Modeling of Methyl Isothiocyanate Air Concentrations Associated With Community Illnesses Following a Metam-Sodium Sprinkler Application

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Background *A series of illnesses occurred in Earlimart, CA on November 13, 1999 following a metam-sodium sprinkler application to a potato field on the southern edge of the town.*

Methods *Case information was obtained from direct interviews, from illness complaints filed with state or county agencies, or from illness reports filed by community physicians for 173 subjects. Air concentrations of the by-product methyl isothiocyanate (MITC) during the episode were estimated based upon the Industrial Source Complex Short-Term (ISC3) air dispersion model, estimates from prior metam-sodium monitoring studies, and data from weather stations in two adjoining communities.*

Results *Weather station data indicated that the November 13th incident corresponded with a temperature inversion at approximately 5:00 p.m. and a shift in the direction of prevailing wind from northwest to southeast. On the edge of Earlimart, 1-hr time weighted average (TWA) MITC concentration estimates ranged from 0.5 to 1 ppm. Of the 173 subjects, 170 had MITC-compatible symptoms, including eye or upper respiratory irritation (77.6%), non-specific systemic symptoms (64.7%), and lower respiratory symptoms (20.0%). 78.2% were exposed within 0.5 miles of the northern edge of the treated field where average 1 hr MITC concentrations exceeded the 800 ppb ocular irritation threshold.*

Conclusions *ISC3 modeling is a useful means of evaluating MITC concentrations for illness episodes when industrial hygiene assessment is not possible.* Am. J. Ind. Med. 46:1–15, 2004. © *2004 Wiley-Liss, Inc.*

KEY WORDS: fumigant; methyl isothiocyanate (MITC); metam-sodium; eye irritation; respiratory irritation; ISC3 air modeling

INTRODUCTION

This report describes a series of illnesses that occurred in Earlimart, CA on November 13, 1999 following a metam-sodium sprinkler application to a potato field on the southern edge of the town. Emergency response to the episode was complicated by initial difficulty in identifying the cause of residents' irritant symptoms and by a lack of medical resources in the community. Cases transported to neighboring communities were identified through the California pesticide illness registry and from complaints filed with

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Accepted 4 April 2004
DOI 10.1002/ajim.20037. Published online in Wiley InterScience (www.interscience.wiley.com)

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Tulare County government agencies. Because no air monitoring was conducted at the time of the incident, air concentrations of the metam by-product methyl isothiocyanate (MITC) during the episode were estimated using a standard air dispersion model, data from a local weather station, and air monitoring studies conducted following applications of metam-sodium in California between 1992 and 1999. For areas with reported cases, this allowed qualitative comparison of model estimates with lowest-observed-effect-levels for MITC related irritant symptoms. Insufficient information was available to quantitatively correlate individual exposures with the presence or absence of symptoms.

Background

The soil fumigant metam-sodium, a pre-plant treatment for control of soil pests and fungi, had increasing use in California between 1989 (351,114 pounds in 250 applications) and 2001 (11,346,620 pounds in 3,232 applications) in 2001. Peak use occurred in 1999 (17,310,844 pounds in 3,844 applications) [California Department of Pesticide Regulation, 1991, 2001]. This increase coincided with a series of cluster illnesses associated with exposure to its airborne degradation products (Table I).

A separate cluster involving several hundred cases of eye and respiratory irritation occurred in association with a metam-sodium spill into the Sacramento River after a train derailed near the town of Dunsmuir, CA in July, 1991. Twenty cases associated with the Dunsmuir spill had reactive airways dysfunction syndrome (RADS) documented with positive methacholine challenge tests [Cone et al., 1994]. Measured air concentrations, beginning 3 days after the spill, ranged from 0.2 to 37 ppb [Alexeeff et al., 1994]. Modeling studies were conducted, but peak air concentrations associated with the spill remain a matter of controversy. Difficulties included variable terrain over a 45 mile section of the Sacramento River, an unknown rate of decomposition of the active ingredient, an unknown rate of dispersion into the river water, and uncertain estimates of atmospheric dispersion [Baskett et al., 1992; Alexeeff et al., 1992, 1994; Kreutzer et al., 1994].

Occupational illness associated with the use of metam-sodium has been reported from several governmental jurisdictions. These include skin and respiratory irritation in metam-sodium applicators in Germany during the 1970s [Jung and Wolff, 1970; Jung, 1975] and more recently from the US State of Washington [Burgess et al., 2000]. Non-occupational ocular and respiratory symptoms similar to those reported here have also occurred following agricultural use of metam-sodium in Nicaragua. The problems were of sufficient magnitude to prompt changes in buffer zones and recommended changes in application practices [Ministerio de Salud DdH, Republica de Nicaragua, 1999].

Metam-sodium is used in many jurisdictions that do not conduct surveillance for pesticide illnesses. In 1997, for example, states without active surveillance programs reported use of 29 million (49%) of the 60 million pounds used in the US, raising the possibility of unreported illnesses in the application work force and in communities adjacent to the application sties. Additional episodes may also be expected as metam-sodium is promoted as an alternative to the use of methyl bromide [Haglund, 1999; Medeiros, 2000]. This is especially likely for pre-plant applications to strawberries [Lopez-Aranda, 2002] and other high value crops that are raised on very limited acreage in mixed agricultural and urban areas [Environmental Working Group, 1997].

The degradation products (or by-products) of metam-sodium include a complex mixture of irritant compounds (Fig. 1): the primary pesticidal agent MITC, as well as methyl isocyanate (MIC), carbon disulfide, hydrogen sulfide, and methyl amine. The specific mix of products depends upon soil pH and other environmental conditions [Wofford et al., 1994; Wales, 2002]. The toxicologic effects of the mixture have not been characterized, although at least limited information is available for each of the individual compounds.

For MITC, a 1996 industry-sponsored human experimental study identified acute eye irritation as a critical acute toxicity endpoint [Russell and Rush, 1996]. This study, submitted to both the US and California EPAs, is not available in the public literature, but a detailed summary is available in a recently completed risk characterization document [Rubin, 2002, 2003]. The intent was to establish the relation between the odor and irritation thresholds for MITC. The odor threshold for 38 subjects ranged from 200 to 8,000 ppb with a geometric mean of 1,700 ppb. Subjects with pregnancy, smell dysfunction, and symptoms of allergy or upper respiratory infection were excluded from the study.

In the eye irritation study, 70 subjects between 18 and 67 received exposure to MITC by means of specially fitted goggles that eliminated respiratory exposure. Possibly sensitive subjects with asthma or ocular allergy were excluded. Volunteer subjects reported increased perception of eye irritation (designated on an analog scale) and increased blink rate at a lowest observed effect level (LOEL) concentration of 800 ppb over a period of 1 hr. The no observed effect level (NOEL) was 220 ppb. For 4-min exposures, the LOEL for eye irritation was 1.9 ppm and the NOEL 600 ppb. The actual threshold could have been overestimated because some signs of ocular irritation, for example, tearing, involve a trigeminofacial reflex from either direct (eye) or indirect (nasal) stimulation.

Although the volunteer study described above did not evaluate the irritating effect of MITC on the respiratory tract, animal studies with the compound demonstrate clear

TABLE I. Priority Illness Episodes in California Involving Off-Site Movement of Metam-Sodium By-Products*

Case number or priority number	Number of cases	Application method	Estimated distance (miles) from area of application to location of person injured	Comment
1987-2610	8	Sprinkler	1.9	Members of two families complained of a strong odor eye and nasal irritation possibly from a pre-plant metam-sodium application to a parsley field down the road, but did not seek medical treatment
90-FRE-88	15	Shank	0.006	Residents across the street from a metam-sodium treated field sought treatment for eye irritation following the application
5-SB-92	6	Shank	0.005	Residents/visitors at a house on Northwest corner of parsley field treated with metam-sodium. They developed mostly eye irritation and throat irritation
13-RIV-93	6	Sprinkler	0.008	Sprinkler application across the street from a field treated with metam-sodium complained of a foul odor, like propane gas, and experienced eye irritation, itching, sore throat, nausea, and headache
66-SJ-93	5	Rotovator	2	Residents approximately 2 miles from the site of application complained of eye irritation, itching, sore throat, nausea, and headache
47-SJ-95	14	Sprinkler	0.13	Fourteen employees of a small manufacturing company developed irritation of the eyes and respiratory tract after a sprinkler application across the highway from their plant east of Stockton. An inversion layer apparently trapped the metam-sodium by-products near the ground
51-SJ-95	19	Sprinkler	0.51	Eight employees and 11 wards at a California Youth Authority facility developed symptoms after smelling an odor that resembled sulfur. It came from a sprinkler application of metam-sodium to a field about a 1/2 mile away. Symptoms included headache, irritated eyes and skin, sore throat, dizziness, and respiratory irritation
52-SJ-95	6	Sprinkler	0.85	Six employees at a manufacturing plant were examined and released after they smelled an odor from a metam-sodium sprinkler application to a field nearby. This was a re-exposure of workers involved in 47-SJ-95. The re-exposure occurred 42 days after the last episode. Symptoms included eye irritation, irritated nose and throat
36-SJ-96	11	Sprinkler	0.8	Workers at California Youth Authority noticed an odor from a metam-sodium application nearby and complained of eye irritation
40-FRE-96	29	Shank	0.13	Waiting for a bus, 28 students and one adult woman were exposed to metam-sodium application 1/8 mile away. Symptoms involved were mostly eye irritation
34-KER-97	38	Shank	$<0.1-0.2$	Off-site movement of metam-sodium by-products affected a mixed agricultural, residential, and commercial business neighborhood in Bakersfield. Telephone or in-person interviews were conducted with 28 residents or visitors to the neighborhood and with ten emergency responders. Symptoms included eye and upper respiratory irritation, aggravation of pre-existing asthma, and non-specific systemic symptoms
25-SB-99	22	Sprinkler	0.8	The odor from various metam-sodium applications affected two business owners and their children while at the shop. Two days later, similar odors affected children and staff at a nearby elementary school. Symptoms included nausea, eye irritation, and sore throats
2000-441	6	Shank	0.16	Six people, including a family of four, noticed odor and developed ocular and respiratory irritation after a nearby field treated with metam-sodium in preparation for planting endive. Most of their symptoms improved when they left the area

*Data Source: California Pesticide Illness Surveillance Program Database.

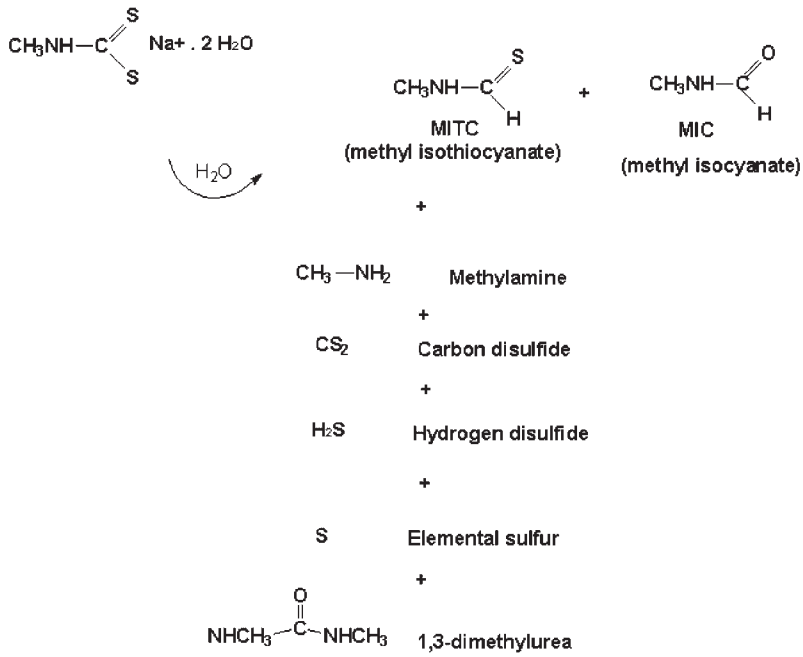

FIGURE 1. Generation of metam-sodium degradation products following reaction with water, with specific mixture of products dependent upon soil pH and other environmental conditions.

respiratory toxicity. For example, Sprague–Dawley rats show a 1-hr median lethal concentration (LC_{50}) of 633 ppm [Clark and Jackson, 1977]. Clinical signs observed included eye irritation, and dyspnea, and hyperactivity, followed by hypoactivity. Convulsions also occurred at concentrations of 733 ppm. The corresponding LC_{50} value for a 4-hr exposure was 180 ppm [Jackson et al., 1981]. Both studies demonstrated predominantly respiratory pathology.

Nesterova also described the inhalation toxicity of MITC, but did not report sufficient experimental detail to allow direct comparisons with the more fully documented studies. Most critically, the study of ocular irritation on cats, identifying a 67 ppb LOEL for a 4 hr exposure, employed an ill-defined colorimetric analytical methodology and did not describe the means of grading the feline irritation response. The irritant gas tested was generated by heating wetted soil treated with metam-sodium, and probably included the full range of metam-sodium by-products and not simply MITC [Nesterova, 1969].

Human volunteer studies demonstrate that MIC, like MITC, is a potent irritant. Eye irritation and tearing occur after 1–5 min exposure to 2 ppm MIC, with more severe responses at 4 ppm [American Conference of Governmental Industrial Hygienists, 1986]. Odor detection is inconsistent from subject to subject. Exposures to 5 ppm provoke eye irritation and tearing after 50 s and exposures to 21 ppm cannot be tolerated for any length of time [National Institute for Occupational Safety and Health, 2002]. In inhalation toxicity studies, MIC demonstrates a 6 hr rat LC50 of 14 ppm and a 2 hr LC50 of 34 ppm. If projected to 1 hr (LC50 of 68 ppm) or 4 hr time intervals (LC50 of 17 ppm) comparable to those used with MITC, the acute inhalation studies indicate an approximately tenfold greater toxicity for MIC.

The irritant symptoms produced by hydrogen sulfide and carbon disulfide [Vicas and Whitcraft, 1990], and methylamine have a high degree of overlap with those produced by MIC and MITC, but quantitative human experimental data on dose response for the sulfide and amine compounds do not exist. For example, the data supporting the 10 ppm threshold limit value (TLV) for hydrogen sulfide include only case reports and observational studies. The standard ignores any possible irritant or nuisance effects occurring at concentrations near the odor threshold of hydrogen sulfide (variously reported as low as 0.5 ppb [Hoshika et al., 1993] and as high as 20 ppb [Vicas and Whitcraft, 1990]. These concentrations are equivalent to 0.005–0.2% of the TLV. For carbon disulfide, the 10 ppm TLV for carbon disulfide is based largely on observational studies of neurotoxicity and cardiotoxicity [American Conference of Governmental Industrial Hygienists, 1996] For methylamine, the TLV documentation does not cite any primary data but states that the 5 ppm standard is based upon human eye irritation [American Conference of Governmental Industrial Hygienists, 1996].

The limitations of the TLVs as standards has been well documented and the process by which they are

developed has been a subject of great controversy. They have particular limitations as environmental standards [Paustenback and Langner, 1986; Castleman and Ziem, 1988; Davis, 1988; Frank, 1988; Ziem and Castleman, 1989; Roach and Rappaport, 1990; Robinson et al., 1991; Robinson and Paxman, 1992; Rappaport, 1993; Egilman and Reinert, 1995; Ruden, 2003].

Regulatory Status of Metam-Sodium in California at the Time of the Earlimart Episode

The label for Sectagon™ 42, the product involved in this episode, permits application with a proprietary Ro-To-Vate & Roll Applicator®, by soil injection, sprinklers, flood (or check), or drip irrigation, disc application, a power roll seal method for beds or rows, and a soil covering method. California application guidelines emphasize methods for minimizing off-site movement as indicated by the presence of by-product odors. Attempts have been made to standardize the implementation of this practice by requiring that the odor monitoring be done by an employee with a ''fresh nose'' [Kings County Department of Agriculture, 2001].

Sprinkler applications are allowed between 500 feet and 1/2-mile of occupied structures if the following conditions are met (otherwise, sprinkler applications are prohibited within 1/2-mile of an occupied structure).

- During time periods when air temperatures commonly exceed 90°F, application must occur during the coolest portion of the day.
- During the application, the irrigation system shall be operated at the lowest possible pressure.
- Water shall be applied immediately after the application to an equivalent depth of 1/4 inch.
- Water is to be applied the day after the application to insure that the soil does not dry out.
- If strong odors occur, application of more water is required.

Individual counties have the authority to set wider buffer zones and establish other application requirements inaccord with local meteorological and geographic conditions. To date additional permit conditions have been established in Kern, Kings and Santa Barbara counties (see Note 1).

State-wide mitigation measures are currently being considered based upon the Russel and Rush 1996 study. A decision not to implement the 22 ppb 1 hr REL recommended in the risk characterization document [Rubin, 2003] was based upon the ''reversible, mild health effects'' demonstrated in the 1996 study [Gosselin, 2002].

MATERIALS AND METHODS

Background on the Application at Earlimart California and the Reported Incident

Demographics and medical services

The community of Earlimart is an unincorporated area in Tulare County, located on California highway 99 about 75 miles south of Fresno. The 2000 population of the Earlimart designated census area was 6,583, with approximately 92% of Hispanic origin, compared to 33% statewide. The median household income at the time of the census was $21,290 annually, compared to $47,493 for the entire state. Approximately 50% of the workforce was employed in agriculture, but unemployment in the community was approximately 20%, while the overall California unemployment rate was 4.3% [US Bureau of Census FF, 2000].

Medical services are limited. There is a community clinic in the center portion of town, operating 5 days a week, but emergency medical treatment is only available in the adjacent communities of Porterville (30 miles north and east), Visalia (33 miles north), and Delano (9 miles south).

Application information and initial report of outbreak

Avenue 44 forms the northern edge of the treated field, approximately 1,000 feet from Spruce Avenue, the southern edge of the populated area of town. The application consisted of a total of six sprinkler sets applied to approximately 75 acres at 42 gallons of Sectagon 42 (4.22 pounds metam-sodium/gallon) for each acre spread out over 5 days (November 9 through November 13, 1999) [Barry, 2000a]. This amounted to an estimated 2,216 pounds of active ingredient (AI)/sprinkler set and 13,293 pounds for the entire application period. The maximum application rate permitted by the product label is 80 gallons/acre [CDMS, 2003].

During this period, the minimum air temperature varied from 42 to 45°F and the maximum air temperatures varied from 65 to 72°F. Soil temperatures were relatively constant, varying from 58 to 61°F. Minimum relative humidity varied from 57.2 to 63.5% and maximum relative humidity each day was 100%. Similar conditions were observed at other nearby weather stations in Lindcove (50 miles distant) and Visalia (36 miles distant) [University of California Statewide Integrated Pest Management Project, 2001].

The final two sprinkler sets were applied sequentially on November 13 between 7:30 a.m. and 7:30 p.m. Sunset occurred at 4:51 p.m. According to information from the Famoso weather station, about 20 miles south of Earlimart, the wind direction shifted from NNW (273°) to SSE (150°)

between 5 and 6 p.m. In addition, both the air temperature and the wind speed dropped after sunset and an inversion condition developed. After 5:00 p.m. the atmospheric conditions (stability class[1]) were characterized as moderately to strongly stable [Barry, 2000a]. About that time, residents began to call emergency services to complain about an odor from a suspected natural gas leak.

Investigation of the reported leak led to realization that the odor derived from the field being treated at the south end of town. Because many people were reporting symptoms, an evacuation was ordered for residents who lived south of Armstrong Avenue. A decontamination center was set up at Earlimart Middle School (599 Sutter Avenue) in the northern portion of town and a number of residents were taken to area hospitals for evaluation. There is no hospital in Earlimart, so residents visited emergency medical services and medical providers in Delano, McFarland, Porterville, and Tulare. Problems experienced by residents at the decontamination center and at the hospitals were discussed at a community meeting on November 18. Residents complained of lack of privacy and decontamination procedures that required disrobing [Schulz, 1999]. They also complained about bills of $6,000 or more received from hospitals providing emergency medical services [Hanley, 1999; United Farmworkers, 2000].

Interviews and records review

Clinical interviews were conducted with 20 community residents on November 19 at the community church on Elm and Spruce Avenues (Fig. 2) after residents were contacted by church staff. Interviews were conducted in either English or Spanish, depending upon the preferred language of individual residents and focused on symptoms experienced on Saturday, November 13, and resident's location at the time of the fumigant application. Information on 14 additional subjects was obtained by proxy interview. These were principally children under 12 with descriptions of their problems supplied by parents. Of the 34 subjects, 22 (64.76%) had supplemental information available from either complaints or medical records (described below).

The interviews were supplemented by a review of 128 complaints submitted to the Tulare County Department of Agriculture. Qualitatively these records provided narrative information similar to that available from direct interviews, but often with fewer details. Complaints submitted in Spanish were translated by one of the authors (MO).

There were an additional 25 proxy complaints, principally describing illnesses in children in the same household. For 119 subjects, the complaints or proxy complaints were the sole source of information on their illness. Records of emergency medical treatment or pesticide illness reports filed by physicians were available for 28 subjects, but were the sole source of information in only six cases.

Symptom classification

Categories used in classifying illnesses were not unique for metam-sodium but resembled those routinely used in categorizing illnesses reported to the California pesticide illness registry [Mehler, 2003] and the registries reporting to the NIOSH SENSOR program [National Institute for Occupational Safety and Health, 2003]. Symptoms reported on complaint forms, in medical records, or recorded during interviews were grouped as follows.

- Odor only or no complaint: No symptoms recorded, or noted presence of odor without experiencing symptoms.
- Irritation of the eyes or upper respiratory tract: Burning or irritation of the eyes, nose, or throat.
- Respiratory: Asthma or lower respiratory irritation (e.g., ''burning of the lungs''), shortness of breath, chest pain, or difficulty breathing, cough, presence of wheezing recorded on medical examination, reported use of inhaler following exposure.
- Non-specific systemic symptoms: Headache, nausea, abdominal pain, diarrhea, and malaise.
- Dermatitis: Presence of skin rash.
- Other: Unrelated symptoms.

Air concentration estimation

None of the local agencies responding to the November 13 incident had resources to collect air samples for MITC or other metam-sodium degradation products. We, therefore, employed the US Environmental Protection Agency (USEPA) Industrial Source Complex Short-Term Version 3 (ISC3), a steady-state Gaussian plume model, to estimate MITC air concentrations. The ISC3 model is a preferred regulatory model, used to estimate pollutant air concentrations following emissions from a wide variety of source types. These include agricultural applications and other area sources [US EPA OAQPS, 1995].

The ISCST and ISC3 have been employed by CDPR since 1992. A 1995 validation study for ISC2 modeling soil fumigation conducted using methyl bromide showed good correspondence between measured and modeled air concentrations. Model estimated flux overestimated the measured flux by no more than 13% [Ross et al., 1995].

The Gaussian plume model [US EPA OAQPS, 1995] estimates the concentration (χ) at downwind distance x (m)

[1] Under stable atmospheric conditions vertical mixing of the atmosphere is damped out, resulting in little or no dilution of a plume of material in the vertical direction.

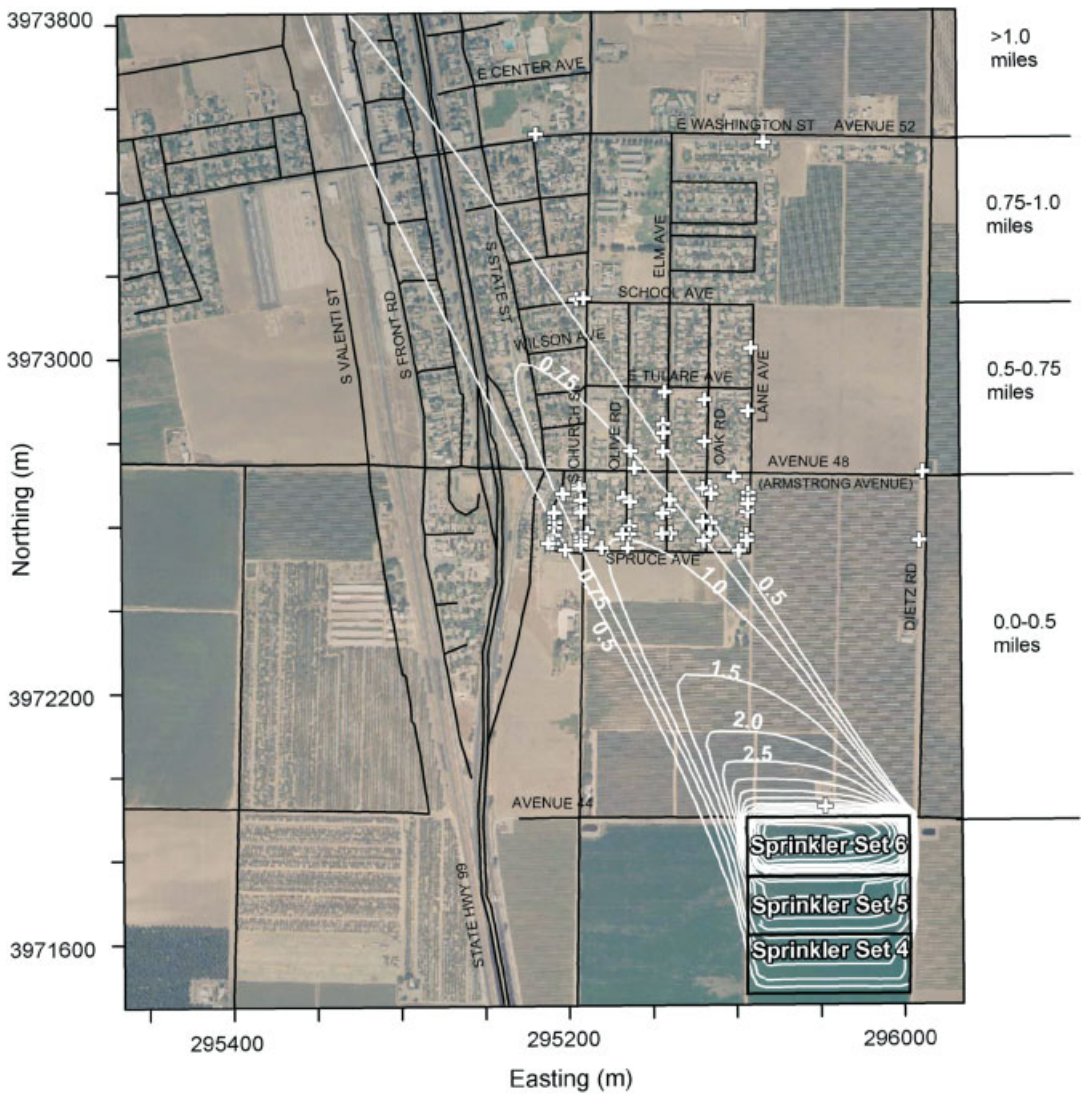

FIGURE 2. Estimated 1-hr time weighted average (TWA) methyl isothiocyanate (MITC) concentration isopleths at Earlimart, CA between 1700 and 1800 hr on November 13, 1999 (Peak 1 and 3 minute concentrations are not shown). Crosses indicate households or submitting illness reports. Universal Tranverse Mercator (UTM) map coordinates: Northing indicates distance in meters from equator; Easting indicates distance from artificial reference point 500 km west of the UTM zone 10 central meridian (123°). *Street name is Armstrong Avenue west of Lane Ave.

and crosswind distance y (m) as follows:

$$\chi = \left(\frac{QK}{2\pi\mu_s\sigma_y\sigma_z}\right) \times \exp\left[-0.5\left(\frac{y}{\sigma_y}\right)^2\right]$$

Q, pollutant emission rate (mass per unit time), or flux; K, a scaling coefficient to convert calculated concentrations to desired units (default value of 1×10^6 for Q in g/s and concentration in $\mu g/m^3$); σ_y, standard deviation of lateral concentration distribution, dependent upon the distance downwind (m); σ_z, standard deviation of vertical concentration distribution, dependent upon the distance downwind (m); μ_s, mean wind speed (m/s).

The values of σ_y and σ_z quantitatively characterize the degree of mixing in the atmosphere, depending upon atmospheric stability. The atmospheric condition present during the evening of November 13, 1999 at Earlimart was stability class F (moderately stable). See Turner [1994] for further information on the determination of the values for σ_y and σ_z.

The Earlimart application was conducted as six sprinkler sets of 20 acres each made over 5 days. The sprinkler sets were rectangular, each with approximate dimensions of 388 m east-to-west by 140 m north-to-south. Flux (variable Q in the model) was estimated using results from two field studies [Wofford et al., 1994; Merrick, 1999]. For the last three

TABLE II. Sprinkler Set Identification, Date and Time of Application, and Estimated Flux Values Used for the ISC3 Air Dispersion Simulation Modeling of Methyl Isothiocyanate (MITC) Concentrations Associated With the Earlimart Evacuation Incident, California

Sprinkler set ID	Date of application	Time of application (hr)	Estimated flux ($\mu g/m^2/s$)[a]
Set 6	11/13/1999	1500–1930	204
Set 5	11/13/1999	0730–1330	74
Set 4	11/12/1999	0845–1455	30

[a]Estimated flux is the flux assumed for each sprinkler set between the hours of 1700–1900 on 11/13/1999.

sprinkler sets, details by date and time of application are given in Table II.

Initial MITC air concentration estimates from the ISC3 model are presented here as 1-hr time-weighted-average (TWA) concentrations, in accordance with the 1 hr intervals used for reporting the available weather data. Peak-to-mean estimation techniques were used to obtain estimates of air concentrations over shorter averaging periods (e.g., 3- and 1-min) [Barry, 2000b]. Because mean concentrations are the result of integration over many short-term peak conditions during the sampling interval, a definable relationship exists between the peaks and the mean [Singer, 1961]. The definable relationship is the mean concentration adjusted by a correction factor accounting for the relationship between the duration of the mean sample and the duration of the desired time interval as shown below:

$$C_{peak} = C_{mean}\left(\frac{\text{sampling time}_{peak}}{\text{sampling time}_{mean}}\right)^{-0.5}$$

where, C_{mean}, the ISC3 estimated 1-hr TWA concentration; sampling time_{peak}, the desired shorter time interval (less than 60 min); sampling time_{mean}, the 1-hr TWA time interval (60 min).

The value of the exponent (−0.5) is commonly used for mean sampling intervals of 10 min to 5–6 hr, based upon the work of Hino [1968]. This peak to mean estimation technique is a standard method to estimate short-term air concentrations when only 1-hr estimates are available.

Weather data

Weather data from the Famoso California Irrigation Management Information System (CIMIS) station (station #138) were used to represent the conditions at Earlimart during the 1700–1900 hr period on November 13, 1999. The station lies approximately 20 miles to the south of Earlimart, separated by flat open terrain. Data from the Visalia station (# 33), 33 miles to the north, were also reviewed to evaluate similarity of weather conditions across Tulare County and Northern Kern County. The terrain between Earlimart and Visalia is also flat and open. The wind speed and direction at Visalia were 0.53 m/s (1.1 mph) and 145° while the wind speed and direction at Famoso were 0.54 m/s (1.2 mph) and 150°, emphasizing the meteorological similarity during the event between these two locations.

Since the evacuations occurred at approximately 1800 hr, the wind speed of 0.5 m/s and wind direction of 150° for 1800 hr at Famoso was used in the simulation. It should be noted that the ISC3 model automatically adjusts wind speeds of less than 1 to 1 m/s, a standard procedure in air dispersion modeling. This adjustment is made to avoid unrealistically high air concentration estimates [US EPA, 2000] because wind speed (μ_s) is in the denominator of the Gaussian Plume model (see the ISC3 equation above).

Graphics

Graphic comparison of the distribution of cases with the ISC3 MITC concentration isoforms employed software from ArcGIS, version 8.2 (ESRI, Inc.) and a true color, high resolution aerial photograph taken 12/1/2002 purchased from a commercial source (TerraExplorer.com, 2004). No aerial photo was available from a date closer to the time of the Earlimart incident, but residential and landscape features were similar to a US Geological Survey aerial photo dated 1994. The ArcGIS shape file allowed direct overlay of MITC concentration isopleths and community street map on to the aerial photograph. Concentration isopleths were produced from the ISC3 plotfile output using the contour map feature of Surfer7 (Golden Software, 1999). Individual cases were mapped to their residence addresses, except for a single resident who reported visiting the field site and reported receiving his greatest exposure there rather than at his residence.

RESULTS

Exposure Description, Odor, and Illness Complaints

The residents did not describe the exposure and its accompanying odor in a uniform manner. Many noted the presence of a foul odor, described as similar to "cooking chiles," "cooking crank," "burning rubber," "Filipino food," "chemicals," "rotten eggs," or "propane gas." Several cases, identified below by illness registry case number, described a distinct, visible mist or fog, or noted the effect of changing weather conditions on the presence of odor.

1999-1252: "It was around 5 o'clock when I saw some kind of fog coming in but with a strong smell. Then less than a minute a cop came knocking on my door and told us to leave our homes because of a chemical going on."

1999-1234: At about 5:00 p.m. on Saturday, November 13, a 39-year-old, living on Dietz south of Avenue 48,

reported noticing an odor like burning rubber, and experienced eye irritation, and irritation of the nose and throat. The material coming from the fields resembled a cloud or a fog.

1999-1239: A resident of East Armstrong, 0.6 miles from the treated field, noticed a strong odor in the late afternoon on Saturday, November 13, corresponding with a change in wind direction—from south and east instead the usual direction, from north and west.

1999-1494: A resident of Spruce Avenue noted that there was an aroma ''like garlic—my eyes started to burn. There was a cloud-like when it's foggy. My head started to ache around 6:00 pm. The police came to my house and I was told to leave.''

Summary of Illness Complaints by Clinical Syndrome and Distance From the Treated Field

Between the on-site interviews, pesticide illness reports, and complaint forms, information was evaluated on 171 community residents or visitors and two emergency response personnel (Table III). These included 170 with MITC-compatible symptoms. Eye irritation or upper respiratory irritation (typically, burning of the eyes, nose, or throat) were present in 132 (77.6%) of the symptomatic cases. Non-specific systemic symptoms (including complaints of headache, nausea, dizziness, shortness of breath, abdominal pain, vomiting, and weakness) were present in 110 (64.7%); 34 (20.0%) had respiratory complaints, principally cough and dyspnea. However, there were six cases with asthma or other lower airway problems, described in more detail below.

By distance, 133 (78.2%) were exposed in the area south of Armstrong Avenue, within 0.5 miles of the treated field, representing 51 separate households and one business. These included one resident who reported being exposed at or near the edge of the field on Avenue 44, and 132 exposed between Spruce and Armstrong avenues (0.45–0.5 miles from the north end of the treated field) (Fig. 2). Nineteen (11.2%), representing ten separate households, were exposed in the area between Armstrong and School Avenue (0.5–0.75 mile from the treated field). Ten (5.9%), from two separate households, were exposed in the area between East School Avenue and East Washington (0.75–1.0 miles from the treated field), and four (2.4%) in the area north of East Washington (>1.0 miles from the treated field). These included one household and three who reported their only exposure at the emergency treatment center (not shown in figure).

The location of four residents at the time of the incident could not be specifically determined. These included one without any specified address and three listing only a post office box. Based upon the distribution of residence addresses for all reported cases, these individuals were most likely to have been exposed in the area south of Armstrong Avenue.

Respiratory Complaints

Six cases of asthma or other lower respiratory tract problems were reported in community residents in conjunction with the November 13 exposure (Table IV). There were 28 additional cases of possible lower respiratory symptoms (Table III). These included 21 cases with dyspnea or chest

TABLE III. Total Number of Cases by the Type of Symptoms and Their Distance From the Metam-Sodium Treated Field on November 13, 1999, California

Symptom complex	≤0.5 miles	0.5–0.75 miles	0.75–1.0 miles	>1.0 miles	PO box only	Total
Odor only or no complaint[a]	2	0	0	1	0	3
Irritant[b] symptoms	51	2	0	0	0	53
Non-specific systemic[c] symptoms	22	9	5	0	0	36
Irritant/systemic	45	6	2	0	0	53
Respiratory[d]	0	0	0	0	1	1
Respiratory/irritant	6	0	0	1	1	8
Respiratory/non-specific systemic	0	0	0	2	0	2
Respiratory/irritant/non-specific systemic	9	2	3	1	2	17
Total	135	19	10	5	4	173

[a]Odor only or no complaint: no symptoms recorded, or noted presence of odor without experiencing symptoms.
[b]Irritation of the eyes or upper respiratory tract: burning or irritation of the eyes, nose, or throat.
[c]Non-specific systemic symptoms: headache, nausea, abdominal pain, diarrhea, malaise.
[d]Respiratory: asthma or lower respiratory irritation (e.g., burning of the lungs, shortness of breath, chest pain or difficulty breathing, cough, presence of wheezing recorded on medical examination, reported use of inhaler following exposure.

pain and no other lower respiratory complaints, six cases with cough, and one case with both cough and dyspnea. Because some residents (e.g., 1999-1539, Table IV) could not afford medical care, it is possible that some additional cases may have occurred, but went undocumented.

Other Complaints

There were eight cases of rash reported by community residents. In one case, the reported rash was limited to the face (1999-1572), and in another the rash was reported to be generalized (1999-1619). In the four remaining cases, the distribution of the reported rash was not specified. One case (1911-1311) was a possible case of varicella (chickenpox). Other complaints that did not fit the predominant symptom pattern included aggravation of a case of peptic ulcer disease (1999-1247) and a case of hypertension (1999-1253). In a case of moderately severe hypertension (1999-1250), it was uncertain whether the condition was due to the effects of evacuation-related anxiety or lack of routine access to medical care.

Air Concentration Estimates

1-hr TWA and 1–3 minute estimates

Figure 2 shows the results of the air dispersion simulation using MITC flux from Sprinkler Sets 4, 5, and 6 and CIMIS weather from the Famoso Station (#138) for 1800 hr on November 13, 1999. These air concentration estimates

TABLE IV. Cases of Lower Respiratory Irritation Following Exposure to Metam-Sodium By-Products on November 13, 1999; Earlimart, California

Case number	Description
Cases of probable lower respiratory irritation with medical history of asthma or COPD $\leq$0.5 miles from the treated field	
1999-1239	This 71-year-old resident had a history of chronic emphysema, asthma, and bronchitis. He normally was unable to walk any distance because of his lung disease and regularly used oxygen at home. At the time, he was evacuated at 8 p.m. on November 13, the odor was bothering him and causing him some mild increased difficulty breathing, shortness of breath and some eye irritation. Medical records from the Delano Regional Medical Center indicate that he had a respiratory rate of 22 when seen in the emergency department, but had normal oxygen saturation (98%) with use of supplemental oxygen. He did not require treatment with albuterol
1999-1241	An 8-year-old girl, with a history of asthma was evaluated for eye irritation, nausea, and shortness of breath. Her symptoms subsided in the emergency room and she did not require treatment with a bronchodilator
1999-1252	This 47-year-old resident, with a past history of hypertension and asthma, reported inhaling a foul odor after seeing a kind of fog come into her neighborhood. She vomited and developed transient shortness of breath. On evaluation at the decontamination center, she was noted to be in moderate respiratory distress, but improved when seen at the Tulare District Hospital. She did not require treatment beyond removal from exposure. On follow-up with her own physician, in McFarland, she was noted to have a persistent cough over the next few days and required extra medication for treatment of her blood pressure
1999-1503	This resident had a history of asthma. At approximately 5 p.m., she had watery eyes and had to use her inhaler because she found it hard to breathe. She went to her doctor, but details of the medical findings and treatment are not available
>1.0 mile from the treated field	
1999-1249	A 60-year-old woman worked as a security officer at Earlimart Middle School, site of the Hazmat decontamination and evacuation center. She had a history of asthma, diabetes, and hypertension. She had nausea, shortness of breath, and chest pain while working at the school Saturday evening, November 13. On examination at the Delano Regional Medical Center, she had elevated blood pressure and scattered wheezing on examination that responded to treatment with nebulized albuterol. Because of the chest pain, she had a cardiogram, and a serum troponin (cardiac muscle protein) measurement, which both proved normal
Undetermined address	
1999-1242	An 18-year-old, whose address was not specified in the available information, was treated at Delano Regional Medical Center for throat irritation and exacerbation of her pre-existing asthma. A few scattered wheezes were recorded on initial physical examination, but cleared after treatment with nebulized albuterol. Oxygen saturation on room air was 98%. She was improved on discharge from the emergency department
Other cases of possible lower respiratory irritation	
1999-1237	This 18-year-old resident, whose only address was a post office box, experienced chest pain, shortness of breath, and vomiting. She was evaluated at the Delano Regional Medical Center, but did not have any wheezing on auscultation and did not require any treatment beyond avoiding re-exposure
1999-1559	A 35-year-old woman who lived more than 1 mile from the treated field, returned to Earlimart at approximately 9 p.m. on Saturday after spending the evening in Delano. She started feeling slight chest tightness, a headache, and a rash; symptoms resolved over the next 1–2 days
1999-1539	An adult male, who lived approximately 0.5 miles from the treated field, complained of problems breathing, but did not go to the doctor, because he did not have any money or health insurance

should be interpreted as 1-hr time weighted averages (TWA). Estimates of the 1-hr TWA air concentrations in the evacuated neighborhood ranged from 0.5 ppm to just over 1 ppm. The estimated maximum 3 min peak concentration in this area was 4 ppm and the estimated maximum 1 min peak concentration was 7 ppm. In the areas of the 0.75 ppm isopleths, estimated 1- and 3-min concentration estimates were 5 and 3 ppm, respectively. In the areas of the 0.5 ppm isopleths, the estimated 1- and 3-min concentration estimates are 3 and 2 ppm, respectively.

The area between Spruce and School Avenues (Fig. 2) had the highest local concentration of MITC and also the large majority of the reported cases. Few complaints were received from populated area bordered by Washington, Tulare, Church, and Lane. However, ten cases were reported from two addresses on East Washington. Three cases were reported from the evacuation center on Sutter Avenue (1.4 miles north of the treated field) and a single case from a street 1 block to the north.

Model estimates are most uncertain in locating the plume from east to west and the location of the centerline of the plume in Figure 2 should not be taken as exact. A shift in the plume to the east could have accounted for the cases along Washington Avenue, but not the cases that occurred at the evacuation center and on the adjacent street north.

DISCUSSION

Limitations of Case Series Data

We evaluated cases associated with off-site movement of metam-sodium by-products in the community of Earlimart on November 13, 1999. Our case series was reported through California's pesticide illness registry, with information derived from heterogeneous sources. These included direct interviews, complaint forms, and illness reports filed by emergency care providers. Because no clinical tests are available for MITC or other metam-sodium by-products, no source of information provided direct proof of exposure.

We did not have the resources to conduct a complete survey of community residents or employ an external control group. It was, therefore, not possible to estimate from our data either the total number or the incidence of exposure related cases.

Although some residents had only transient symptoms, we were unable to conduct systematic follow-up examinations of affected community residents and cannot determine from our data whether persistent cases of reactive airway disease occurred, similar to those reported following the 1991 spill into the Sacramento River near Dunsmuir [Cone et al., 1994]. Because medical resources in Earlimart are so limited, it is uncertain whether potentially affected residents had access to pulmonary function laboratories capable of performing testing (methacholine challenge) necessary to document presence of RADS.

Some recorded complaints were probably not related to the episode, except by time of occurrence. One case of skin rash (e.g., 1999-1311), for example, was reported to resemble varicella. Other skin conditions unrelated to the outbreak may also have been reported. It appears doubtful that airborne degradations products reached concentrations high enough to produce skin reactions comparable to those resulting from direct dermal contact [Jung and Wolff, 1970; Jung, 1975; Koo et al., 1995; Penagos et al., 2000]. Some anxiety-related conditions probably occurred, perhaps related to the disruption caused by the hazardous materials response and evacuation [Schulz, 1999].

The reports could have been biased by the controversy surrounding the incident and the lack of privacy during decontamination procedures at the evacuation center. Nevertheless, the evacuation and emergency response was initiated in response to complaints from community members and was not responsible for initiating the complaints per se. The irritant ocular and respiratory symptoms reported by community residents were consistent with expected effects of metam-sodium degradation products.

The difficulty of differentiating exposure related complaints from those produced by evacuation-related anxiety would not have been reduced by a more complete survey of the community. It is also extremely likely that a comparison survey of an adjacent community would have demonstrated a prevalence of eye, respiratory, and non-specific systemic complaints lower than that present in Earlimart on November 13, 1999. Even such a study would have provided only descriptive information insufficient to meet the epidemiologic criteria for a causal association between the symptoms reported by Earlimart residents and the presence of metam-sodium degradation products.

The single most helpful aid to evaluating the reported complaints would have been air measurements taken during the evening of November 13. No such measurements are available because the agricultural industry and local agricultural agencies have as a matter of policy relied upon odor monitoring to detect offsite movement of MITC and other by-products. The limitations of this approach are illustrated by Russell and Rush [1996] study demonstrating that the average odor threshold for MITC is approximately twice the LOEL value for irritation.

To evaluate possible exposures resulting from the metam-sodium applications just south of Earlimart, we employed a Gaussian technique applied by the California EPA for estimating buffer zones for methyl bromide and the US EPA for modeling exposures to industrial pollutants (see Note 2).

Conditions associated with the event at Earlimart satisfied the assumptions of the Gaussian model. The time

scale of the initial event was on the order of minutes to 1 hr, the terrain flat, and the distance from the source to the receptors relatively small and observations of persons in the area substantiate the assumption that the wind speed and direction were essentially constant. The estimates generated for the Earlimart incident, unlike those done following the 1991 Sacramento River spill, benefited from a flux estimate obtained from a DPR monitoring study conducted under conditions reasonably similar to those during the incident [Wofford et al., 1994].

Limitations of the Modeling Data

Although the ISC2 validation study reported by Ross et al. [1995] used weather station data from the site of application, we observed little variation in the data obtained from the Famoso weather station 20 miles south of Earlimart and data from the Visalia station 33 miles north. We, therefore, do not believe the absence of an Earlimart weather station materially affected the estimates of MITC flux from the ISC3 model.

The location of the model-generated 1-hr TWA plume should not be interpreted as the exact location of the actual plume during the event (Fig. 2). Under the meteorological conditions present at the time of the episode, the instantaneous pollutant plume would tend to be narrow and subject to significant crosswind meander. Therefore, at any particular moment during 1800 hr, the location of the instantaneous plume may have been displaced by some degree to the east or west relative to the centerline position of the 1-hr TWA plume. Observations made by community residents described above (e.g., case 1999-1234, near Dietz and Armstrong) confirmed that local variations in concentration of the plume did occur.

Our 1 hr estimates represent the minimum MITC concentrations in the affected area of Earlimart: up to 1 ppm along Spruce Avenue and 0.75 ppm between Spruce and Armstrong. Short-term concentrations calculated by peak-to-mean estimation ranged as high as 7 ppm. Local temperature inversions may have further intensified peak exposures. Atmospheric condition at the time of the incident indicate that inversion conditions were present [Barry, 2000a]. However, the height of the inversion was not measured by the remote weather stations at Famoso and Visalia.

The 1-hr TWA levels approximate the 0.8 ppm LOEL for ocular irritation in the 1996 Russell and Rush study. It is, therefore, plausible to assume that the level of MITC present in the area south of School Avenue was high enough to cause most of the reported complaints. Other metam-sodium degradation products may also have contributed to the reported symptoms. Estimated MITC concentrations at the evacuation center (1.4 miles north of the treated field) and the adjacent street were probably too low to account for the reported cases in those areas.

Relationship Between Ocular and Respiratory Irritation Thresholds

Although it was not possible to evaluate respiratory effects from the 1996 experimental study, the case series reported here included eye and upper respiratory irritation, non-specific systemic symptoms, and exacerbation of pre-existing cases of lower respiratory disease. This suggests that the respiratory and irritant thresholds may be similar, although the data here do not allow more than qualitative inferences to be made. Nevertheless, these data are not consistent with the assumption that MITC causes only mild ocular symptoms.

Prevention of Future Outbreaks

Although odor was prominently associated with the plume at Earlimart, it remains uncertain to what extent odor monitoring could have prevented or lessened the impact of the outbreak. Because the 1.7 ppm mean odor threshold for MITC is well above the mean irritation threshold [Russell and Rush, 1996], it may be necessary to consider other strategies for preventing future outbreaks. Inexpensive direct reading tools to monitor other metam-sodium by-products include colorimetric tubes for H_2S and CS_2. Nevertheless the tubes evaluate concentrations of these contaminants in the range of their TLVs, far above the range associated with metam-sodium applications. Wofford, for example, documented a 77 ppb peak concentrations of H_2S during a metam-sodium application in Kern County in 1994. [Wofford et al., 1994] This corresponds to less than 1% of the 10 ppm TLV. The most sensitive direct reading instruments for hydrogen sulfide can detect concentrations of 1 ppb, but might be subject to interference from dairies, poultry farms, and other ambient sources common in rural areas [Livestock and Poultry Environmental Stewardship Curriculum, 2003].

Measures adopted by individual counties to date have included 1mile buffer zones for sprinkler applications [San Luis Obispo County Department of Agriculture, 2001; Santa Barbara County Department of Agriculture, 2001]. Prohibitions against night-time applications, when temperature inversions are most likely to occur, will also likely have some impact. Nevertheless, this outbreak illustrates the margin-of-safety for use of metam-sodium is precariously low. The application of 13,293 pounds of metam-sodium over a 5-day period, suggests the likelihood of off-site movement into adjacent populated areas unless ideal weather conditions occur throughout the entire period. Even higher air concentrations of MITC might have occurred had a maximum application rate (80 gallons/acre instead of the 42 gallons/acre) actually been used.

CONCLUSIONS

The ocular, respiratory, and non-specific systemic symptoms reported by community residents corresponded with complaints associated with prior episodes associated with metam-sodium by-products. No direct measurements of air concentration were available. However, estimates of air concentrations based upon air contaminant dispersion modeling showed average 1 hr air concentrations in good agreement with the distribution of illness complaints in the community. ISC3 modeling is a useful means of evaluating air-contaminant concentrations associated with community illness episodes when direct industrial hygiene assessment is not possible. In the investigation described, the model allowed integration of weather data, contaminant flux and recorded illnesses. This was not done in prior reported episodes associated with the agricultural use of metam-sodium.

Note 1

Kern County modified permit conditions in response to a 1997 episode in the Rosedale neighborhood of Bakersfield (see Table I, priority episode 34-Ker-97) [Kern County Department of Agriculture, 2001]. These required a 1/2-mile buffer zone between the site of sprinkler applications and sensitive areas.[2] Similar restrictions were put in place at the same time in neighboring Kings County. Santa Barbara [Santa Barbara County Department of Agriculture, 2001] established a 1-mile buffer zone for sprinkler applications following a 1999 incident at Cuyama School (see Table I, priority episode 25-SB-99). In June 1999, San Luis Obispo County, immediately to the north of Santa Barbara, also adopted a similar 1-mile buffer zone [San Luis Obispo County Department of Agriculture, 2001].

Note 2

Basic assumptions include: (1) continuous and constant emission rate (flux) constant over the modeling period, effectively a steady state condition, (2) relatively flat terrain, (3) no degradation of the pollutant, (4) ground reflection of the pollutant (e.g., no absorption), (5) constant wind speed and direction, in time and in elevation, from the source to the receptor, (6) normal (Gaussian) distribution of the pollutant concentration, and (7) only crosswind and vertical dispersion (no downwind dispersion) [Beychock, 1999].

[2] For example, as defined on the Sectagon label as ''residential areas, labor camps, businesses, day care centers, hospitals, in-patient clinics, nursing homes, or any public areas such as schools, parks, playgrounds, or other public facilities not including public roads.''

ACKNOWLEDGMENTS

The authors gratefully recognize the assistance and expertise of Johanna Walters in preparing the map graphics for this article.

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