

Effect of Films on 1,3-Dichloropropene and Chloropicrin Emission, Soil Concentration, and Root-Knot Nematode Control in a Raised Bed

Lifang Luo,^{†,‡} Scott R. Yates,^{*,†} Daniel J. Ashworth,^{†,‡} Richeng Xuan,^{†,‡} and J. Ole Becker[#]

[†]U.S. Salinity Laboratory, Riverside, 450 West Big Springs Road, Riverside, California 92507, United States

[‡]Department of Environmental Sciences, University of California, Riverside, California 92507, United States

[#]Department of Nematology, University of California, Riverside, California 92521, United States

ABSTRACT: Soil fumigation is an important component of U.S. agriculture, but excessive emissions can be problematic. The objective of this study was to determine the effects of agricultural films (e.g., tarps) on soil fumigant atmospheric emissions and spatiotemporal distributions in soil, soil temperature, and plant pathogen control in the field using plastic films with various permeabilities and thermal properties. A reduced rate of 70% InLine (60.8% 1,3-dichloropropene (1,3-D) and 33.3% chloropicrin (CP)) was applied via drip line to raised soil beds covered with standard high-density polyethylene film (HDPE), thermic film (Thermic), or virtually impermeable film (VIF). 1,3-D and CP emission rates were determined using dynamic flux chambers, and the concentrations in soil were measured using a gas sampler. The pest control efficacy for the three treatments was determined using bioassay muslin bags containing soil infested with citrus nematodes (*Tylenchulus semipenetrans*). The results show that the Thermic treatment had the highest emission rates, followed by the HDPE and VIF treatments, and the soil concentrations followed the reverse order. In terms of pest control, covering the beds with thermic film led to sufficient and improved efficacy against citrus nematodes compared to standard HDPE film. Under HDPE, >20% of nematodes survived in the soil at 30 cm depth at day 12. The VIF treatment substantially reduced the emission loss from the bed (2% of the Thermic and 6% of the HDPE treatments) and eliminated plant parasitic nematodes because of its superior ability to entrap fumigant and heat within soils. The findings imply that not only the film permeability but also the synergistic ability to entrap heat should be considered in the development of new improved films for fumigation.

KEYWORDS: fumigant, plastic films, emission, pest control

INTRODUCTION

Soil fumigants have been widely used to control weeds, plant-parasitic nematodes, soilborne fungi, insects, and other disease organisms in high-value agricultural production systems.¹ However, after being applied to soils, fumigants evaporate and rapidly escape into the atmosphere due to their high volatility. This imposes potential health risks to nearby residents and increases the level of volatile organic compounds in the air.

Covering the treated field with plastic films following fumigation is a widely used strategy to reduce fumigant emission loss and retain high fumigant concentrations in the soil for pest control. Polyethylene tarpaulins (tarps), such as low-density or high-density polyethylene (LDPE or HDPE) films, are commonly used for fumigant emission reduction. However, they did not prove to be highly effective in reducing emissions of methyl bromide (MeBr),^{2,3} 1,3-dichloropropene (1,3-D),⁴ and other MeBr alternatives.⁵ Compared with polyethylene films, virtually impermeable films (VIF), a group of coextruded films combining multiple film materials (ethylene vinyl alcohol or polyamide), have much lower permeability to fumigant vapors.⁵ Research has demonstrated that VIF provided better emission reduction of MeBr,² 1,3-D, and chloropicrin (CP),^{4,6} as well as methyl iodide (MeI).⁷ Generally, a film's vapor permeability is a critical factor for its effectiveness in emission reduction.^{5,8}

Besides containing fumigants in the soil, tarps also greatly affect soil temperature, which can be an important and

synergistic factor for controlling soilborne pests.^{9,10} For example, Porter and Merriman¹¹ observed that maximum soil temperatures at 5, 10, and 20 cm increased about 14, 12, and 9 °C, respectively, after covering the soil with a 50 μm polyethylene film. When soils are above a certain temperature level, heat alone can suppress many pests and pathogens.^{9,12} Consequently, soil solarization has been promoted as a method for controlling soil pests, primarily in high-value crops in sunny arid regions.¹³ When soil is tarped for fumigation, soil solarization simultaneously plays a role in pest survival. Previous studies have also demonstrated synergistic effects between soil fumigant and temperature; that is, lower application rates were required at higher temperatures for adequate pest control.^{14–16} However, the effects of fumigation films on increasing soil temperature and improving pest control are often neglected in the development of fumigant emission reduction methods. Additionally, temperature influences fumigant transport and fate. For example, increasing temperature increases the diffusion rate of fumigant vapors through the plastic film and in the soil and increases the fumigant degradation rate in soil.³ Therefore, thermal properties of films should be also considered for fumigant practices in agriculture.

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One method to potentially reduce emissions of fumigants is to apply soluble formulations of fumigants through subsurface drip irrigation systems established to irrigated crops. The surface is also often covered with plastic film following fumigation. Compared with conventional shank methods of injection, application of fumigants through drip irrigation systems could be more economical and environmentally friendly.¹⁷ The objective of this study was to determine the ability of different types of films to reduce atmospheric emission losses of 1,3-dichloropropene (1,3-D) and chloropicrin (CP), retain them in soils, enhance soil temperature, and aid parasitic nematode control at a reduced rate of fumigation under one of the following three plastic film treatments: covered with a standard high-density polyethylene film (HDPE), a thermic film (Thermic), or a virtually impermeable film (VIF) in a field site.

METHODOLOGY

Study Site and Plot Description. The field experiment was conducted at the Agricultural Experiment Station, University of California, Riverside, in September 2009. The soil type was an Arlington sandy loam (coarse-loamy, mixed, thermic Haplic Durixeralf), consisting of 64% sand, 29% silt, 7% clay, and 0.92% organic matter, with a pH of 7.2. The soil bulk density was about 1.42 g cm⁻³ in the upper 16 cm of soil and about 1.57 g cm⁻³ below.

Soil beds were constructed to simulate the typical growing system of tomato, bell pepper, and other vegetable crops. Before bed construction, the soil was irrigated to obtain moist conditions and then tilled. The initial water content was about 0.05 g g⁻¹ in the surface layer and 0.10 g g⁻¹ below 10 cm. Each bed was 42 cm wide at the top, 56 cm wide at the bottom, and 20 cm high, and each furrow was about 44 cm wide (Figure 1). The bed length for each treatment

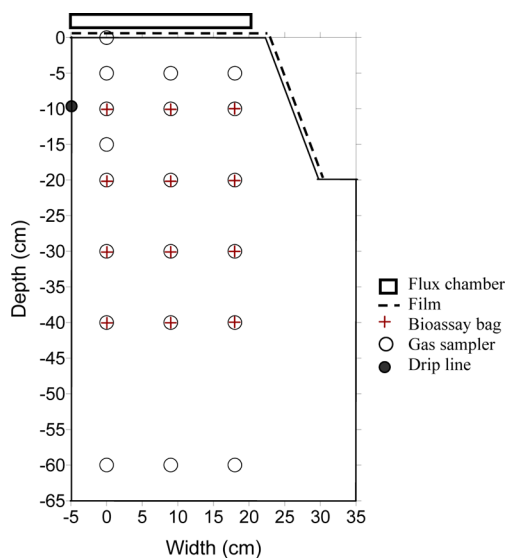


Figure 1. Schematic diagram of half-bed furrow system including the dimensions of the bed and furrow, placements of the plastic film, bioassay bags, gas samplers, and drip irrigation line in a soil cross-sectional view.

was 457 cm. A drip tape (Ro-Drip, 20 cm emitter spacing, flow rate of 250 L h⁻¹ 100 m⁻¹) was mechanically installed along the center of each bed at a depth of 10 cm.

Chemical and Plastic Films. The commercial product InLine containing 60.8% 1,3-dichloropropene (1,3-D) and 33.3% chloropicrin (CP) (Dow Agrosciences, Indianapolis, IN, USA) was used for fumigation. The properties of 1,3-D and CP were previously reported.²³ *cis*-1,3-D, *trans*-1,3-D, and CP have dimensionless Henry's

law constants of 0.074, 0.043, and 0.1, respectively, at 25 °C. The solubilities are 2.3 and 2.2 g L⁻¹ at 25 °C for *cis*-1,3-D and *trans*-1,3-D and 1.6 g L⁻¹ at 20 °C for CP.

Three types of plastic films were selected as tarps for the trial: a 1 mil (a thousandth of an inch), clear, high-density polyethylene film (HDPE, Dow Chemical Co., Midland, MI, USA), a 1.5 mil, clear, virtually impermeable film (VIF, Klerk's Plastics, Hoogstraten, Belgium), and a 1 mil, clear, "thermic" tarp (Pliant Corp., Washington, GA, USA). Among these three types of films, HDPE is commonly used for fumigation and other agricultural practices; as aforementioned, VIF has high efficiency in reducing fumigant volatilization; the thermic tarp is manufactured primarily to enhance soil temperature. The effects of three types of films on soil temperature were examined in a separate experiment conducted during August 24–30, 2010. The temperature sensors were installed in triplicate at 5, 10, 15, 20, 30, 40, and 50 cm deep in the soils covered by HDPE, VIF, and thermic films. The cumulative heat index (CHT₃₀, °C h) was calculated with a threshold temperature of T₀.¹⁸

$$HT_{T_0} = \begin{cases} 0, & T < T_0 \\ [T(t) - T_0]\Delta t, & T \geq T_0 \end{cases} \quad (1)$$

$$CHT_{T_0}(t) = \int_0^t HT_{T_0}(\tau) d\tau \quad (2)$$

where HT_{T₀} is the heat stress index, T₀ is a threshold temperature, Δt is the time interval, and τ is an integration variable. The heat suppression on soil pathogen increases with greater HT_{T₀} value.¹⁸ A value of 30 °C was used for T₀ because the survival of nematodes began to decrease when this temperature was exceeded.¹⁶ VIF had the greatest CHT₃₀ as a function of soil depth, followed by Thermic and HDPE; therefore, the ranking in efficiency to improve soil temperature was VIF > Thermic > HDPE (Figure 2).

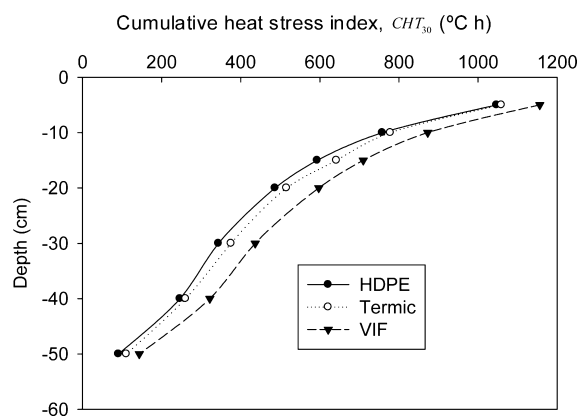


Figure 2. Change of the cumulative heat stress index along depths for the soils covered by a high-density polyethylene film (HDPE), thermic film (Thermic), and virtually impermeable film (VIF) during August 24–30, 2010.

Bioassay Bag Preparation. In previous research, little difference was detected in the dose–response of MeI fumigation among three economically important plant parasitic nematode species, *Meloidogyne incognita*, *Heterodera schachtii*, and *Tylenchulus semipenetrans*.^{19,20} Consequently, the citrus nematode (*T. semipenetrans*) served as a representative plant parasitic nematode in a bioassay to determine the efficacy of each treatment.¹⁹ Feeder roots of a *T. semipenetrans*-infested citrus orchard were collected at the University of California Riverside Citrus Research Center and carefully washed free of soil. The roots containing mature females with egg masses were cut into small pieces, passed through a 2 mm sieve, and mixed thoroughly with soil obtained from the field trial location. The citrus nematode density in the soil was determined according to a modified Baermann funnel technique²¹ with an extraction efficacy of approximately 35%. Each muslin bag

(Hubco Inc., Hutchinson, KS, USA) was filled with 50 cm³ *T. semipenetrans*-infested soil and kept in a 16 °C cold room for the following day field trial.

Installation and Drip Fumigation. In the middle of each bed, a narrow trench was excavated, and gas samplers and bioassay bags were placed in the soil profile before the trench was backfilled (Figure 1). A 10-port air sampler was used to simultaneously withdraw air samples through 10 Teflon tubes (o.d. = 1.8 mm, i.d. = 0.71 mm) from the soil for fumigant concentration measurement. For each bed, two sets of air samplers were used. The inlet ports of Teflon tubes were buried at three positions along the bed: center (at 0, 5, 10, 15, 20, 30, 40, and 60 cm depths), shoulder (at 5, 10, 20, 30, 40, and 60 cm depths), and middle of the above two positions (at 5, 10, 20, 30, 40, and 60 cm depths) (Figure 1). The outlets of Teflon tubes were mated with a quick connect (Small Parts, Inc., Miami Lakes, FL, USA) and connected to 10 50-mL gastight syringes. A sorbent tube (XAD 4 80/40 mg; Supelco Inc., Bellefonte, PA, USA) was used to trap the fumigant gases (1,3-D and CP) in the drawn air. Two replicated bioassay bags were buried at the same three positions as the gas samplers (i.e., center, shoulder of the bed, and middle of the two) at 10, 20, 30, and 40 cm depths. This provided 1,3-D and CP concentration measurements close to the nematodes and enabled a direct measurement of nematode exposure to the fumigant.

Onto each bed was placed a dynamic flux chamber (20 cm × 60 cm) as shown in Figure 1. The inlet to each chamber was connected to a pipe running to a point approximately 30 m upwind of the fumigated area. The mass flow rate of the clean air through the chamber was maintained at 17.5 L min⁻¹. To achieve this, the outlet from each chamber was connected to a central manifold attached to an industrial vacuum pump. For determination of fumigant concentrations within the air flow, the mass flow was subsampled at the chamber outlet at a rate of 80–100 mL min⁻¹ and directed through an XAD-4 sorbent tube. A charcoal tube was used as a backup to check for fumigant breakthrough. A system of solenoid valves controlled by a datalogger was used to sequentially sample up to four consecutive sorbent tubes per chamber. Hourly averaged subsample flow rates were also recorded by the datalogger.

Each treatment bed had a plastic barrel placed at one end that acted as a reservoir for the fumigant solution. An outlet from each barrel was connected to the drip line running through the relevant bed. Into each barrel, running from a central manifold, was a pressure source capable of maintaining around 8–11 psi within each barrel. Thus, application of the fumigants was controlled off-site using an air compressor. At the time of fumigation, 38 L of tap water was added to each barrel followed by a known mass of InLine (49.7 g). On the basis of the surface area of the raised bed (top and sidewalls), this equated to an application rate of 134 kg ha⁻¹ (70% of typical agricultural application rate). Immediately following the addition of the fumigant, the barrels were sealed and shaken to aid mixing. Fumigation was initiated at 11:30 a.m. on September 22, 2009 ($t = 0$), by starting the air compressor. It took about 4 h for a barrel to be emptied by the drip application.

To measure fumigant concentration in the soil, 50 cm³ of the soil pore air was drawn at 0.22, 0.35, 0.96, 1.29, 1.96, 2.96, and 5.29 days after fumigation. Once the experiment started, the sorbent tubes for flux chambers were sampled at an interval of 3 h. After day 6, an interval of 6 h was used due to the decrease in the emission intensity. After 12 days, the experiment was terminated. The films were removed and the bags were pulled out for nematode survival analysis in the laboratory.

Due to the high variation in the nematode population, geometric mean was used for calculation.²² A concentration–time index (CT), cumulative concentration over time, was calculated from the measured concentrations of 1,3-D and CP:

$$CT = \sum_{i=0}^n C_i \Delta t \quad (3)$$

We assumed the combined effects of 1,3-D and CP on nematodes were additive. Therefore, the sum of concentration–time index of 1,3-D and CP was used when it was associated with nematode survival.

Chemical Analysis. Sampled charcoal and XAD 4 charcoal tubes were stored in –60 °C until extracted. The method in Ashworth et al.²³ was followed to extract 1,3-D and CP and analyze the solute concentration.

RESULTS AND DISCUSSION

Atmospheric Emissions. Most of the emission losses (more than 82% for 1,3-D and 95% for CP) occurred during the first 3 days of the field experiment with (Figure 3). Because

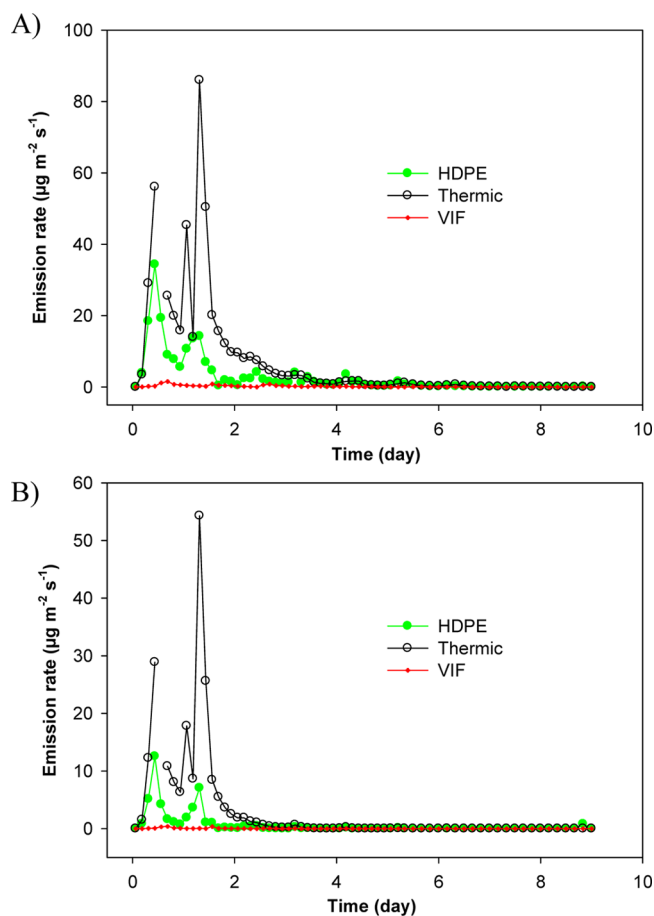


Figure 3. (A) 1,3-D and (B) CP emission rates over time for the bed covered by a high-density polyethylene film (HDPE), thermic film (Thermic), and virtually impermeable film (VIF).

the fumigants were applied relatively close to the soil surface (i.e., 10 cm), they can be rapidly emitted into the atmosphere due to high summer soil temperatures in southern California. The high temperatures facilitated the rapid conversion of liquid fumigants to a gaseous form and led to high emissions on the first day. After 3 days, the emission rates of Thermic and HDPE treatments became relatively very low, about 3% of their peak rates. The emission rates for the Thermic treatment were the highest among three treatments, and the maximum peak reached 86 and 54 $\mu\text{g m}^{-2} \text{s}^{-1}$ at day 1.3 for 1,3-D and CP, respectively (Table 1). As expected, the emission rates were much lower for the VIF treatment, and the maximum rates were only about 1.55 and 0.39 $\mu\text{g m}^{-2} \text{s}^{-1}$, respectively, occurring at day 0.7 for both 1,3-D and CP. Consequently, the VIF treatment had the lowest total mass losses, about 3 and 1%

Table 1. Peak Emission Rates and Total Emissions from the Bed Top for Three Experimental Treatments

treatment	peak emission rate ($\mu\text{g m}^{-2}\text{s}^{-1}$)		total emission (%)	
	1,3-D	CP	1,3-D	CP
HDPE	34.4	12.5	13.5	5.6
Thermic	86.0	54.3	33.4	25.5
VIF	1.6	0.4	0.9	0.3

of those from the Thermic treatment for 1,3-D and CP, respectively. Under HDPE, the peak emission rate was reached at 0.45 day. Both the peak emission rate and total mass losses from the HDPE treatment were about 40 and 20% of those from the Thermic treatment for 1,3-D and CP, respectively. The peak time for the VIF and HDPE treatments was earlier than that observed previously in a field experiment, with the maximum rate observed more than 1 day after fumigation when 1,3-D and CP were applied at 30–46 cm depth in the summer.⁶ In a laboratory chamber experiment,²³ it was noted that earliest and highest emissions of 1,3-D and CP for the HDPE treatment occurred relatively early when the application depth was 10 cm deep (about 17 h for 1,3-D and 21 h for CP). This suggests that the shallow application depth might be related to the earlier peak time in this study.

The Thermic film is manufactured primarily to enhance soil heating for the purposes of soil solarization. The data suggest that as a barrier for fumigants, the Thermic film is less effective than HDPE. Compared to Thermic, HDPE reduced total emission losses from the bed by 60% for 1,3-D and 78% for CP. The emission reduction relative to the bare soil would be even greater if emission loss for Thermic was assumed to be less than that of a bare condition. The total emission reduction for CP is greater than that previously reported when it was applied using a drip irrigation system (40% reduction),²⁴ but it is consistent with the previous findings that HDPE provided better emission reduction for CP than for 1,3-D and that VIF is much more effective in emission reduction than HDPE.^{4,6} However, it was previously noted that, for the VIF treatment, more 1,3-D and

CP could be released from the uncovered furrow than from the bed surface.²³ Because both the bed and sidewall were covered with VIF, the furrow was the only region from which emissions could readily occur. In contrast, being covered by HDPE, the majority of fumigant emissions occurred from the bed surface due to its relatively high permeability. Nevertheless, VIF was still more effective than HDPE in reducing the overall total emission.²³ Therefore, in the present study, although emissions from the furrow region of the VIF-covered bed might have occurred, it seems likely that the level of emissions would not have altered the conclusion that the VIF led to very low emission loss.

Spatiotemporal Distribution of Soil Gas-Phase Concentration. Observation of fumigant concentration over time is essential for determining pest exposure. After day 0.96, 1,3-D was widely spread throughout the bed, and measurable concentrations were found up to about 50 cm deep, suggesting significant gas-phase diffusion occurred soon after injection (Figure 4). Overall, compared with the two other treatments, the soil covered with the VIF had the highest 1,3-D concentration in the profile (maxima $> 2 \mu\text{g mL}^{-1}$), especially at the soil surface. Furthermore, 1,3-D was more uniformly distributed in the lateral direction for the VIF treatment. This lateral distribution of 1,3-D indicates that VIF effectively inhibited the vertical volatilization at the soil surface and that emission could have occurred from the furrow. Both the HDPE and Thermic treatments had higher concentrations near the drip line and decreased with distance. Generally, soil 1,3-D concentrations for the Thermic treatment were the lowest among the three treatments, consistent with the emission data that more mass losses occurred for this treatment. The spatiotemporal change for CP had a pattern similar to that for 1,3-D (not shown).

Gas-phase concentration measurements show that 1,3-D concentration was maximal up to $9 \mu\text{g mL}^{-1}$ at 0.22 day at 5 cm depth and rapidly decreased during the first 3 days (not shown). At day 5.29, the soil concentration became very low. The rapid decrease of fumigant concentration over time was

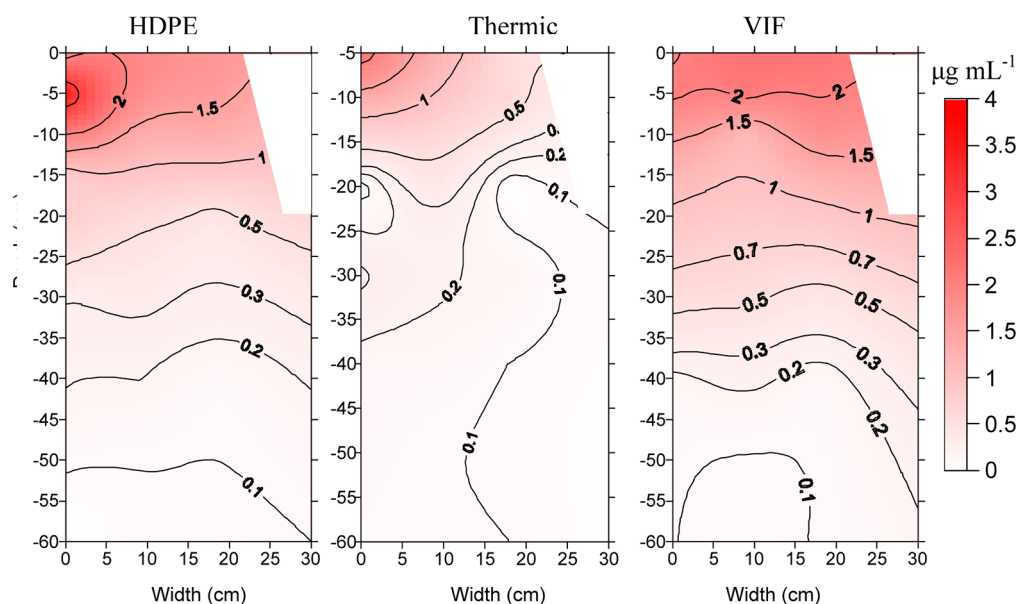


Figure 4. Spatial distribution of 1,3-D soil gas-phase concentration at 0.96 day after fumigation in the half-bed furrow system for the beds covered by a high-density polyethylene film (HDPE), thermic film (Thermic), and virtually impermeable film (VIF).

consistent with the observed trend of the emission fluxes for three treatments. In a laboratory study, Ashworth et al.⁷ observed that VIF was able to retain fairly high soil concentrations of MeI for at least 1 week when soil was fully covered. For this study, in addition to soil degradation and emission loss from the bed and bedside, emission losses from the uncovered furrow might have also contributed to the rapid decrease in the soil concentration.²³ The effect of the bare furrow would be particularly important for beds with a relatively small width (42 cm). Another factor contributing to a rapid decrease in soil concentrations would be an increase in the film permeability^{5,8} and fumigant degradation rate in the soil²⁵ due to high soil temperatures in the field under the film. The half-life of 1,3-D is about 6.3 days at 20 °C and decreases to 1.3 days at 40 °C for the soil in this study.²⁵ According to the temperature measurements in 2010, VIF might have had the highest soil temperature and thus temperature effects on fumigant degradation, followed by the Thermic and HDPE treatments (Figure 2).

Pest Survivability. The spatial distributions of the ratio of surviving nematodes, at day 12 after fumigation, are presented in Figure 5 for three treatments. Overall, the nematodes in the soil profile were effectively controlled in the VIF and Thermic treatments. Only a very small number of nematodes survived in the soil 30 cm below the bed shoulder. Compared with the above two treatments, HDPE was less effective in reducing nematode populations. The area of best efficacy coincided with the region of higher fumigant concentration, that is, the area surrounding the drip tape. However, considerable numbers of nematodes survived in the deeper soil. The highest nematode survival ratio was 49%, found at a depth of 40 cm near the bed shoulder. In contrast, the corresponding values for the Thermic and VIF treatments were much lower, 7.5 and 0.8%, respectively.

Figure 6 shows a plot of concentration–time (CT) values and nematode mortality for the three treatments. Generally speaking, the CT value required to achieve a nearly 100% efficacy in nematode elimination was about $12 \mu\text{g h mL}^{-1}$, the same as the value found by Wang and Yates.²⁶ However, for the same level of CT, the corresponding mortality was relatively lower for the HDPE treatment, compared to those for the VIF and Thermic treatments, indicating that for the HDPE treatment, a greater CT value (i.e., fumigant exposure) was required to fully control nematodes. Although the Thermic treatment had lower soil fumigant concentrations than those of the HDPE treatment (Figure 4), nematode mortality was higher. As Figure 2 shows, the VIF treatment had the highest cumulative heat stress in the soil profile, followed by the Thermic and HDPE treatments. In the top 10 cm of soil, the cumulative heat stress of the Thermic treatment was only slightly higher than that of the HDPE treatment. Nevertheless, due to the intensive heating, the nematodes could be nearly fully controlled by temperature alone under all three treatments in the top 10 cm of soil, according to our unpublished data in other experiments. This agrees with the previous studies that solarization is highly effective in controlling surface soil pests.⁹ The nematode survival data also showed that few nematodes survived within 10 cm of surface soil (Figure 5). The difference in the cumulative heat stress became greater between 10 and 40 cm. For example, the CHT_{30} values at 15 cm during August 24–30, 2010, were 593, 642, and 710 °C h for the HDPE, Thermic, and VIF treatments, respectively. Fumigant efficacy and heat stress on nematodes would greatly increase with

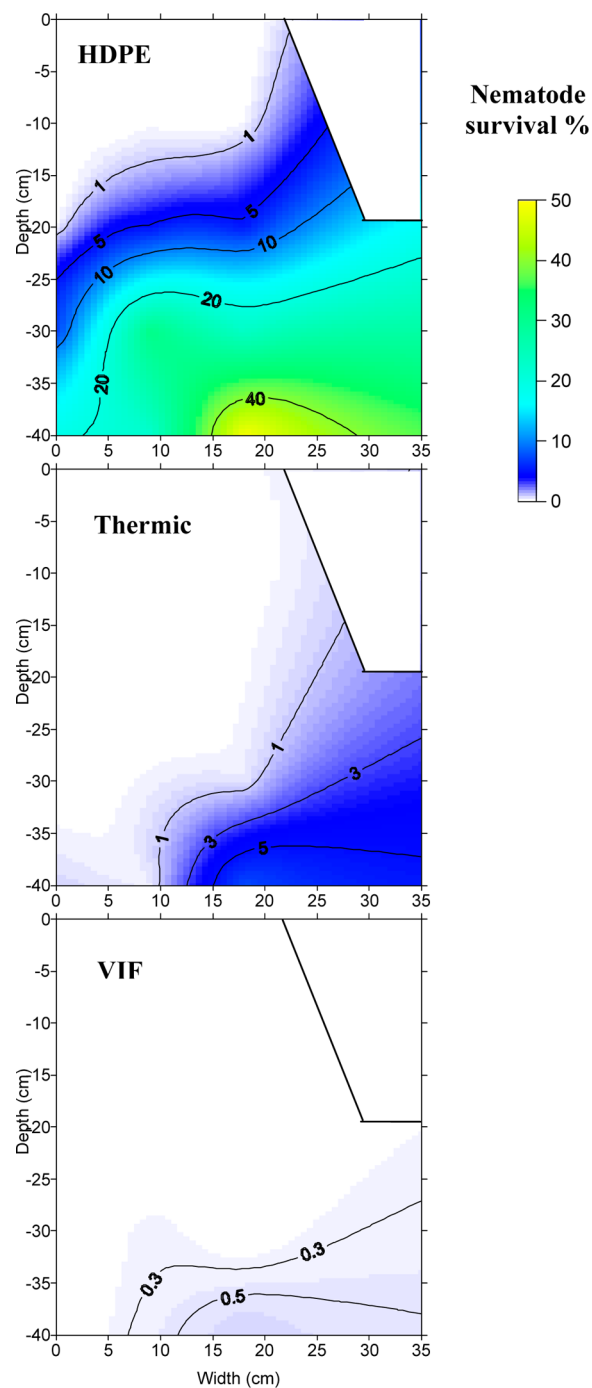


Figure 5. Nematode survival at 12 days after fumigation in the soils covered by a high-density polyethylene film (HDPE), thermic film (Thermic), and virtually impermeable film (VIF).

CHT_{30} . Xue et al.¹⁶ proved that CT values required for adequate nematode control rapidly decreased with temperatures. At 30 °C, the lethal CT values of 1,3-D for nematodes were only about 50% or less of that at 20 °C. At 40 °C, little or no fumigant was needed to kill *T. semipenetrans* because temperature alone became lethal. Similar temperature limits were reported for *M. incognita*.^{10,27} The time required to kill *M. incognita* by temperature alone exponentially decreased with subacute lethal temperature.¹⁰ The higher cumulative heat stress values for the Thermic treatment likely enhanced InLine efficacy against nematodes relative to the HDPE treatment,

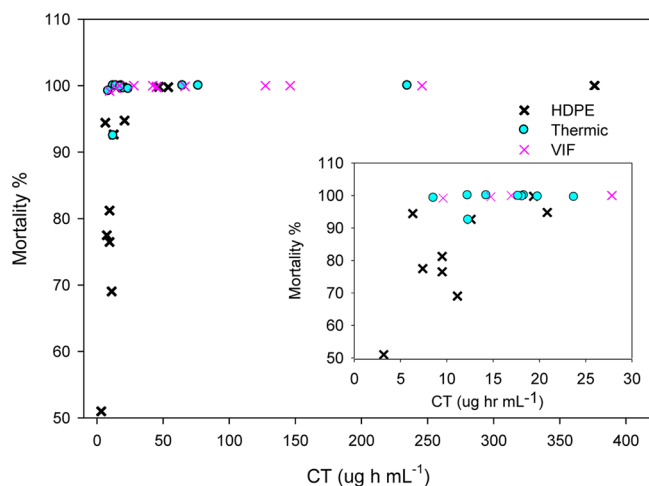


Figure 6. Relationship between the concentration–time index (CT) and nematode mortality for the soils covered by a high-density polyethylene film (HDPE), thermic film (Thermic), and virtually impermeable film (VIF). The portion with CT < 30 $\mu\text{g h mL}^{-1}$ is magnified (inset).

particularly within 10 and 40 cm of soil. For the VIF treatment, the higher fumigant concentrations together with the higher soil temperatures would have had a synergistic effect that led to the lowest nematode survival ratios among the three treatments.

Therefore, among the three types of films, the VIF treatment was the most effective in reducing emission loss and controlling citrus nematodes because it provided the best fumigant containment within soil and also the most elevated soil temperatures. The Thermic treatment also had sufficient efficacy toward the parasitic nematodes in soils due to the increases in soil temperature. However, this treatment also had the highest fumigant emission loss with potential risk to the environment and public. The standard HDPE together with the same reduced rate of InLine was the least effective in killing nematodes, especially at >20 cm soil depths.

When the plastic film is used for fumigation, it not only serves as a barrier for fumigant transport but also improves retention of heat from soil solarization. The findings imply that both film vapor permeability and the thermal properties of the film material may contribute to pest control. Fumigant efficacy controlling soil pests can be improved in soils covered with the films by reducing emission loss and producing higher soil temperatures such as VIF in this study. Therefore, both the vapor permeability and ability to entrap heat should be considered in the development of new improved films.

AUTHOR INFORMATION

Corresponding Author

*E-mail: Scott.Yates@ars.usda.gov. Fax: 1 (951) 342-4964. Phone: 1 (951) 369-4803.

Notes

The authors declare no competing financial interest.

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