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Monte Carlo-based dispersion modeling of off-gassing releases from the fumigant metam-sodium for determining distances to exposure endpoints

D.A. Sullivan*, M.T. Holdsworth, D.J. Hlinka

Sullivan Environmental Consulting, Inc., 1900 Elkin Street, Suite 240, Alexandria, VA, USA

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Abstract

Fumigants, such as metam-sodium, can help control pests in the soil and improve the quality and yields for a wide range of crops. Enhanced control of field loss, or off-gassing, to the atmosphere is an important means of promoting fumigant efficacy and sound environmental management of fumigants. Metam-sodium has been used for over 40 years and is one of the most widely used fumigants in the United States. This paper describes a modeling method, the fumigant emissions modeling system (FEMS), which was developed to promote more realistic assessment of downwind concentrations in the vicinity of an applied field. Two models of the US Environmental Protection Agency, i.e., the industrial source complex short-term (ISCST3) model and the toxic modeling system short-term (TOXST) model are used, in conjunction with a pre-processing program, PCRAMMET, and the FEMS programming code to provide a Monte Carlo treatment for all key model inputs. Start times for applications are triggered on a Monte Carlo basis consistent with the annual application frequency. Typically, 1000–10,000 years of applications are simulated, outputting data to display average exceedances per year of selected concentration endpoints as a function of distance from the edge of the treatment zone. A sensitivity analysis is displayed for key meteorological variables, showing uncertainty in emission rates to be a significant input parameter. Some comparisons also are made between ambient concentrations and modeled personal and indoor exposures, showing perspective on the benefits of the indoor environment to buffer peak concentrations.

Keywords: Monte Carlo; Dispersion modeling; Randomization; Uncertainty; Metam-sodium; Methyl isothiocyanate; ISCST3; TOXST; Fumigants; Pesticide; Air quality; Agriculture; Exposure assessment

1. Introduction

Growers use fumigants, such as metam-sodium, to reduce pest pressures on crops from weeds, soil-borne diseases, and nematodes. With the phase-out of methyl bromide because of concerns involving stratospheric ozone depletion, there is enhanced interest in alternative fumigants. Metam-sodium, along with 1,3 dichloropropene and chloropicrin, are currently the major alternatives to methyl bromide, alone or in combination, in the form of pre-plant treatments. Fumigants can help to control pests in the soil, thereby, increasing the quality and yields in a wide range of crops, including carrots, strawberries, tomatoes, potatoes, melons, peppers, peanuts, cut flowers, and many other important crops.

A great deal of field research has been conducted on metam-sodium over the past several years, which has been directed towards the minimization of volatile losses from the treatment zone of the principal degradation product, i.e. methyl isothiocyanate (MITC) (Merricks 2001, 2002a,b; Sullivan et al., 2000a,b, 2001).

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*Corresponding author.
E-mail address: sull_env@ix.netcom.com (D.A. Sullivan).

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The principal benefits of this research strategy are as follows: (1) reduced volatilization loss acts to increase the dose in the treatment zone and promote more efficient, and potentially more effective, use of the pesticide and (2) reduced volatilization acts to reduce health and safety concerns.

The primary focus of this paper is on the evaluation of downwind airborne exposures to MITC from metam-sodium applications. There are two primary factors that need to be analyzed to estimate these exposures. First, emission rates as a function of time need to be estimated, including coverage of the substantial diurnal variability and day-by-day trends in off-gassing rates that have been observed in many field studies (Merricks 2002a,b, 2001; Sullivan et al., 2000a,b, 2001; Van den Berg et al., 1999, Van den Berg et al., 1993; Woodrow, et al., 2001). Second, these emission rates need to be evaluated via dispersion modeling in order to estimate airborne concentrations of MITC in the vicinity of applied fields, including the identification of maximum expected concentrations as a function of averaging time and distance from the edge of an applied field.

The companion paper, *Control of off-gassing rates of methyl isothiocyanate from the application of metam-sodium by chemigation and shank injection*, summarizes a method to estimate expected emission rates as a function of time after the completion of an application. The emission fitting procedure is based on extensive ambient monitoring of MITC airborne concentrations in the range of 150–800 m from the edge of applied fields, followed by statistical analysis to fit the emissions data to the observed concentration fields. Dispersion modeling is used to bridge the measured concentrations with expected emission rates, normalized to an emission rate of $1\,\mu g\,m^{-2}\,s^{-1}$.

A field applied by metam-sodium is a complex source of air pollutants that has substantial variability in off-gassing rates throughout the diurnal cycle and decreasing amplitudes in peak daily emissions with time. The characteristics of the releases often can be visualized as a damped wave that generally is attenuated each day, rapidly approaching background levels, usually within 2–4 days after an application. The characteristics of this type of source introduce several important complications. Examples of these complications, followed by a recommended solution for each, are summarized below.

- *Emission rates involve transformed chemicals with complex release characteristics*. Emission rate variability over time is best addressed at this time using empirical methods, similar to an approach developed by the California Department of Pesticide Regulation (Johnson et al., 1999). *Recommended solution*: Until more definitive emissions data become available to specifically address a wider range of soil types and conditions, it is appropriate to represent the release pattern of all fields using conservative emission rates that err on the side of overestimating ambient concentrations for each application method. Most of the empirical studies that were used as the basis for the emissions assessment for metam-sodium were based on summertime conditions in Bakersfield, California, which is expected to produce upper-end exposures because of high air and soil temperatures and sandy soil types. As more field studies become available to represent a wider range of soil types and conditions, the conservatism of using data from hot and arid climates can be reduced. More field studies also would provide a stronger basis to support the development of physical/chemical models for soil dose and off gassing.
- *Intermittent source*. Metam-sodium is applied only one time every 1–4 years at typical fields. With only several days per year of off-gassing rates per application that are significantly above background levels, this source of air pollution is much different than traditional industrial sources. If the standard modeling approach of assuming constant emission rates were to be used, for example, the distribution of concentrations would be unrealistically skewed toward higher concentrations, and would substantially overstate the impacts from a fumigation source, such as a metam-sodium application. *Recommended solution*: The preferred approach to model an infrequent source is to evaluate the source on a Monte Carlo basis, setting the probability for the start of an application based on operational practice, e.g. statistically one time per year or less. Once an application is initiated on a Monte Carlo basis, the modeling analysis proceeds through a sequential series of empirically-based emission rates, with updates of emission rates each hour for the first four days post application.
- *Need to consider the range of possible meteorological and emission conditions*. Even though only several days of impacts are typically associated with an application, it is important to consider the range of potential meteorological conditions and emission rates that could be anticipated throughout a long period of record. *Recommended solution*: Through establishing distributions for emission rates and key meteorological inputs to the modeling to account for the uncertainty in each parameter, the expected range of input parameters can be specifically represented and incorporated specifically into the modeling analyses to provide risk managers with sufficient perspective to make informed decisions.
- *Identify input parameters with the greatest uncertainty*. A challenge in any dispersion modeling analysis involves identifying the sensitivity of key model inputs. *Recommended solution*: Through the use of distributions of key model inputs, uncertainty

can be addressed by isolating key variables one at a time, and holding the selected variable at the best estimated value for each hour, i.e. not drawing from the applicable distribution. Sensitivity testing on this basis for emission rates, wind speed, wind direction and atmospheric stability presents a more definitive description of the principal areas of uncertainty in this analysis.

2. Technical approach

The fumigant emissions modeling system (FEMS) uses several of the US Environmental Protection Agency programs. The dispersion modeling approach used in FEMS is based on a standard US dispersion model, i.e. the US Environmental Protection Agency's (EPA) industrial source complex short-term (ISCST3; US EPA, 1995) model, which is used in conjunction with a post-processing model designed to support Monte Carlo treatments of the modeled concentrations, i.e. the EPA TOXST model (Grosch et al., 1993). By drawing input variables (wind speed, wind direction, atmospheric stability, and emission rates) from a Monte Carlo pre-processing program incorporated into FEMS, the ISCST3 model is used to model multiple-year simulations (e.g. 200 simulated years of operation) based on a 5-year base dataset of hourly meteorological data. In this system, 200 base years of input files are modeled specifically in ISCST3 to create the dataset for the TOXST analysis. TOXST is then used to draw samples of application periods to match the selected number of applications per year. Running 1000–10,000 simulations in FEMS is generally recommended because an analysis comparing results using different simulation periods (up to 20,000 simulation years) showed that 1000–10,000 simulations produce reasonably stable results (see Fig. 1).

FEMS was designed to model 200–100,000 simulated years of operation per model run based on 1–5 years of hourly meteorological data. Since FEMS was designed to account for the uncertainty in both the emissions and meteorological input data, 200 annual input files are output from ISCST3 and input to TOXST as the basis to draw long-term simulations. One limitation of the current version of TOXST is the non-random seed used to begin the Monte Carlo sequence; therefore, each of the 200 annual input files to TOXST is based on sequential meteorological files extracted from the 5-year base meteorological data set with a randomly selected starting day for each year (8760 hourly records for a non-leap year and 8784 records for a leap year). This approach maintains the sequential nature of the meteorological data set while promoting a random selection of days that are analyzed in each simulation.

The uncertainty in the emissions data and meteorological inputs are represented in distributions, from which the input files to ISCST3 are drawn. The TOXST model is used to select the specific start days for the applications at a field, and also adjacent independent fields if desired, using an independent Monte Carlo basis for the start times of each application.

3. Identification of distributions of input data to ISCST3

If there were perfect correlation between the measured and modeled concentrations, the emission rates would

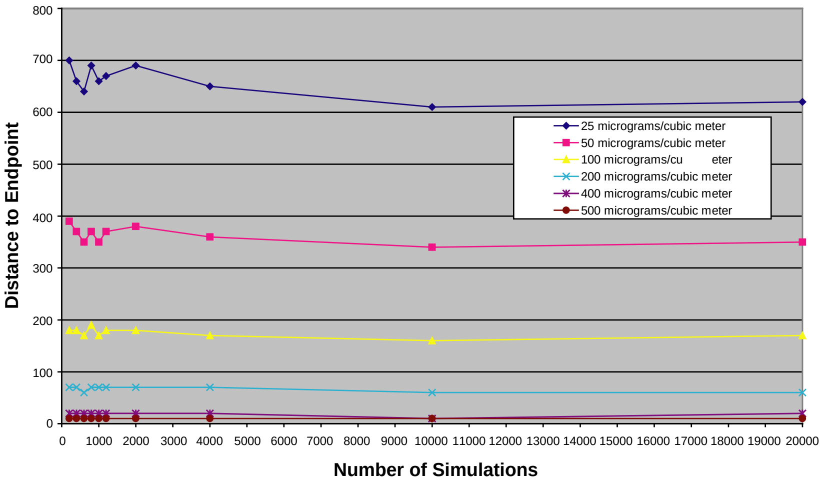

Fig. 1. Comparison of the number of simulated years versus MITC endpoints using the Kern 2001 intermittent seal study emission rates (Merricks, 2002), 4-h averaging, and 1.49 exceedances.

be known with certainty for the specific days used to conservatively represent emission rates as a function of time. Since there is scatter in the relationship between measured and normalized modeled concentrations; however, there also is uncertainty in specifying the slope, and thereby, the estimated emission rates. By computing the standard error of the estimate in logarithmic space, a distribution of emission rates for each 4-h empirical period was computed, providing a more appropriate distribution of emission rates (Cullen and Frey, 1999; Bortnick and Stetzer, 2002).

Distributions of emission rates in original units were developed using the Land method to estimate confidence intervals for natural log-normalized medians (Land, 1971; American Mathematical Society, 1975). These values estimate the confidence intervals of the estimated best-fit emission rates for each 4-h sequential monitoring period used in the analysis.

The following procedure was used to compute emission distributions based on the measured air quality data collected during field studies (Merricks, 2002a,b, 2001), which were conducted in accordance with Good Laboratory Practice (GLP) requirements (US EPA, 1989). In order to display the distribution of emission rates in the original, non-natural log-transformed units, the Land procedure was used to compute "*H*" values that were needed to estimate the distribution in the original units.

Best estimate for Emission Rate in original units is

$$E_{50} = \mathrm{EXP}(y + 0.5 \times \mathrm{SE}_\mathrm{b}^2).$$

Confidence values for emission rate were computed as follows: (Gibbons and Coleman, 2001; Berthouex and Brown, 1994)

$$E_\alpha = \mathrm{EXP}(y + 0.5 \times \mathrm{SE}_\mathrm{b}^2 + (H_\alpha \times \mathrm{SE}_\mathrm{b}/(n-1)^{0.5})),$$

where E_α is the emission rate to the confidence level α in original units (non-natural log transformed), y the median emission rate in natural log scale ($\mu\mathrm{g\,m^{-2}\,s^{-1}}$), SE_b the standard error of the estimate of the median emission rate in natural log scale, H_α the "*H*" coefficient from Land tables for selected confidence intervals from 2.5% to 97.5% confidence, and, n the number of paired observations of measured and modeled concentrations.

The final emission rates for each period in the ISCST3 modeling analysis were selected by Monte Carlo sampling from the emission distributions. By using a randomized selection of emission rates each hour, drawing from the sequential emissions distributions for each application method, the uncertainty in the emission fitting procedure was incorporated into the analysis. An example of the emission rate distributions for a selected range of confidence intervals is shown in Table 1 and Fig. 2. The values in Table 1 are representative of the emissions distributions computed for a chemigation application with intermittent water seals (Merricks, 2002b). This range in confidence intervals (2.5, 5.0, 10, 25, 40, 50, 60, 75, 90, 95, and 97.5 percentile confidence levels) was selected to cover a large range in emission rates. In order to avoid artifacts in the extremes of the distribution, the lower and upper bounds were set at 2.5 and 97.5 percentile levels (Hanna et al., 1998). In future refinements, the emissions distributions for each period may be parametrically defined rather than discretely computed as in Table 1.

4. Meteorological data

A different set of analyses had to be performed on the wind speed, wind direction, and stability data to account for the uncertainty in the meteorological conditions input to the modeling analysis. Generally, the National Weather Service data inventory of weather monitoring stations is used as a basis for the 5-year meteorological dataset required as input into FEMS (on-site meteorological data for such a long period of time generally is not feasible). The most representative meteorological monitoring station should be used to minimize differences in meteorology between the off-site monitoring station and on-site conditions at the field in question. Anemometer and wind vane exposure heights in the range of 6–10 m are most consistent with the standard input requirements for a ground-level source such as a fumigation application. Once a representative 5-year data set is acquired, the procedure to account for uncertainty within the hourly averages of the meteorological data is incorporated into 200 years of meteorological files (40 sets of 5 simulated years of meteorological data based on drawing from the estimated distribution that represents the uncertainty in the measured values). These 200 files are created on a Monte Carlo basis from the 5-year hourly datasets to account for the uncertainty in the meteorological input parameters (wind speed, wind direction, and stability).

FORTRAN programs were developed to account for the uncertainty in wind direction, wind speed, and atmospheric stability, similar to the development of emissions distributions. These key meteorological terms are treated as Monte Carlo variables in order to more fully account for the expected range of model input conditions. In each case, a probability density function was parametrically defined (type of distribution and quantitative range for the 95th percentile confidence; Hanna et al., 1998) based on expert elicitation methods, which are commonly used to incorporate uncertainty in terms of probability distributions when direct data to define the probability distribution are scarce or unavailable (Cullen and Frey, 1999). Appropriate limits were set to avoid artifacts associated with the tails of the distribution, with the Monte Carlo draw being limited to the 95th percentile range centered on the median. Wind

Table 1
Emission distribution analysis for the Kern 2001 chemigation intermittent seal study[a]

		Kern 2001 chemigation intermittent seal study emissions results (Merricks, 2002b)													
Day	Period	2.50%	5.00%	10.00%	25.00%	40.00%	50.00%	60.00%	75.00%	90.00%	95.00%	97.50%	Standard error	Pairs	Emissions method employed
1	5:00–9:00	7.36	7.64	7.98	8.41	8.85	9.28	9.88	10.48	11.07	11.76	12.47	0.45	> = 3	Log-Normalized
1	9:00–13:00	6.74	6.86	6.99	7.14	7.3	7.45	7.63	7.81	7.99	8.17	8.34	0.19	> = 3	Log-Normalized
1	13:00–17:00	48.82	53.5	59.74	70.76	81.78	92.8	124.06	155.32	186.57	245.43	325.94	1.15	> = 3	Log-Normalized
1	17:00–21:00	11.48	12.31	13.37	15.04	16.7	18.36	21.81	25.27	28.72	34	40.36	0.89	> = 3	Log-Normalized
1	21:00–1:00	4.61	4.79	5	5.28	5.56	5.84	6.24	6.65	7.05	7.53	8.03	0.46	> = 3	Log-Normalized
2	1:00–5:00	2.87	3	3.15	3.36	3.56	3.77	4.07	4.38	4.68	5.05	5.44	0.53	> = 3	Log-Normalized
2	5:00–9:00	4.58	4.7	4.85	5.02	5.2	5.38	5.6	5.83	6.05	6.29	6.53	0.31	> = 3	Log-Normalized
2	9:00–13:00	3.24	3.28	3.32	3.37	3.42	3.47	3.52	3.58	3.63	3.69	3.74	0.13	> = 3	Log-Normalized
2	13:00–17:00	46.48	49.7	53.78	60.11	66.45	72.78	85.72	98.66	111.59	131.1	154.43	0.85	> = 3	Log-Normalized
2	17:00–21:00	14.78	15.35	16.03	16.92	17.82	18.72	20.01	21.31	22.6	24.14	25.75	0.46	> = 3	Log-Normalized
2	21:00–1:00	3.03	3.15	3.29	3.47	3.66	3.84	4.11	4.37	4.64	4.95	5.28	0.46	> = 3	Log-Normalized
3	1:00–5:00	2.86	2.89	2.93	2.97	3.01	3.05	3.09	3.14	3.18	3.23	3.27	0.12	> = 3	Log-Normalized
3	5:00–9:00	2.58	2.61	2.65	2.69	2.73	2.77	2.81	2.86	2.9	2.94	2.98	0.13	> = 3	Log-Normalized
3	9:00–13:00	1.54	1.57	1.62	1.67	1.72	1.77	1.83	1.9	1.96	2.03	2.09	0.27	> = 3	Log-Normalized
3	13:00–17:00	11.37	11.67	12.06	12.52	12.97	13.43	14.01	14.58	15.16	15.78	16.4	0.32	> = 3	Log-Normalized
3	17:00–21:00	21.09	21.74	22.49	23.46	24.42	25.39	26.68	27.96	29.25	30.69	32.15	0.36	> = 3	Log-Normalized
3	21:00–1:00	4.57	4.74	4.94	5.2	5.47	5.73	6.09	6.45	6.81	7.22	7.65	0.44	> = 3	Log-Normalized
4	1:00–5:00	4.26	4.37	4.5	4.65	4.81	4.96	5.15	5.34	5.53	5.74	5.94	0.29	> = 3	Log-Normalized
4	5:00–9:00	1.56	1.62	1.69	1.77	1.86	1.95	2.07	2.19	2.31	2.45	2.58	0.43	> = 3	Log-Normalized
4	9:00–13:00	2.01	2.05	2.1	2.15	2.2	2.25	2.31	2.37	2.43	2.49	2.55	0.21	> = 3	Log-Normalized
4	13:00–17:00	3.56	3.67	3.81	3.98	4.16	4.33	4.56	4.79	5.03	5.29	5.56	0.38	> = 3	Log-Normalized
4	17:00–21:00	6.84	7.16	7.55	8.09	8.63	9.17	10.02	10.87	11.71	12.79	13.94	0.57	> = 3	Log-Normalized
4	21:00–1:00	5.37	5.49	5.63	5.79	5.96	6.12	6.32	6.52	6.73	6.94	7.15	0.25	> = 3	Log-Normalized
5	1:00–5:00	2.48	2.58	2.69	2.84	2.96	3.13	3.33	3.53	3.73	3.97	4.2	0.45	> = 3	Log-Normalized

[a] Periods are 4-h averaged and sequential based on 96 h (4 days) of downwind measured concentrations, with a sampling frequency of 4 h. Note that pairs are valid if the measured and modeled concentrations are both greater than 0.1 $\mu g\,m^{-3}$.

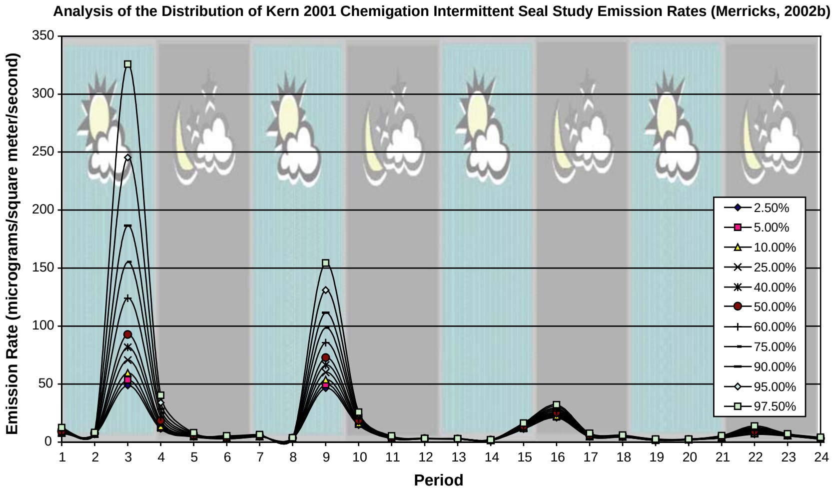

Fig. 2. Analysis of the distribution of Kern 2001 chemigation intermittent seal study emission rates (Merricks, 2002b). Note that our uncertainty peaks each day at 13:00–17:00 (convective peak conditions). These high emissions with good dispersion may not translate into higher impacts because favorable dispersion conditions are present.

Table 2
Assumed distributions for meteorological parameters (Hanna, 1998)[a]

Term	Shape of distribution	95% confidence range (±1.96 sigma)	Lower limit	Upper limit
Wind speed	Natural log normal	$\pm 1.5\,\mathrm{m\,s^{-1}}$	0	NA
Wind direction	Normal	$\pm 45\,\mathrm{ubar^{-1}}\ (\mathrm{m\,s^{-1}})$	0	360
Stability class	Normal	±1	1	6

[a] The wind direction and stability class terms, which were not log-normalized, were not affected by negative numbers because the wind direction cross-over at 360 was accounted for and A stability (1) was not further reduced.

speed is assumed to have a natural log-normalized distribution while wind direction and stability are assumed to be normally distributed (Hanna et al., 1998). Table 2 summarizes the type of distribution (normalized or log-normalized) and the 95% confidence range (±1.96 standard deviations). A randomized starting day for each of the 40, 5 year meteorological data-sets is used to ensure that the selection sequence in the Monte Carlo sampling is random each run.

Meteorological data can vary substantially between regions; therefore, the FEMS program has an option to allow the user to choose meteorological data from up to 10 different meteorological regions. The most representative 5 years of meteorological data can be selected for the region of interest.

5. Set-up and execution of ISCST3

The ISCST3 input files include a link to randomized hourly emission files for the year and randomized meteorological data (wind speed, wind direction, stability), and a receptor grid placed in the format of rings centered on the field at distances, such as the default set of 100, 200, 300, 500, 750, 1000, 1500, 2000, 2500, and 3000 m. Natural logarithmic interpolation is used to calculate the distances to exposure endpoints between these receptor rings (see Results section). An example of the field layout and receptor grid (entire receptor grid and a close-in grid) is provided in Figs. 3 and 4. Area source treatments of the emitting field are used with hourly emission rate files to produce the output concentrations at each receptor.

Hourly emission rates are established by drawing from the empirically fitted emissions distributions calculated for the 24, 4-h periods associated with each application and sealing method of interest, which are cycled continuously in ISCST3 and applied over the 200 yearly runs that are the basis for the long-term simulations. In addition, the model user can account for a fraction of the maximum application rate when modeling reduced application rates, e.g. 37.5 gallons per acre rate (50% of maximum 75 gallons per acre rate). It is assumed as a default that the emission rates can be

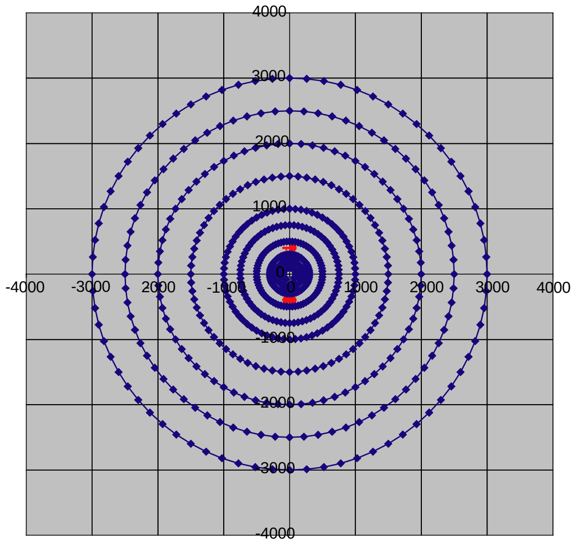

Fig. 3. Sample field layout and receptor grid (entire 100–3000 m polar grid).

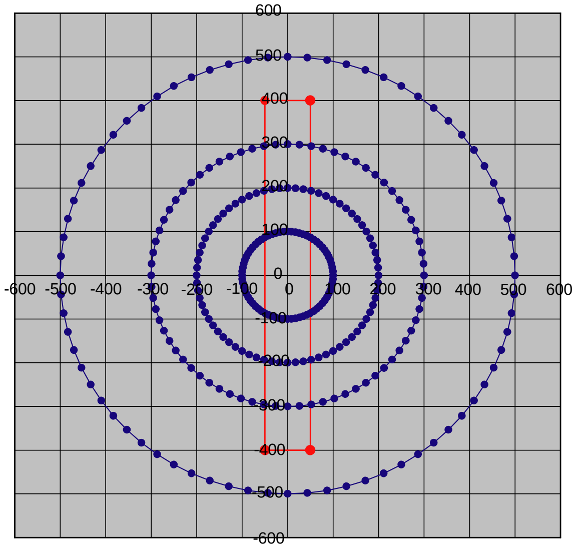

Fig. 4. Sample field layout and close-in receptor grid (100, 200, 300 and 500 m polar grid).

scaled on a linear basis as a function of the maximum application rate.

The ISCST3 air dispersion model is run using receptor heights of 1.5 m and a default height of 0.0 m for the emitting source. Model output is needed for 1-h average concentrations, which are input to the TOXST program. The averaging times that can be used in TOXST are limited to 1, 2, 3, 4, 6, 8, 12, and 24 h.

6. Set-up and execution of TOXST

Once the ISCST3 runs are completed, the TOXST program is run for all 200 ISCST3-generated TOX files (one per year) to determine the average number of exceedances per year at each receptor for a particular concentration threshold. In addition to the TOX files, the TOXST program requires the user to input several variables including the exposure endpoint concentrations (up to six endpoint concentrations can be used; e.g., 10, 50, 100, 500, 1000, and 2000 $\mu g\,m^{-3}$), the number of total years to be simulated (e.g., 1000 total simulations equals 5 simulations for each of the 200 TOX files), the averaging time (e.g., 4 h), the probability of an application sequence beginning at any 1 h (e.g., 0.0001154 for one 96-h application sequence per year), the length of time the source is active (e.g., 96 h), and the background concentration of the pollutant (e.g., simplified to 1 $\mu g\,m^{-3}$ for these analyses). Several studies performed to date have shown the background MITC airborne concentrations, for example, were on the order of 1.0 $\mu g\,m^{-3}$ averaged over an entire year (Seiber et al., 1999; Lompoc Interagency Work Group, 1999; California Air Resources Board, 1994). On this basis, the background concentrations are orders of magnitude lower than typical peak measured concentrations.

TOXST is used to initiate the start of an application on a Monte Carlo basis. Current analyses are based on probabilities for one application per year. The analysis also can be set up for different number of applications per year, e.g. 0.25–4. If adjacent fields are analyzed, each one would be treated as an independent source, as appropriate. Each application start time, once triggered through Monte Carlo selection in the TOXST model, is diurnally matched to the sequence established by the applicable field study. The full emissions loop is executed in sequence until each of the 24, 4-h periods of the 96-h total off-gassing period per application is evaluated in TOXST. Once the full emissions cycle is completed, the emissions in TOXST are effectively set to zero until the next time an application is triggered and the process is repeated again.

7. Results for modeling ambient concentrations

A FORTRAN program was developed to calculate the maximum distance to meet specific endpoint concentrations based on the TOXST output. Maximum concentrations as a function of downwind distance typically are evaluated using the 1.49 exceedance level as the point of compliance (i.e. there will not be more than an average of one exceedance per year of the specific concentration endpoint under review at any receptor

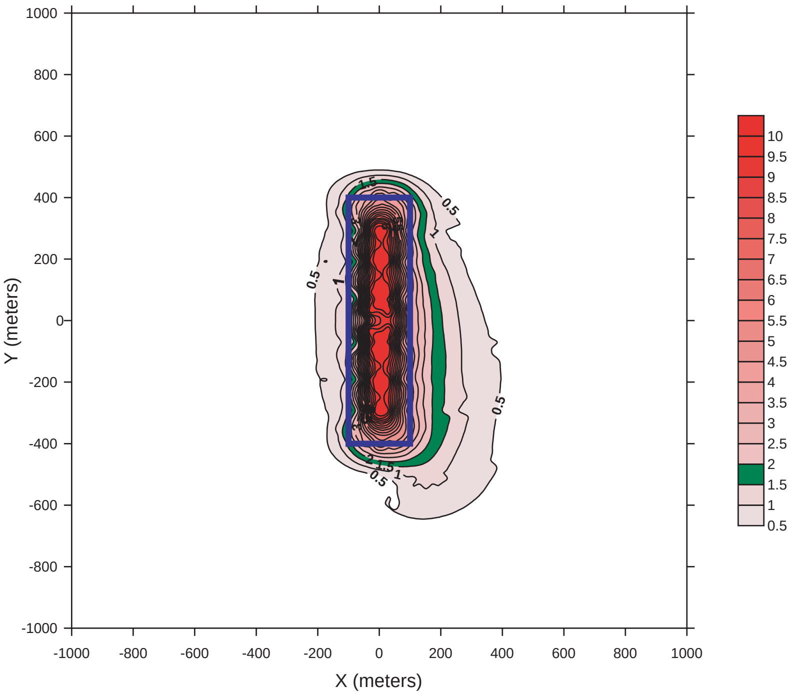

Fig. 5. TOXST analysis of the number of exceedances per year with distance from the edge of a field using a hypothetical 100 μg m^{-3} endpoint, 4-h averaging, and 1000 simulated years of randomized emission rates and meteorological data except atmospheric stability for the Kern 2001 metam-sodium intermittent seal application study (Merricks, 2002).

beyond the identified distance). Fig. 5 shows an example of an isopleth analysis of average exceedances per year using 4-h averaging, a hypothetical 100 μg m^{-3} concentration threshold, and an emission rate sequence developed previously based on a chemigation application of metam-sodium using chemigation and intermittent water sealing (Merricks, 2002b).

8. Evaluation of uncertainty and variability

If model input parameters, such as emission rate, wind speed, wind direction, and atmospheric stability, could be measured with complete accuracy, and for a sufficiently long number of years to fully capture the variability in each parameter, then there would be no reason to select input parameters for each hour from probability density functions. Of course, in reality these inputs are uncertain and incompletely cover the full range in variability. The best estimated values are generally input to deterministic modeling analyses for each hour. The actual value of an input at any specific hour in a probabilistic modeling analysis, on the other hand, could be higher or lower than the best-estimated emission rate or measured meteorological parameter.

How much uncertainty[1] to the model output is contributed by each input parameter? A series of sensitivity analyses were modeled by selecting, on a Monte Carlo basis, all key model input parameters from their respective distributions using just the best-estimated value for one selected input parameter. The results of the sensitivity analyses for a hypothetical scenario[2] are presented in Table 3 for key variables.

The randomization of emissions rates allows for consideration of upper-end emissions, which can be substantially higher than typical values (see Fig. 2). Non-randomization would tend to decrease peak short-term exposures if there are significant spikes in the emission rate distributions. As emissions distributions include a wider range in emissions and averaging time decreases (e.g., 1-h averaging as compared with 4-h

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[1] For the end use of these analyses, it is appropriate to combine uncertainty and variability into the term uncertainty. There are insufficient data to accurately separate these terms for fumigant applications of this nature.

[2] The scenario was based on running the Kern 2001 intermittent seal study (Merricks, 2002b) emission rate distribution, 4-h averaging, 1000 simulated years, and 1.49 exceedances per year.

Table 3
Sensitivity analysis table

Scenario	Distance (m) to 50 $\mu g\,m^{-3}$ endpoint where average exceedances per year = 1.49
Full set of randomized distributions	350
No randomized distributions	340
All randomized distributions except emission rate	330
All randomized distributions except wind direction	370
All randomized distributions except wind speed	370
All randomized distributions except atmospheric stability	350

averaging), the sensitivity to emissions would be expected to increase.

As shown in this example, the greatest uncertainty in the fitted emission rates is during the mid to late afternoon periods. Two reasons for this observation are hypothesized as follows:

(1) During the afternoon period winds often are stronger and steadier in terms of wind direction flow. This often can lead to smaller sample sizes that meet the condition that paired measured and normalized modeled concentrations are both $> 0.1\,\mu g\,m^{-3}$, and
(2) The Gaussian distribution assumed in the ISCST3 dispersion model may not represent vertical dispersion as accurately as during non-convective periods. This may be a more significant factor for sampling sites farthest down-wind, e.g. in the 500–800 m range from the field.

The most realistic consideration of all parameters would be to use randomized emission rates, randomized wind direction, randomized wind speed, and non-randomized stability (to reduce potential nocturnal bias).

8.1. Indoor air assessment

Since most people spend the majority of their time indoors, actual exposures would be expected to be quite different than those computed when implicitly assuming a hypothetical person is always stationary on their "front porch" breathing ambient air. This factor can be especially important for short-term averaging times on the order of several hours or less. Typical infiltration rates can act to buffer the peak ambient concentrations. As a means of providing perspective, an indoor component was added to this fumigant modeling approach. Indoor exposures were assessed based on Chaloulakou and Mavroidis (2002).

8.2. Indoor air concentration

The following equation calculates indoor concentrations based on several factors:

$$C_i = ((2 - b_1)/(2 + b_1)) * C_i + ((2 * t)/(2 + b_1)) * \times (C_{oi} + C_o)/2)$$

k	Mixing factor (typically ranges from 0.3 to 1.0. A default value of 0.65 was used in this analysis)
am	Infiltration flow rate (air changes per hour). A value of 2.4 was used if inside with partially open windows or 1.0 if windows were closed (Chaloulakou and Mavroidis, 2002).
b_1	(k) (am)
C_i	initial indoor air concentration ($\mu g\,m^{-3}$)
C_{oi}	initial outdoor air concentration ($\mu g\,m^{-3}$)
C_o	present outdoor air concentration ($\mu g\,m^{-3}$)
t	timing interval (h)

The probability of the windows of a selected structure being open or closed varies on a seasonal basis. Probabilities are assigned for: (a) the probability of an individual being inside the structure (e.g. place of residence or work)[3] and (b) the probability of the windows being open or closed. Assignment of these terms in the model is made on an hourly basis.

The execution of the modeling analyses for indoor exposures is conducted sequentially. First, the ambient concentrations based on ISCST3 are linked with the indoor model to update the concentrations (indoor and personal) on an hourly basis.[4] The computed ratio of indoor/ambient concentrations and personal/ambient concentrations are then used to compute scalars to adjust the hourly emissions input to ISCST3 to correspond to those needed to match the indoor and personal concentrations. These new emission rates are then run through the ISCST3 dispersion model and the results input to TOXST. The resulting average number of exceedances per year can be output for: (a) ambient exposures only, (b) indoor exposures only, (c) personal

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[3] This method assumes the subject is at one location throughout the day or conservatively assumes that both the work location, if indoors, and indoor residential exposures are the same.

[4] Personal exposure is an estimate of exposures that would be measured by placing a lapel sampler intake on an individual. The personal exposures are simply based on the indoor or ambient exposures for the hour depending on whether the subject is inside or outside. Refer to Table 5 for a summary of the assumptions for the indoor and personal exposure estimates.

Table 4
Assumptions for times spent indoors versus diurnal and seasonal periods

	Windows open (%)	Windows closed (%)	Indoors (%)	Outdoor "on porch" (%)	Outdoor far away or in transit[a] (%)
Daytime spring/summer/fall	20	80	87	3	10
Daytime winter	1	99	89	1	10
Nighttime spring/summer/fall[b]	10	90	99	1	0
Nighttime winter	0	100	100	0	0

[a] Note: For outdoor time periods, exposure levels were conservatively set to the 1 $\mu g\,m^{-3}$ background level.

[b] Nighttime is defined as from midnight—6 a.m.

Table 5
Comparison of ambient, indoor, and personal exposure distance to endpoint runs for intermittent sealed chemigation study for a hypothetical 100 $\mu g\,m^{-3}$ endpoint (Merricks, 2002b)

	Averaging times (h)		
	1	4	8
Ambient	530	170	90
Indoor	410	180	90
Personal	390	150	80

exposures only, and (d) all of the above, or any combination of the three options the user wants. A total of 720 receptors representing individual locations can be modeled using this method. Table 4 summarizes the assumptions that were made, which were adapted from the indoor analysis in Jenkins et al. (1992).

As an example, the same scenario as displayed in Fig. 5 was used to compare ambient, indoor, and estimated personal exposures. Table 5 shows the distances from the edge of the field to the hypothetical 100 $\mu g\,m^{-3}$ threshold using differing averaging times and 1.49 exceedances per year. As shown, the indoor environment can provide a reduction in computed personal exposures, which becomes a more substantial factor as averaging time or infiltration rate is reduced.

Although it is anticipated that most regulatory agencies will consider ambient exposures when evaluating regulatory options, review of indoor air personal exposures provides a more complete perspective on expected margins of exposure relative to those conservatively computed based on ambient assessment only, and thereby, can improve the basis for making regulatory decisions.

9. Summary and conclusions

In terms of methodology, the approach shown in this paper and the companion paper could be applied to other agricultural fumigants, as well as to other sources with intermittent releases, especially for those with emission trends that are difficult to characterize. The expanded uses of Monte Carlo methods to represent the range in meteorological and emissions data provides a basis to more definitively and quantitatively express the effects of uncertainty and variability in the input data on the results of exposure assessments. This approach provides decision makers with the benefits of distribution of exposures, rather than single point estimates, to support regulatory decisions.

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