Pesticide Science *Pestic Sci* **55**:154–160 (1999)

Spatial and temporal distributions of 1,3-dichloropropene in soil under drip and shank application and implications for pest control efficacy using concentration–time index

Dong Wang* & Scott R Yates

US Salinity Laboratory, USDA-ARS, 450 West Big Springs Road, Riverside, CA 92507, USA

Abstract: A field experiment was conducted to study the spatial and temporal distributions of (*EZ*)-1,3-dichloropropene (1,3-D) in the soil and effects on pest control efficacy. An emulsifiable concentrate formulation of 1,3-D (Telone EC) was applied with drip irrigation at 47 kg AI ha^{-1} to two different depths (2.5 and 20.3 cm, respectively). Comparisons were made between the two drip treatments and a direct shank injection of 1,3-D (Telone II) at 112 kg AI ha^{-1}. Concentrations of 1,3-D in soil air were measured at several locations over time to determine the spatial and temporal characteristics, and to calculate the concentration–time index (*CT*). Citrus nematodes (*Tylenchulus semipenetrans*) were placed in the fumigated soil at 25 cm depth and their mortality rates were compared to the calculated *CT*. Distributions of 1,3-D were found to be relatively uniform in both the drip irrigation and the shank injection treatment. An application rate of 47 kg ha^{-1} with drip irrigation was sufficient to achieve significant concentration levels in soil beds. Applying 1,3-D with direct shank injection at 112 kg ha^{-1} extended the measurable concentration levels to the furrows between the soil beds and to a depth of 1 m below the soil surface. Effective control of *T. semipenetrans* was achieved with both the drip irrigation and the shank injection. A threshold soil 1,3-D *CT* value of 12 μg h cm^{-3} was needed to reach a 100% efficacy for *T. semipenetrans*. The study indicates that 1,3-D fumigation may be carried out with drip irrigation at very low rate, and a *CT* index may be derived to aid in the determination of a minimum effective dosage.

Keywords: fumigant; 1,3-dichloropropene; distribution; efficacy; concentration–time index

1 INTRODUCTION

Modern soil disinfestation for controlling soil-borne plant pathogens and parasitic nematodes has relied heavily on the use of chemical fumigants.[1] Atmospheric emission of these volatile chemicals from the soil is often a major source of air pollution, and regulatory measures have been applied to restrict or suspend their use. The fumigant nematicide 1,3-dichloropropene (1,3-D) has been used widely in the past to control plant parasitic nematodes. Because of the large potential emission loss,[2] off-site air pollution[3,4] and concerns of groundwater contamination,[5] agricultural use of 1,3-D in California and other regions of the world is currently restricted to very low rates and under restricted conditions. Recent research also indicates that soil fumigation with methyl bromide, another major soil fumigant, may result in 21–87% of the latter being lost to atmospheric volatilization.[6–9] Furthermore, because of its high potential for depleting stratospheric ozone, agricultural fumigation with methyl bromide is scheduled to be phased out in 2001 in the US. To meet the urgent demand of locating an alternative soil fumigant, there is a need to investigate different management methods that may be used to apply 1,3-D which would reduce its emission loss while maintaining effective pest control.

Field studies on methyl bromide have indicated that fumigant emission can be reduced by increasing application depth,[9,10] covering the field with plastic films,[10] or reducing the application dosage.[11,12] Because fumigant movement in soil is predominantly by gas diffusion, the increased application depth would lengthen the time needed for the chemical to reach the soil surface, and increase the residence time for degradation in the soil.[13] In addition, use of a plastic cover may reduce fumigant loss into the atmosphere after it reaches the soil surface. The cover is effective only if the plastic material is relatively impermeable to the chemical.[11] The use of an

* Correspondence to: Dong Wang, US Salinity Laboratory, USDA-ARS, 450 West Big Springs Road, Riverside CA 92507, USA
E-mail:dwang@ussl.ars.usda.gov

(*Received 21 August 1998; revised version received 2 September 1998; accepted 12 October 1998*)

 Pestic Sci 0031–613X/99/$17.50

effective cover film may allow a reduction in the application dosage, reducing the overall potential emission loss.

Fumigant 1,3-D is a very effective soil disinfectant in controlling many plant pathogens and parasitic nematodes.[1] For example, it has been used effectively in controlling *Pratylenchus crenatus* in potato.[14] One way of determining the minimum effective dosage is to use the concentration–time index (CT) which has been used for evaluation of phosphine against *Ephestia elutella*[15] or sulfuryl fluoride against *Isoptera rhinotermitidae*.[16] In soil fumigation, the determination of CT for a particular fumigant—pest combination requires detailed measurement of the chemical concentrations over space and time[17] and a corresponding efficacy assessment.

The purpose of this study was to characterize the spatial and temporal distributions of 1,3-D in a field soil and to determine the implications of these distributions on pest control efficacy. In the study, 1,3-D was applied with drip irrigation to two different depths and compared with direct shank injection. Concentrations of 1,3-D in soil air were measured at several locations over time to obtain the spatial and temporal characteristics, and to calculate CT. Citrus nematodes (*Tylenchulus semipenetrans*) were introduced to known locations in the fumigated soil and their mortality rates were compared to the calculated CT.

2 MATERIALS AND METHODS

2.1 Plot description and 1,3-D application

To simulate pre-plant soil fumigation to tomato, bell pepper, and other vegetable crops, soil beds were constructed in a 1.2-ha plot located on a University of California Agricultural Experimental Station near the Riverside campus. Each bed was 102 cm wide, 19 cm high and 36 m long; and was constructed following procedures similar to a commercial field operation. The soil at the research site is an Arlington fine sandy loam (coarse-loamy, mixed, thermic, Haplic Durixeralf) and consists of approximately 64% sand, 29% silt and 7% clay. The organic carbon content is less than 0.5% for the surface 20 cm soil and decreases rapidly to less than 0.01% at deeper depths. Prior to bed construction, the plot was irrigated and tilled to about 30 cm, resulting in an initial soil water content of about 13% and bulk density of 1.5 g cm^{-3}.

The beds were divided into three treatments, each consisting of three beds, with an additional buffer bed (no 1,3-D) between each treatment. Fumigant 1,3-D was applied to the treatments by (i) commercial continuous shank injection at 30.5 cm below the bed center, (ii) deep drip irrigation at 20.3 cm depth and (iii) shallow drip irrigation at 2.5 cm depth. To reduce emission, the beds for surface drip were covered with a layer of high-density polyethylene film (HDPE, ~0.025 mm thick) before 1,3-D application. In the two drip irrigation treatments, a single drip tape, located below the center of each bed, was used to deliver 1,3-D with the irrigation water. The drip tapes were installed during the last process of bed construction (i.e. bed shaping), with a shank mounted on the back of the tractor. A mixing tank was used to premix, dilute, and inject 1,3-D (Telone EC), a water-dispensable formulation with a Z/E ratio of 52 : 48, into the drip irrigation system at an application rate of 47 kg AI ha^{-1}, a typical rate for this area. Chemical injection was made between 1555 and 2235 h on 15 July 1997. Water application was continued for an additional 1.5 h after 1,3-D injection. This was to flush the drip system so that no residual 1,3-D would remain in the drip lines. For the shank injection treatment, unformulated 1,3-D (Telone II, with a Z/E ratio of 58 : 42) was injected, between 1510 and 1515 h, below the center of the beds at 112 kg AI ha^{-1}, using slanted shanks mounted behind a tractor. The three treatments and 1,3-D application methods are summarized in Table 1.

2.2 Sampling and analysis of 1,3-D in soil air

Three soil-air profile samplers were installed in each treatment for measuring 1,3-D gas concentration distribution. The three profiles were located (i) directly below the center of each bed, (ii) 25 cm from the bed center and (iii) 50 cm from the bed center or in the furrow between two adjacent beds. In the shank injection, sampling ports were located at 0, 5, 15, 25, 36, 43, 50, 61, 76, and 100 cm depths. In the deep drip treatment, soil air samples were collected

Formulation	*Application*[a] *method*	*Depth (cm)*	*Rate (kg AI ha^{-1})*	*Surface porosity control*
Telone II (unformulated)	Shank injection	30.5	112	Bare
Telone EC	Drip	20.3	47	Bare
Telone EC	Drip	2.5	47	Tarp[b]

Table 1. Summary of 1,3-D treatments in a field experiment conducted at Riverside, California

[a] Application for shank injection on 15 July 1997, between 1510 and 1515 hours; drip application with water on 15 July 1997, between 1555 and 2235 h, and continued with water only until ~2400 h to flush the drip system.

[b] Tarp = high density polyethylene film; thickness ~0.025 mm.

at 0, 5, 10, 15, 20, 25, 35, 50, 76, and 100 cm depths. In the shallow drip application, 1,3-D samples were taken at 0, 2.5, 5, 10, 20, 25, 30, 50, 76, and 100 cm depths (Fig 1). The locations of sampling ports were different between the three treatments since 1,3-D was applied at different depths. The intentional concentration of sampling locations near the 1,3-D source was to provide a better characterization of 1,3-D distribution, especially the early-time two-dimensional diffusion. Because the beds were 19 cm high, the first sampling port in the furrow started near the surface or 19 cm from the top of the beds. This reduced the total number of sampling ports from the furrow to six for the two drip treatments and seven for the shank application.

To prevent chemical adsorption, narrow Teflon tubing (ID = 1.07 mm; OD = 1.88 mm) was used in the two drip treatments for 1,3-D gas sampling. The tubing for the two drip irrigation treatments was installed to the preselected depths after bed construction and drip tape installation, but before 1,3-D application. To prevent 1,3-D loss from gaps between the small tubes, all the gaps were sealed with silicone glue and the soil surrounding the tubes compacted to near to or little more than the original soil bulk density. In the shank treatment, stainless steel tubing was installed to the preselected locations and depths immediately after 1,3-D injection. The ten or six Teflon tubes from each location were connected to a ten-port quick coupling (Small Parts Inc, O-212101), so that soil air from each location was sampled at the same time with a ten-port semi-automated soil gas sampler. This semi-automated soil gas sampler was constructed by mounting ten 60 cm^3 syringes on a frame with a common pull handle. Each needle was connected, through the adsorption media or activated charcoal tubes (ORBO-32, Supelco Inc., Supelco Park, Bellefonte, PA 16823, 100 mm long and 8 mm in outside diameter, containing 600 mg of charcoal granules), to one of the ten or six Teflon tubes. By presetting the common pull handle to a fixed distance, the volume of soil air sampled would automatically be controlled to the range of 0 to 60-cm^3, and be consistent between sample locations. Except for the first sampling sequence (where 20 cm^3 was taken), 40 cm^3 soil air from each sampling point was drawn through the activated charcoal tubes. The sampling was made three times during the first day after 1,3-D application, twice on the second day, and once daily from day 3 to day 9.

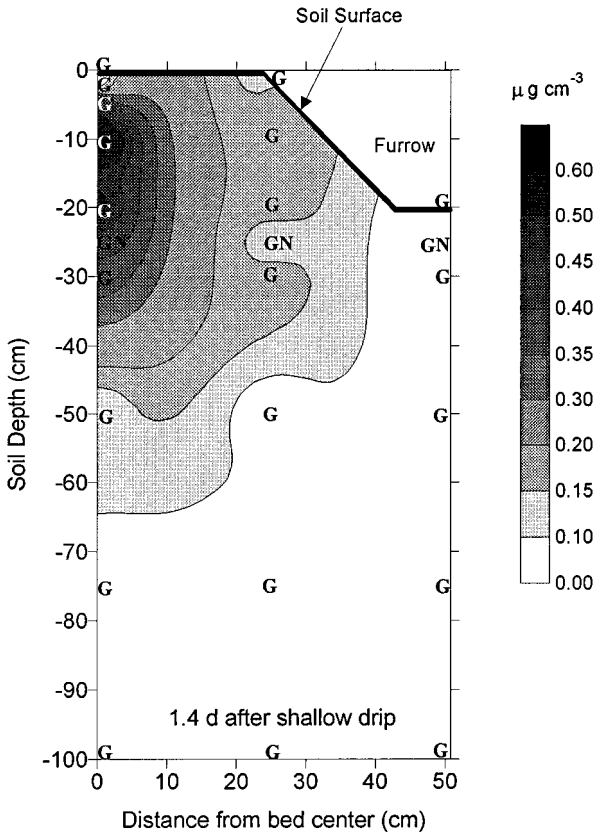

Figure 1. 1,3-D concentration distribution at 1.4 days after application (47 kg ha^{-1}) with shallow drip irrigation at 2.5 cm depth. Letter G represents soil gas sampling port and letter N shows the location of nematode placement.

To recover fully the adsorbed 1,3-D from each charcoal tube, an analytical procedure that can produce a near 100% recovery efficiency for 1,3-D was followed.[18,19] In the method, the charcoal granules were first transferred into 9-cm^3 headspace vials, then 1.0 cm^3 benzyl alcohol was added. The total 1,3-D mass from each tube was then analyzed with a Hewlett-Packard 5890 gas chromatograph with an electron-capture detector. Because of the large number of samples, injections were made from a Tekmar 7000 headspace autosampler equipped with a 7050 sample carousel.

2.3 Pest control assessment and concentration–time calculations

For each treatment, two replicated samples of citrus nematodes (*Tylenchulus semipenetrans*) were buried at three locations: at 25 cm depth below (i) the bed center (ii) laterally 25 cm from the bed center, and (iii) at 5 cm depth in the furrow (Fig 1). These depths were chosen because they are where the highest nematode populations are likely to occur. The nematodes were contained in cloth bags porous to 1,3-D, but which would prevent nematodes from escaping. An average of 3095 nematodes was included in each of the three sample bags from each location. The placement of the nematode samples was coordinated with the installation of soil air samplers, so that one soil air sampler was placed along with the nematode samples. This provided 1,3-D concentration measurement close to the nematodes, which resulted in a direct measurement of the concentrations to which the nematodes were exposed. The nematode sample bags were removed from the plot for analysis of mortality rate at 5.3 days after 1,3-D application. We chose the 5.3 days for nematode mortality count since over 90% of the emission loss would then have occurred.[19] Replicated samples

were also buried in plots without 1,3-D treatment, and were used as controls for the efficacy assessment.

The concentration-time index was calculated as the integral of 1,3-D concentration with respect to time. This may be mathematically described as:

$$CT(t_o) = \int_0^{t_o} C_{x,y}\, dt \qquad (1)$$

where $CT(t_0)$ is the concentration–time index ($\mu g\, h\, cm^{-3}$) up to time t_0 (h); $C_{x,y}$ is the chemical concentration at a given location (x, y) in the soil air ($\mu g\, cm^{-3}$); and t is the time (h). Because chemical concentrations could only be measured at finite time increments, the integration was evaluated numerically using the trapezoidal rule or a linear interpolation between two adjacent sampling points. Measured 1,3-D concentrations from the sampling ports next to the nematode samples were used to calculate the CT by integrating to time t_0 (=5.3 days), and the CT values were correlated to the mortality rates of these nematodes.

3 RESULTS AND DISCUSSION

3.1 Spatial distribution of 1,3-D in soil air

Measured 1,3-D concentrations decreased uniformly with distance from the source to the fumigated beds in shallow drip application (Fig 1). The concentration contours indicate that at 1.4 days after application, significant levels of 1,3-D had moved 40 cm horizontally from the source and 65 cm vertically from soil surface, reaching the bed bottom. The highest concentration was between 0.6 and 0.7 $\mu g\, cm^{-3}$, and had moved downward to 10 cm from the original application depth of 2.5 cm. When 1,3-D was applied at 20.3 cm with drip irrigation, the center mass remained at a relatively higher concentration, ranging from 1.8 to 2.2 $\mu g\, cm^{-3}$, than in the shallow drip plot, and moved downward only slightly to about 25 cm (Fig 2). Nearly the entire bed received significant levels of 1,3-D for at least 1.4 days, indicating a potentially more effective pest control than the shallow drip.

Comparing the concentration levels in the two drip treatments (ie Fig 1 vs Fig 2) clearly shows that more 1,3-D remained in the soil in the deep than in the shallow drip at 1.4 days after application. Because the same amount of 1,3-D was applied in the two drip treatments, more 1,3-D was probably lost to emission from the shallow than from the deep drip application, as expected. The relatively low 1,3-D concentrations in the shallow drip also indicate that the HDPE film used for covering the beds was not very effective in containing 1,3-D emission loss. This is consistent with laboratory studies on 1,3-D permeability through HDPE films.[19] These concentration plots are only snapshots of a continuous 1,3-D redistribution process. For situations where degradation is slow, it is likely that the emission from the deep drip will persist longer than from the shallow drip, and the total emission loss at longer times may be similar to that from the shallow drip.

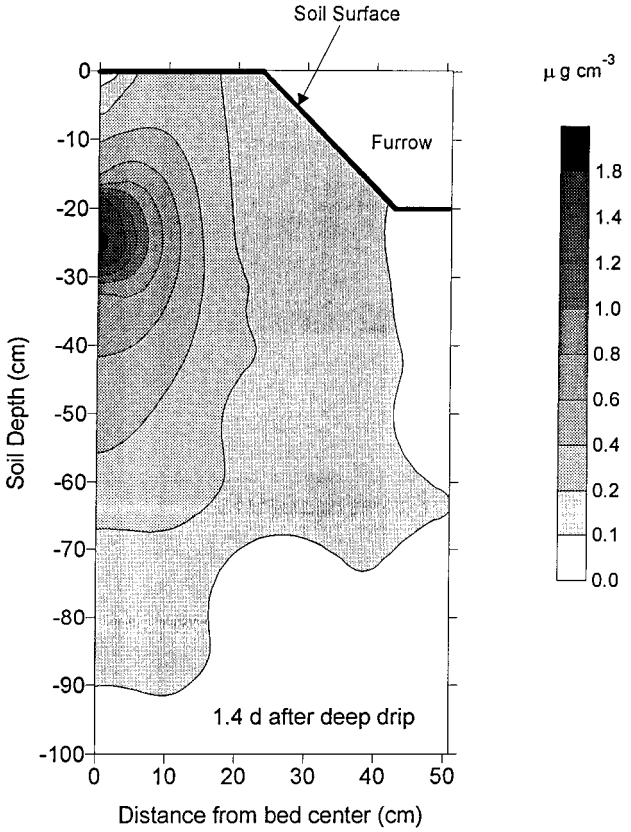

Figure 2. 1,3-D concentration distribution at 1.4 days after application (47 kg ha^{-1}) with deep drip irrigation at 20.3 cm depth.

In both drip treatments, 1,3-D traveled more in the vertical than in the lateral direction. The difference is caused by the effect of gravity on the 1,3-D applied with the irrigation water. In addition to diffusion, 1,3-D moves with water subjected to both capillary and gravity forces as convective flow.

The gas phase 1,3-D concentrations in the shank injection treatment were significantly higher than those in the two drip treatments (Fig 3). Significant concentration levels reached the whole soil profile and below 1 m depth. The highest concentrations remained at levels greater than 2.0 $\mu g\, cm^{-3}$ between 10 and 50 cm depths, at 1.2 days after 1,3-D injection. The elevated levels of 1,3-D are primarily attributed to the higher application rate (Table 1), and to a lesser extent to the 4.8 h earlier sampling time than for the drip treatments. Because only nonformulated 1,3-D was injected, the redistribution or movement would be controlled by gas-phase diffusion and would not be significantly affected by water movement. The lack of convective transport kept the center mass at the original location where the chemical was applied (ie near 30 cm depth). Consistent with measurements in field beds planted with pineapples,[17] 1,3-D gas distributions decreased uniformly with distance from the source when the

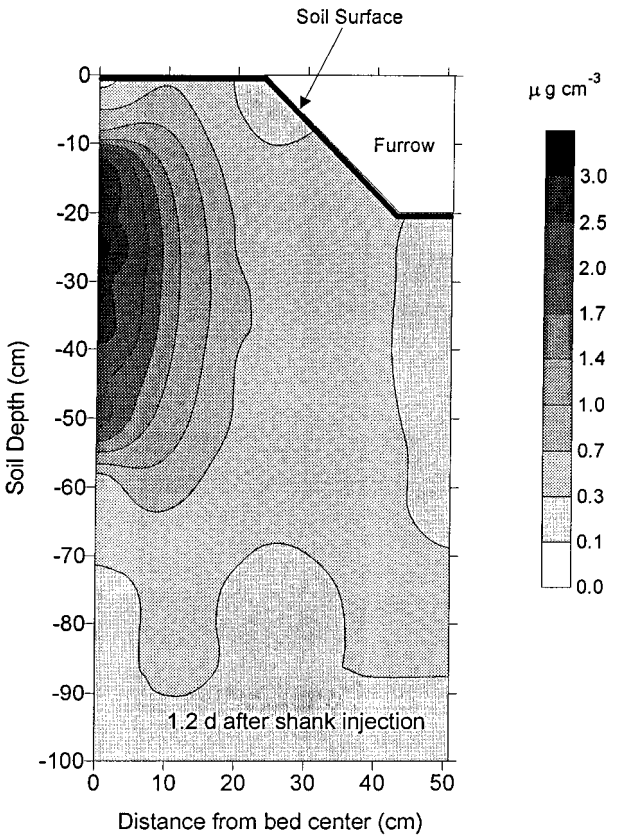

Figure 3. 1,3-D concentration distribution at 1.2 days after application (112 kg ha^{-1}) with direct shank injection at 30.5 cm depth.

chemical was applied with either drip irrigation or direct shank injection at the 2.5 to 30 cm depth range.

3.2 Temporal distribution of 1,3-D in soil air

Directly below bed center, measured maximum 1,3-D concentration reached 8 μg cm^{-3} at 10 cm depth at 0.625 days (15 h) after the shallow drip application (Fig 4). At the soil surface, the concentration was about 0.4 μg cm^{-3} due to some effect of the plastic cover film or a high water content at the soil surface. The concentration decreased quickly to less than 1 μg cm^{-3} at 1.4 days after application, and no significant amount of 1,3-D penetrated below 50 cm. This is consistent with earlier observations on the spatial distributions in the treated beds since most of the applied 1,3-D was lost by emission during the first 2 days after application. With drip application at 20.3 cm, significant amounts of 1,3-D reached 76 cm depth and the concentration decreased slowly over time (Fig 5). The highest concentration was at about 25 cm depth where 1,3-D concentration remained at about 0.8 μg cm^{-3} at 3.32 days after application. The persistence is caused by the longer distance to the soil surface and possibly a lower 1,3-D degradation rate due to the low population of micro-organisms in the subsurface soil.[20] Because of the absence of surface cover, no significant 1,3-D was measured at the soil surface.

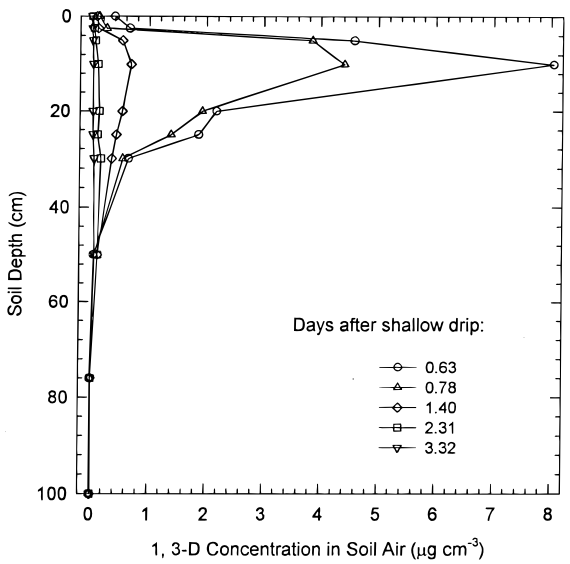

Figure 4. 1,3-D concentrations directly below bed center at selected times after application with drip irrigation at 2.5 cm depth.

In the shank-injection-treated beds, the top 60 cm soil was exposed to high 1,3-D concentrations for more than 3 days (Fig 6). The high concentration for a long duration was attributed to the increased application rate (112 kg ha^{-1}) and depth (30.5 cm), as compared to the two drip treatments. The slow dissipation may have been caused by the absence of transport by irrigation water as in the drip application. Diffusion was the only mechanism responsible for 1,3-D redistribution when non-formulated 1,3-D was applied with direct shank injection. No significant amount of 1,3-D was measured at the soil surface, due to the bare soil surface. Comparison of concentration distributions from the drip irrigation and the shank injection indicated that both methods of application would result in reasonably similar

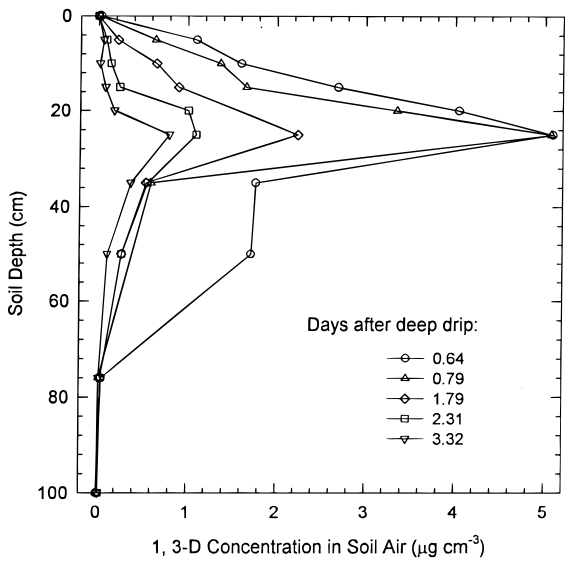

Figure 5. 1,3-D concentrations directly below bed center at selected times after application with drip irrigation at 20.3 cm depth.

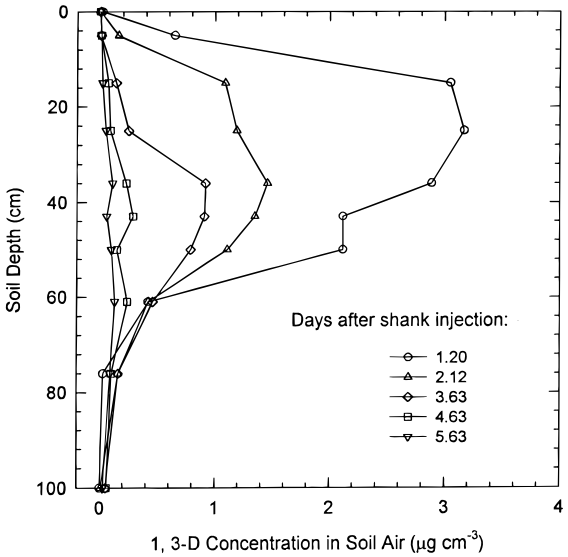

Figure 6. 1,3-D concentrations directly below bed center at selected times after application with shank injection at 30.5 cm depth.

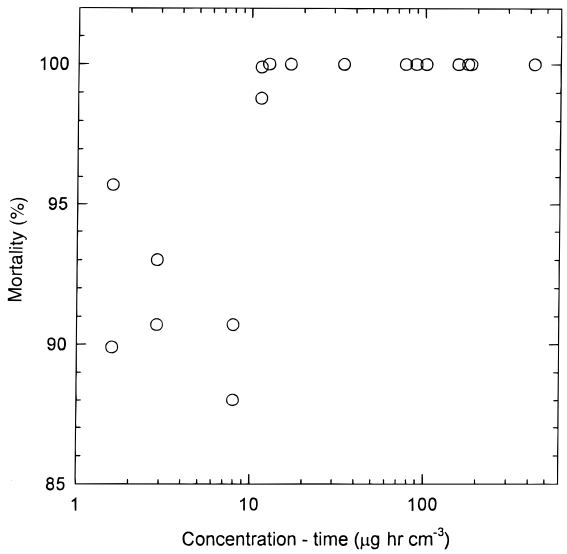

Figure 7. 1,3-D concentration–time index in relation to mortality of citrus nematodes (*Tylenchulus semipenetrans*).

spreading of 1,3-D gas in the treated beds. Corresponding pest control in the field beds may be sufficient regardless of the application method and the dosage may be as low as 47 kg ha^{-1} when applied with drip irrigation.

3.3 Concentration–time as an index of pest mortality

The integrated concentration–time (CT) index at locations where nematode samples were placed ranged from about 2 μg h cm^{-3} at shallow drip treatment to 425 μg h cm^{-3} in the shank injection plot. The lowest nematode mortality was 88%, found in the deep drip treatment where the CT at time of nematode removal was 8 μg h cm^{-3}. From the six replicated samples, measured nematode mortality (mean ± standard deviation) was respectively 97.4 (±3.7), 93.7 (±4.7), and 100 (±0)% for the shallow drip, deep drip, and shank injection treatment. A scatter plot of CT against nematode mortality from each individual sample location indicated that a near 100% efficacy was achieved from $CT \geq 12$ μg h cm^{-3} (Fig 7). This information is useful because it establishes a threshold CT value (ie 12 μg h cm^{-3}) for a particular chemical–pest combination. Although other environmental factors such as temperature may also contribute to pest mortality, direct toxicity from a pesticide such as 1,3-D would be the single most important factor for pest control.

To further explore the CT concept, concentrations of methyl bromide from a previous study[10] on a fungus (*Rhizoctonia solani*) were used to obtain the CT values to correlate to the mortality of *R. solani*. A plot of methyl bromide CT against *R. solani* mortality indicated that a threshold CT value of about 1600 μg h cm^{-3} is required to reach an effective control of *R. solani* (Fig 8). Because *R. Solani* is more difficult to control than *T. semipenetrans*, methyl bromide CT values of less than 1300 μg h cm^{-3} resulted in insufficient efficacy (<20% mortality). The approach of establishing a threshold CT value for a particular pesticide–pest combination provides a more quantitative determination of pesticide management. Following this framework, mathematical modeling may be used to determine the minimum effective application rate, reducing the risk of environmental contamination from excessive pesticide use.

4 CONCLUSIONS

Similar distributions of 1,3-D gas were obtained in field beds when the fumigant was applied with either drip irrigation or direct shank injection. Increasing application depth probably reduced the rate of 1,3-D dissipation into the atmosphere, resulting in higher CT or longer exposure time to potential soil-dwelling pests than application near the surface.

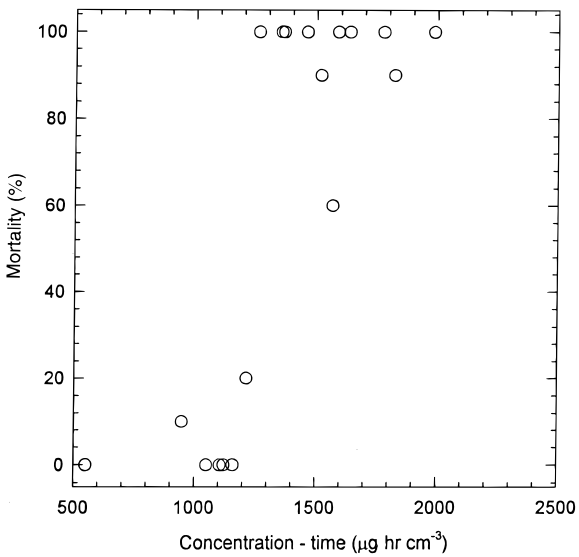

Figure 8. Methyl bromide (MeBr) concentration–time index in relation to mortality of fungus (*Rhizoctonia solani*).

Because of gravity effect on water movement, redistribution of 1,3-D was enhanced in the downward direction when applied with irrigation water. Applying 1,3-D with drip irrigation at 47 kg ha^{-1} produced significant concentration levels in the treated beds, whereas application with direct shank injection at 112 kg ha^{-1} extended the detectable concentration range to the whole soil profile, including furrows, and to depths below 1 m.

Effective control of *T. semipenetrans* was obtained in either the shallow drip, deep drip, or shank injection treatment at 25 cm depth, confirming the potential of reducing 1,3-D application dosage. To achieve a near 100% efficacy for *T. semipenetrans*, a threshold 1,3-D *CT* was found to be about 12 μg h cm^{-3}.

ACKNOWLEDGEMENTS

The authors thank R LeMert and P Zhang for analyzing 1,3-D samples in the laboratory, F Ernst for help in collecting 1,3-D gas samples in the field, and O Becker and J Darsow for analyzing pest control efficacy.

REFERENCES

1 Noling JW and Becker JO, *J Nematol* **26**:573–586 (1994).
2 Chen C, Green RE, Thomas DM and Knuteson JA, *Environ Sci Technol* **29**:1816–1821 (1995).
3 van den Berg F, *Pestic Sci* **36**:195–206 (1992).
4 van den Berg F, Roos AH, Tuinstra LGMTh and Leistra M, *Water Air Soil Pollut* **78**:247–264 (1994).
5 Szeto SY Grove G, Liebscher H, Hii B and Zebarth BJ, *J. Environ Qual* **23**:1367–1370 (1994).
6 Yagi K, Williams J, Wang NY and Cicerone RJ, *Science* (*Washington*) **267**:1979–1981 (1995).
7 Yates SR, Gan J, Ernst FF., Mutziger A and Yates MV, *J Environ Qual* **25**:192–202 (1996).
8 Majewski MS, McChesney MM, Woodrow IE, Prueger JH and Seiber JN, *J Environ Qual* **24**:742–752 (1995).
9 Yates SR, Wang D, Ernst FF and Gan J., *Environ Sci Technol* **31**:1136–1143 (1997).
10 Wang D, Yates SR, Ernst FF, Gan J, Gao F and Becker JO, *Environ Sci Technol* **31**:3017–3022 (1997).
11 Wang D, Yates SR, Ernst FF, Gan J and Jury WA *Environ Sci Technol* **31**:3686–3691 (1997).
12 Gamliel A, Grinstein A, Peretz Y, Klein L and Katan J, *Plant Disease* **81**:469–474 (1997).
13 Leistra M, Groen AE, Crum SJH and van der Pas LJT *Pestic Sci* **31**:197–207 (1991).
14 Kotcon JB and Loria R, *Plant Disease* **71**:1122–1124 (1987).
15 Bell CH, *Pestic Sci* **35**:255–264 (1992).
16 Su NY Osbrink WLA and Scheffrahn RH, *J Econ Entomol* **82**:156–158 (1989).
17 Schneider RC, Green RC, Wolt JD, Loh RKH, Schmitt DP and Sipes BS, *Pestic Sci* **43**:97–105 (1995).
18 Gan J, Yates SR, Spencer WF and Yates MV, *J Chromatogr* **684**:121–131 (1994).
19 Gan J, Yates SR, Wang D and Ernst FF, *J Environ Qual* **27**:432–438 (1998).
20 Smelt JH, Teunissen W, Crum SJH and Leistra M, *Netherlands J Agric Sci* **37**:173–183 (1989).