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Pesticides in Air
Part II:

Development of Predictive Methods for Estimating Pesticide Flux to Air

CALIFORNIA ENVIRONMENTAL PROTECTION AGENCY

AIR RESOURCES BOARD
Research Division

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Final Report
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Prepared for:

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Abstract

This project consisted of two parts: (1) A literature review of post-application pesticide volatilization flux values that have been measured or modeled; and (2) development of predictive tools for estimating post-application flux derived from correlations of pesticide physicochemical properties and measured flux reported in the literature (residue drift during application was not considered in this project). Part (2) was the primary goal of the project since a method for predicting pesticide flux would enable Air Resources Board personnel to predict the levels at which pesticides are likely to be found in the ambient air. This information would allow the prioritization of pesticides that pose potential health problems and for which monitoring should be considered. A review of the literature reporting on pesticide volatilization flux from soil, water, crop surfaces, generated using lab chambers, field measurements, or various modeling and other approaches, is divided into four main sub-headings: **Category I** -- field/laboratory flux chambers; **category II** -- meteorological methods; **category III** -- indirect residue analysis; and **category IV** -- mathematical modeling. Of the field/laboratory studies of pesticide volatilization flux, about 53% dealt with residues on soil, while 32% dealt with residues on foliage and the remaining 15% with residues in water. The references fall primarily in **categories I, II**, and **IV** (37%, 27%, and 26%, respectively), with **category III** constituting only 10% of the references. Our own efforts toward developing predictive flux tools took information from **categories I-III** for pesticides applied to soil, incorporated into soil, applied to crop surfaces, and applied to water and correlated the physicochemical properties of those pesticides with their published evaporative flux values, resulting in a set of mathematical correlations. These correlations were derived from the following pesticide physicochemical properties: (1) Soil adsorption [K_{OC}], water solubility [S_W], and vapor pressure [VP] for residues on soil; (2) K_{OC}, S_W, VP, application rate [AR], and depth of incorporation [d] for residues incorporated into soil; (3) VP only for residues on crop surfaces; and (4) VP and S_W for residues dissolved in water (in this case, evaporative flux was normalized to water concentration). In all cases, chemical properties and evaporative flux correlated well. These correlations could be used to estimate flux for chemicals that have known physicochemical properties. Using these estimated flux values as source strengths in the EPA's SCREEN-2 model, we calculated downwind concentrations for carbofuran, oxydemeton-methyl, methidathion, azinphosmethyl, and molinate that compared well with CARB-measured concentrations for these pesticides applied to field crops, orchards, and a rice field.

Acknowledgments

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Summary and Conclusions

Among the necessary inputs for assessing risks associated with pesticides in air is some estimate of exposure. Ideally, this would begin with a straightforward method for estimating volatilization flux for pesticides applied to soil, plants, and water. In searching the literature, we found much pesticide volatilization data, measured under both laboratory and field conditions, for residues originating on soil, plant surfaces, and in water. These data could be used as a basis for estimating flux. We also found a number of mathematical models that ranged in complexity from the "effusion" type, which assumes that volatilization is occurring from a non-adsorptive surface or from a well-mixed body of water (only pesticide vapor pressure and Henry's Law constant are determining factors for these two cases), to descriptive differential equations that include all possible factors that together will significantly affect net volatilization (e.g., diffusion coefficients in water and vapor, mass-transfer coefficients, soil adsorption, Henry's Law constant, vapor pressure, wind speed, etc.). Furthermore, these more complex models were designed to describe dissipation (i.e., volatilization, degradation) essentially over the lifetimes of the pesticides. Our search of the literature garnered about ninety references that describe field, laboratory, and mathematical modeling work.

Rather than attempting to derive yet another complex model from first principles, our objective was to correlate published volatilization flux data that we found in our literature search for pesticides with their vapor pressures, modified by a combination of terms, including pesticide physicochemical properties, to reflect environmental conditions. Furthermore in these correlations, we would not include off-target drift during application, although substantial, but short-term, losses can occur by this route. Instead, the focus of this study was on the volatilization flux data measured within the first few hours after application when post-application volatilization losses are typically the greatest, representing the worst-case scenario for pesticide losses to the atmosphere other than during application. The terms in the correlations would operate on pesticide vapor pressure

[VP], as the underlying driving force for evaporation, and would include soil adsorption coefficient [K_{OC}], water solubility [S_W], application rate [AR], and depth of soil incorporation [d]. Wind speed and temperature were not included as explicit terms since these conditions were implicit in the field data (i.e., field wind speeds and temperatures when the volatilization flux values were measured fell in the ranges 1-4 m/sec and 20-30°C, respectively). However, these environmental conditions are dealt with separately below to illustrate how they can affect volatilization. It was our intention to derive three basic correlations for pesticide residues (1) on soil, including the special case of soil incorporation, (2) on plants, and (3) dissolved in water to develop predictive tools for estimating flux from these treated matrices. We recognize the fact that under typical field conditions, residues of compounds applied to planted and flooded fields will often fall on other surfaces as well (i.e., soil for planted fields and soil and plants for flooded fields) and contribute to the overall observed evaporative flux. While flux measured in the field will reflect all sources, the errant residues often amount to a very small fraction of the total and are probably not significant contributors to overall flux. In our correlations discussed below, we related measured flux to the target surface only. However, this is not to say that for special cases it might be necessary to take errant residues into account to fine-tune a model.

Measured volatilization flux reported in the literature was correlated with compound physicochemical property terms in Ln-Ln relationships. The Ln-Ln relationship was used for two reasons: 1) The flux values and compound physicochemical properties ranged over orders of magnitude (e.g., 10^{-7} to 1 Torr for vapor pressure and 10 to 10^7 $\mu g/m^2/hr$ for flux), making it difficult to visually display the data and to assess the positions of the data points relative to the regression line; and 2) the regression correlation of a linear plot for data spanning orders of magnitude would be heavily weighted by the largest data points. The physicochemical property terms used in the correlations were the following: (1) Residues on soil -- VP/[K_{OC} x S_W]; (2) residues on plants -- VP; and (3) residues dissolved in water -- VP/S_W (for good correlations in this case, field volatilization flux had to be normalized to field water concentration). In the case of residues incorporated into soil, AR was added to the term for residues on soil and a new correlation was derived (i.e., measured flux was correlated with the term $[VP \times AR]/[K_{OC} \times S_W]$). Then this latter term was multiplied by 1/d, where d is depth of incorporation, for calculating flux values of soil-incorporated residues (our results were 74-84% of measured flux values under laboratory and field conditions for triallate [Jury et al., 1980] and trifluralin [White et al., 1977]). This soil incorporation approach assumes that the soil column from the surface to the depth of incorporation contains a uniform distribution of the pesticide and that volatilization is occurring from the surface of this column. This is in contrast to the situation where the pesticide is injected at depth, as for a soil fumigant such as methyl bromide, leading to concentration gradients declining from the point of injection to the soil surface. In this case where flux from the soil surface is controlled by fumigant diffusion to the surface, the soil incorporation correlation would greatly over estimate evaporative flux. For residues in water, the calculated flux normalized to water concentration would be multiplied by this concentration to obtain the absolute flux. The correlation lists contained 12-15 entries each, and had regression coefficient (r^2) values that fell in the range 0.93-0.99. Table S-1 contains the list of the chemicals with their properties and evaporative flux values used in all of the correlations. The graphed correlations are shown in Figures S-1 through S-3.

Measured vs modeled downwind concentrations. In the following discussions concerning the use of estimated flux in the EPA's SCREEN-2 dispersion model to predict downwind concentrations, it should be pointed out that we have not included any factors for chemical degradation in air. If atmospheric degradation were occurring at a significant rate, as can be the case for the organophosphate insecticide parathion and for the dinitroaniline herbicide trifluralin (Woodrow et al., 1978), downwind concentrations of the parent compounds could be markedly reduced over those predicted by the SCREEN-2 model.

Table S-1. Properties (20-30°C) of reference compounds for evaporative flux correlations.

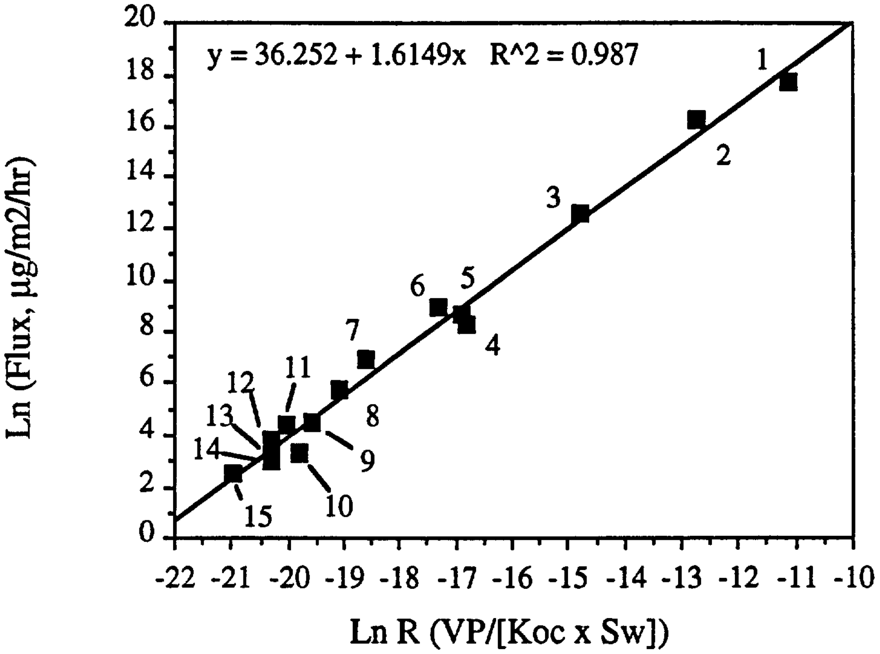
Compound	K_{oc}, ml/g	S_w, mg/L	VP, torr[a]	Ln Flux ($\mu g/m^2/hr$)[b]
Beacon oil	1,000	100	1.5 (S, P)	17.79(S)/17.15(P)
Chevron oil	1,000	100	0.3 (S, P)	16.27(S)/14.84(P)
Dodecane	1,000	100	0.14 (P)	13.80
n-Octanol	--[c]	590	0.13 (P)	14.12
Eptam	240	375	3.4×10^{-2} (S, W)	12.58(S)/9.39(W)
PCNB	5,000	0.44	1.1×10^{-4} (S)	8.29
Trifluralin	8,000	0.3	1.1×10^{-4} (S, P)	8.70(S)/7.37(P)
Fonofos	870	13	3.4×10^{-4} (S)	8.99
Lindane	1,100	7	6.45×10^{-5} (S)	6.92
Dieldrin	9,817	0.2	1×10^{-5} (S) 4.94×10^{-6} (P)	5.77 (S) 5.15 (P)
Chlorpyrifos	6,070	0.982	1.87×10^{-5} (S)	4.52
Diazinon	1,000	48.6	1.2×10^{-4} (S) 1.12×10^{-5} (P) 4.8×10^{-5} (W)	3.92 (S) 6.81 (P) 5.87 (W)
Atrazine	100	33	6.75×10^{-6} (S)	4.43
Dacthal	3,200	0.5	2.5×10^{-6} (S)	3.00/3.14
p,p'-DDT	139,959	0.00335	7.2×10^{-7} (S) 3.3×10^{-7} (P)	3.82 (S, P)
Prometon	150	720	8.26×10^{-5} (S)	2.53
Tridiphane	5,600	1.8	2.2×10^{-4} (P)	8.87
Pendimethalin	5,000	0.3	3×10^{-5} (P)	6.95
2,4-D (iso-octyl)	500	8	2×10^{-5} (P)	6.51
Toxaphene	1,514	3	4×10^{-6} (P)	5.29
Deltamethrin	22,000	0.002	1.5×10^{-8} (W)	8.23[d]
Et-Parathion	5,000	15	5.2×10^{-6} (W)	5.15
Me-Parathion	5,100	25	6.3×10^{-6} (W)	4.35
Mevinphos	44	6×10^5	2.2×10^{-3} (W)	0.83
Molinate	89	688 800	5.6×10^{-3} (W) 3.1×10^{-3} (W)	7.42/7.69 6.44
Thiobencarb	900	30	1.5×10^{-5} (W)	5.64/5.44

[a] S = soil; P = plant; W = water. The multiple vapor pressure values reflect the varying environmental conditions under which evaporative flux was measured.

[b] For flux from water, the values have been normalized to compound water concentration.

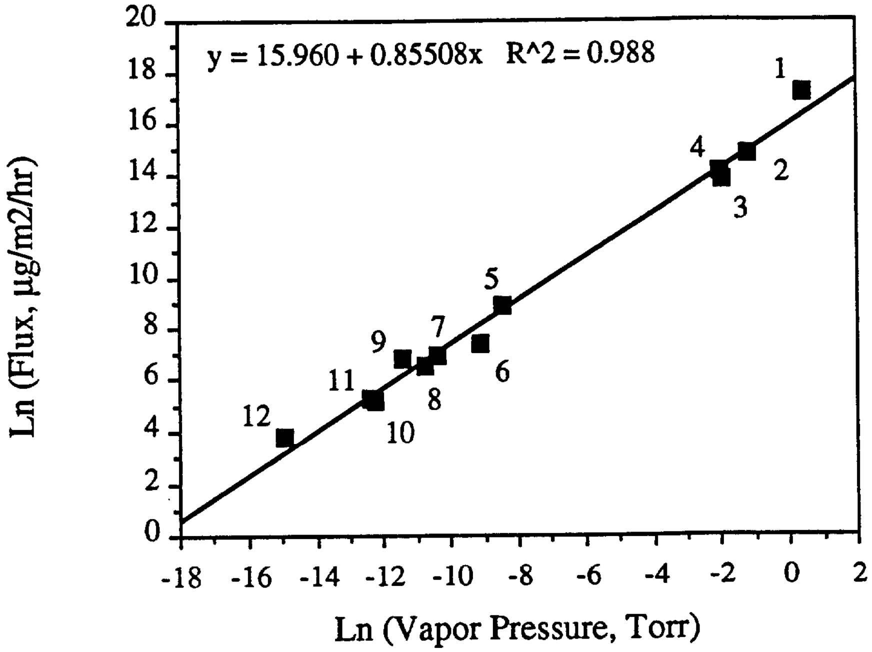
[c] Unavailable.

[d] Two separate fields were studied, but the same normalized flux was measured.



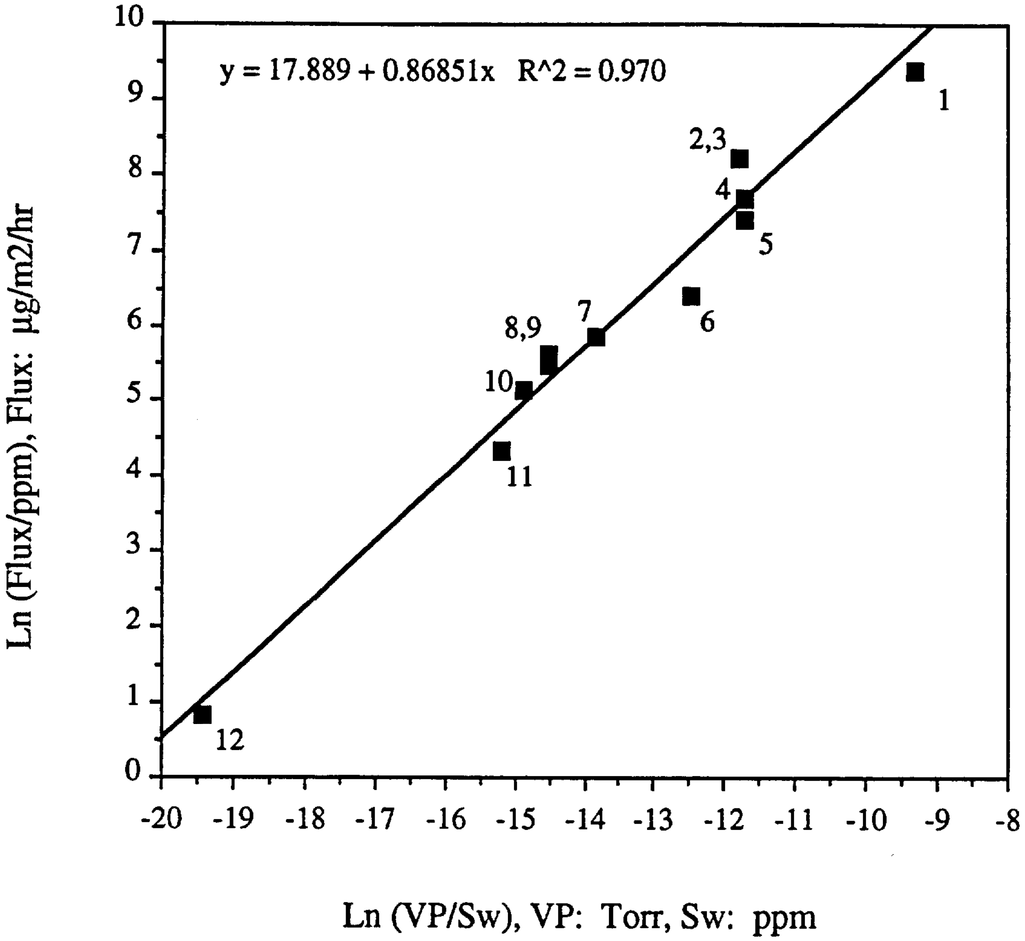
1. Beacon oil (Woodrow et al., 1983)
2. Chevron oil (Woodrow et al., 1983)
3. Eptam (Nash, 1983)
4. PCNB (Nash and Gish, 1989)
5. Trifluralin (Glotfelty et al., 1984)
6. Fonofos (Whang et al., 1993)
7. Lindane (Majewski et al., 1990)
8. Dieldrin (Nash, 1983)
9. Chlorpyrifos (Majewski et al., 1990)
10. Diazinon (Majewski et al., 1990)
11. Atrazine (Nash and Gish, 1989)
12. p,p'-DDT (Willis et al., 1983)
13. Dacthal (Majewski et al., 1991)
14. Dacthal (Ross et al., 1990)
15. Prometon (Spencer and Cliath, 1990)

Figure S-1. Correlation of pesticide flux from soil with chemical properties.



1. Beacon oil (Woodrow et al., 1983)
2. Chevron oil (Woodrow et al., 1983)
3. Dodecane (Roberts, 1981)
4. n-Octanol (Tinsley, 1979)
5. Tridiphane (McCall et al., 1986)
6. Trifluralin (Nash and Hill, 1990)
7. Pendimethalin (Jenkins et al., 1993)
8. 2,4-D (iso-octyl) (Grover et al., 1984)
9. Diazinon (Glotfelty et al., 1990)
10. Dieldrin (Nash and Hill, 1990)
11. Toxaphene (Willis et al., 1983)
12. p,p'-DDT (Willis et al., 1983)

Figure S-2. Correlation of pesticide flux from "inert" surfaces (plants, glass, plastic) with vapor pressure.



1. Eptam (field) (Cliath et al., 1980)
2,3. Deltamethrin (field) (Muir et al., 1985)
4. Molinate (field) (Soderquist et al., 1977)
5. Molinate (field) (Ross and Sava, 1986)
6. Molinate (lab) (Seiber et al., 1986)
7. Diazinon (lab) (Sanders and Seiber, 1983)
8. Thiobencarb (field) (Ross and Sava, 1986)
9. Thiobencarb (lab) (Seiber et al., 1986)
10. Ethyl Parathion (lab) (Sanders and Seiber, 1983)
11. Methyl Parathion (lab) (Sanders and Seiber, 1983)
12. Mevinphos (lab) (Sanders and Seiber, 1983)

Figure S-3. Correlation of evaporative flux from water with chemical properties.

Treated foliage. The correlation for residues on plant ("inert") surfaces was used to derive estimated flux values which were then used in the EPA's SCREEN-2 dispersion model to calculate pesticide residue concentrations in air downwind of treated fields for comparison with measured downwind concentrations. The pesticides selected by Air Resources Board personnel for comparison were the following: (1) Carbofuran applied to alfalfa; (2) oxydemeton-methyl applied to broccoli/cauliflower; (3) methidathion applied to mature orange trees; and (4) azinphosmethyl applied to mature walnut trees. Table S-2 summarizes the model results and the measured values. In all cases, the modeled and measured results compared well. For carbofuran, concentration was measured at a distance of 20 meters from the field, while the model could not estimate the concentration closer than 26 meters. Even so, the results compared to within a factor of two. For those cases where measured concentrations were less than the limit of quantitation (LOQ), the model predicted values were also less than or equal to the LOQ.

The estimated flux values for carbofuran and oxydemeton-methyl that were derived from the correlation for residues on plant surfaces were used in the dispersion model without modification. The dispersion model assumes that the source is a square-planar surface. This assumption is a good approximation for fields that are close to being planar (e.g., alfalfa, row crops, etc.). However, for residues on mature, leafed-out trees in an orchard, the dispersion model will underestimate downwind concentrations because residues would be volatilizing from a surface area considerably greater than that for the square-planar surface containing the orchard. Therefore, for methidathion and azinphosmethyl, the estimated flux values had to be adjusted by the total leaf surface area (m^2/tree) and the orchard tree density (tree/m^2). For example, the estimated flux for methidathion derived from the correlation for residues on plant surfaces was 4.96×10^{-8} g/m^2/sec. Using this value in the SCREEN-2 model gave a concentration of about 0.12 μg/m^3 15 meters downwind of the field, compared to a measured concentration of 1.4 μg/m^3. The modeled value of 1.6 μg/m^3 reported in Table S-2 was derived in the

Table S-2. Comparison of measured and modeled downwind pesticide concentrations.

		Concentration in air, µg/m^3	
Pesticide	Commodity	Measured[a]	Model
Carbofuran	alfalfa	0.60[b]	0.39[b]
Oxydemeton-methyl	broccoli/cauliflower	<LOQ[c]	11[c]
Methidathion	oranges	1.4[d]	1.6[d]
Azinphosmethyl	walnuts	<LOQ[e]	0.23[e]

[a] Provided by the Air Resources Board.

[b] Sampler was 20 meters from field. Model distance was 26 meters.

[c] Sampler and model distance from field was 15 meters.
LOQ (limit of quantitation) = 70 µg/m^3.

[d] Sampler and model distance from field was 15 meters.

[e] Samplers were 15-25 meters from field. Model distance was 27 meters.
LOQ = 0.23 µg/m^3.

following way using the total leaf surface area per tree of 500 m^2 for mature orange trees (Spencer et al., 1973) and the typical orchard tree density of 0.028 tree/m^2 (~36 m^2 are required for each orange tree [Bienz, 1993]):

$$(4.96 \times 10^{-8}\ g/m^2/sec) \times (500\ m^2/tree) \times (0.028\ tree/m^2) = 6.94 \times 10^{-7}\ g/m^2/sec$$

This adjusted flux value was then used in the SCREEN-2 model. The same adjustment was also made for azinphosmethyl on walnuts, but using the leaf surface area and orchard tree density for oranges. By doing this gave only an approximation for walnuts, since walnut trees are typically larger than orange trees, so that orchard tree density is probably less, and, furthermore, total leaf surface area for walnuts may not be as great.

While adjustment for leaf surface area gave reasonable results for the particular orange and walnut tree examples, it should be noted that the results for other examples will be affected by total leaf surface area, which will vary as a function of the type and size of the orchard and of the stage of leaf development. Furthermore, an assumption in the approach described above is that leaf surface area is uniformly coated with pesticide. In actuality, this would not be the case, and pesticide distribution would depend on the application method. However averaged over the large surface area of a typical orchard, pesticide distribution would probably not affect evaporative flux as much as the meteorological conditions and pesticide physicochemical properties.

Treated water. The correlation for residues volatilizing from treated water was used to derive estimated flux values for use in the SCREEN-2 model to calculate downwind concentrations for comparison with measured downwind concentrations for the rice herbicide molinate. For daytime and nighttime sampling, we used molinate vapor pressures and water solubilities at 30°C (9.92 x 10^{-3} torr and 576 ppm) and 20°C (3.1 x 10^{-3} torr and 800 ppm), respectively. The ratios of the vapor pressures and water solubilities were used in the correlation for residues volatilizing from treated water to derive flux values normalized to water concentration. These normalized flux values were then multiplied by the range of molinate concentrations in water (2.34-4.68 ppm, derived from a range of water depths [12-24 cm] and the application rate [5.6 kg/ha]) to derive absolute flux values used in the SCREEN-2 dispersion model. Table S-3 summarizes the sampling periods post-application, the pertinent field data required by the model, such as the wind speed and atmospheric stability factors that existed during the sampling periods, and the measured and modeled downwind concentrations. The modeled concentrations for the actual downwind sampling distances were derived from extrapolation of concentration vs distance plots and the modeled concentrations were adjusted for wind speed and fraction of molinate remaining in the field water. In all cases, measured and modeled concentrations compared well, which was surprising for the first sampling period at least since during this time, when granular molinate would be dissolving in the field water, much of the airborne residue would be expected to originate from chemical intercepted by pad dikes and other surfaces and general contamination from drift during application. However, molinate dissolution in water can be rapid, leading to concentration maxima on the day of application.

Uncertainty in estimated flux. The calculated or estimated flux values and downwind air concentrations should be regarded as screening tools, suitable for a first-cut ranking of chemical-surface combinations which might lead to downwind exposures of concern from a risk assessment viewpoint. Needless to say, there will be uncertainty in any such simplified approach to estimating exposures to airborne residues. While the extent of the uncertainty is not clear, its origin would reside in the measured flux data and the compound physicochemical property data used to derive the correlations. Much of the flux data were obtained using micrometeorological methods, such as aerodynamic-gradient, theoretical profile shape, energy balance, eddy correlation, and integrated horizontal flux, which have been found to be statistically equivalent (Majewski et al., 1990). The aerodynamic-gradient is the most commonly used method, but it may underestimate evaporative losses (Glotfelty et al., 1984; Grover et al., 1988) and do so by as much as 50% in some cases (Seiber and McChesney, 1987). Furthermore, associated uncertainties

Table S-3. Modeled vs measured air concentrations downwind of a molinate-treated rice field.[a]

Sample ID	Sampling Period	Downwind Distance, m	Atmospheric Stability	Molinate in water (fraction)[b]	Wind speed, m/s (*u*)	Air concentration, $\mu g/m^3$	
						Measured	Modeled[c]
3N1	3(D)	9.14	B	0.99	3.6	11.3	6.2-12.5 (ave = 11.2)
4N1	4(D)	9.14	C	0.98	5.4	22.6	10.3-20.5 (ave = 18.5)
5N1	5(N)	9.14	E	0.93	3.1	10.1	7.4-14.9 (ave = 13.4)
6N1	6(D)	9.14	B	0.85	3.6	8.6	5.3-10.6 (ave = 9.5)
8S1	8(D)	22.9	B	0.72	4.0	6.8	4.4-8.9 (ave = 8.0)
10S1	10(D)	22.9	B	0.61	4.5	6.0	3.8-7.6 (ave = 6.8)

[a] Field sample data provided by the California Air Resources Board.

[b] Seiber et al., 1986; Seiber and McChesney, 1987.

[c] Adjusted for wind speed (*u*/3) and fraction molinate remaining in the field water.

in flux values using this method can be as much as 50% (Majewski et al., 1995). Taken together, the uncertainties inherent in the flux measurement methods and the physicochemical property data were reflected in the absolute percent variance of the individual data points relative to the correlation regression lines. Average variance was 8.2±5.6%, 5.2±5.6%, and 5.4±4.3% for soil, plant, and water, respectively. These variances were not significantly different at the 95% confidence level indicating that the results were independent of the treated matrix.

Outliers. Several outliers that did not fit the correlations included chlorpyrifos (Whang et al., 1993), triallate and trifluralin (Grover et al., 1988), chlorpropham (Turner et al., 1978), and dieldrin (Willis et al., 1972) on soil, and dieldrin and heptachlor (Taylor et al., 1977) on pasture grass. Overall, observed evaporative flux was 3 times (triallate and trifluralin incorporated into soil) to almost 4,000 times (chlorpropham on wet soil) flux values predicted using the correlations. By assuming that chlorpropham and dieldrin volatilized from water on soil that was maintained moist and that soil-incorporated triallate and trifluralin were not evenly distributed along the soil column (i.e., residues were in the upper 40-50% of the soil column), it was possible to achieve agreement between observed and predicted flux to well within a factor of two. For chlorpyrifos residues on soil and dieldrin and heptachlor residues on pasture grass, the 1-2 order of magnitude greater observed flux remains unexplained. No outliers have been found for residues in water, although we have not made an exhaustive search of the literature.

Effect of wind speed and temperature. Limited information is available in the literature regarding the effect of wind speed and temperature on residue evaporative flux from soil. What information there is suggests that temperature has a greater effect on volatility than does wind speed and that volatility increase may be greater for temperature increases at higher initial temperatures. It is the temperature of the treated surface that would affect volatility more than air temperature, although published studies often only report air temperature assuming that the treated surface is in thermal equilibrium with the atmosphere. However, soil surface temperatures can be 25°C greater than those of the air (Woodrow et al., 1983). The behavior of residues on "non-interactive" surfaces, such as glass, is similar, except that a somewhat greater increase in wind speed is necessary to achieve the same increase in volatility compared to soil. This implies that surface texture may be important, where, for a given wind speed, a rough surface may aid in the transfer of residues to the atmosphere through turbulence of the air moving over the surface causing a decrease in the thickness of the stagnant layer at the surface. By contrast, air flow over a relatively smooth surface (e.g., glass) would be close to laminar, so increased pesticide volatility would require a greater increase in wind speed to affect the stagnant layer at the surface in the same way as for the soil case.

The effect of wind speed and temperature changes on pesticide evaporative flux is assumed to occur through changes in the pesticide vapor pressure. Wind speed and temperature terms are essentially operators acting directly on vapor pressure. The wind speed term would have the form $[1 + n\mathbf{u}]$ (Woodrow et al., 1983), where **u** is wind speed (m/sec) and $n \approx 3\text{-}6$, depending on the compound's heat of vaporization. The effect of temperature on the vapor pressure is expressed in the integrated form of the Clausius-Clapeyron equation

$$\mathrm{Ln}\ (\mathrm{VP}_2/\mathrm{VP}_1) = -(\Delta H/R)\ (1/T_2 - 1/T_1)$$

where ΔH is the heat of vaporization (cal/mole) and R is the gas constant (1.99 cal/deg mole). To account for both wind speed and temperature together, the vapor pressure of a pesticide on a treated surface would be operated on by the term

$$e^{-(\Delta H/R)\ (1/T2-1/T1)} \times [1 + n\mathbf{u}]$$

where T_1 corresponds to the initial vapor pressure. Since the correlations developed in this study implicitly included a range of temperatures (20-30°C) and wind speeds (1-3 m/sec), this term may be used in cases where the pesticide vapor pressure is known only at some temperature other than the temperature(s) implicit in the correlations and under different wind conditions that would deviate considerably from those implicit in the correlations.

Other methods for estimating flux. We used the Eureka Laboratories (EL) models (Leung et al., 1978) and various forms of the Knudsen effusion (KE) equation (Tinsley, 1979) to estimate flux for residues on soil and plant surfaces and dissolved in water. The results are listed in Table S-4 along with estimated flux values calculated using our correlation regression equations (Figures S-1 through S-3). Comparing our regression results with results from the other methods indicated that these two methods overestimated flux from soil (~2-300 times for both EL and KE) and underestimated flux from water (50-200 times for EL and 5-10 times for KE). However for residues on plant surfaces, EL and KE both agreed with the regression results to within a factor of two. These results lend support to the assumption that plant surfaces are essentially non-interactive, at least for freshly applied pesticides, and vapor pressure (included in the EL and KE approaches) would be the predominate compound property affecting volatilization. For soil and water, on the other hand, the EL and KE approaches, since they are derived from simple concepts concerning water flux (EL) and molecular effusion (KE), do not take into account all the myriad interactions possible for chemical residues in these matrices. Estimation methods that include chemical flux data measured under a variety of actual field and laboratory conditions (e.g., the correlation approach used in this study), because they would implicitly contain the variety of chemical-matrix interactions, stand a better chance of providing useful predictive data.

Recommendations

Based on the results of this study, we make the following recommendations:

1). We recommend correlating the physicochemical properties of pesticides with their measured evaporative flux values as a direct and relatively simple way of estimating flux for chemicals applied under similar conditions. This type of correlation will implicitly contain the range of environmental conditions under which the evaporative flux values were measured. This is an advantage since explicit, detailed terms describing environmental conditions are not required, as would be the case with the more descriptive models. The

Table S-4. Comparison of regression-calculated flux values with values estimated using the Eureka Laboratories (EL) and Knudsen effusion (KE) methods.

		Flux, µg/m^2/hr[a]		
Compound	Matrix	EL[b]	KE[c]	Regression[d]
Eptam	Soil	2×10^6	1×10^6	2×10^5
Trifluralin		2×10^4	5×10^3	8×10^3
Fonofos		4×10^4	1×10^4	4×10^3
Lindane		1×10^4	3×10^3	0.5×10^3
Dieldrin		488	466	224
Chlorpyrifos		3×10^3	836	103
Diazinon		2×10^4	5×10^3	70
Atrazine		853	237	51
Dacthal		392	109	33
p,p'-DDT		35	32	32
Prometon		322	3×10^3	10
Tridiphane	Plants	--	1×10^4	6×10^3
Trifluralin		1×10^4	5×10^3	4×10^3
Pendimethalin		2×10^3	1×10^3	1×10^3
2,4-D (isooctyl)		2×10^3	897	819
Diazinon		984	471	499
Toxaphene		405	196	207
Dieldrin		480	232	248
p,p'-DDT		31	15	24
Deltamethrin	Water	2×10^{-2}	0.1-0.5	1-4
Eptam		215	8×10^3	4×10^4
Molinate		60	$(2\text{-}3) \times 10^3$	$(4\text{-}7) \times 10^3$
Thiobencarb		1	14	111

[a] Values rounded to the nearest whole number.

[b] EL = Eureka Laboratories (Leung et al., 1978).

[c] KE = Knudsen effusion.

For soil and plant surfaces: Flux = $1.98 \times 10^{-5}\ P\ (M/2\pi\mathcal{R}T)^{1/2}$.

For water: $C_t = C_o \exp[-(\beta H f/d)t]$, where C_t is solute concentration at time t, H = Henry's law constant, d = depth (cm), and $f = (M/2\pi\mathcal{R}T)^{1/2}$.

[d] From regression equations (Figures S-1 through S-3) using chemical properties in Table S-1.

exception that would require explicit temperature and/or wind speed terms would be for pesticides used under conditions not included in the correlations. However, since many pesticides are applied under similar conditions, the correlations in this study should give reasonable flux estimations for a wide range of different chemicals.

2). We recommend using the flux values estimated from these correlations as source strength inputs to the SCREEN-2, or other (e.g., ISCST-II), dispersion model for order-of-magnitude calculations of downwind concentrations in air. This proved to be a promising approach because of the good comparison between calculated and measured concentrations for a set of pesticides applied to field crops and orchards. We feel this approach worked partly because the environmental conditions for the non-correlated chemicals fell in the range implicit in the correlations described in 1). It might be prudent for pesticide screening evaluations to use a wide range of possible meteorological conditions as input data to the SCREEN-2, or other, model to obtain conservative, overestimated downwind concentrations for regulatory purposes.

3). If the environmental conditions for non-correlated pesticides are outside the range of conditions for the pesticides used to derive the correlations, then we recommend modifying the vapor pressures of the non-correlated pesticides by the multiplicative factor

$$e^{-(\Delta H/R)\,(1/T2-1/T1)} \times [1 + n\mathbf{u}]$$

to compensate for temperature and/or wind speed differences. However, this recommendation is contingent on further refinement and validation of this multiplicative factor to see if it will actually give reasonable results.

4). Future work dealing with the estimation of downwind pesticide concentrations using dispersion models should also include deposition and reaction rate constants for those compounds that might settle out and/or undergo significant chemical conversion between the time the residues are released from the treated fields and the time they reach the downwind sampling sites. Furthermore, it might be important to consider conversion on the treated surface, especially for those cases where the conversion products are also volatile and might pose a threat to human health.

Introduction

Volatilization of pesticides after application to soil, water, and plant foliage is often a significant pathway for losses to the atmosphere, with subsequent movement to non-target areas. Losses can also occur as particulates through the action of wind erosion. However, losses of pesticides from treated surfaces typically occur in the form of vapors for those compounds that are volatile. Factors that influence evaporative losses from treated surfaces include pesticide vapor pressure along with environmental conditions that control movement away from treated surfaces (e.g., molecular diffusion and eddy dispersion) (Thornthwaite and Holzman, 1939; Parmele et al., 1972). A volatilizing pesticide will first diffuse through a stagnant air boundary layer immediately above the treated surface and then be carried away by turbulent flow of the air. As the wind speed increases, the depth of the stagnant air boundary layer will decrease, resulting in increased volatilization rate of the applied pesticide. The roughness of the surface will influence volatilization rate by leading to greater turbulence closer to the surface as the roughness increases, resulting in greater turbulent transfer of deposited residues to the atmosphere.

Other factors that influence volatilization include sorption to soil and Henry's law constant (ratio of pesticide vapor pressure and water solubility). The effective vapor pressure of a pesticide can be greatly reduced by soil sorption, which is primarily determined by soil organic matter content. However, moisture content of the soil can off-set the effect of sorption on volatilization by enhancing pesticide volatility in moist soils, compared to these soils when dry, because water will compete for sorption sites. Often the observation is that when pesticides are applied to soil surfaces, pesticide flux remains high until the surface dries. Flux will then be influenced by soil sorption, and when surface residues become depleted diffusion of pesticides from within the soil penetrated by the spray then becomes important. In this regard, depth of soil incorporation might also be considered a factor influencing volatilization, and movement through soil to the surface is in part accomplished by diffusion through soil vapor spaces and through soil moisture.

So, diffusion coefficients for pesticides in water and vapor are important here, as well as soil porosity. Furthermore, the movement of water to the soil surface can also carry pesticide residues to the surface (wick effect caused by capillary action). In this regard, the pesticide's Henry's law constant becomes important. This has been borne out by studies that have compared the volatilization of a number of incorporated pesticides for similar soils and found that volatilization depended primarily on the Henry's law constant (Jury et al., 1984b). These investigators found that when the water solubility was relatively high (low Henry's law constant), pesticide residues would accumulate at the surface faster than they would evaporate, resulting in increased flux over time. In contrast to this, pesticides with relatively low water solubilities (high Henry's law constant) would show declining flux over time regardless of the movement of water to the surface.

A pesticide's Henry's law constant is also important for moderately soluble residues in treated water, since distribution between the air-water interface is determined by this term. Depending on the conceptual model used to describe evaporation from water, flux is directly proportional to the Henry's law constant. However, complicating this may be situations where flux is also a function of concentration gradients in the water so that component diffusion to the water surface controls flux. The latter would be characteristic of deep water where mixing may not be efficient in contrast to turbulent water with efficient mixing and large surface-to-volume ratios where flux would be greater (e.g., flooded rice fields) (Tinsley, 1979). For water-miscible residues, volatilization of each component dissolved in water may be simply controlled by the component's partial pressure (derived from a component's saturation vapor pressure, mole fraction, and solution activity coefficient [Raoult's law]), assuming no concentration gradients and minimal interaction between the solution components.

In recent years, many investigators have attempted to correlate pesticide volatilization with the various influencing factors mentioned in the foregoing discussion in descriptive equations that can be used to predict losses from treated surfaces. These equations, or models, range in complexity from the "effusion" type, which assume that volatilization is occurring from a non-adsorptive surface or from a well-mixed body of water (only pesticide vapor pressure is the determining factor), to descriptive differential equations that include all possible factors that together will significantly affect net volatilization (e.g., diffusion coefficients in water and vapor, mass-transfer coefficients, soil adsorption, Henry's law constant, vapor pressure, wind speed, etc.). Furthermore, these models were formulated to describe volatilization behavior of pesticides over prolonged periods of time (i.e., days and weeks) after application.

Our approach in this study was based on the fundamental idea that pesticide saturation vapor pressure is the underlying driving force for evaporation, and that other factors that influence volatilization, such as soil adsorption, depth of soil incorporation, or water solubility, can be viewed as operators on the saturation vapor pressure to give a reduced, or effective, vapor pressure under a particular set of conditions. So, in line with this thinking, we decided that instead of deriving another complex model from first principles, we would correlate published volatilization flux data for pesticides with their vapor pressures, modified by a combination of terms, including pesticide physicochemical properties, to reflect environmental conditions. Furthermore in these correlations, we decided to focus on the volatilization flux data measured within the first few hours after application because losses are typically the greatest during this time period (except for certain types of soil applications that show a volatilization delay [e.g., soil-injected Telone and metham sodium]) and would represent the worst-case scenario for pesticide losses to the atmosphere. The terms in the correlations that would operate on pesticide vapor pressure include soil adsorption coefficient, water solubility, application rate, depth of soil incorporation, wind speed, and temperature. It was our intent to derive three basic correlations for pesticide residues (1) on soil (with the special case of soil incorporation), (2) on plants, and (3) dissolved in water.

In addition to these correlations, our search for volatilization flux values resulted in a literature data base that is also included in this report as an important part of the study. We were able to garner about ninety references that describe field, laboratory, and mathematical modeling work. However, while we acknowledge the fact that pesticide residues will move from target sites primarily as spray drift during application and primarily as volatilized vapors post-application, our focus, both for the correlations and the literature search, was exclusively on post-application volatilization, and particularly within the first few hours after treatment when volatilization would be the greatest.

The following report covers three main project objectives:

1) Literature review. The list of references in the review contains, if not all, at least the most pertinent studies covering a range of approaches to the estimation of pesticide volatilization flux.

2) Derivation of correlations from which estimated flux values would be obtained. The primary intent of the correlations is to provide a way of ranking pesticides, as well as to provide flux values for specific pesticides that can be used as input data for dispersion models.

3) Use of estimated flux values as source terms in a dispersion model to predict downwind concentrations. Flux values calculated using the correlations derived in this study will be used as source terms in the EPA's SCREEN-2 model to calculate downwind concentrations for comparison with concentrations measured in the field at or near the same downwind distances. This is a major goal of the study that would provide a way of estimating downwind concentrations for exposure assessment without having to conduct a field study.

Literature Review

In searching the literature, it was interesting to note that of the field/laboratory studies of pesticide volatilization flux that we have found, about 53% dealt with residues on soil, while 32% dealt with residues on foliage and the remaining 15% with residues in water. This distribution may only reflect agricultural practices where many pesticides are used to prepare soil for planting and where pesticides applied to planted fields leave residues in the soil that outlast the crop, and where the crop-related residues are harvested with the crop and become a food-source problem rather than an environmental problem. While the crop remains in the field, however, it provides a surface for volatilization flux and a medium for chemical reaction that would affect the flux term for the parent compound. A notable example of the application of pesticides to water is rice culture which exists only in a few regions of the U.S.

With regard to techniques for determining volatilization flux, the literature references can be divided into four sub-categories, where some of the references that cover more than one technique are included in more than one category:

Category I -- field/laboratory flux chambers.

Category II -- meteorological methods.

Category III -- indirect residue analysis.

Category IV -- mathematical modeling.

The references fell primarily in **categories I, II,** and **IV** (37%, 27%, and 26%, respectively), with **category III** falling far behind (10%). Flux chambers (**category I**) are used to enclose treated surfaces so that volatilizing residues can be easily trapped and quantified for flux estimation (*1-36*), as opposed to open-field methods where pesticide concentration, temperature, and wind speed gradients are measured above a treated surface and used to estimate flux (**category II**) (*37-62*). These two approaches represent direct measurement techniques, in contrast to indirect techniques which involve quantifying residues in the treated matrix (e.g., soil, water, foliage) and deducing volatilization from the results (**category III**) (*63-72*). The latter appears not to be a popular technique primarily because it works only for stable residues, or for residues where volatilization is the predominant loss route. For example in an earlier study involving rice culture, volatilization of methyl parathion from the treated water, as determined by the analysis of air samples, was about 1-2 orders of magnitude less than volatilization determined from water analysis (*24*). This difference was due to the fact that other routes of dissipation, in addition to volatilization, existed for the water-borne residues of this organophosphate. Finally, the use of mathematical models (**category IV**) is growing in popularity (*73-98*). The high level of interest in the use of models to estimate flux is not surprising in light of ever decreasing budgets (in general, less funding is required for models than for a field study) and increasing pressures to regulate pesticide use, where there may not be the time for a full-blown field or laboratory study. However, the integrity of the models can only be assured by basing them on data obtained using techniques from the other three categories.

Of the experimental techniques available for the determination of volatilization flux, the costliest are those described under **category II**, where expensive equipment and substantial labor and time investments are required. However, data obtained using **category II** techniques can be very reliable, partly because these techniques, based on equilibrated wind, temperature, and concentration gradients above the field, will give flux values averaged over the entire treated field (*40-44*). This is in contrast to **category I** techniques, where chambers in the field can give only localized point-source flux terms. Extrapolating data obtained in this way to the entire field is risky at best because of the wide variations point-to-point in field concentrations (*35*). However, this problem can be overcome by using model "fields" entirely contained within a chamber (*8, 10, 17, 21, 22, 28-30, 34*), where most of the residues volatilizing from the contained treated surface can be trapped for analysis, and where the walls of the chamber can be rinsed to obtain the remaining residues for mass balance purposes. Because of the relatively low cost and ease of use, chamber techniques remain popular, and can be a fairly reliable source of data for regulatory purposes.

Partly because of relative cost and partly due to the predictive ability of computer-based models, **category IV** techniques are viewed by regulatory agencies with some interest. During their development, these techniques require validation by using them in combination with experimental data as input terms for the models. It is hoped that once the experimental data have been used to "adjust" or "train" (*74*) the models, they can be used to predict the outcome for new flux scenarios without having to do the field work. However, the most realistic approach would probably involve a combination of minimal field/laboratory measurements and computer-based models for the estimation of flux.

A case in point to illustrate a combination of field and computer efforts comes out of two recent studies (1992 and 1994) where we measured methyl bromide volatilization and fate in Monterey County, California. Using techniques described in **category II**, we measured air concentrations of methyl bromide above fumigated fields to determine its flux and downwind from the fields to determine its fate using the methyl bromide concentration and meteorological data collected. In cooperation with personnel from the California Department of Pesticide Regulation, downwind concentration data from one of these studies (1992, *44*) were used in the Industrial Source Complex (ISC) model to back-calculate to the flux term for methyl bromide and the result was compared with the measured term. The ISC-calculated result, based only on a handful of downwind samples, agreed to within a factor of two with volatilization flux that we determined using **category II** techniques. Our 1994 data have yet to be evaluated in this way. But, if this last study upholds the factor of two comparison between dispersion model calculated and measured flux for methyl bromide, we may recommend this as one possible approach, that would require minimal effort and materials, for the determination of pesticide flux. All that would be involved would be to take a few air samples at one downwind station close to a treated field, use the concentration data as input to a dispersion model to calculate a flux term, and then use this flux term to calculate concentrations in air at any distance downwind for risk assessment purposes. This approach would also allow us to evaluate and validate a particular dispersion model (e.g., ISC, SCREEN-2) as a basis for making recommendations.

Modeling Volatilization Flux

The literature review gave us a data base from which to draw compound/flux combinations for our correlations discussed below. Table 1 is a composite of all the compounds used to derive these correlations for treated soil, plant, and water matrices under both field and laboratory conditions. We have included the form of the compound (e.g., pure, technical, emulsifiable concentrate [EC] and wettable powder [WP] formulations), application rates (0.01-1,040 kg/ha), test conditions (i.e., wind speed and temperature), the method used to determine evaporative flux (i.e., residue analysis of the treated matrix or of the air sweeping the treated matrix in enclosed chambers, and meteorological methods), and the time of year and locations for field applications. The flux values used in the correlations below were the "worst-case" situations, which frequently occurred in the field within a few hours after application. These flux values were correlated with compound vapor pressures, modified by compound properties appropriate to the treated matrix (e.g., soil adsorption coefficients [K_{OC}] and water solubility [S_W] for treated soil and S_W for treated water). These correlations for treated soil, plant, and water matrices were formulated as Ln-Ln plots for two reasons: 1) The flux values and compound physicochemical properties ranged over orders of magnitude (e.g., 10^{-7} to 1 Torr for vapor pressure and 10 to 10^7 µg/m^2/hr for flux), making it difficult to visually display the data and to assess the positions of the data points relative to the regression line; and 2) the regression correlation of a linear plot for data spanning orders of magnitude would be heavily weighted by the largest data points. We recognize the fact that under typical field conditions, residues of compounds applied to planted and flooded fields will often fall on other surfaces as well (i.e., soil for planted fields and soil and plants for flooded fields) and contribute to the overall observed evaporative flux. While flux measured in the field will reflect all sources, the errant residues often amount to a very small fraction of the total and are probably not significant contributors to overall flux. In our correlations discussed below, we related measured flux to the target surface only. However, this is not to say that

Table 1. Compilation of compounds used to derive flux relationships for residues on soil and plants and in water.

Compound	Form Applied	Surface Treated	App'n Rate, kg/ha	Test Conditions (ws[m/s]/temp[°C])	Basis for Flux Measurement	Season/Location (field app'ns)
Beacon oil (Woodrow et al., 1983)	neat	soil	656	0.5-1.2/10-40	medium residues[a]	July/Davis, CA
	neat	glass	810	<1/25	medium residues	--
Chevron oil (Woodrow et al., 1983)	EC	soil	20	1-3/26-40	medium residues	Aug/Helm, CA
	neat	glass	1,040	<1/25	medium residues	--
Eptam (EPTC) (*14, 37*)	ns[b]	soil	2.0	0.08/27	medium residues	
	EC	water	3.04	1-4/15-30	micrometeorologic[c]	May/Brawley, CA
PCNB (*15*)	technical (28.9%)	soil	2.5	0.08/5-35	medium residues	--
Trifluralin (*38, 87*)	EC	soil	2.84	ns/19-30	micrometeorologic	Aug/Beltsville, MD
	ns	weedy turf	2.5	ns/ns	medium residues	--
Fonofos (*57*)	EC	soil	5.3	ns/11-32	micrometeorologic	Apr/Beltsville, MD
Lindane (*41*)	EC	soil	1.5	0.8-1.3/ns	micrometeorologic	Sept/Davis, CA
Dieldrin (*14, 87*)	pure (100%)	soil	2.5	0.08/5-35	medium residues	--
	ns	weedy turf	2.5	ns/ns	medium residues	--
Chlorpyrifos (*41*)	EC	soil	1.5	0.8-1.3/ns	micrometeorologic	Sept/Davis, CA
Diazinon (*21, 41, 63*)	EC	soil	1.5	0.8-1.3/ns	micrometeorologic	Sept/Davis, CA
	WP	dormant peach	4.5	0.8-1.1/ns	medium residues	Jan/Parlier, CA
	technical (87.2%)	water	ns	0-2.5/21-24	medium residues	--
Atrazine (*15*)	technical (96.5%)	soil	2.5	0.08/5-35	medium residues	--
Dacthal (*42, 47*)	WP	soil	7.1	<1-6/8-29	micrometeorologic	Apr/Davis, CA
	WP	soil	7.0	<1/22	micrometeorologic	Apr/Davis, CA
p,p'-DDT (*60*)	EC	soil	1.3	1.6-1.7/30-34	micrometeorologic	Aug/Clarksdale, MS
	EC	cotton	1.3	1.6-1.7/30-34	micrometeorologic	Aug/Clarksdale, MS
Prometon (*93*)	pure std	soil	ns	0.3/25	medium residues	--
Dodecane (*19*)	neat	plastic	440	1/15-35	medium residues	--
n-Octanol (*97*)	neat	glass	ns	4/20	medium residues	--
Tridiphane (*11*)	EC	giant foxtail	0.56	0.2/12-30	medium residues	--
Pendimethalin (*7*)	EC	turfgrass	3.4	ns/22	micrometeorologic	May/So Deerfield, MA
2,4-D (iso-octyl)*64*	EC	wheat	0.5	2.6/23-31	micrometeorologic	June/Regina, SK
Toxaphene (*60*)	EC	cotton	3.73	1.6-1.7/30-34	micrometeorologic	Aug/Clarksdale, MS
Deltamethrin (*67*)	EC (^{14}C)	water	0.01	<1-3/ns	medium residues	July/Winnipeg, Man.
Et-Parathion (*21*)	analytical (98%)	water	ns	0-2.5/21-24	medium residues	--
Me-Parathion (*21*)	analytical (99%)	water	ns	0-2.5/21-24	medium residues	--
Mevinphos (*21*)	technical (60%)	water	ns	0-2.5/21-24	medium residues	--
Molinate (*24, 69, 72*)	WP	water	3.14	1.4/28	micrometeorologic	May/Willows, CA
	WP	water	3.36	ns/18-35	medium residues	May/Sutter Co, CA
	pure std	water	ns	0-2.5/21-24	medium residues	--
Thiobencarb (*24, 69*)	WP	water	4.48	1.4/28	micrometeorologic	May/Willows, CA
	pure std	water	ns	0-2.5/21-24	medium residues	--

[a] Residue analysis of treated matrix. [b] ns = not specified. [c] e.g., aerodynamic and theoretical profile shape (trajectory simulation).

for special cases it might be necessary to take errant residues into account to fine-tune a model.

Residues on soil surfaces. The literature review indicated that about 53% of the field/laboratory studies of pesticide volatilization dealt with residues on soil, and many of these residues resulted from surface applications. From this literature base, we compiled a list of pesticides (Table 2) for which evaporative flux had been accurately measured under field conditions and for which physicochemical property information (i.e., soil adsorption coefficient [K_{OC}], water solubility [S_W], and vapor pressure [VP]) was available (Suntio et al., 1988; Taylor and Spencer, 1990; Wauchope et al., 1992; Montgomery, 1993). Furthermore, the flux values listed in Table 2 were determined immediately after soil treatment when volatility was greatest, therefore representing a "worst-case" scenario for evaporation.

We have used the Knudsen effusion equation ($Q = 1.98 \times 10^{-5} P [M/2\pi \mathcal{R}T]^{1/2}$; Q = flux, P = vapor pressure, M = molecular weight, $\mathcal{R}$ = gas constant, and T = absolute temperature) and have obtained fairly good correlations of flux with compound vapor pressure for a number of surface-applied pesticides. Reasonable, order-of-magnitude, results with this relationship would be expected for those situations where pesticides are volatilizing from a moist soil surface during the first few hours after application when flux is usually the greatest because of minimal interaction of the pesticides with the moist soil. After the soil surface dries and surface residues are depleted, flux characteristically declines as pesticides volatilize from residues within the soil and from residues adsorbed to the soil surface. In a Knudsen-type approach, relating flux to vapor pressure for the compounds in Table 2 resulted in fairly good correlations (Figure 1), which were due primarily to the fact that many of the listed flux values were determined immediately after application when volatilization rate was probably the greatest.

However, an improved correlation was achieved when soil adsorption (K_{OC}) and water solubility (S_W) for these compounds were combined with vapor pressure to allow for

Table 2. Properties of reference compounds to correlate flux with soil adsorption (K_{OC}), water solubility (S_W), and vapor pressure (VP) at 25-30°C.

Compound	K_{OC} (ml/g)	S_W (mg/L)	VP (torr)	Ln R[a]	Ln Flux
Beacon oil	1,000	100	1.5	-11.107	17.786[b]
Chevron oil	1,000	100	0.3	-12.717	16.274[b]
Eptam	240	375	$3.4\text{x}10^{-2}$	-14.789	12.578[c]
PCNB	5,000	0.44	$1.1\text{x}10^{-4}$	-16.811	8.292[d]
Trifluralin	8,000	0.3	$1.1\text{x}10^{-4}$	-16.898	8.700[e]
Fonofos	870	13	$3.4\text{x}10^{-4}$	-17.320	8.987[f]
Lindane	1,100	7	$6.45\text{x}10^{-5}$	-18.598	6.916[g]
Dieldrin	9,817	0.2	$1\text{x}10^{-5}$	-19.095	5.660[d]
Chlorpyrifos	6,070	0.982	$1.87\text{x}10^{-5}$	-19.580	4.525[g]
Diazinon	1,000	48.6	$1.2\text{x}10^{-4}$	-19.819	3.292[g]
Atrazine	100	33	$6.75\text{x}10^{-6}$	-20.008	4.426[d]
Dacthal	3,200	0.5	$2.5\text{x}10^{-6}$	-20.277	2.996/3.144[h]
p,p'-DDT	139,959	0.00335	$7.2\text{x}10^{-7}$	-20.294	3.825[c]
Prometon	150	720	$8.26\text{x}10^{-5}$	-20.991	2.526[i]

[a] $R = VP/[K_{OC} \text{ x } S_W]$.

[b] Woodrow et al., 1983.

[c] Nash, 1983; Willis et al., 1983.

[d] Nash and Gish, 1989.

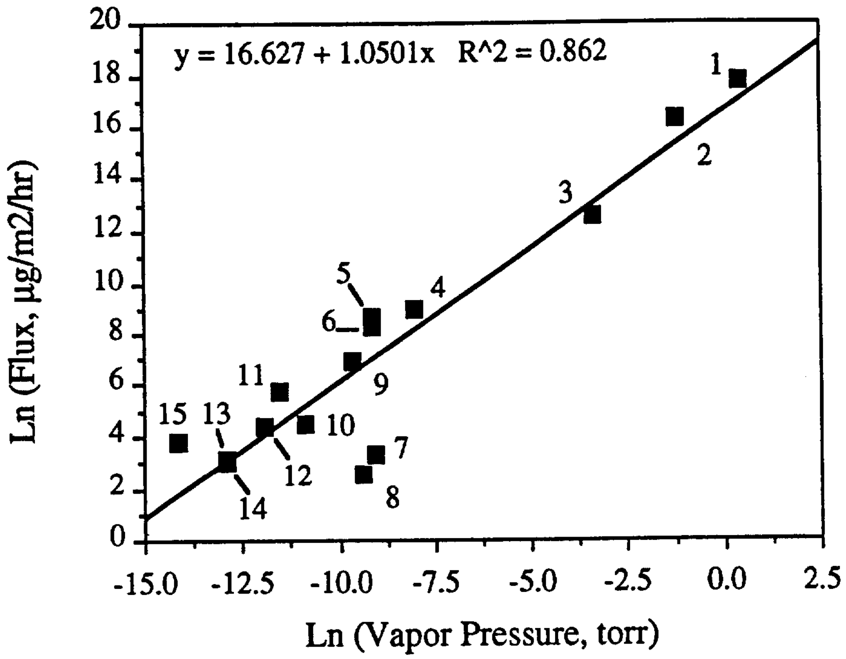
[e] Glotfelty et al., 1984.

[f] Whang et al., 1993.

[g] Majewski et al., 1990.

[h] Ross et al., 1990; Majewski et al., 1991.

[i] Spencer and Cliath, 1990.



1. Beacon oil (Woodrow et al., 1983)
2. Chevron oil (Woodrow et al., 1983)
3. Eptam (Nash, 1983)
4. Fonofos (Whang et al., 1993)
5. Trifluralin (Glotfelty et al., 1984)
6. PCNB (Nash and Gish, 1989)
7. Diazinon (Majewski et al., 1990)
8. Prometon (Spencer and Cliath, 1990)
9. Lindane (Majewski et al., 1990)
10. Chlorpyrifos (Majewski et al., 1990)
11. Dieldrin (Nash, 1983)
12. Atrazine (Nash and Gish, 1989)
13. Dacthal (Majewski et al., 1991)
14. Dacthal (Ross et al., 1990)
15. p,p'-DDT (Willis et al., 1983)

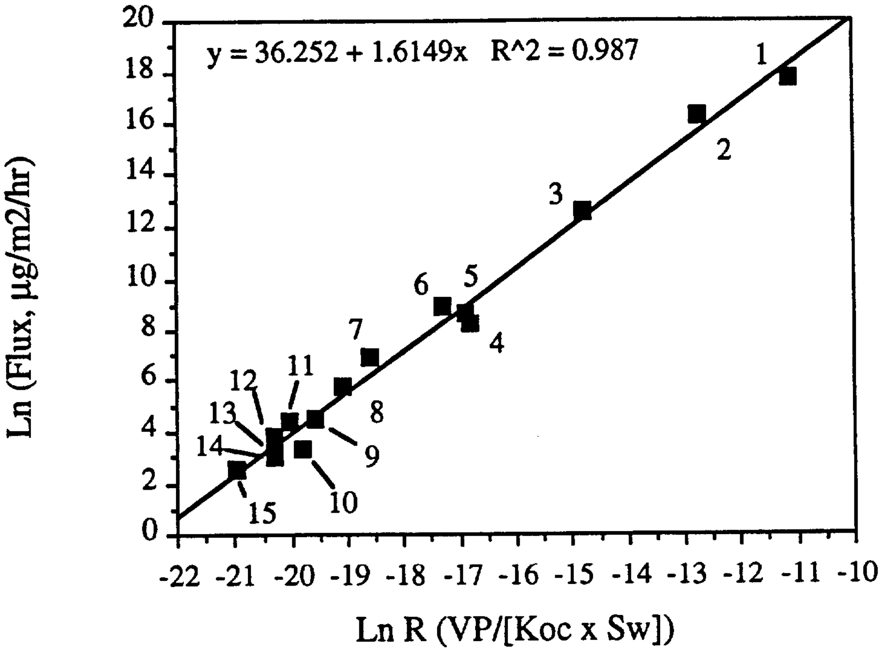
Figure 1. Correlation of vapor pressure with pesticide evaporative flux from soil.

the distribution of the pesticides among the various compartments of the soil environment. To achieve this, we related flux to the ratio R = (VP/[K_{oc} x S_w]) (Thomas, 1990) in a Ln-Ln plot for each compound, which resulted in less scatter about the regression line (Figure 2) compared to Figure 1. For several of the compounds, not all of the sources we consulted gave the same values for a given physicochemical property; in some cases, we adopted the majority values and in other cases we selected values that seemed most reasonable to us based on our knowledge and experience. Furthermore, as much as was possible, the selection of data was made maintaining consistency with regard to temperature (e.g., S_w and VP determined at the same temperature).

Soil-incorporated residues. Other investigators have demonstrated, under controlled laboratory conditions, a clear correlation between soil concentration and evaporative flux for soil-incorporated pesticides (Farmer et al., 1972; Spencer et al., 1973; Jury et al., 1980). We assumed that evaporative flux would be directly proportional to the application rate (AR), which would be related to soil concentration, and we found that for a linear data correlation Ln (flux) was proportional to Ln AR. For most of the compounds listed in Table 2, we were able to find AR data. We used these data to correlate evaporative flux with the ratio R = [(VP x AR)/(Koc x Sw)], where AR has the units kg/ha, resulting in the following correlation:

$$\mathrm{Ln}\ (\mathrm{Flux}) = 24.5 + 1.0533\ (\mathrm{Ln}\ R),\ r^2 = 0.93$$

For a given application rate, the depth of incorporation will determine the concentration of pesticide residue in the soil column and, therefore, the flux rate of the residues. Combining application rate with depth of incorporation should give a term related to the residue concentration in soil. To see how this might work, we can add a depth term (d, cm) to the above ratio R, giving [(VP x AR)/(Koc x Sw x d)], and use this in the regression equation above for application rate to calculate volatilization rate. As an illustration, Jury et al. (1980) treated two types of soil with triallate (9.4 and 13.4 kg/ha) and thoroughly mixed them to give a concentration of 10 ppm. Volatilization of triallate was measured from soil



1. Beacon oil (Woodrow et al., 1983)
2. Chevron oil (Woodrow et al., 1983)
3. Eptam (Nash, 1983)
4. PCNB (Nash and Gish, 1989)
5. Trifluralin (Glotfelty et al., 1984)
6. Fonofos (Whang et al., 1993)
7. Lindane (Majewski et al., 1990)
8. Dieldrin (Nash, 1983)
9. Chlorpyrifos (Majewski et al., 1990)
10. Diazinon (Majewski et al., 1990)
11. Atrazine (Nash and Gish, 1989)
12. p,p'-DDT (Willis et al., 1983)
13. Dacthal (Majewski et al., 1991)
14. Dacthal (Ross et al., 1990)
15. Prometon (Spencer and Cliath, 1990)

Figure 2. Correlation of pesticide flux from soil with chemical properties.

columns 10 cm deep under slight air movement, giving flux values in the range 517-708 μg/m^2/hr. Using the application rate, depth, and property data for triallate in the regression equation above, calculated flux rates for the two soils were in the range 383-557.

An important test of our estimation method for soil-incorporated pesticides would be to compare calculated flux with flux measured under typical field conditions and application regimes. However, few examples from the literature of field measurements for soil-incorporated residues have been located, and even fewer examples have the requisite data for making flux calculations. One of the few examples that had the requisite data is a field study (White et al., 1977) that looked at the dissipation of trifluralin incorporated into soil to a depth of 7.5 cm. The investigators found that by taking soil cores at different depths, 90% of the incorporated herbicide (AR = 1.12 kg/ha) resided in the top 2.5 cm. Based on this information and using the regression equation from the previous paragraph, we calculated a flux rate of about 312 μg/m^2/hr, which compared well with the measured value of about 370. While these results, together with those from the previous paragraph, are encouraging, it is obvious that more field-derived data are needed to refine and validate the estimation method for soil-incorporated pesticide residues.

The soil incorporation approach assumes that the soil column from the surface to the depth of incorporation contains a uniform distribution of the pesticide and that volatilization is occurring from the surface of this column. This is in contrast to the situation where the pesticide is injected at depth, as for a soil fumigant such as methyl bromide, leading to concentration gradients declining from the point of injection to the soil surface. In this case where flux from the soil surface is controlled by fumigant diffusion along the gradient to the surface and diffusion away from the surface, the soil incorporation correlation would greatly over estimate evaporative flux. For example for methyl bromide, maximum measured evaporative flux under field conditions was about 400 μg/m^2/sec for a non-tarped field (Majewski et al., 1995), while calculated flux using the soil incorporation approach would be 2-3 orders of magnitude greater. It is obvious that another type of model is needed for soil-injected fumigants, for example one that might include gaseous diffusion coefficients (Brown and Rolston, 1980; Rolston and Glauz, 1982).

Pesticide residues on plants. As a first approach to modeling evaporative flux from plant surfaces, we assumed that pesticide residues would be essentially evaporating from a non-interactive surface in the period just after application. This approach led to a very good correlation between evaporative flux and vapor pressure for a dozen compounds (Table 3 and Figure 3). For four of these compounds, evaporative flux was measured for residues on "inert" surfaces, such as glass and plastic. Since these four correlated well with the other eight compounds on plant surfaces, our assumption concerning the non-interacting nature of plant surfaces, at least for times just after application, is supported. Furthermore, we used the Knudsen effusion equation ($Q = 1.98 \times 10^{-5} P [M/2\pi \mathcal{R}T]^{1/2}$) to calculate flux values for the twelve compounds, assuming non-interactive surfaces. All of the calculated results (Table 3) compared with the observed flux values to within an order-of-magnitude, and for seven of the twelve compounds the flux values compared to within a factor of two. These results lend further support to the assumed non-interactive nature of plant surfaces for freshly applied pesticides where vapor pressure would be the predominate compound property affecting volatilization.

Pesticide residues in water. In correlating evaporative flux of pesticides from treated water, we assumed that the compound vapor pressures would be modified primarily by water solubility. This situation would be analogous to the one where residues were incorporated into soil. In this case, the deeper the incorporation (i.e., the lower the soil concentration) the less the effective vapor pressure, and the less the volatilization rate. For pesticide residues in treated water, the less the residue concentration the less the effective vapor pressure (through mole fraction and activity), and, thus, the less the volatilization rate. Therefore, evaporative flux from water should correlate with the ratio VP/S_W, where S_W is water solubility. This approach led to a very good correlation for about a dozen cases, but only when measured evaporative flux was normalized to actual water

Table 3. Evaporative flux of pesticide residues on non-interactive (plant, glass, plastic) surfaces at 20-30°C.

			Flux, µg/m^2/hr	
Compound	Surface	VP, torr	Obs. (Fig. 3)	Knudsen
Beacon oil[a]	glass	1.5	2.82x10^7	4.44x10^7
Chevron oil[a]	glass	0.3	2.80x10^6	9.84x10^6
Dodecane[b]	plastic	0.14	0.99x10^6	4.40x10^6
n-Octanol[c]	glass	0.13	1.35x10^6	3.64x10^6
Tridiphane[d]	giant foxtail	2.2x10^{-4}	7.13x10^3	9.50x10^3
Trifluralin[e]	weedy turf	1.1x10^{-4}	1.59x10^3	4.85x10^3
Pendimethalin[f]	turfgrass	3x10^{-5}	1.04x10^3	1.21x10^3
2,4-D (iso-octyl)[g]	wheat	2x10^{-5}	670	897
Diazinon[h]	dormant peach orchard	1.12x10^{-5}	909	471
Toxaphene[i]	cotton	4x10^{-6}	199	196
Dieldrin[e]	weedy turf	4.94x10^{-6}	172	232
p,p'-DDT[i]	cotton	3.3x10^{-7}	45.8	15.0

[a] Woodrow et al., 1983.

[b] Roberts, 1981.

[c] Tinsley, 1979.

[d] McCall et al., 1986.

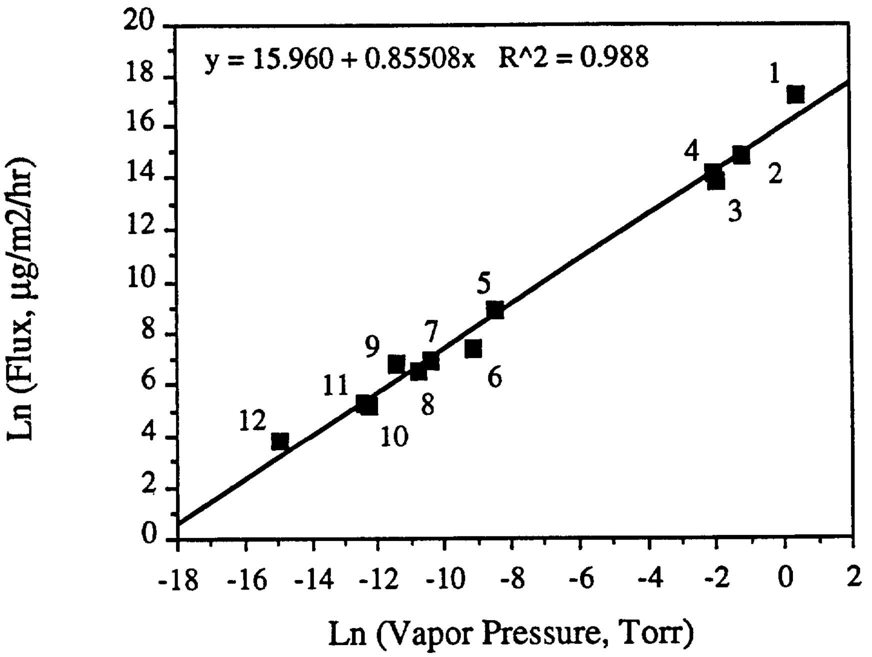
[e] Nash and Hill, 1990.

[f] Jenkins et al., 1993.

[g] Grover et al., 1984.

[h] Glotfelty et al., 1990.

[i] Willis et al., 1983.



1. Beacon oil (Woodrow et al., 1983)
2. Chevron oil (Woodrow et al., 1983)
3. Dodecane (Roberts, 1981)
4. n-Octanol (Tinsley, 1979)
5. Tridiphane (McCall et al., 1986)
6. Trifluralin (Nash and Hill, 1990)
7. Pendimethalin (Jenkins et al., 1993)
8. 2,4-D (iso-octyl) (Grover et al., 1984)
9. Diazinon (Glotfelty et al., 1990)
10. Dieldrin (Nash and Hill, 1990)
11. Toxaphene (Willis et al., 1983)
12. p,p'-DDT (Willis et al., 1983)

Figure 3. Correlation of pesticide flux from "inert" surfaces (plants, glass, plastic) with vapor pressure.

concentration (Table 4 and Figure 4). This correlation was especially encouraging in view of the fact that the data base contained a mix of laboratory and field volatilization measurements.

While a good correlation of the data required normalizing evaporative flux to water concentration, this normalized flux may be misleading. For example, deltamethrin has the lowest vapor pressure (1.5×10^{-8} torr at 25°C) of the compounds in our data base, but also the lowest water solubility (0.002 ppm at 20°C), placing it in the correlation in Figure 4 near eptam, which has about a six order-of-magnitude greater vapor pressure, but also a five order-of-magnitude greater water solubility. Furthermore, mevinphos, whose vapor pressure is about 1/10 that of eptam, but about five orders-of-magnitude greater than that of deltamethrin, has the lowest normalized volatilization rate of the group, primarily because of its extremely high water solubility ($\sim 6 \times 10^5$ ppm). In other words, since the water solubility of deltamethrin is so exceedingly low, the concentration in water will also be low in field situations (Table 4). Thus, we would not expect the absolute evaporative flux for this compound to be very high, on just a mass/area basis. This is borne out by the fact that the absolute evaporative flux values measured in the field for deltamethrin, eptam, and mevinphos were 1.3-6.4, 2.6×10^4, and 399 $\mu g/m^2/hr$, respectively (Table 4). It is our intention that the correlation in Figure 4 be used to derive absolute evaporative flux values from chemical properties and measured water concentrations. It is the absolute values that would give a true measure of the mass movement of chemical from water to air.

Downwind Concentrations Based on Estimated Flux

In the following discussions concerning the use of estimated flux in the EPA's SCREEN-2 dispersion model to predict downwind concentrations, it should be pointed out that we have not included any factors for chemical degradation in air. If atmospheric degradation were occurring at a significant rate, as can be the case for the organophosphate insecticide parathion and for the dinitroaniline herbicide trifluralin (Woodrow et al., 1978),

Table 4. Chemical property data and evaporative flux rates for pesticides applied to water.

Compounds	Vapor Pressure (VP), torr	Water Solubility (S_W), ppm	Ln (VP/S_W)	Flux, µg/m²/hr	Water Conc., ppm	Ln (Flux/ppm)
Deltamethrin[a]	1.5×10^{-8}	0.002	-11.80	1.3-3.5	0.00034-0.00095	8.23
Deltamethrin[a]	1.5×10^{-8}	0.002	-11.80	1.3-6.4	0.00034-0.00173	8.23
Diazinon[b]	4.8×10^{-5}	48.6	-13.83	4.8×10^{3}	13.5	5.87
Eptam[c]	3.4×10^{-2}	375	-9.31	2.6×10^{4}	2.17	9.39
Et-Parathion[b]	5.2×10^{-6}	15	-14.88	605	3.5	5.15
Me-Parathion[b]	6.3×10^{-6}	25	-15.19	1.12×10^{3}	14.5	4.35
Mevinphos[b]	2.2×10^{-3}	6×10^{5}	-19.42	399	174	0.83
Molinate[d]	5.6×10^{-3}	688[g]	-11.72	5.75×10^{3}	3.43	7.42
Molinate[e]	5.6×10^{-3}	688[g]	-11.72	3.93×10^{3}	1.80	7.69
Molinate[f]	3.1×10^{-3}	800[g]	-12.46	1.58×10^{4}	25.1	6.44
Thiobencarb[d]	1.5×10^{-5}	30	-14.51	160	0.57	5.64
Thiobencarb[f]	1.5×10^{-5}	30	-14.51	3.88×10^{3}	16.3	5.47

[a] Muir et al., 1985.

[b] Sanders and Seiber, 1983.

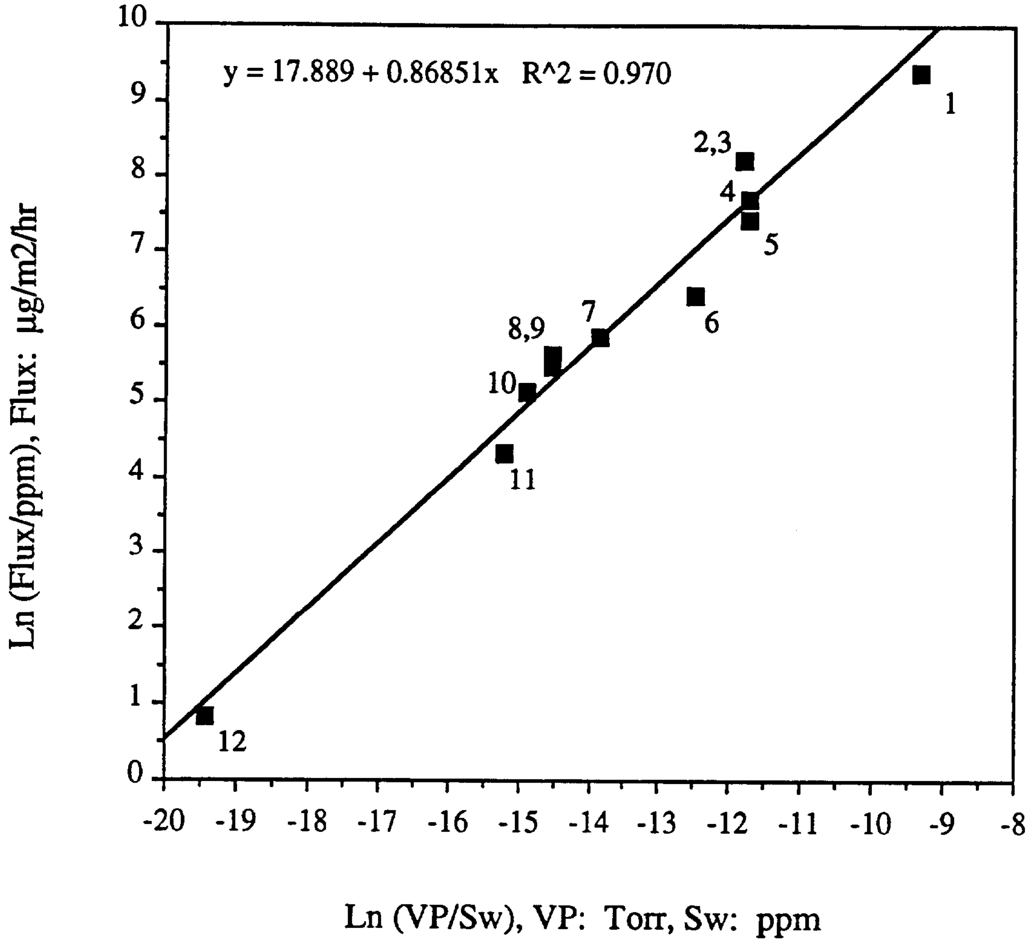
[c] Cliath et al., 1980.

[d] Ross and Sava, 1986. Vapor pressure under field conditions.

[e] Soderquist et al., 1977. Vapor pressure under field conditions.

[f] Seiber et al., 1986. Vapor pressure under laboratory conditions.

[g] Seiber and McChesney, 1987.



1. Eptam (field) (Cliath et al., 1980)
2,3. Deltamethrin (field) (Muir et al., 1985)
4. Molinate (field) (Soderquist et al., 1977)
5. Molinate (field) (Ross and Sava, 1986)
6. Molinate (lab) (Seiber et al., 1986)
7. Diazinon (lab) (Sanders and Seiber, 1983)
8. Thiobencarb (field) (Ross and Sava, 1986)
9. Thiobencarb (lab) (Seiber et al., 1986)
10. Ethyl Parathion (lab) (Sanders and Seiber, 1983)
11. Methyl Parathion (lab) (Sanders and Seiber, 1983)
12. Mevinphos (lab) (Sanders and Seiber, 1983)

Figure 4. Correlation of evaporative flux from water with chemical properties.

downwind concentrations of the parent compounds could be markedly reduced over those predicted by the SCREEN-2 model.

Treated foliage. The correlation for residues volatilizing from treated plant surfaces was used to derive estimated flux values which were used in the EPA's SCREEN-2 dispersion model to calculate pesticide residue concentrations in air downwind of treated fields for comparison with measured downwind concentrations. The pesticides selected for comparison are listed in Table 5, along with pertinent field data required by the model, such as the atmospheric stability factors that existed during the sampling periods. Table 6 summarizes the model results and the measured values. In all cases, the modeled and measured results compared well. For carbofuran, concentration was measured at a distance of 20 meters from the field, while the model could not estimate the concentration closer than 26 meters. Even so, the results compared to within a factor of two. For those cases where measured concentrations were less than the limit of quantitation (LOQ), the model predicted values were less than or equal to the LOQ. For oxydemeton-methyl in particular, the modeled downwind concentration was derived from the most stable conditions, which would favor high downwind concentrations for a "worst-case" situation.

The estimated flux values for carbofuran and oxydemeton-methyl that were derived from the correlation for residues on plant surfaces were used in the dispersion model without modification. The dispersion model assumes that the source is a square-planar surface. This assumption is a good approximation for fields that are close to being planar (e.g., alfalfa, row crops, etc.). However, for residues on mature, leafed-out trees in an orchard, the dispersion model will underestimate downwind concentrations because residues would be volatilizing from a surface area considerably greater than that for the square-planar surface containing the orchard. Therefore, for methidathion and azinphosmethyl, the estimated flux values had to be adjusted by the total leaf surface area (m^2) per tree and the orchard tree density (tree/m^2). This adjustment was based on the assumption that all the leaves were uniformly coated by the pesticides, but in actuality this

Table 5. Field data for use in the SCREEN-2 model to compare measured and modeled downwind concentrations.

Pesticide	Commodity	Temp., °C	Wind speed (m/sec)[a]	Field, ha[b]	Downwind dist. (m)[c]	Stability[d]
Carbofuran	alfalfa	15-25	1-3	28.3	15-20	B
Oxydemeton-me	broc/caul	15-25	--	6.48	15-20	F
Methidathion	oranges	20-30	1-3	6.07	15-20	A
Azinphosmethyl	walnuts	15-30	1-3	47.8	15-25	B

[a] Wind speeds used in the model were the following: Carbofuran -- 2.5; oxydemeton-methyl -- 1; methidathion -- 2; azinphosmethyl -- 2.

[b] Field dimensions for square planar fields used in the model were the following: Carbofuran -- 376 m; oxydemeton-methyl -- 255 m; methidathion -- 174 m; azinphosmethyl -- 402 m.

[c] Carbofuran: Sampler and model distances were 20 and 26 m, respectively; oxydemeton-methyl: Sampler and model distances were both 15 m; methidathion: Sampler and model distances were both 15 m; azinphosmethyl: Sampler and model distances were 15-25 and 27 m, respectively.

[d] For sampling periods of three hours (carbofuran) and four hours (oxydemeton-methyl, methidathion, and azinphosmethyl).

Table 6. Comparison of measured and modeled downwind pesticide concentrations.

Pesticide	Estimated Flux ($g/m^2/sec$)	Concentration in air, $\mu g/m^3$	
		Measured[a]	Model
Carbofuran	1.39×10^{-7}	0.60	0.39
Oxydemeton-methyl	3.08×10^{-7}	<LOQ[b]	11
Methidathion	6.94×10^{-7}	1.4	1.6
Azinphosmethyl	6.72×10^{-8}	<LOQ[c]	0.23

[a] Provided by the Air Resources Board.

[b] LOQ (limit of quantitation) = 70 $\mu g/m^3$.

[c] LOQ = 0.23 $\mu g/m^3$.

will depend on the method of application. Furthermore, the total leaf surface area will vary as a function of the type and size of the orchard and the stage of leaf development. The combined effect of application method and orchard maturity will introduce some uncertainty in flux estimation.

The estimated flux for methidathion derived from the correlation for residues on plant surfaces was 4.96×10^{-8} g/m^2/sec. Using this value in the SCREEN-2 model gave a concentration of about 0.12 μg/m^3 15 meters downwind of the field, compared to a measured concentration of 1.4 μg/m^3. The modeled value of 1.6 μg/m^3 reported in Table 6 was derived in the following way using the total leaf surface area per tree of 500 m^2 for mature orange trees (Spencer et al., 1973) and the typical orchard tree density of 0.028 tree/m^2 (~36 m^2 are required for each orange tree [Bienz, 1993]):

$$(4.96 \times 10^{-8}\ \text{g/m}^2\text{/sec}) \times (500\ \text{m}^2\text{/tree}) \times (0.028\ \text{tree/m}^2) = 6.94 \times 10^{-7}\ \text{g/m}^2\text{/sec}$$

This adjusted flux value was then used in the SCREEN-2 model. The same adjustment was also made for azinphosmethyl on walnuts, but using the leaf surface area and orchard tree density for oranges. By doing this gave only an approximation for walnuts, since walnut trees are typically larger than orange trees, so that orchard tree density is probably less, and, furthermore, total leaf surface area for walnuts may not be as great.

While adjustment for leaf surface area gave reasonable results for the particular orange and walnut tree examples, it should be noted that the results for other examples will be affected by total leaf surface area, which will vary as a function of the type and size of the orchard and of the stage of leaf development. Furthermore, an assumption in the approach described above is that leaf surface area is uniformly coated with pesticide. In actuality, this would not be the case, and pesticide distribution would depend on the application method. However averaged over the large surface area of a typical orchard, pesticide distribution would probably not affect evaporative flux as much as the meteorological conditions and pesticide physicochemical properties.

Treated water. The correlation for residues volatilizing from treated water (Figure 4) was used to derive estimated flux values for the rice herbicide molinate under a variety of conditions. These flux values were used in the SCREEN-2 dispersion model to calculate molinate residue concentrations in air downwind of a treated rice field for comparison with measured downwind concentrations. Table 7 summarizes the sampling periods post-application, the pertinent field data required by the model, such as the wind speed and atmospheric stability factors that existed during the sampling periods, and the measured and modeled downwind concentrations.

Since the concentration of molinate in the field water was not measured and the variation in water depth was not recorded, we used the application rate for the subject field (~5.6 kg/ha molinate) and water depth data from other studies (e.g., 12-24 cm; ave = 15 cm [Ross and Sava, 1986; Seiber and McChesney, 1987]) to calculate a range of water concentrations for molinate (2.34-4.68 ppm). Furthermore, for daytime and nighttime sampling we used molinate vapor pressures and water solubilities at 30°C (9.92×10^{-3} torr and 576 ppm) and 20°C (3.1×10^{-3} torr and 800 ppm), respectively, keeping in mind the fact that molinate solubility in water decreases with increasing temperature (Seiber and McChesney, 1987). The ratios of the vapor pressures and water solubilities were used in the correlation for residues volatilizing from treated water to derive flux values normalized to water concentration. These normalized flux values were then multiplied by the range of molinate concentrations in water to derive absolute flux values used in the SCREEN-2 dispersion model to calculate downwind concentrations in air. For sampling periods about 24 hours or greater post-application, we also factored in the decline in water concentration with time, which for molinate has a half-life of about 4 days (Seiber et al., 1986; Seiber and McChesney, 1987). Since water concentration has a direct effect on flux, the flux values derived from the correlation in Figure 4 were adjusted by simply multiplying by the fraction of molinate remaining in the water for the particular sampling period post-application. These adjusted values were then used in the SCREEN-2 model to calculate

Table 7. Modeled vs measured air concentrations downwind of a molinate-treated rice field.[a]

Sample ID	Sampling Period	Downwind Distance, m	Atmospheric Stability	Molinate in water (fraction)[b]	Wind speed, m/s	Air concentration, µg/m^3	
						Measured	Modeled[c]
3N1	3(D)	9.14	B	0.99	3.6	11.3	6.2-12.5 (ave = 11.2)
4N1	4(D)	9.14	C	0.98	5.4	22.6	10.3-20.5 (ave = 18.5)
5N1	5(N)	9.14	E	0.93	3.1	10.1	7.4-14.9 (ave = 13.4)
6N1	6(D)	9.14	B	0.85	3.6	8.6	5.3-10.6 (ave = 9.5)
8S1	8(D)	22.9	B	0.72	4.0	6.8	4.4-8.9 (ave = 8.0)
10S1	10(D)	22.9	B	0.61	4.5	6.0	3.8-7.6 (ave = 6.8)

[a] Field sample data provided by the California Air Resources Board.

[b] Seiber et al., 1986; Seiber and McChesney, 1987.

[c] Adjusted for wind speed and fraction molinate remaining in the field water.

downwind concentrations. Since the sampling distances from the treated field were too close for the SCREEN-2 model to accommodate, the model was used to calculate concentrations at several distances downwind and the concentration vs distance plots were extrapolated to the sampling distances to derive the modeled concentrations for those distances. Finally, the effect of wind speed (>3 m/s) on molinate flux from water was assumed to be linear and proportional (Seiber and McChesney, 1987). In other words, since the correlation in Figure 4 implicitly contained wind speeds up to about 3 m/s, modeled air concentrations were adjusted for wind speeds, *u*, above this value by multiplying by the factor *u*/3. For example, the non-adjusted modeled concentration range for sampling period 4(D) was 5.7-11.4 $\mu g/m^3$; multiplying this range by the wind speed factor 5.4/3 (= 1.8) gave the range listed in Table 7.

In most cases, the modeled concentration ranges bracketed measured values (Table 7). However, for the first sampling period, and perhaps the second period as well, high concentrations of molinate in air may have reflected non-equilibrium conditions with much airborne residue originating from chemical intercepted by pad dikes and perhaps other surfaces and general contamination of the air from drift during application. In other words, since some time is required for the granular molinate to dissolve in the rice field water, air concentrations immediately after application could not be solely attributed to evaporative flux of dissolved residues from water. Even so, modeled air concentrations compared well with measured values for the first two sampling periods. However, molinate dissolution in water can be rapid and have rates varying from concentration maxima appearing on the day of application (Seiber et al., 1986; Ross and Sava, 1986) to as much as 17 hours post-application (Seiber and McChesney, 1987).

Uncertainty in Estimated Flux

The calculated or estimated flux values and downwind air concentrations should be regarded as screening tools, suitable for a first-cut ranking of chemical-surface combinations which might lead to downwind exposures of concern from a risk assessment viewpoint. Needless to say, there will be uncertainty in any such simplified approach to estimating exposures to airborne residues. While the extent of the uncertainty is not clear, its origin would reside primarily in the measured flux data and the compound physicochemical property data used to derive the correlations. Much of the evaporative flux data used in this study for the treated soil, plant, and water correlations were obtained using micrometeorological methods. These methods include aerodynamic-gradient, theoretical profile shape, energy balance, eddy correlation, and integrated horizontal flux. While each method has its associated uncertainty, these various methods taken together appear to be statistically equivalent (Majewski et al., 1990). However, it is not certain if these methods give accurate descriptions of what is actually occurring in the field because several of these methods are based on similar assumptions about compound mass transfer and reliable validation techniques are lacking. The aerodynamic-gradient is the most commonly used method, but it may underestimate evaporative losses (Glotfelty et al., 1984; Grover et al., 1988) and do so by as much as 50% in some cases (Seiber and McChesney, 1987). Furthermore, flux values determined using the aerodynamic-gradient method can have associated uncertainties of as much as 50% (Majewski et al., 1995). While our correlations based on a combination of micrometeorological data and data derived from direct analysis of the treated matrices were quite good, this may have been due in part to the fact that the correlations were logarithmic rather than linear, as discussed above.

Taken together, the uncertainties inherent in the flux measurement methods and the physicochemical property data were reflected in the absolute percent variation of the individual data points relative to the correlation regression lines. We found that for soil, plant, and water matrices the ranges in variation were 1.7-22.5%, 0.6-19.5%, and 0.7-16.2%, respectively, for the data sets. Average variation was 8.2±5.6%, 5.2±5.6%, and 5.4±4.3% for soil, plant, and water, respectively. Combining all the data sets gave a range of 0.6-22.5%, with an average of 6.4±5.3%. Using the statistical test for comparison of means, the average variations were not significantly different at the 95% confidence level indicating that the results were independent of the treated matrix.

Outliers

While much of the literature data for evaporative flux from soil, plant, and water surfaces worked well to give reasonable correlations for flux estimation purposes, there were a few examples from the literature that did not fit these correlations well and were, therefore, not included. Most of these outliers involved applications of pesticides to soil, followed by two instances of residues on plant surfaces. All of the data points that we found in the literature for residues in water were used to derive the correlation; no outliers have been found for residues in water, although we have not made an exhaustive search of the literature.

Chlorpropham on wet soil (Turner et al., 1978) exhibited an evaporative flux rate that was almost 4,000 times greater than what would be predicted by the correlation for soil. Similarly for dieldrin on soil that was maintained moist, evaporative flux was about 10 times that predicted by the soil correlation, while flux for dieldrin from relatively dry soil was only about 1.4 times that predicted by the correlation (Willis et al., 1972). If it is assumed that volatilization for the two wet soil situations occurred primarily from water rather than from soil, predicted and observed flux agreed to within a factor of two. For soil-incorporated triallate and trifluralin (Grover et al., 1988), observed flux was about 3 times greater in both cases than that predicted for residues incorporated to a depth of 5 cm. However if incorporation was not uniform throughout the depth, as discussed earlier (see Soil-incorporated residues above), but residues resided in the upper 40-50% of the soil column, then predicted and observed flux for both compounds would almost agree exactly (factor of 1-1.3). For chlorpyrifos residues on soil (Whang et al., 1993) and dieldrin and heptachlor residues on pasture grass (Taylor et al., 1977), the 1-2 order of magnitude greater observed flux remains unexplained. However for atrazine and fonofos, which were applied along with chlorpyrifos, observed flux agreed quite well with the correlation for residues on soil.

Effect of Wind Speed and Temperature on Evaporative Flux

Soil. Limited information is available in the literature regarding the effect of wind speed and temperature on residue evaporative flux. One study (Farmer et al., 1972) measured the volatility of soil-incorporated dieldrin as affected by temperature and air flow. These investigators found that, overall, volatilization rate increased by a factor of 1.8 for a wind speed increase factor of 3.6, while volatilization rate increased by a factor of about 3 for a temperature increase of 10°C (20°C to 30°C; increase factor of 1.5). In another study under more field-like conditions (Nash and Gish, 1989), the investigators found that, for a series of pesticides applied to soil surface, volatilization rate increased by a factor of 1.8 for each 10°C increase in temperature. For dieldrin in particular, volatilization rate increased by a factor of about 2 for each 10°C increase in temperature (initial temperature = 5°C; increase factor of 3). Taken together, these results suggest that temperature has a greater effect on volatility than does wind speed for residues on soil, and that volatility increase may be greater for temperature increases at higher initial temperatures. It is the temperature of the treated surface that would affect volatility more than the air temperature. Published studies often only report the air temperature, assuming that the treated surface is in thermal equilibrium with the surrounding air. However, we have found that under intense sunlight conditions in the field soil surface temperatures can be as much as 25°C greater than those of the air (Woodrow et al., 1983).

Plants (i.e., non-interactive) surfaces. Volatilization rates for dieldrin on glass plates and quartz sand (1,000 ppm) were measured in different studies (Phillips, 1971; Farmer and Letey, 1974) and combined into one relationship correlating with wind speed at 30°C (Spencer and Cliath, 1977). In general for dieldrin residues on glass, a 3-fold increase in volatilization rate was caused by an 8-fold increase in wind speed, while for residues on quartz sand a 1.5-fold increase in wind speed resulted in less than a 1.2-fold increase in volatilization rate. For dodecane on polypropylene pans (Roberts, 1981), a five-fold increase in volatilization rate was caused by imposing a wind speed of about 1.12 m/s (21-23°C), compared to no measurable air movement. This increased volatilization rate was doubled again with an approximate doubling of the wind speed.

In one of our earlier studies (Woodrow et al., 1983), we used the dodecane data to develop a wind speed term ([1 + 3u], u is wind speed [m/s]) to modify the evaporation rate constants of pure hydrocarbons and their mixtures so that change in volatilization rate was directly related to change in wind speed. The chemical properties terms that we have developed in the current study for generating correlations with volatilization rates are directly related to the evaporation rate constants. In fact, other investigators have attempted to use chemical property data to calculate volatilization rate constants for pesticides on soil, for example (Thomas, 1990). Therefore, our wind speed term, or similar expression, should be useful in estimating the effect of wind speed on the volatilization of pesticide residues applied to relatively non-interactive surfaces. For 1,000 ppm dieldrin on quartz sand at 30°C (Farmer and Letey, 1974), measured volatilization rate was in the range (2.24-2.69) x 10^3 μg/m^2/hr for wind speeds in the range 0.89-1.33 m/s. Using our correlation for residues on plant surfaces (Figure 3) and the vapor pressure for dieldrin at 30°C (~1 x 10^{-5} torr), the calculated volatilization rate without wind is about 453 μg/m^2/hr. Imposing a wind speed of about 1.11 m/s (average of the range stated above), by using the term [1 + 3u] as a multiplicative factor for the vapor pressure and the correlation for residues on plant surfaces, gave a volatilization rate of about 1.6 x 10^3 μg/m^2/hr, which compares within a factor of two with the measured volatilization rates. We suspect that in the term [1 + 3u], the multiplicative factor for wind speed (u) is related to heat of vaporization, and, therefore, will vary with the class and type of compound. For compounds that have lower volatility than dodecane, such as dieldrin, the multiplicative factor should probably be greater. For example, by using the measured volatilization rate of 2.46 x 10^3 μg/m^2/hr (average of 2.24 and 2.69) at a wind speed of 1.11 m/s (average of

0.89 and 1.33) and our correlation for residues on plant surfaces, the wind speed term for dieldrin would be more like [1 + 6u].

In addition to air movement, it would also be expected that temperature would have a noticeable effect on volatilization of residues from non-interactive surfaces. This temperature effect would act through the vapor pressure, which is related to temperature in the integrated form of the Clausius-Clapeyron equation

$$\mathrm{Ln}\ (VP_2/VP_1) = -(\Delta H/R)\ (1/T_2 - 1/T_1)$$

where ΔH is the heat of vaporization (cal/mole) and R is the gas constant (1.99 cal/deg mole). Much work has been done to determine heats of vaporization for pesticides, and compilations exist (Kim, 1985). However, if this type of information is not available, all that is needed is at least two vapor pressure values at two different temperatures to plot Ln VP vs 1/T to be used to calculate the vapor pressure at a different temperature; the slope of such a plot will contain the heat of vaporization. The vapor pressure at the new temperature can, then, be used to calculate the volatilization rate at the new temperature using the properties/flux correlation for residues on non-interactive surfaces (Figure 3).

The effect of wind speed and temperature changes on pesticide evaporative flux is assumed to occur through changes in the pesticide vapor pressure. Wind speed and temperature terms are essentially operators acting directly on vapor pressure. Since evaporative flux is directly correlated with pesticide vapor pressure for residues on non-interactive surfaces, such as plant surfaces for a while after application, changes in vapor pressure will result directly in changes to evaporative flux. To account for both wind speed and temperature together, the vapor pressure of a pesticide on a non-interactive surface would be operated on by the term

$$e^{-(\Delta H/R)\ (1/T2-1/T1)} \times [1 + nu]$$

where T_1 corresponds to the initial vapor pressure and $n \approx 3\text{-}6$, or greater, depending on the compound's heat of vaporization. Since the correlations developed in this study implicitly included a range of temperatures (20-30°C) and wind speeds (1-3 m/sec), this term may be used in cases where the pesticide vapor pressure is known only at some temperature other than the temperature(s) implicit in the correlations and under different wind conditions that would deviate considerably from those implicit in the correlations.

Other Flux Estimation Methods

Eureka Laboratories (EL). The flux estimation methods developed by EL are all related to water flux (Leung et al., 1978). For soil, plants, and water the evaporation expressions recommended by EL are as follows:

$$\text{Soil: } -(dm_i/dt) = (E/[1-RH])(P_i[M_i]^{1/2}/P_w[18]^{1/2})$$

$$\text{Plants: } -(dm_i/dt) = (ET_j/[1-RH])(P_i[M_i]^{1/2}/P_w[18]^{1/2})$$

$$\text{Water: } -(dm_i/dt) = M_{io}\Big[\exp(-K_l[t-1/L]) - \exp(-K_l[t/L])\Big]$$

where $-(dm_i/dt)$ is the monthly loss of compound i per acre, E = inches of water evaporated from soil, ET_j = inches evapotranspiration for crop j, RH = relative humidity, M_{io} = number of pounds applied, K_l = the liquid exchange constant (Liss and Slater, 1974) for the evaporating compound, t = months, and L = water depth (meters). For the compounds used in this study, we derived E, ET_j, RH, and K_l from data provided in the EL report. However for many cases, these terms could only be approximated.

Using EL's relationships for residues in water, on plants, and on soil we calculated evaporative flux rates for comparison with the measured values used to derive the correlations in this study and with values calculated from our regression equations listed in Figures 2-4. We found that while there was reasonable agreement in some cases (agreement within a factor of 2), in general these relationships overestimated flux for residues on plants and soil surfaces (2 times greater for plants and 2-286 times greater for soil) and underestimated flux for residues dissolved in water (50-200 times less) compared to the flux values calculated using our regression equations (Regression, Table 8). In all fairness, it should be pointed out that EL's methods give monthly averages for chemical flux while the flux values used in our estimation methods were measured within a few hours after application. Furthermore, some lack of agreement may have been due to the

Table 8. Comparison of measured and regression-calculated flux values with values estimated using Eureka Laboratories' models.

		Flux, $\mu g/m^2/hr$[a]		
Compound	Matrix	Measured[b]	EL[c]	Regression[d]
p,p'-DDT	Soil	46	35	32
Eptam		3 x 10^5	2 x 10^6	2 x 10^5
Prometon		12	322	10
Dieldrin		287	488	224
Chlorpyrifos		92	3 x 10^3	103
Lindane		1 x 10^3	1 x 10^4	502
Trifluralin		6 x 10^3	2 x 10^4	8 x 10^3
Fonofos		8 x 10^3	4 x 10^4	4 x 10^3
Diazinon		27	2 x 10^4	70
Dacthal		22	392	33
Atrazine		84	853	51
Trifluralin	Plants	2 x 10^3	1 x 10^4	4 x 10^3
Pendimethalin		1 x 10^3	2 x 10^3	1 x 10^3
2,4-D (isooctyl)		670	2 x 10^3	819
Dieldrin		172	480	248
Toxaphene		199	405	207
Diazinon		909	984	499
p,p'-DDT		46	31	24
Molinate	Water	(4-6) x 10^3	60	(4-7) x 10^3
Eptam		3 x 10^4	215	4 x 10^4
Deltamethrin		1-6	2 x 10^{-2}	1-4
Thiobencarb		160	1	111

[a] Values rounded to the nearest whole number.

[b] Tables 2-4.

[c] EL = Eureka Laboratories (Leung et al., 1978).

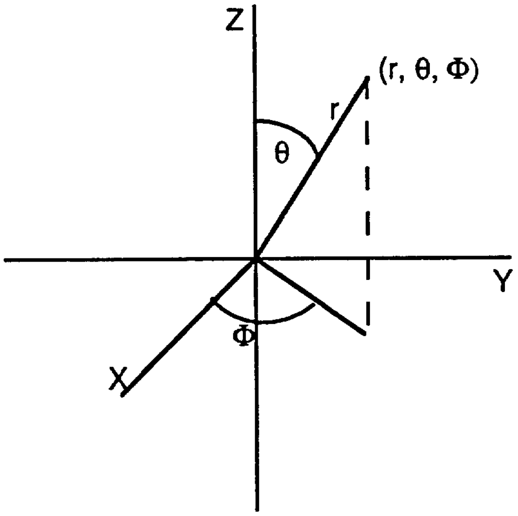
[d] From regression equations (Figures 2-4) using chemical properties in Tables 2-4.

fact that EL's water flux (E) data for residues on soil were compiled for a particular county in California and probably should not be used universally, and that for residues on plants evapotranspiration (ET_j) is a plant-driven process unrelated to the behavior of chemical residues on plant surfaces. Also, using the gas and liquid exchange constants for carbon dioxide and water to estimate these constants for chemical residues dissolved in water may not be appropriate in every case.

Knudsen Effusion (KE). Using polar coordinates to describe molecules hitting a unit surface, the following equation can be derived by integrating over the limits for ϕ and θ of 0 to 2π and 0 to $\pi/2$, respectively:

$$dN'/dt = (1/4)N\bar{c}$$

where dN'/dt is the total number of molecules striking from all directions, N is the number of molecules per cc, and $\bar{c}$ is the mean molecular velocity. If ρ is the gas density, the mass of gas that effuses in unit time (dm/dt) is



$$dm/dt = (1/4)\rho\bar{c}$$

or

$$dm/dt = \rho(\mathcal{R}T/2\pi M)^{1/2}$$

since

$$\bar{c} = (8\mathcal{R}T/\pi M)^{1/2}$$

Furthermore, since

$$P = (n/V)\mathcal{R}T$$

then

$$MP = \rho\mathcal{R}T$$

and

$$dm/dt = (MP/\mathcal{R}T)(\mathcal{R}T/2\pi M)^{1/2}$$

The final result is the well-known KE equation:

$$W/t = P(M/2\pi\mathcal{R}T)^{1/2}$$

where W is weight of loss by effusion. However, this expression is reliable only for effusion of molecules into a vacuum. For evaporation from a source into the atmosphere, the following modified expression is used:

$$Q = \beta\, P\, (M/2\pi\mathcal{R}T)^{1/2}$$

where Q is flux (g/cm^2•sec), $\beta = 1.98 \times 10^{-5}$, and P is the vapor pressure of the compound. For this expression to work, the gas constant ($\mathcal{R}$) must have the value of 8.31×10^7 g•cm^2/sec^2•°K•mole. This requires that the vapor pressure have the units of dynes/cm^2.

The modified form of the KE equation has been used to describe evaporative flux of chemicals from non-interactive surfaces (Tinsley, 1979), and this method of estimating flux has been dealt with briefly earlier in this report in the sections that discuss residues on soil and plants. For residues dissolved in water, the expression has the same form, except that $\beta = 2.48 \times 10^{-5}$ to take into account the fact that water surfaces can be distorted by wind (Tinsley, 1979). A more precise form of the KE equation for residues in water includes the Henry's law constant (H) in a term that serves as the rate constant in a first-order expression:

$$C_t = C_0 \exp[-(\beta H f/d)t]$$

where C_t is solute concentration at time t, d = depth (cm), and $f = (M/2\pi\mathcal{R}T)^{1/2}$.

Table 9 summarizes a comparison of measured and regression-derived flux values for pesticides on soil, plants, and in water with flux values estimated using the various forms of the KE equation. For residues on soil, the KE equation overestimated flux for most cases (2-300 times greater for 9 out of the 11 regression examples), while for residues in water, flux was underestimated for most cases (5-10 times less for 3 out of the 4 regression examples). However, for residues on plant surfaces the KE-calculated flux values exceeded the regression values by a factor of only 1-2 for 4 of the 8 examples, and for the remaining examples the regression values exceeded the KE values by a factor of only 1-2. The relatively good agreement for plant residues lends support to the assumed non-interactive nature of plant surfaces, at least for freshly applied pesticides where vapor pressure would be the predominate compound property affecting volatilization.

The above results for the EL and KE methods indicate that vapor pressure, a major term in these approaches, would be the predominate compound property affecting volatilization of residues from plant surfaces and would explain the good agreement between these methods and the regression-calculated flux values in Tables 8 and 9. For soil and water, on the other hand, the EL and KE approaches, since they are derived from simple concepts concerning water flux (EL) and molecular effusion (KE), do not take into account all the myriad interactions possible for chemical residues in these matrices. Estimation methods that include chemical flux data measured under a variety of actual field and laboratory conditions (e.g., the correlation approach used in this study), because they would implicitly contain the variety of chemical-matrix interactions, stand a better chance of providing useful predictive data.

Table 9. Comparison of measured and regression-calculated flux values with values estimated using the Knudsen effusion equations.

		Flux, µg/m^2/hr[a]		
Compound	Matrix	Measured[b]	Knudsen[c]	Regression[d]
Eptam	Soil	3×10^5	1×10^6	2×10^5
Trifluralin		6×10^3	5×10^3	8×10^3
Fonofos		8×10^3	1×10^4	4×10^3
Lindane		1×10^3	3×10^3	0.5×10^3
Dieldrin		287	466	224
Chlorpyrifos		92	836	103
Diazinon		27	5×10^3	70
Atrazine		84	237	51
Dacthal		22	109	33
p,p'-DDT		46	32	32
Prometon		12	3×10^3	10
Tridiphane	Plants	7×10^3	1×10^4	6×10^3
Trifluralin		2×10^3	5×10^3	4×10^3
Pendimethalin		1×10^3	1×10^3	1×10^3
2,4-D (isooctyl)		670	897	819
Diazinon		909	471	499
Toxaphene		199	196	207
Dieldrin		172	232	248
p,p'-DDT		46	15	24
Deltamethrin	Water	1-6	0.1-0.5	1-4
Eptam		3×10^4	8×10^3	4×10^4
Molinate		$(4\text{-}6) \times 10^3$	$(2\text{-}3) \times 10^3$	$(4\text{-}7) \times 10^3$
Thiobencarb		160	14	111

[a] Values rounded to the nearest whole number.

[b] Tables 2-4.

[c] For soil and plant surfaces: Flux = $1.98 \times 10^{-5} P (M/2\pi\mathcal{R}T)^{1/2}$.
For water: $C_t = C_o \exp[-(\beta H f/d)t]$, where C_t is solute concentration at time t,
H = Henry's law constant, d = depth (cm), and $f = (M/2\pi\mathcal{R}T)^{1/2}$.

[d] From regression equations (Figures 2-4) using chemical properties in Tables 2-4.

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