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A comparison between field-estimated and HYDRUS-simulated emission of 1,3-Dichloropropene from agricultural fields

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1. Introduction

1,3-Dichloropropene (1,3-D) is a fumigant used to control nematodes, insects and disease organisms in the soil. It is commonly used as a pre-plant treatment that is injected into soil. It may also be applied through drip irrigation. Regardless of the application method, the possibility of offsite transport of this fumigant due to volatilization may subsequently result in human exposure through inhalation. To mitigate its potential cancer risk, the Department of Pesticide Regulation (DPR) limits the use of 1,3-D on a regional basis (township cap). The current township cap is 136,000 “adjusted” pounds during a calendar year in any township (six-by six-mile area). Adjusted pounds refer to the amount of 1,3-D active ingredient multiplied by an application factor (AF) to account for differences in air concentrations due to application method, region, and season of application.

Application factors are multipliers that DPR originally intended to use to account for variation in cumulative flux density between different application methods (e.g., deep versus shallow shank injection). The basis for determining an AF is the emission ratio (ER, also referred to in some documents as an 'emission rating'), an estimate of the emitted fraction of total applied mass at a given time post-application (ER = cumulative flux / mass applied). Department of Pesticide Regulation derived ER values currently used in AF calculations from a small selection of field-estimated ER values obtained from field flux studies. However, these initial field studies are small in number due to various reasons including financial challenges. Therefore, DPR combines computer modeling with field studies to expand its ability to estimate ER for different application methods and under different environmental conditions.

HYDRUS (Simunek et. al., 2008) is the primary model used by DPR to simulate the emission of fumigant from soil to the atmosphere. HYDRUS is a numerical model that solves the Richards equation for saturated and unsaturated water flow and convection-dispersion type equations for heat and solute transport. For the purpose of simulating processes involved in fumigation, the flow equation simulates dynamics of water within the soil profile, leaching, and evaporation. The heat transport equation accounts for heat transfer caused by conduction as well as by convection with flow of water. The convection-dispersion solute transport equations consider reactions between the solid, liquid, and gas phases that are required for solutes that exist in both adsorbed and volatile phases such as pesticides. The solute transport equations also simulate zero-order production, first-order degradation, and first-order decay/production reactions. In addition, it simulates solute transport simultaneously in both the liquid and gaseous phases.

The HYDRUS model has been successfully employed to simulate the fate of fumigants including 1,3-D for different application methods and under different conditions ranging from bare ground broadcast to partially totally impermeable film (TIF) covered chemigated bed application (Spurlock et al. 2013). In this report we further evaluate the ability of the HYDRUS model to estimate 1,3-D flux under different application methods. The key difference between this evaluation and that of Spurlock et al. (2013) is the availability of detailed site-specific input data for HYDRUS. Therefore, the objective of this report is to evaluate how well HYDRUS model results compare to field-estimated flux values in conditions when a limited amount of site-specific input data is available and the HYDRUS model relies on input parameters from previously peer-reviewed publications.

2. Materials and methods

2.1. Simulation of field experiments

A total of 23 experiments was available for the purpose of modeling from the 1,3-D emission database compiled by Kandelous (2018) (See appendix A for detail). The HYDRUS model requires specific information about the experiment, such as the extent of modeling domain; soil physical, hydraulic, thermal, and chemical properties; solute properties such as diffusion, reaction, degradation, absorption, and their temperature dependency; initial status of water, solute, and temperature in the soil profile (so-called initial condition); boundary conditions in respect to water (e.g., irrigation, precipitation, evaporation), heat (temperature at soil surface and temperature of irrigation water); and solute (concentration of applied solute in irrigation water in case of chemigation, or tarp permeability for gas diffusion) and their temporal variation.

There were no geographical restrictions and all experiments (from four states of California, Texas, Georgia, and Florida) with minimum required input data of domain properties, soil properties, initial, and boundary conditions were considered for HYDRUS modeling. A total of 13 experiments from the dataset of 23 experiments available, met the minimum input requirements and were used for HYDRUS modeling. Minimum input data from the 13 experiments was extracted and the remaining required input data was assumed based on data in published literature and from previous HYDRUS simulations conducted by Brown (2018) (See appendix B for detail). Table 1 shows a brief description of experiments used for HYDRUS modeling that cover a broad range of field fumigation methods (FFM) including FFM codes 1201, 1202, 1206, 1209, 1242, and 1259.

Table 1. List of field experiments used for evaluation of HYDRUS model to simulate 1,3-D flux from fumigated fields.

	Study	FFM code	Application type	Application depth (in)	Application rate (lb/ac)	Duration (day)	Tarp type	Tarp cut (day)
1	Gillis and Dowling, 1999	1201	Broadcast	14	121.51	14	NA	NA
2	Gillis and Dowling, 1999	1201	Bed	12	67.41	14	NA	NA
3	Ajwa and Sullivan, 2012b	1202	Broadcast	12	122.80	14	PE	6
4	Ajwa, et al., 2012	1202*	Broadcast	8	153.50	10	HDPE	No cut
5	Ajwa, et al., 2012	1202*	Broadcast	8	146.25	10	HDPE	No cut
6	Knuteson, et al. 1992b	1206	Broadcast	18	121.21	14	NA	NA
7	Knuteson, et al. 1995	1206	Bed	21	119.24	21	NA	NA
8	van Wesenbeeck and Philips, 2000	1209	Drip	1	67.23	14	PE	No cut
9	Ajwa and Sullivan, 2012a	1242	Broadcast	12	230.80	18	TIF	16
10	Ajwa and Sullivan, 2012a	1242	Broadcast	12	221.00	12	TIF	10
11	Ajwa and Sullivan, 2012a	1242	Broadcast	12	239.80	7	TIF	5
12	Ajwa and Sullivan, 2012b	1242	Broadcast	12	126.39	14	TIF	6
13	Ajwa, 2015	1259	Drip	1	173.11	13	TIF	No cut

*These two studies had an application depth shallower than required minimum of 12 inches.

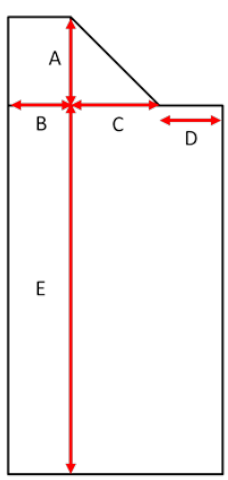
It should be noted that although the minimum depth for FFM 1202 is 12 inches, two experiments included in this study were conducted at a depth of only 8 inches. However, for the purpose of evaluating HYDRUS' ability to produce results comparable to the field-estimated flux values, these two FFM 1202 experiments were included in this study. These are noted in Table 1.

Domain

Information about application method and associated field preparation, application equipment specifications, injection depth, and soil layering were extracted for each individual experiment and a representative simulation domain was created in HYDRUS. The simulation domain represents a vertical cross-section of soil profile perpendicular to the injection line (i.e., chisels or drip-lines). The width of the domain was set to a distance half-way between adjacent injection lines. While the width of simulation domain (B+C+D) varied between different studies, the length of the domain (A+E) was set to 125 cm to eliminate any potential effect of bottom boundary condition on simulation results. Figure 1 shows a general shape of the HYDRUS domain with associated dimensions for the individual studies.

Initial condition

Soil moisture and temperature measured at the beginning of experiment were used to generate soil moisture and temperature profiles for initial conditions in the HYDRUS domain. Fumigant concentration of entire profile, except the injection area, was assumed to be zero. The concentration of fumigant (C) within the injection area was determined as C = M / V, where M is the amount of fumigant applied to the soil surface area of HYDRUS domain, and V is the chisel's volume of influence (i.e. volume of soil that was covered by fumigant at injection time). An illustrative example is presented in appendix C.



	Study	Application depth (in)	A	B	C	D	E
1	Gillis and Dowling, 1999	14	0	25	0	0	125
2	Gillis and Dowling, 1999	12	15	30	10	10	110
3	Ajwa and Sullivan, 2012b	12	0	60	0	0	125
4	Ajwa, et al., 2012	8	0	15	0	0	125
5	Ajwa, et al., 2012	8	0	15	0	0	125
6	Knuteson, et al. 1992b	18	0	22.5	0	0	125
7	Knuteson, et al. 1995	21	20	20	10	20	105
8	van Wesenbeeck and Philips, 2000	1	20	32.5	7	35.5	105
9	Ajwa and Sullivan, 2012a	12	0	60	0	0	125
10	Ajwa and Sullivan, 2012a	12	0	60	0	0	125
11	Ajwa and Sullivan, 2012a	12	0	60	0	0	125
12	Ajwa and Sullivan, 2012b	12	0	60	0	0	125
13	Ajwa, 2015	1	35	32.5	7	35.5	90

Figure 1. General shape of the HYDRUS domain with associated dimensions for individual studies. Parameters A-E are in cm.

Boundary condition

The bottom boundary for water flow was set as “free drainage” and the “third-type” boundary condition for both solute and heat transport. For non-tarpaulin soil surface, the top boundary was set as atmospheric boundary condition for water flow, “volatile type” for solute transport, and “first type” for heat transport. Similar boundary conditions were used for tarpaulin soil surface in broadcast application in which the entire soil surface was covered.

In bed applications including drip (bed chemigation), “atmospheric boundary” was used for the part of soil surface covered with tarpaulin and the remaining area was assigned the “variable flux 1” boundary condition. The top boundary for solute transport was set as “volatile type” and for heat transport as “first type.” However, a different set of boundary temperatures were assigned for soil surface covered by tarpaulin in compare with the bare soil. For drip application, the boundary around dripline was set as “variable flux 2” for water flow, and the “third-type” boundary condition for both solute and heat transport.

Evaporation for tarpaulin soil surface was set to zero, while evapotranspiration data from nearby California Irrigation Management Information System (CIMIS) stations were used for non-tarpaulin soil surface. A sine-wave shape temperature curve was assumed to represent diurnal variation of temperature. The magnitude of average and peaks as well as timing of peaks’ occurrence of soil surface (depth < 5cm) measured in the field were considered for generating sine-wave shape soil surface temperature for both bare soil and under tarpaulin conditions.

Soil properties

Soil profile and layering information provided by field studies was used as the basis to characterize the HYDRUS domain profile for each simulation. Soil textural properties and bulk density were used as inputs for ROSETTA to estimate soil hydraulic properties required by HYDRUS.

Unless specified in this report, soil, solute, and heat specific parameters were the same as those employed by Brown (2018).

2.2. Determination of Emission Ratio and Peak Flux Values

Emission ratio

Emission ratios for both field study estimates and HYDRUS simulations were determined by dividing the total amount of 1,3-D emitted from the application area within the monitoring period by the total amount of applied 1,3-D.

Peak flux

HYDRUS simulations were constructed such that emission values were simulated over the same time intervals as in the corresponding field experiment, thereby allowing for pair-wise comparisons of fluxes. For example, a field experiment collecting air samples in 6-hour intervals would be modeled here with flux output simulated over the corresponding 6-hour intervals. The sampling intervals varied within and between experiments ranging from 2-hour to 24-hour periods. For both field study and HYDRUS simulation, 24- and 72-hour rolling averages of fluxes for the entire monitoring period were calculated. It should be noted that for those instances in which the rolling periods of either 24 or 72 hours were not available, the closest rolling period was chosen such that it did not exceed 25 and 73 hours for 24- and 72-hour rolling averages, respectively (see Kandelous 2018, Appendix C). In the next step, maximum 24- and 72-hour rolling averages fluxes were determined.

2.3. Statistical indicators

The HYDRUS model's performance for simulating the flux of 1,3-D from the soil surface was evaluated by comparing field-estimated fluxes to HYDRUS-simulated fluxes and calculating a range of error estimates, dimensionless efficiency tests, and a test of significance in linear relation.

The error estimates included mean error (ME), mean absolute error (MAE), and root mean square error (RMSE). Linear regression analysis was used to calculate the coefficient of determination (R^2) and to test the level of linear relationship between field-estimated and HYDRUS-simulated fluxes. Linear model assumptions were evaluated using a global validation criteria introduced by Peña and Slate (2006). Lastly, the HYDRUS model's performance in simulating 1,3-D fumigant flux was evaluated by two indicators of model efficiency: one proposed by Nash and Sutcliffe (1970) (Nash and Sutcliffe model efficiency or NSE) and the other introduced by Willmot (1981), the index of agreement (IA). The NSE ranges between $-\infty$ and 1 (perfect fit), where a value between 0 and 1 is considered to indicate acceptable performance. The IA ranges from 0 to 1, where a value of 1 means a perfect agreement between model and observation. See appendix D for equations used to calculate these statistical indicators.

3. Results and discussion

Figure 2 shows the comparison between simulated and field-estimated emission ratio and peak 24-hour and 72-hour rolling average fluxes. The regression analysis for the slope resulted in r-square values of 0.76, 0.77, and 0.83 for emission ratio and peak 24-hour and 72-hour rolling average fluxes, respectively (See appendix E for detailed information of regression analysis and evaluation of linear model assumptions). These relatively high r-square values are indications of the strength in linear relationship between simulated and field-estimated values. It is also shown that the 95% confidence interval around the regression line includes the slope of one. The close proximity of slope values to one and relatively high values for the coefficient of determination show that simulated values are close to those calculated from field data. The error estimate indicators of ME, MAE, and RMSE show relatively small values for all three indices indicating small differences between simulated and field values. In order to provide a comprehensive evaluation of model performance, NSE and IA were calculated for all three of the emission ratios and peak 24-hour and 72-hour rolling average fluxes (Fig.2). Nash and Sutcliffe model efficiency values were all greater than 0.75, which shows that the HYDRUS model is highly efficient in recreating field-estimated data. In addition, IA was calculated for all three indices, and all IA values were above 0.92. These values show that there are strong agreements between simulated and field-estimated emission ratios and peak 24-hour and 72-hour rolling average fluxes.

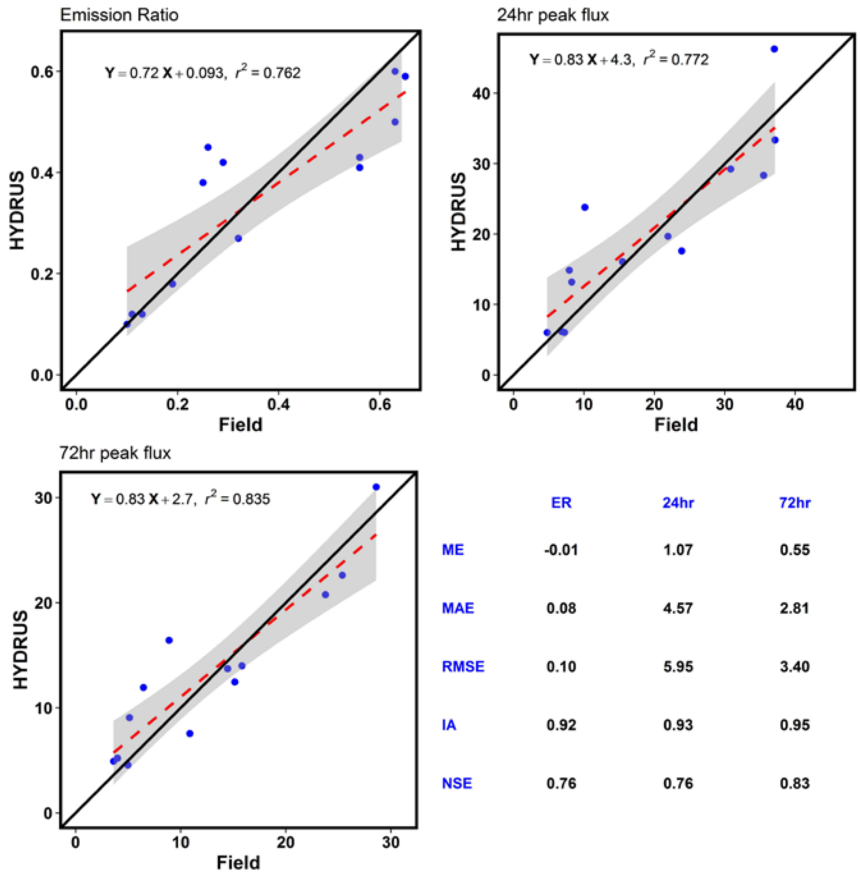


Figure 2. Comparison between simulated and field-estimated emission ratios, peak 24-hr rolling average fluxes, and peak 72-hr rolling average fluxes. The red dashed line shows the linear regression line, the gray shaded area shows the 95% confidence interval, and the black line represents the 1:1 ratio line. RMSE is root mean square error, MAE is mean absolute error, ME is mean error, NSE is Nash and Sutcliffe model efficiency, and IA is index of agreement.

Figure 3 shows a paired comparison of simulated and field-estimated emission ratios and peak 24-hour and 72-hour rolling average fluxes. In general, it shows that the simulated values are slightly lower for fumigation methods with shallow injection depths (i.e., FFM 1201 and 1202) and are slightly higher than those estimated from field studies for fumigation methods with deep injection depths (i.e., FFM 1206 and 1209). It also shows generally good agreement between simulated and field-estimated values when soil surface is covered with TIF (i.e., FFM 1242 and 1259).

4. Conclusion

As stated in the objective, the goal of this report was to evaluate the applicability of the HYDRUS model to estimate 1,3-D emissions using the conventionally collected data. The final comparison is not between simulated data and observed data, as the field scale fluxes were estimated using Aerodynamic, Integrated Horizontal Flux, or Back-Calculation approaches. Therefore, the analysis is a comparison between two approaches to estimate 1,3-D emission rate (flux).

Given the statistical comparisons conducted in this report, we conclude that simulated fluxes by the HYDRUS model are comparable to those estimated using submitted field study data. Therefore, HYDRUS can provide an alternative or assistive tool to estimate the emission rate of 1,3-D from the application area.

Field variability in application rate, soil type, soil moisture, tarp properties and possible tarp perforation provide considerable spatiotemporal variation in emission rate at field scale. While the variability in most of these parameters cannot be realistically quantified, a comprehensive HYDRUS modeling study should consider those variations whenever possible.

The advantage of using HYDRUS is that it can demonstrate the uncertainty in fumigant emissions on a field scale while other approaches report a single emission value despite the existence of considerable uncertainty in the estimated value.

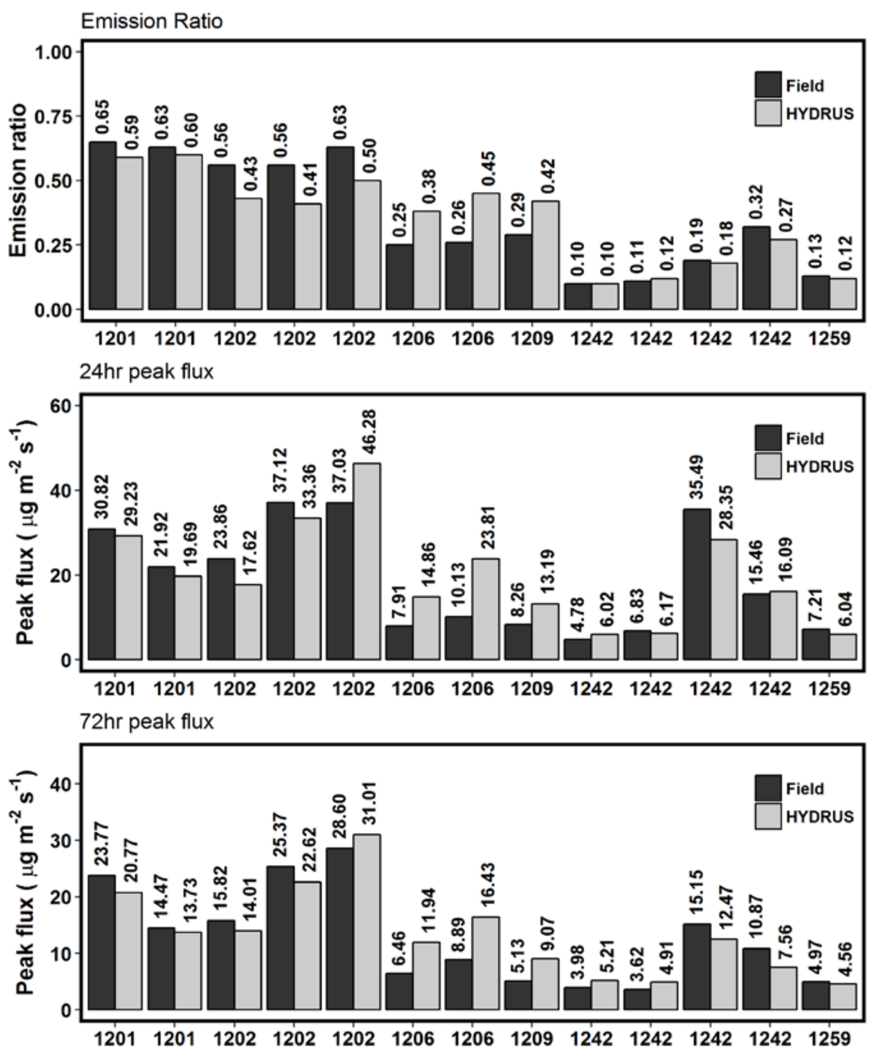


Figure 3. Bar plots of comparison between simulated and field-estimated emission ratio, peak 24-hr rolling average fluxes, and peak 72-hr rolling average fluxes. The X-Axis show FFM code associated with each bar and in the same order as in Table 1. FFM 1201 represents non-tarped shallow broadcast, FFM 1202 represents tarped shallow broadcast, FFM 1206 represents non-

tarped deep broadcast, FFM 1209 represents chemigated tarped bed, FFM 1242 represents TIF shallow broadcast, and FFM 1259 represents chemigated TIF bed.

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