

## Determining Off-Site Concentrations of Volatile Pesticides Using the Trajectory-Simulation Model

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### ABSTRACT

An environmental transport model is described and illustrated that will enable the prediction of the mass and/or concentration of volatile pesticides at any depth in the soil profile and at any height in the atmosphere relatively far distanced downwind from agricultural fields. The theoretical profile shape model is used to determine transport in the atmosphere and is coupled to the behavior assessment model (BAM) to describe transport in soils. The model outputs include the soil-water concentrations, the surface volatilization rate, the concentration of pesticides in the atmosphere above the field, and the atmospheric concentration at specified distances downwind from the field. The results from this approach can be used in exposure assessment studies to determine the risk for individuals living near agricultural fields.

**T**HE OCCURRENCE OF PESTICIDES in the atmosphere or in groundwater is an important national issue. Since pesticide movement through soil is closely related to both pesticide application and water management practices, it is imperative to adopt management practices that reduce the movement of pesticides outside of the root zone. In areas of rain-fed agriculture, much effort and concern is being devoted to determining the effects of conservation tillage and related practices on groundwater quality. In these areas, natural factors affect the rate and timing of water application, often giving the farmer little control over the movement of pesticides. During intense storms, soil characteristics such as high infiltration rates greatly increase the potential for pesticides to reach groundwater; also, variations in the moisture content of a soil can affect the loading to the atmosphere of volatile pesticides (Taylor and Spencer, 1909; Spencer and Cliath, 1990). In irrigated areas, the application of water is highly controlled; hence, given an adequate understanding of how water management affects pesticide transport, contamination by pesticides can be minimized.

To do this, we need to answer questions such as: How do we reduce pesticide leaching while preventing the build-up of salt in the root zone of irrigated soils?; What are optimal timing of irrigation and pesticide applications that will reduce pesticide loadings both to groundwater and into the atmosphere?; and Under what conditions can pesticides be safely applied in irrigation water without excessive losses to the atmosphere? To address these questions, subsurface transport studies must be coupled to atmospheric transport to assess the effects of the water application methods in characterizing pesticide fate and transport.

One method for determining effective management practices is to use models to compare the environmental impacts resulting from various management alternatives (Jury et al., 1984). This approach requires an understand-

ing of, and the ability to predict, all routes of pesticide transport and dissipation. An often neglected route of dissipation for pesticides with large vapor pressures (or large Henry's constants) is volatilization. Although this process reduces the potential for groundwater contamination, it also reduces the efficiency of the pesticide in performing its assigned task and causes an increased potential for contaminating the atmosphere. Contamination of the atmosphere can pose severe risk to persons living downwind from the treated field, because many pesticides are considered to be carcinogenic (Doull, 1989). Consequently, pesticide transport models should have the capability to predict the rate of volatilization of pesticides under field conditions and subsequent transport in the gaseous phase. An additional advantage of modeling the volatilization process is that it provides scientists studying atmospheric pollution a means for determining the loading of agricultural chemicals into the atmosphere. This information is critical if quantitative models of atmospheric pollution and transport of toxic chemicals are to be developed.

In California, exposure to volatile pesticides has been blamed for increased occurrences of cancer in communities that are located near fields where pesticides are used. It is difficult to show whether transport of volatile pesticides through the atmosphere is to blame for the observed increases in cancer or whether they are due to some other cultural or environmental factor. Methods that will enable the prediction of the concentration of pesticides at any height in the atmosphere a relatively far distance downwind from an agricultural field (i.e., at habitable structures such as homes) are desirable. The proposed atmospheric transport model may be coupled to existing models describing the transport of pesticides in the subsurface, allowing the extent of atmospheric and groundwater contamination from pesticides to be determined.

The purpose of this article is to describe a transport model that will characterize the environmental fate of volatile pesticides. The model can simulate pesticide leaching to groundwater, loading of pesticides into the atmosphere, and subsequent transport of volatile pesticides downwind from agricultural fields. This is achieved by coupling the atmospheric transport model to an existing solute transport model that allows the pesticide concentration in the atmosphere and soil-root zone to be determined. The theoretical profile shape model (Wilson et al., 1982) is used to determine transport in the atmosphere and is coupled to the behavior assessment model (Jury et al., 1983) to determine soil-water concentrations and pesticide loading to the atmosphere.

The theoretical profile shape method has been recently described by Wilson et al. (1982) as a method to determine the gaseous mass transfer from field experiments. This approach is based on the trajectory simulation model described by Wilson et al. (1981a,b,c). Wilson et al. (1983), Majewski et al. (1989), and Majewski et al.

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(1990) have used the theoretical profile shape method, among others, to determine the rate of pesticide and ammonia (NH<sub>3</sub>) volatilization from field experiments. Although this method has been shown to be well suited for such endeavors, the method can also be used to determine the atmospheric concentration anywhere over the field or at any specified distance downwind from the field, if the pesticide flux into the atmosphere is known or can be simulated. Although the approach is very general, allowing the use of site-specific parameters obtained from field experimentation, the purpose of the research described herein is to develop a simplified screening model. Therefore, several simplifying assumptions concerning both the transport and trajectory simulation model have been adopted and are described in the following sections.

## METHODS

To obtain an environmental transport model for volatile pesticides, several component models must be combined. The approach described here requires information on the pesticide loading into the atmosphere from treated soil, the measurement or prediction of the wind speed with respect to height and a method for simulating pesticide movement through the atmosphere.

### Subsurface Transport and Pesticide Loading to the Atmosphere

The model used to characterize the subsurface transport of volatile pesticides and the flux of pesticides into the atmosphere is the behavior assessment model (BAM) described by Jury et al. (1983). When a more detailed description of the volatilization process is required, more complex numerical models could be used. The BAM assumes that transport occurs in one dimension, that the pesticide is subject to linear equilibrium adsorption, first-order decay, and vapor partitioning follows Henry's Law. The water movement is assumed to be constant and induced by a constant evaporation rate. The equation that describes transport under these idealized conditions is

$$\frac{\partial C}{\partial t} = D_E(\partial^2 C/\partial z^2) - V_E(\partial C/\partial z) - \mu C \quad [1]$$

where  $D_E$  and  $V_E$  are the effective dispersion and advection coefficients,  $\mu$ , is a first-order degradation coefficient and initially the pesticide is incorporated into the soil following

$$\begin{aligned} C(z,0) &= C_o & 0 < z < z_d \\ C(z,0) &= 0 & z > z_d \end{aligned} \quad [2]$$

and at the surface and lower boundaries

$$\begin{aligned} -D_E \frac{\partial C}{\partial z} \Big|_{z=0} + V_E C \Big|_{z=0} &= -H_E C \Big|_{z=0} \\ C(z, \infty) &= 0 \end{aligned} \quad [3]$$

where  $H_E$  is the effective mass transfer coefficient describing vapor transport across a stagnant boundary layer located at the soil surface. From the solution to this equation, the rate of volatilization of pesticides into the atmosphere can be obtained (see Jury et al., 1983, Eq. [25]).

## Wind Profiles

Wind profiles were obtained using existing wind data collected at the University of California's Moreno Valley field station between 14 and 20 Dec. 1989. The data were collected as wind run for a 10-min interval at heights 20, 40, 80, 160, 240, and 360 cm. Hourly values were obtained by averaging. A regression analysis was used to determine the friction velocity,  $u_*$ , for each hourly interval during the period by using the following log-linear wind speed profile.

$$U(z) = \frac{u_*}{k} \left( \ln \frac{z}{z_o} + 4.7 \frac{z - z_o}{L} \right) \quad [4]$$

where  $k \equiv 0.4$  is von Karman's constant,  $z_o$  is the roughness length, and  $L$  is the Monin-Obukhov length characterizing the atmospheric stability. For the purposes of the screening model,  $L$  was assumed to be 500 cm, representing stable atmospheric conditions. To use this approach for a site- and time-specific simulation, both  $u_*$  and  $L$  could be obtained from a regression analysis using measured hourly values of the wind speed.

## Trajectory Simulation

The Trajectory Simulation Model is used to determine the pesticide transport in the atmosphere. This approach is a method that tracks the movement of particles in the atmosphere under conditions of inhomogeneous turbulence. Because the method is described in detail by Wilson et al. (1981a,b,c), only a brief description and necessary extensions to the method will be described here.

The simulation of a particle of air mass proceeds in an atmosphere that possesses the following properties.

1. The Eulerian velocity  $\sigma_w(z)$ ; time,  $\tau(z)$ ; and length,  $\Lambda(z) = \sigma_w(z)\tau(z)$ , scales are assumed to vary only in the vertical direction. The velocity and time scales relate, respectively, to the effective or root-mean-square value of the vertical velocity and the persistence of the vertical velocity as a function of height.
2. The horizontal wind speed varies only in the  $z$  direction, and the time average value of the vertical wind speed is zero.
3. The source of pesticides is spatially uniform in the treated region (i.e., field). Off-site, however, there is no source of pesticide and no degradation of pesticide occurs once it is in the atmosphere.
4. A transformed coordinate system exists that is related to the Lagrangian length scale. In this transformed coordinate system, the turbulence is homogeneous (i.e., constant length, velocity, and time scales).
5. The boundary condition at the soil surface is reflective. Therefore, once a particle is emitted from the soil surface, it remains in the air stream throughout the simulation.
6. The atmosphere is assumed to be stable (i.e., the Monin-Obukhov length is positive) throughout the simulation. This is not a requirement of the method, rather stable atmospheric conditions represent an undesirable (i.e., and in a sense "worst") case since the mass of pesticides remains larger near to the surface compared with neutral or unstable conditions. Another advantage of this assumption is that the Eulerian velocity scale is constant; reducing, slightly, the number of computations required.
7. At locations where there are changes in the surface roughness,  $z_s$ , it is assumed that logarithmic profiles develop immediately downwind from the change in the surface property.

The simulation is conducted by discretizing the vertical dimension into  $N$  layers, where the height of each layer is  $Z_i^* = (i-1)AZ^*$  and  $Z^*$  represents transformed coordinates. The actual vertical height is related to the transformed height through

$$dz = (\Lambda_L(z)/\Lambda_L(H)) dz^* \quad [5]$$

where  $A(z) = 1/2z/(1 + 5z/L)$  is the Lagrangian length scale for stable atmospheric conditions,  $L$  is the Monin-Obukhov length, subscripted  $L$  indicates a Lagrangian frame of reference, and  $H$  is a reference height. Associated values for  $z_i$  and  $A_k$ , are calculated once using Eq. [5] and [6]

$$\Delta x_i = u(z_i)[\tau_L(z_i)/\tau_L(H)]\Delta t_H \quad [6]$$

The simulation is conducted in the  $Z^*$  coordinate system using

$$AZ^* = \left( w_L(t_H) + \sigma_w(H)\tau_L(z_i)\frac{\partial\sigma_w}{\partial z} \right)\Delta t_H \quad [7]$$

where

$$w_L(t_H + \Delta t_H) = w_L(t_H)e^{-\frac{\Delta t_H}{\tau_H}} + \left( 1 - e^{-\frac{2\Delta t_H}{\tau_H}} \right)^{1/2} \sigma_w(H)r \quad [8]$$

is a Markov chain representing the random nature of the particle's movement and  $r$  is a random variable with a standard-normal distribution. When the atmosphere is stable, the Eulerian velocity scale is approximately constant and can be described using  $\sigma_w = 1.25 u^*$ . Therefore, the second term on the right-hand side of Eq. [7] is zero and the vertical displacement increment for any timestep is  $AZ^* = w_L(t_H) \Delta t$ .

The simulation is conducted by discretizing the soil surface in the source area, which is defined to extend from  $0 \leq x \leq X_{\max}$ , into  $M$  sections. A large number of particles are emitted from each section and tracked until they reach the collector located at  $X_{\max}$ . The instantaneous horizontal position is determined from Eq. [4] and [6] using the current vertical position,  $z$ , obtained using Eq. [7] and [8]. For each particle, the final height results from repeated applications of Eq. [4] and [6] through [8]. Once a particle has reached the collector, the count of the appropriate level,  $Z_i^*$ , is incremented by one. This procedure is repeated to obtain multiple realizations of the distribution of particles due to a uniform source extending from  $0 \leq x \leq X_{\max}$ .

Once a large number of realizations have been obtained, the statistical character of the vertical distribution of particles becomes fixed, and this distribution can be used to simulate the time-dependent scaled concentration as a function of height resulting from a time-varying source and wind velocities. A critical feature of this method is that the ratio of the vertical to horizontal flux [i.e.,  $\bar{u}\bar{C}/F_z(0)$ ] is independent of the friction velocity and hence the magnitude of the wind speed. This

occurs because any increase in  $u^*$  causes a proportional decrease in the concentration of particles, which is determined by dividing the number of particles at any specified height by the volume of air sampled at that height.

### Off-Site Transport

The concentration of pesticides a specified distance from the source area can be obtained by continuing the movement of the particles from the collector located at the edge of the field to a new collector a specified distance from the field. It is also possible to change the surface roughness conditions to simulate a different surface. If a new surface roughness is used, a new transformed vertical coordinate system must be devised and the particles placed into the appropriate level,  $Z_i^*$ , such that the real height does not change. Once this is accomplished, the movement of the particles from the field boundary to the second collector can be obtained, which results in a new distribution of particles with respect to height. This procedure implicitly assumes that logarithmic profiles develop instantaneously downwind from a change in the surface roughness. In general, a transition zone would occur (Rosenberg et al., 1983), but is neglected to simplify the model.

## RESULTS

For purposes of this example, the soil is assumed to be at a steady state; thus the soil moisture content, evaporation rate, and water velocity are constant. These assumptions allow a simple model like the BAM to be used. The surface volatile flux for lindane (gamma 1,2,3,4,5,6-hexachloro-cyclohexane) with an application rate of 1 kg/ha as a function of time is shown in Fig. 1. The model parameter values used for the simulation are given in Table 1. Commonly observed time series of pesticide flux would have a cyclic and somewhat erratic behavior with the peak flux values occurring at approximately the same time of the day (Harper et al., 1976; Cliath et al., 1980). This kind of time series would require either extensive experimentation to obtain hourly values of pesticide flux or the use of highly complex models. Since the proposed approach is intended for screening purposes, the use of simple models such as BAM is assumed to be adequate.

A major factor affecting the transport of pesticides in the atmosphere is wind velocity. To incorporate the wind speed into the model, some method for determining the magnitude of the wind speed as a function of height is

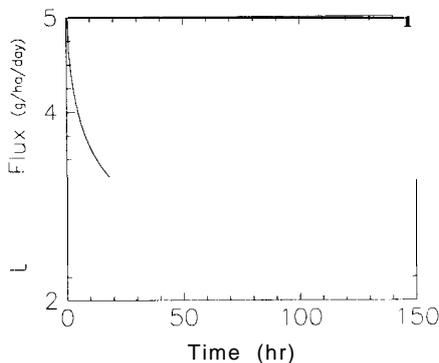


Fig. 1. Lindane surface flux with respect to time when the application rate is 1 kg/ha. The BAM model was used and assumes evaporation was 0.5 cm/d.

Table 1. Soil and pesticide properties used in simulation.

Soil-pesticide property	Value	Dimensions
Soil porosity	0.5	cm <sup>3</sup> /cm <sup>3</sup>
Soil bulk density	1.325	g/cm <sup>3</sup>
Soil water content	0.3	cm <sup>3</sup> /cm <sup>3</sup>
Organic C fraction, $F_{oc}$	0.0125	%
Air diffusion coefficient	4320	cm <sup>2</sup> /d
Liquid diffusion coefficient	0.432	cm <sup>2</sup> /d
Boundary layer thickness	0.475	cm
Incorporation thickness	10	cm
Henry's Law constant	0.000133	(mg/L)/(mg/L)
Organic C partition coefficient	1300	mL/(g $F_{oc}$ )
Half-life	266	d

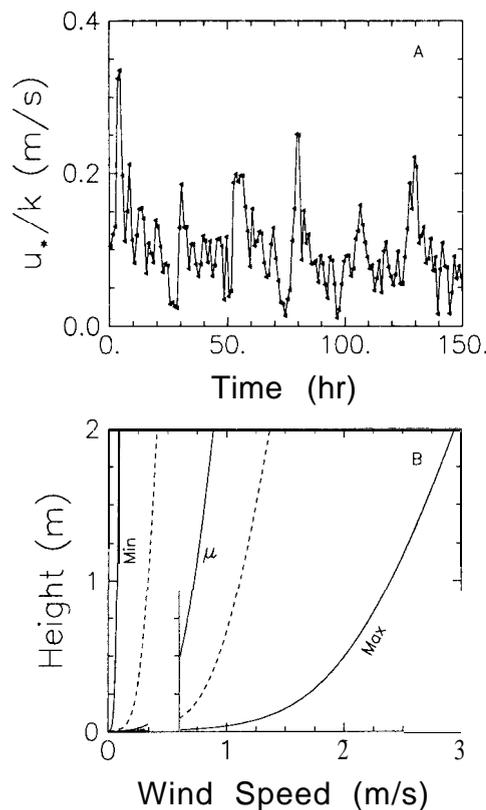


Fig. 2. (A) Time series of the friction velocity,  $u_*$ , at the Moreno Valley Field Station during the period 14-20 Dec. 1989. (B) The minimum, maximum, mean, and  $\pm 1$  SD from the mean wind velocity profiles.

needed. Using the log-linear wind profile to express the vertical dependence of the wind speed, a value for the friction velocity is required to obtain values for the wind velocity. Using existing experimental wind profile data, the variation of friction velocity,  $u_*$ , over the 151 h of the simulation can be obtained and is shown in Fig. 2A. It is apparent from this figure that the friction velocity is highly variable throughout the experimental period. Shown in Fig. 2B is the maximum, minimum, mean, and  $\pm 1$  SD from the mean wind speed. This demonstrates the wide range of wind velocities that occurred during the sampling period and provides somewhat realistic behavior in the wind speed.

Shown in Fig. 3 is the horizontal/vertical flux ratio as a function of the vertical position. This profile was obtained for a plane source 100 m in length where the roughness length for the field was assumed to be 0.2 cm. The curve marked stable and unstable, respectively, were obtained using an atmosphere stability parameter,  $L$  (i.e., the Monin-Obukhov length), of 500 and  $-500$  cm, where values for  $L > 0$  represents stable atmospheric conditions. To develop this curve, the horizontal length was discretized into 1000 sections, and 2500 particles were released from each section for stable atmospheric conditions and 100 for unstable conditions. The particles were tracked as they moved through the atmosphere under the influences of turbulence until they passed into the collector, located at  $x = 100$  m. This curve gives a weighted distribution of particles with height and provides a theoretical profile shape. Figure 4 shows the path

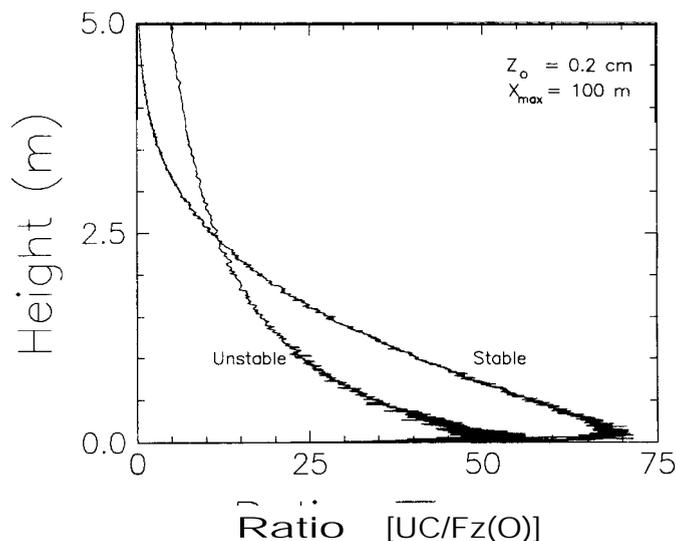


Fig. 3. Ratio of vertical to horizontal flux that results from the trajectory-simulation model. This curve is based on a uniform plane source 100 m in length, a surface roughness length of 0.2 cm, and a Monin-Obukhov atmospheric stability length of 500 cm (stable) and  $-500$  cm (unstable).

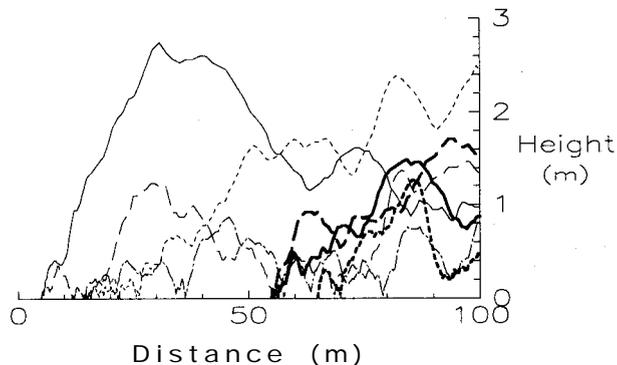


Fig. 4. The paths taken by seven particles. Two particles were released at  $x=5$  m, 15 m, and 55 m, and one released at  $x = 65$  m.

taken by seven particles released at various distances along the field. Two were released at  $x = 5$  m, 15 m, and 55 m and the 7th released at 65 m.

Given known values for the average hourly wind speed with respect to height,  $U(z)$ , and the hourly surface flux of pesticides  $F_z(0)$ , it is possible to use the theoretical profile shape shown in Fig. 3 to obtain the mass or concentration of pesticides at any height and time within the surface boundary layer. This is shown in Fig. 5 where the mass of lindane per unit cm of field width is shown in Fig. 5A and the cumulative lindane concentration at the edge of the field (i.e.,  $x = 100$  m) is shown in Fig. 5B after 24, 48, 96, and 151 h have passed. For the simulated conditions, the maximum mass of lindane occurs at a height of 1 m. The mass can also be expressed as a concentration using  $C = \text{mass}/(\text{unit width})(\Delta z)(\text{wind run})$ , where wind run =  $\int_0^t u(z, \bar{t}) d\bar{t}$ .

The off-site transport of lindane was determined by assuming that the land surface adjacent to the field for a distance of 100 m has a surface roughness of 0.5 cm.

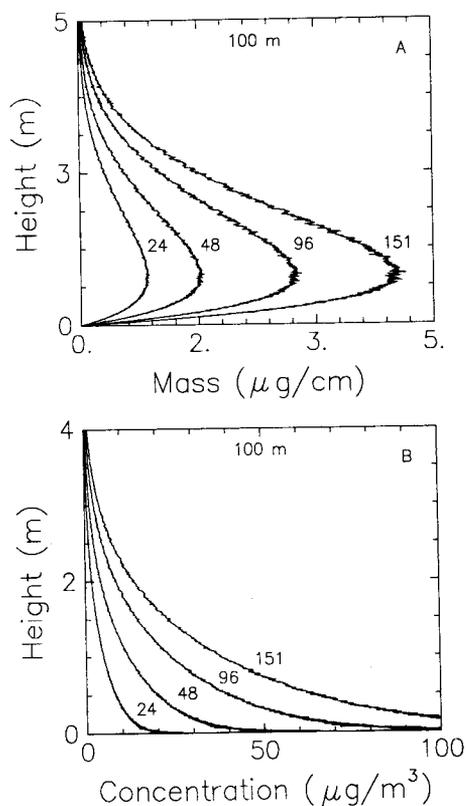


Fig. 5. (A) The atmospheric mass and (B) concentration of lindane in the  $x=100$  m collector after continuously sampling for 24, 48, 96, and 151 h.

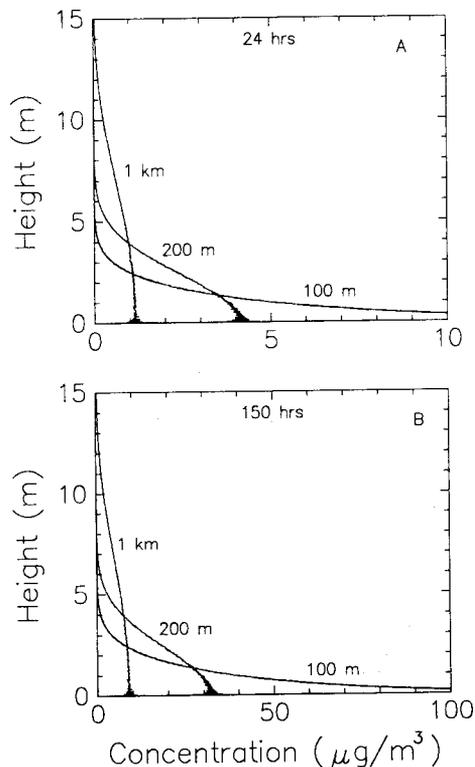


Fig. 6. The atmospheric concentration of lindane at the  $x=100$  m, 200 m, and 1 km collectors after continuously sampling for (A) 24 and (B) 150 h. The surface roughness was assumed to be 0.2 cm for  $0 \leq x \leq 100$  m, 0.5 cm for  $100 \text{ m} \leq x \leq 200$  m, and 1.0 cm for  $200 \text{ m} \leq x \leq 1 \text{ km}$ .

Each particle was allowed to continue its movement through this zone and another distribution of the ratio of horizontal to vertical flux with respect to height was determined at a collector located at  $x = 200$  m. For the zone between  $200 \text{ m} \leq x \leq 1 \text{ km}$ , it was assumed that the roughness length was 1 cm and a third distribution of horizontal to vertical flux was produced. Since these two curves are similar in shape to Fig. 3, they are not reproduced here. Shown in Fig. 6 are the lindane air concentrations at the 100 m, 200 m, and 1 km distances after accumulating in the collector for 24 h (Fig. 6A) and 150 h (Fig. 6B) since pesticide application. Clearly, at the edge of the treated area (i.e., 100 m) the maximum concentration occurs at the soil-air interface. At greater distances from the treated surface, the concentration front steepens with increasing distance as the mixing process continues. At the 1-km distance, the concentration of lindane in the atmosphere below the 5-m height is approximately  $10 \mu\text{g}/\text{m}^2$  per (m field width). This corresponds to an exposure to a total mass of between  $2.5 \mu\text{g}/\text{cm}$  at 1 m,  $4.4 \mu\text{g}/\text{cm}$  at 2 m, and  $6.4 \mu\text{g}/\text{cm}$  at 5 m high when determined as the total mass in a 2-cm interval centered on the specified height. This represents a fraction 0.1 to 0.4% of the total mass emitted (i.e.,  $1768 \mu\text{g}/\text{cm}$ ) during the simulation.

Although the present simulation allows highly site-specific conditions and a wide range of processes to affect the transport process, there are several important factors that have been ignored, for simplicity. The lateral diffusion of pesticides has not been considered. The method could easily include this, but the number of ad-

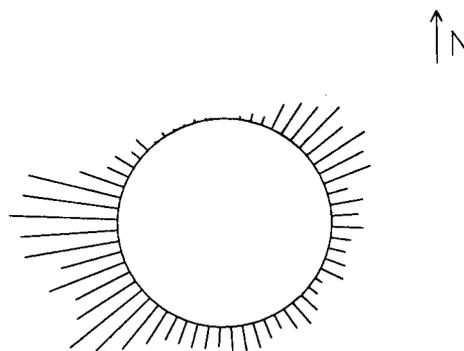


Fig. 7. Wind direction frequency diagram for 151-h time interval. Vector length is proportional to the amount of time wind was blowing in a particular direction.

ditional computations might make this prohibitive. Also, compared with the movement in the downwind direction, lateral movement would be relatively small, especially in the interior regions of the field. Near the field boundaries, however, the concentration gradients may be large enough to cause significant lateral diffusion.

Another factor that has been ignored is wind direction. Although the simulated results assume a constant wind direction, variable direction can be incorporated into the approach without difficulty and would provide more realistic results. The importance of wind direction is readily visible in Fig. 7, which shows the wind direction frequency during the simulation. The frequency is de-

terminated by counting the number of 10-min intervals that the wind direction points into 1 of 60  $6^\circ$  increments. The vector length in Fig. 7 is proportional to the number of times the wind was blowing in the specific direction. This figure clearly demonstrates that the wind direction is highly variable and covers almost every direction, at least during this 151-h sampling period. It would be fairly easy to determine the average wind run and the corresponding flux emitted during each 10-min interval to obtain a picture of the mass or concentration that occurs at each angle from the source area. This would also allow the animation of the plume providing a time-dependent concentration distribution as a function of the wind direction. The researcher could watch the pesticide plume track around the source area in response to changes in the wind speed and direction.

The method also assumes that the sampling interval (here 1 h) is much larger than the time necessary for the air to move from the source area to the collector. Adopting this assumption allows the time delay for movement between the source area and the collector to be ignored. If this is not the case, the delay would have to be incorporated into the model.

Atmospheric degradation described using a first-order degradation process could be added to the model, since the time required for an air mass to travel from the source area to the collector is known. However, since the travel time between the source and collector is relatively small for the distances considered, it is unlikely that degradation would be important. Also, in terms of developing a screening or regulatory model, a more conservative estimate would occur if degradation is not considered. If a pesticide has a very short half-life, however, the degradation process would have to be included, if the simulation is to approximate reality.

## CONCLUSIONS

A model describing the environmental fate of volatile pesticides has been developed by combining the BAM, experimental data describing the wind speed as a function of height, and the trajectory simulation model of atmospheric transport. This model can be used to determine both the pesticide concentrations in the subsurface as well as the mass or concentration of pesticides in the air.

The atmospheric transport simulation is based on the distribution of particles at the collector resulting from the trajectory simulation model. The number of pesticides and their positions are used to determine the ratio of the horizontal to vertical flux. This is crucial information that allows the atmospheric mass or concentration to be obtained. In terms of the computations necessary to obtain this information, it is also the most numerically intensive part of the simulation. For the examples shown herein (i.e., with 2.5 million particles), 12 to 60 h of computer time (on a 486/33) is needed to determine the

flux ratios at the collectors. Clearly, as computer processing speeds increase, problems of this sort will be possible on much shorter time frames. Also, for routine simulations, it may be possible to use far fewer number of particles (i.e., 100 000 to 1 000 000) which would significantly reduce the computational time. A large number of particles was used herein to provide assurance that the particle distribution was close to the theoretical distribution of the process, although comparing the curves marked stable and unstable in Fig. 3 demonstrates that minor differences would occur if only 1 million particles were used.

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