calculated values of geometric mean P/O ratios in Table 6 were estimated using stack parameters listed in Table 2 for Stack 3020 and using relevant values of the stack height (87.3), the stack diameter (1.5 m) and the stack exit velocity (13 m s^-1) for Stack 7911 (Binford et al. 1970). The interpolated values of geometric mean P/O ratios in Table 6 were estimated using stack parameters listed in Table 2 for Stack 3020 for both Stacks 3020 and Stack 7911.

The geometric mean P/O ratios in Table 6 do not show significant differences between the interpolated and directly calculated estimates of concentrations at the nine monitoring stations. The wet and dry deposition rates are highest for the elemental form of iodine followed by those for the particulate and organic forms. Therefore, removal of elemental 131I from the plume by dry and wet deposition would result in the lowest concentrations of 131I at a receptor location if all of the 131I were released in elemental form. The highest concentrations of 131I at a receptor location would result if all of the 131I were released in organic form, because of its extremely low deposition velocity. The geometric means of the P/O ratios presented in Table 6 clearly reflect these observations. It is clearly seen from Table 6 that most predictions fall within a factor of two of the observations.

These results show that the SORAMI model provides reasonable predictions (within a factor of 2) for most locations, and, therefore, its use for the reconstruction of doses and risks is justified. There are a few locations where the model consistently provides under- or overestimates of the measured concentrations. However, these predictions are still within a factor of two.

Model validation for short-term releases

Release data on 131I were available for three weekly periods in the 1960’s along with reported measured weekly average concentrations of 131I in air at one location for each of the releases. During the week ending 17 January 1965, 1 × 10^11 Bq of 131I was released (Lasher 1965), and the maximum weekly average concentration of 131I was measured at Station HP-34 (Fig. 1).
as 0.0074 Bq m\(^{-3}\) (ORNL 1966). About 90% of the release was from Stack 3039. During the week ending 15 May 1966, \(3.7 \times 10^{10}\) Bq of \(^{131}\)I was released (Lasher 1966), and the maximum weekly average concentration of \(^{131}\)I was measured at Station HP-38 (Fig. 1) as 0.0044 Bq m\(^{-3}\) (ORNL 1967). About 94% of the release was from Stack 3039, 3.5% was from Stack 2026, and 2% was from Stack 3020. Finally, during the week ending 9 July 1967, \(1.5 \times 10^{10}\) Bq of \(^{131}\)I was released (Lasher 1967d), and the maximum weekly average concentration of \(^{131}\)I was measured at Station HP-33 (Fig. 1) as 0.0081 Bq m\(^{-3}\) (ORNL 1968). About 74% of the release was from Stack 3039, 21% was from Stack 2026, and 5% was from Stack 3020. 

Since the meteorological parameters were already developed for each of the specified months, it was possible to estimate the ground-level concentrations of \(^{131}\)I using the meteorological data for each month separately. The results of this validation analysis are presented in Table 7. Again, the chemical form(s) in which iodine was released at the source was not known. Therefore, three separate analyses were conducted, as in the case of the validation for routine releases, with the assumption that all of the iodine was released (a) in elemental form, (b) in organic form, or (c) in particulate form. For this exercise, all of the releases were assumed to have occurred from Stack 3020. The 400 alternate values of 7-d, time-integrated \(\chi/Q\) ratios were developed from the hourly meteorological data in a similar manner to that used to develop the 31-d time-integrated ratios for routine releases; from these, the weekly average \(\chi/Q\) ratios were estimated. These ratios were then multiplied by the constant release rate using eqn (1), to obtain the 400 alternate realizations of the weekly average concentration of \(^{131}\)I in air.

Results for January 1965 and May 1966 clearly show that the measured concentrations are contained within the lower and upper bounds of the 95% subjective confidence intervals of the predicted concentrations. However, the predicted concentrations grossly underestimate the measured concentrations for July 1967. The reasons for this are not clear. Results for routine releases,
presented in Table 6, also indicate that the SORAMI model typically under-predicts at the Gallaher Gate Station (HP-33). Because more short-term monitoring and release data were not available, further short-term validation could not be performed. However, it is clear that the model can be used for predicting ground-level concentrations of $^{131}$I from the accident on 29 April 1954.

**RESULTS**

**Routine releases**

Distributions of ground-level $\chi/Q$ ratios were developed in the same manner as described for the validation study. Using these $\chi/Q$ ratios, the source terms for each year between 1944 and 1956 (Apostoaei et al. 1998), and eqn (1), concentrations of $^{131}$I in the three forms were estimated for each of the 400 nodes. Contours of the lower- and upper-bound estimates of the 95% subjective confidence interval of the annual average ground-level concentrations of total $^{131}$I ($^{131}$I in all 3 forms) are presented in Figs. 2 and 3, respectively, for 1953. Because the source terms were generated on an annual average basis and the $\chi/Q$ ratios used are applicable to all years, the shape of the contours would be very similar to those in Figs. 2 and 3 for all years. For that reason the annual average source terms for the three forms of $^{131}$I are

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**Fig. 2.** Contours for the lower bound estimates (2.5 percentiles) of the 95% subjective confidence interval for the annual average, ground-level concentrations of total $^{131}$I ($^{131}$I in all three chemical forms) in air (Bq m$^{-3}$). The actual concentration at a given location is highly likely to be greater than the value presented in the figure.
not presented in this paper; the reader is referred to Apostoaei et al. (1998) for a detailed discussion of source term development for this dose reconstruction study.

The contours indicate that the concentrations decrease most rapidly as a function of distance along the northwest direction from X-10. It should be noted that the northeast-southwest direction represents the direction of predominant wind flow. The wind flows in these directions are also typically of larger magnitude than those along the northwest and southeast directions. Reasons for the lower concentrations towards the northwest from the X-10 facility than those towards the southeast are not entirely clear although inspection of the annual average wind rose did indicate that the lowest frequency of winds occurs towards the northwest directions from the ORNL. A comparison of these concentration contours with the P/O ratios in Table 6 for Gallaher Gate and Blair Gate indicates that there may be a model-bias towards under-prediction by a factor of 2 towards the west and west-northwest directions. However, the uncertainties in air concentrations do not dominate the uncertainty in the overall estimates of dose and risk to an exposed individual (Apostoaei et al. 1998). Furthermore, the presence of potential bias was determined from just two monitoring stations. Therefore, a factor to correct for potential bias is not included to the estimates of dose and risk. Similarly, Table 6 indicates the potential for a bias of a factor of 2 towards over-estimation for the Hickory

Fig. 3. Contours for the upper bound estimates (97.5 percentiles) of the 95% subjective confidence interval for the annual average, ground-level concentrations of total $^{131}\text{I}$ ($^{131}\text{I}$ in all three chemical forms) in air (Bq m$^{-3}$). The actual concentration at a given location is highly likely to be less than the value presented in the figure.
Creek Bend and Gallaher Bend Stations in the east-southeast and east directions. Again, a factor to correct for the potential for bias is not carried through to the estimates of dose and risk.

Sensitivity analyses conducted for this dose reconstruction study (Apostoaiei et al. 1998; Hoffman et al. 1996) indicated that uncertainty in the atmospheric dispersion component contributed at the most 15% to the uncertainty in the concentration of $^{131}$I in cow’s milk and less than 3% to the estimated risk of thyroid cancer. Since the uncertainty in air concentrations do not dominate the uncertainty in the overall estimates of dose and risk to an exposed individual, a detailed sensitivity analysis for the atmospheric dispersion component was not conducted. Based on past experience, uncertainty in the characterization of source term, $Q$, is expected to be the single-most dominant contributor to the uncertainty in air concentration.

**1954 accident**

Transport simulations for each of the five half-hour periods of the accident that occurred between 4:30 and 7:00 p.m. on 29 April 1954 were conducted using the accident-specific input parameters presented earlier. Results were first developed as $x/Q$ ratios for each half-hour period and for each node. These half-hourly ratios were multiplied with half-hourly release rates (Apostoaei et al. 1998) from appropriate sources (stack or building) and for appropriate chemical forms, using eqn (1). From the hourly concentrations, the time-integrated concentrations of $^{131}$I in all three chemical forms were developed. Time-integrated concentrations were estimated for 464 nodes on a polar grid. The 400 nodes on the polar grid are identical to the ones used in the analysis for routine releases. An additional sixty-four nodes were added to include more locations within the first 2 km to compare the results of the predictions with the onsite radiation measurements made immediately after the accident, as shown in Fig. 4. The lower-, median, and upper-bound estimates of the 95% subjective confidence intervals of the total time-integrated concentrations of $^{131}$I are presented in Figs. 5, 6, and 7, respectively. It should be noted that the concentrations in these figures are representative of emissions from both the stack and the building housing the radioactive lanthanum processing facility.

Two distinct plumes developed in response to the prevailing wind directions during the five half-hour periods of the accident. The northwest direction of contaminant transport is corroborated by the radiation measurements shown in Fig. 4. However, the transport...
towards the north cannot be corroborated by the measurements shown in Fig. 4, primarily because there were no monitoring stations towards the north. The movement of the plume towards the north, as predicted by the SORAMI model, is, therefore, plausible.

It was noted that the 95% subjective confidence intervals span several orders of magnitude for some locations. The primary reason for this was found to be the uncertainty assigned to the wind direction. An uncertainty range of $11.25^\circ$ was assigned to each measured value shown in Table 3. This results in a wide range of cross-wind distances ($y$ in eqn 1) from the plume centerline (along the wind direction) for a given location. The exponential term containing the $y$ and $\sigma$, in eqn (1) accordingly takes a wide range of values from 0 (when the plume centerline is far from the location of interest) to some large finite value (when the location of concern is well within the plume). As seen from Figs. 5, 6 and 7, the directions of plume travel during the five half-hour periods are such that most of the locations of concern do not fall within the plume and some are exposed only to the edge of the plume.

**CONCLUSIONS AND FUTURE REFINEMENTS**

Accurate reconstruction of health impacts from past releases require that uncertainties in the knowledge of
input parameters be propagated through the model and that the model structure incorporate all process-level details. In order to address these requirements for the dose reconstruction at Oak Ridge, a unique Gaussian plume model, SORAMI, was developed and satisfactorily validated using available field data. The reader is referred to Apostoaei et al. (1998) for a discussion of the health impacts from exposures to the releases of $^{131}$I to the atmosphere as a result of the Oak Ridge operations.

For the routine releases, the atmospheric dispersion modeling in this analysis used hourly specifications of all data except the source term. In the next phase of the analysis, it is recommended that the entire analysis from release to health risk be conducted at the scale of one hour.

Better understanding of the spatial characteristics of the wind direction in the Oak Ridge Reservation will be necessary to reduce the uncertainty in the predicted range of time-integrated concentrations of $^{131}$I in air from the accidental release of 29 April 1954. Eckman et al. (1992) used a network of meteorological monitoring stations to analyze the wind data in the Oak Ridge region. In that study the meteorological data were recorded at 15-min intervals. Correlations among the same parameters measured at different stations and among different parameters measured at the same stations can be developed and used along with the meteorological data available from strip charts from various stations in Oak Ridge during the time of the accident. This information can be used to

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**Fig. 6.** Contours of the central estimates (50 percentiles) of the 95% subjective confidence interval for the time-integrated, ground-level concentrations of total $^{131}$I ($^{131}$I in all three chemical forms) in air (Bq h m$^{-3}$) following the accident on 29 April 1954.
establish an interpolated wind-field structure for the Oak Ridge region that prevailed during the time of the accident. This methodology would allow the determination of the distribution of probable directions of the travel of every puff of $^{131}$I released to the atmosphere at specific times during an accidental release.

A combined modeling approach that addresses transport of individual puffs while modeling the Gaussian growth of the puff would allow tracking of several such puffs, each with an alternate path of plume travel. This would be a significant improvement over the current analysis, provided that the development of a probability-based wind-field structure is feasible and defensible.

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