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Simulating emissions of 1,3-dichloropropene after soil fumigation under field conditions

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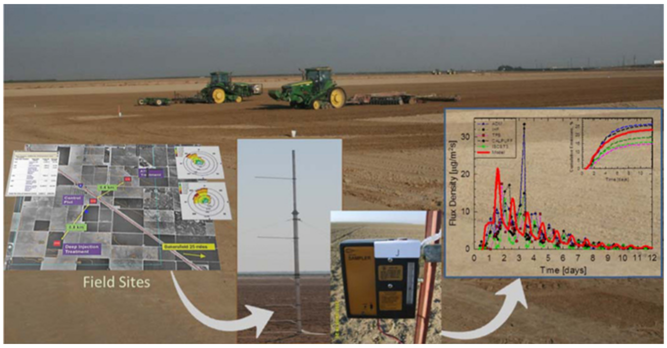
HIGHLIGHTS

- Compares predicted and measured fumigant emissions from five field experiments
- Two independent datasets and five methods were available for measured emissions.
- Total emissions were adequately predicted using a standard modeling approach.
- The ranges of predicted and measured emissions were similar (5.8–29% and 4.3–26%).

GRAPHICAL ABSTRACT



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ABSTRACT

Soil fumigation is an important agricultural practice used to produce many vegetable and fruit crops. However, fumigating soil can lead to atmospheric emissions which can increase risks to human and environmental health. A complete understanding of the transport, fate, and emissions of fumigants as impacted by soil and environmental processes is needed to mitigate atmospheric emissions. Five large-scale field experiments were conducted to measure emission rates for 1,3-dichloropropene (1,3-D), a soil fumigant commonly used in California. Numerical simulations of these experiments were conducted in predictive mode (i.e., no calibration) to determine if simulation could be used as a substitute for field experimentation to obtain information needed by regulators. The results show that the magnitude of the volatilization rate and the total emissions could be adequately predicted for these experiments, with the exception of a scenario where the field was periodically irrigated after fumigation. In addition, the timing of the daily peak 1,3-D emissions was not accurately predicted for these experiments due to the peak emission rates occurring during the night or early-morning hours. This study revealed that more comprehensive mathematical models (or adjustments to existing models) are needed to fully describe emissions of soil fumigants from field soils under typical agronomic conditions.

1. Introduction

Soil fumigant chemicals have the potential to increase atmospheric concentration of photochemical smog precursors, which could react with nitrogen oxides in the atmosphere and can significantly impact regional air quality by emitting toxic substances into the regional air

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stream (van den Berg et al., 1999; Yates et al., 2011b). Photochemical smog has become a problem in California during the summer months and fumigants have been identified as potential contributors. Monitoring indicates that additional efforts are needed to ensure that U.S. EPA's 8-h ozone standard is attained throughout the state (Neal et al., 2009).

Telone II® (1,3-dichloropropene, 1,3-D) is an effective nematicide commonly used for preplant soil fumigation. According to CDPR (2017), 1,3-D was the most highly used fumigant (based on mass applied) in CA from 2011 to 2015 and the third most highly used pesticide (based on mass applied) in 2015. Between 2007 and 2015, 1,3-D use in CA increased from 4.3 × 106 kg (9.5 × 106 lbs) to 7.2 × 106 kg (15.8 × 106 lbs), with the applied land area increasing from 21.8 × 103 ha (53.9 × 103 acres) to 32.2 × 103 ha (79.4 × 103 acres). However, 1,3-D is considered a possible carcinogen and is a Clean Air Act substance (Baker et al., 1996). The high volatility of 1,3-D (vapor pressure 4.3 kPa; Wauchope et al., 1992) facilitates its movement (which increases effectiveness), but can also lead to high atmospheric emissions and the potential for worker exposure (Albrecht, 1987). In 1990, the detection of high 1,3-D concentrations in ambient air samples at multiple sites in California led to a temporary suspension of 1,3-D as a soil fumigant (CDPR, 2002). The suspension remained in effect until 1995 when approaches to mitigate emissions were developed and tested.

There is very little published information on field-scale emissions of 1,3-D. There have been several reported soil column experiments (Basile et al., 1986; Gan et al., 1998a, 1998b; Zheng et al., 2006; Gao and Trout, 2007; McDonald et al., 2008; Ashworth et al., 2009) that placed 1,3-D emission losses from bare soil in the range 20–77% of the applied dosage. These studies also revealed that emission rates obtained using laboratory soil columns can have high variability (i.e., $\mu = 41\%$, $\sigma = 16\%$). Total emission rates have been obtained under field conditions using micrometeorological and flux chamber methods. These studies have shown a similar range in total emissions 12–80% (van den Berg, 1992; Chen et al., 1995; van Wesenbeeck et al., 2007; Chellemi et al., 2010; Gao et al., 2008; Gao et al., 2009), with the highest estimates obtained using flux chambers ($\mu = 56\%$, $\sigma = 22\%$). Flux chambers offer a relatively simple and cost-effective method for determining emission rates. They are also known to suffer from several practical problems such as sampling a small surface area and the presence of the chamber affecting the tested surface (Gao et al., 1997), which can affect the emission measurements. Flux chambers have been shown to provide results consistent with micrometeorological methods when flux chambers are well designed and appropriately used (Gao et al., 1997). Flux chambers may be the only practical method available for measuring emissions for small treated areas or when experiments are conducted with several treatments in close proximity.

It is important to understand emissions from fumigated soil at typical agronomic-scales to reduce the adverse impacts from soil fumigation. Large-scale field measurement of fumigant emissions provides information regulators require at the appropriate scale while serving as a reference baseline for the development and testing of new methods to reduce emissions.

Continued use of soil fumigant chemicals in the future will likely require developing strategies to reduce emission rates. In order to achieve this goal, extensive environmental fate and transport information is needed that can be used to design and test potential mitigation approaches compatible with large farming operations. However, field experimentation is expensive and comparing the results from different field experiments is tenuous unless the experiments are conducted at the same time, in the same location and in the same (or similar) soil type. Meeting these requirements significantly increases the cost and complexity of field experimentation.

Computer modeling provides an alternative to conducting large-scale field experiments. Using modeling, new and potentially cost-effective management practices could be developed and initially tested by comparing simulation results from new vs. standard pesticide management methods. Potential fumigation practices that mitigate emissions could then be tested in the field. However, before this type of approach becomes useful, it is necessary to understand and be able to predict all of the important routes of transport and dissipation, so that models outputs are similar to field measurements.

The objective of this study was to determine if numerical simulation can be used as a substitute for conducting complex, expensive and time consuming 1,3-D field experiments. Field experiments are currently used to quantify regulatory information on the short-term and total emission rates of 1,3-D after shank fumigation in large-scale agricultural fields. Information on emissions is required by state and federal regulators as inputs into risk assessment models for the development of buffer zones to ensure public safety, and also benefits the agricultural community by providing science-based information concerning rates of emission. The simulation results described below were obtained in a predictive mode and did not use any inverse approach.

2. Modeling methods

2.1. Transport equations

The model used to simulate fumigant fate and transport in variably-saturated soils and subject to variable temperatures requires three governing processes: water flow, heat transport, and solute fate and transport. The transport of water was simulated using Richard's Equation (Šimůnek and van Genuchten, 1994; Jury and Horton, 2004):

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left[K_x(h)\frac{\partial h}{\partial x}\right] + \frac{\partial}{\partial z}\left[K_z(h)\frac{\partial h}{\partial z} + K_z(h)\right] - S \qquad (1)$$

where θ is the volumetric water content ($\mathrm{cm}^3\ \mathrm{cm}^{-3}$), h is the pressure head (cm), K is hydraulic conductivity ($\mathrm{cm\ s}^{-1}$), and S is a sink term (s^{-1}); t is time (s), x and z is distance (cm). For all simulations, the sink term was set to zero.

Heat transport was simulated using Eq. (2) (Šimůnek and van Genuchten, 1994):

$$C_h(\theta)\frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left[\lambda_x(h)\frac{\partial T}{\partial x}\right] + \frac{\partial}{\partial z}\left[\lambda_z(h)\frac{\partial T}{\partial z}\right] - C_w q\frac{\partial T}{\partial z} \qquad (2)$$

where T is soil temperature (K), C_h and C_w are the volumetric heat capacity for the porous media ($\mathrm{J\ m}^{-3}\ \mathrm{K}^{-1}$) and liquid, respectively, and λ is the apparent thermal conductivity ($\mathrm{W\ m}^{-1}\ \mathrm{K}^{-1}$).

The transport of soil fumigant requires descriptions of the phase partition between liquid, gas and solid phase, dispersion (convection and diffusion), and degradation processes. Degradation was described using a first-order decay reaction, and included the ability to specify the degradation rate in each phase (liquid, vapor, and solid). The governing transport equations for the fumigant were written as follows (Šimůnek and van Genuchten, 1994):

$$\eta\frac{\partial C_g}{\partial t} + \theta\frac{\partial C_l}{\partial t} + \rho_b\frac{\partial C_s}{\partial t} = \frac{\partial}{\partial x}\left[D_g\frac{\partial C_g}{\partial x} + D_l\frac{\partial C_l}{\partial x} - q_x C_l\right] + \frac{\partial}{\partial z}\left[D_g\frac{\partial C_g}{\partial z} + D_l\frac{\partial C_l}{\partial z} - q_z C_l\right] - \eta\mu_g C_g - \theta\mu_l C_l - \rho_b\mu_s C_s \qquad (3)$$

where C_g, C_l, and C_s are gas-, liquid-, and solid-phase concentrations ($\mu\mathrm{g\ cm}^{-3}$), respectively; D_l and D_g are liquid- and gas-phase diffusion coefficients ($\mathrm{cm}^2\ \mathrm{s}^{-1}$), respectively; μ is a first-order degradation coefficient (s^{-1}); θ, ρ_b, and η, respectively, are water content ($\mathrm{cm}^3\ \mathrm{cm}^{-3}$), bulk density ($\mathrm{g\ cm}^{-3}$), and air content ($\mathrm{cm}^3\ \mathrm{cm}^{-3}$); q is the Darcian flux density; and the subscripts: l, s, and g indicate liquid-, solid-, and gas-phases, respectively. The partitioning between the liquid- and gas-phase was assumed to obey Henry's Law (e.g., $C_g = K_h C_l$) and the partitioning between the liquid- and solid-phase was assumed to be equilibrium adsorption $C_s = K_d C_l$. The temperature dependence of the diffusion coefficients and degradation rates were described using