Correlation To Estimate Emission Rates for Soil-Applied Fumigants

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The emission rates [ER (μg m^{-2} s^{-1})] for subsurface injections and surface chemigations for 15 fumigant applications were combined with the physicochemical properties of the fumigants [vapor pressure, VP (Pa); water solubility, S_w (mg L^{-1}); soil adsorption coefficient, K_{oc} (mL g^{-1})] and with application conditions [application rate, AR (kg ha^{-1}); depth of application, d (cm)]. This resulted in the regression Ln ER = 3.598 + 0.9400 Ln R [R = (VP $\times$ AR)/($S_w \times K_{oc} \times d$)], which can be used to estimate emissions for new applications. Emission rates derived from the linear correlation were used as input to an atmospheric dispersion model to estimate concentrations of fumigants in air at various downwind distances, and the results were compared with concentration values measured in the field near sources. The fumigant correlation along with an atmospheric dispersion model can be used as a rapid screening method by regulatory and enforcement agencies for exposure and risk assessment.

KEYWORDS: Soil fumigants; emission rates; atmospheric dispersion models

INTRODUCTION

The atmosphere is essentially an infinite reservoir and serves as a transport medium for moving pesticide residues from target to nontarget areas. Because of this, there is much concern over the exposure of humans and ecosystems to errant pesticide residues. To address this concern, information is needed regarding emission rates and exposures. Over the past several decades, much effort has been expended in experimentally measuring field and laboratory evaporative flux (emission) rates and downwind concentrations for pesticides. Although not all pesticides have been evaluated in this way, there is enough emission rate information so that models having predictive value can be developed. Because of the relative cost of field and laboratory work and the related time delay before answers are obtained, regulatory agencies in recent years have shown a growing interest in and dependence on predictive models, at least for screening and for estimating environmental behavior. Models that reliably estimate emission rates, when coupled with atmospheric dispersion models for estimating downwind concentrations, can become a powerful tool in the hands of regulatory and enforcement agencies for estimating and assessing exposure.

Fumigants are characterized by high vapor pressures (e.g., ~3−217 kPa) that make them useful for the treatment of soils because of the resultant high diffusivities. Many are applied by drip-irrigation and shank injection at depth (e.g., chloropicrin, metam sodium, 1,3-D, methyl iodide, methyl bromide), followed by plastic tarping, in most cases, to trap the fumigant as much as possible in the soil column so that it can do its work. Others are applied by surface chemigation [e.g., metam sodium, which releases methyl isothiocyanate (MITC) in soil or water; sodium tetrathiocarbonate, which releases carbon disulfide], where the material is sprayed on the soil surface or added to irrigation water. Because of their high vapor pressures, fumigants cannot be completely contained, and evaporative losses will occur. Losses are greatest for surface chemigation, as would be expected.

We have taken published emission rates (μg m^{-2} s^{-1}) for subsurface injection (drip and shank applications) and surface chemigation for a number of different fumigant applications and combined them with fumigant physicochemical properties to obtain a correlation that can be used to estimate emission rates for new application scenarios. We previously reported similar correlations for emission rates of semivolatile pesticides applied to soil, water, and foliage (*1*, *2*). In this paper, we describe how the correlation was developed for fumigants and how it can be used in conjunction with atmospheric dispersion models to estimate concentrations of the fumigants in air at any distance downwind of the source. The evaluation and validation of the correlation were done by comparing estimated air concentrations with those measured by other investigators near treated fields.

MATERIALS AND METHODS

Correlation. Table 1 lists some common soil fumigants and their properties, including application conditions, such as application rate and soil depth. An additional condition for eight of the applications was the use of thin-film (~1+ mil) polyethylene tarps (entries 1, 3, 5−8, 14, and 15). For the remaining seven applications, tarps were not used (entries 2, 4, and 9−13). The effect of tarping on fumigant emission rates was not factored into the correlation as this was outside the scope of this study. Only the emission rates as measured, regardless of tarping, were used.

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Table 1. Fumigants and Their Chemical Properties and Application Conditions

entry	fumigant	chemical properties			application conditions			
		VP, Pa	S_w, mg L^{-1}	K_{oc}, mL g^{-1}	AR, kg ha^{-1}	d, cm	ER, μg m^{-2} s^{-1a}	R^b
1	chloropicrin (*3, 11*) shank	3173	2270	7	372	26.6	98.4	2.79
2	1,3-D (*3, 8*) shank	3866	2250	32	139	38.1	9.03	0.196
3	1,3-D (*3, 9*) drip	3866	2250	32	97.1	15.0	12.8	0.348
4	CS$_2$(*3, 13*) surf chem	47900	2300	298	29.2	NA	64.3	2.04
5	MeI (*6, 7*) lab soil col	53061	14200	11.5	110	30.0	50.0	1.19
6	MeI (*6, 7, 10*) (CA-I) shank	53061	14200	11.5	283	25.4	96.3	3.62
7	7. MeI (*6, 7, 10*) (CA-II) shank	53061	14200	11.5	274	15.2	210	5.86
8	MeI (*6, 7, 10*) (FL) shank	53061	14200	11.5	289	30.5	111	3.08
9	MITC (*3, 4, 12*) drip	2666	7600	32	81	10.0	4.06	0.089
10	MITC (*3, 4, 14*) 2007 shank	2666	7600	32	108	22.9	2.08	0.052
11	MITC (*3, 4, 14*) 2007 surf chem	2666	7600	32	108	NA	43.2	1.18
12	MITC (*3, 4, 15*) 2008 shank	2666	7600	32	139	22.9	2.58	0.066
13	MITC (*3, 4, 15*) 2008 surf chem	2666	7600	32	190	NA	75.0	2.08
14	MeBr (*3, 4, 17, 18*) shank	216645	13200	56	263	30.0	81.0	2.57
15	MeBr (*6, 7*) lab soil col	216645	13200	56	110	30	42.9	1.07

[a] Measured in the field and laboratory. [b] $R = (VP \times AR)/(S_w \times K_{oc} \times d)$.
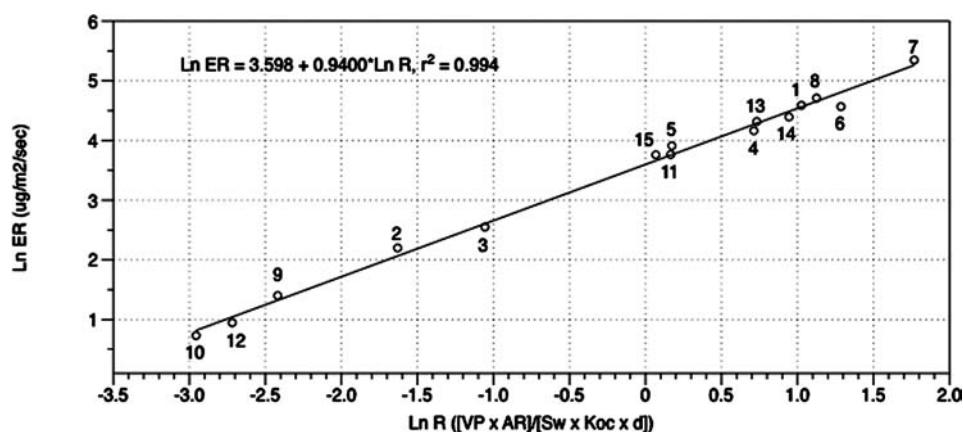
Figure 1. Correlation for surface- and subsurface-applied soil fumigants.

Whereas vapor pressure is the property that primarily determines evaporative losses, it can be attenuated by solubility in water (S_w, mg L^{-1}), adsorption to soil (K_{oc}, mL g^{-1}), and depth of application (d, cm). Vapor pressure (VP, Pa), as the numerator, is first multiplied by the application rate (AR, kg ha^{-1}) and then divided by the chemical properties and soil depth to obtain an R term, where

$$R = (VP \times AR)/(S_w \times K_{oc} \times d) \qquad (1)$$

Equation 1 can be correlated with the measured emission rate ER (μg m^{-2} s^{-1}) (**Table 1**). For surface chemigation, the depth term does not apply. The application rate is included because it determines the total mass of the fumigant applied. In our earlier studies (*1, 2*), we correlated the natural logarithms of ER and R, because emission rates and chemical properties differed by orders of magnitude. We did this so that the ER/R data pairs could have similar weighting factors in the regression. We took the same approach with the fumigants.

For the correlation method to be of value, it is important to use reliable chemical property and emission rate data. For example, a number of different sources, such as reference works, industry and university reports, and published peer-reviewed papers, were consulted to obtain the data summarized in **Table 1** (*3−18*). If there were several values for a particular property, as determined by different investigators under a given set of conditions, the value selected was the one that occurred most often in the scientific literature, or, short of that, was determined by investigators whose work has been recognized as the most reliable by other investigators. A few of the emission rate values listed in **Table 1** (entries 1, 2, 4, and 14) were determined in the field using atmospheric gradient methods that measured fumigant concentrations (c, μg m^{-3}) and wind speeds (u, m s^{-1}) at multiple levels above the fields or at their downwind edge. Emission rates were calculated from integration of the multiple concentration times wind speed ($c \times u$) values over the height of the sampling mast (*17, 19*). This method has the advantage of averaging emission rate over the entire source (*20*). In the laboratory, emission rates were based on evaporative losses from enclosed soil columns under simulated field conditions [entries 5 and 15 (*7*)]. Another method that is often used by regulatory and enforcement agencies involves measuring pesticide concentrations at several downwind distances from a treatment site and then using the results in the U.S. Environmental Protection Agency's Industrial Source Complex-Short-term (ISC-ST) atmospheric dispersion model to back-calculate to estimated emission rates (*21*). Because this method requires less costly and minimal air sampling and meteorological equipment, it has been used in recent years in preference to atmospheric gradient methods, but with equivalent results. The back-calculation method was used to estimate emission rates for eight of the entries in **Table 1** (entries 6−13).

Validation. Field studies conducted by the California Department of Pesticide Regulation, the California Air Resources Board, and the University of Nevada were used as sources of field data (i.e., application conditions and measured air concentrations) (*18, 22, 23*). The application conditions (application method and rate; meteorology) from the field studies and emission rates from the correlation were used with an atmospheric dispersion model [ScreenView (*24*)] to calculate concentrations in air for a number of selected downwind receptors. These calculated concentrations were compared with those measured in the field.

RESULTS AND DISCUSSION

Correlation. The result of the correlation is plotted in **Figure 1**. The correlated data gave a regression with a high correlation coefficient ($r^2 = 0.994$), even when fumigants injected at depth in

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Table 2. Measured versus Calculated Emission Rates

| fumigant | emission rate, μg m^{-2} s^{-1} | | %Δ |
	measured[a]	calculated[b]	
1	98.4	95.8	3
2	9.03	7.89	13
3	12.8	13.5	5
4	64.3	71.4	11
5	50.0	43.0	14
6	96.3	122	27
7	210	192	8
8	111	105	5
9	4.06	3.76	7
10	2.08	2.27	9
11	43.2	42.7	1
12	2.58	2.84	10
13	75.0	72.7	3
14	81.0	88.7	10
15	42.9	38.9	9
av			9

[a] Table 1. [b] Figure 1.

soil, with and without tarping with polyethylene film, were combined with fumigants applied to the soil surface. **Table 2** compares measured emission rates from **Table 1** with those calculated from the regression in **Figure 1**, with percent differences (%Δ) relative to the measured values. The differences fell in the range of 1−27%, with an average of 9% and a median value of 9%.

Methyl bromide (MeBr) and methyl iodide (MeI) were not initially included in the correlation because of some question regarding a reliable soil adsorption (K_{oc}) value for these fumigants. Gan and Yates (*6*) and Gan et al. (*25*) found that many of the published K_{oc} data for MeBr are only apparent and are due primarily to decomposition of the fumigant. After their apparent K_{oc} values were corrected for soil decomposition, there was no longer a measurable K_{oc} for most of the soils tested. MeI, on the other hand, showed greater soil stability, resulting in measurable K_{oc} values for all of the soils tested. The average apparent K_{oc} value was 11.6 mL g^{-1} ($n = 4$); the average value, corrected for some soil decomposition, was 9.8 mL g^{-1}. Various literature sources for MeBr have K_{oc} values falling in the range from 8 to 83 mL g^{-1} (*3, 6, 16*), all uncorrected for soil decomposition. The soil stability results for MeI and MeBr suggest that published K_{oc} values for the remaining entries in **Table 1** are probably apparent and reflect some decomposition. Determination of the relative contribution of soil decomposition to the K_{oc} values for these fumigants was outside the scope of this study.

The correlation without MeI and MeBr gave the following regression:

$$\text{Ln ER} = 3.595 + 0.9445 \text{ Ln } R \quad r^2 = 0.997 \quad (2)$$

Using the measured emission rate data for MeI and MeBr in this correlation (**Table 1**), it was possible to back-calculate to a K_{oc} value for the two fumigants. The regression equation was rearranged and used in the following way:

$$\text{Ln } R = (\text{Ln ER} - 3.595)/0.9445 \quad (3)$$

$R = (\text{VP} \times \text{AR})/(S_w \times K_{oc} \times d)$, with K_{oc} as the unknown. K_{oc} for MeI fell in the range of 9.8−14.9 mL g^{-1}, with an average of 11.5 mL g^{-1} for the four entries in **Table 1**. This is essentially the same as the uncorrected value calculated from data determined by Gan and Yates (*6*). The back-calculated K_{oc} value for MeBr was

about 56 mL g^{-1}, which is close to the high end of the range quoted above.

The dimensionless Henry's constant, K_H, can be expressed as

$$K_H = C_a/C_w \quad (4)$$

and the soil distribution coefficient, K_d, as

$$K_d = C_s/C_w \quad (5)$$

Combining eqs 4 and 5 gives

$$K_d = (C_s/C_a)K_H \quad (6)$$

and

$$K_{oc} = K_d/(0.58 f_{om}) \quad (7)$$

where f_{om} is the fraction of organic matter in the soil (*3*). The terms C_s, C_a, and C_w are concentrations of the adsorbate in soil (g g^{-1}), air (g mL^{-1}), and water (g mL^{-1}), respectively. Using MeBr soil adsorption data from Gan et al. (*25*) and $K_H = 0.3$ (*6*), estimated apparent K_{oc} values were in the range of 30−40 mL g^{-1} for a soil with slightly less than 1% organic matter. Therefore, the back-calculated apparent K_{oc} value of 56 mL g^{-1} is not unreasonable.

MeBr was included in the correlation (**Figure 1**) because it still has some use as a preplant soil fumigant (e.g., about 2.5 million kilograms was used in California in 2008) and field-measured emission data were readily available. But by 2015, all soil fumigations with MeBr will no longer be allowed. Therefore, in 2007−2008, the U.S. EPA gave a conditional registration to MeI as a viable replacement for MeBr (*26*). MeI has a much lower ozone depletion potential (0.016) compared to MeBr (0.6−0.7), and the atmospheric lifetime for MeI is only 4−8 days, compared to 1.5−2.0 years for MeBr (*6*). Most states have already registered MeI as a preplant soil fumigant in anticipation of replacing MeBr.

Validation. To estimate downwind pesticide concentrations using an atmospheric dispersion model, the input data included, in addition to the emission rate estimated from the correlation, field dimensions, downwind distance of the receptor, atmospheric stability (*27*), wind speed, and wind vector angle. The calculated emission rates for soil fumigants (**Figure 1**) and field-measured meteorological and field dimension data were used as input to the atmospheric dispersion model ScreenView (*24*), based on the EPA's SCREEN model, to estimate downwind concentrations for three application scenarios (*18, 22, 23*). ScreenView, which can be used only with single sources, uses the same algorithms as ISC-View ([EPA's ISC-ST model (*28, 29*)]), but allows the input of user-selected meteorological data and distances to downwind receptor sites. Using the **Figure 1** correlation, the field-measured and model-estimated concentrations compared to within 1−35%, with an average of about 14% and a median value of 8%, relative to the field-measured values (**Table 3**). For all cases, estimated downwind concentrations were calculated for air sampling intervals within the first 24 h, assuming that emission rates would be greatest during that period. This is generally true, and the emission rate declines over succeeding days after application. Although the estimated values compared well with the measured values overall, variability was due to the inability to replicate exactly the environmental conditions and the physical layout of the fields. Even so, the two sets of data in **Table 3** compared well within a factor of 2, which may be sufficient for use as a screening application.

The emission rate data in **Table 1** were derived from application methods commonly used in commercial agriculture. Although the data resulted in a good correlation regression

Table 3. Estimated and Measured Fumigant Concentrations at Downwind Receptor Sites

sampling interval[a]	application scenarios	fumigant	concentration in air, $\mu g/m^3$ measured	estimated
1a (22)	surface drip irrigation with	MITC[b]	35.6 (6.2 m)[c]	34.8
1b	aqueous metam sodium, no tarp		49.4 (5.6 m)	48.4
2a (21)	dry, crystalline Dazomet	MITC[d]	179 (18.1 m)	156−227 (193)[e]
2b	applied to soil surface by		157 (18.1 m)	170−247 (209)
2c	ground rig; irrigated by		213 (12.2 m)	200−232 (217)
2d	sprinkler		382−560 (464)[f] (12.2−18.4 m)	558−637 (598)
3a (17)	shank injection with tarp	MeBr[g]	350 (1.4 m)[h]	474
3b			440 (0.8 m)	518
3c			525 (0.4 m)	569
3d			750 (0.12 m)	742

[a] Day/night periods within first 24 h. [b] Metam sodium. [c] Sampling station downwind distance. [d] Dazomet. [e] Range (av) due to different atmospheric stabilities (26). [f] Range (av) for three closely located sampling stations. [g] Tricon 67/33 (67% MeBr, 33% chloropicrin). [h] Height above ground at 6 m from downwind edge of field.
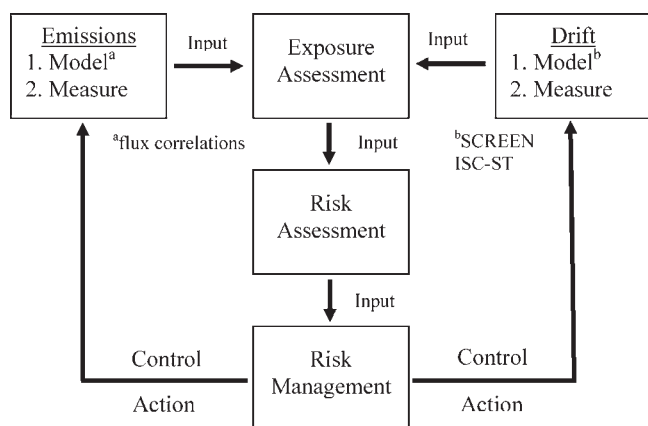
Figure 2. Pesticide exposure and risk assessment flowchart.

(**Figure 1**), any deviation from these common application methods could create outliers in the correlation. A specific example is the use of very impermeable films (VIF) to retard emissions. These films have been shown to reduce emissions to a fraction of what they would be with the more conventional polyethylene film (30, 31). The use of less common application practices, such as VIF, would require the development of new correlations.

The estimated downwind concentrations for MeBr in **Table 3** were based on typical application conditions for this soil fumigant (17, 18). Using the same application conditions [i.e., field dimensions, application rate (AR), and depth of injection (d)] and meteorological conditions as for MeBr, estimated concentrations for MeI in air at the same downwind distance would be only about 1% greater than those for MeBr. However, MeI is a broad-spectrum, acutely toxic chemical and, because of this, is about 1.5−2.0 times more effective as a soil fumigant than MeBr (32, 33). This suggests that less MeI can be applied, leading to lower emission rates and less downwind exposure for farm workers and nearby residents.

The type of regression illustrated in **Figure 1** can be part of a tool kit available to regulatory and enforcement agencies for exposure and risk assessment. As illustrated in **Figure 2**, if actual measurements are not possible, modeled emissions can be used as input to atmospheric dispersion models (e.g., ISC-ST, SCREEN) to estimate downwind concentrations. Emissions and drift estimates will serve as input to an exposure assessment protocol, leading to a risk assessment based on the magnitude of the estimated emissions and downwind concentrations. Although

the best approach is still to measure emissions and concentrations directly, the correlation approach presented here, in combination with atmospheric dispersion models, has sufficient accuracy so that the model package can serve as a relatively rapid screening method for determining potential exposure. Furthermore, the model approach can be used to estimate potential exposure under various conditions of wind speed, wind direction, temperature, etc., which can present combinations difficult to replicate in a field measurement effort. A series of pesticides could easily be evaluated within a day. The ones that showed modeled emissions and downwind concentrations above action levels would be flagged for further evaluation in the field. This has the potential of significantly reducing the workload, especially in the registration process for new pesticides.

The fumigant correlation presented here is an extension of the general method discussed previously (1, 2). In these earlier papers, we presented correlations for semivolatile pesticides applied to soil, foliage, and water surfaces, with soil incorporation in some cases. These correlations were validated by using them, along with an atmospheric dispersion model, to estimate downwind concentrations for a number of commercial applications. In the current paper, we have added to the regulatory tool kit by providing a correlation for highly volatile fumigants applied to soil by surface chemigation and at depth by subsurface injection.

SAFETY

No hazardous chemicals or procedures were used in this study.

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