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Effect of Application Methods on 1,3-Dichloropropene Volatilization from Soil under Controlled Conditions

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ABSTRACT

Emissions of fumigants can be an important source of air pollution at soil fumigation sites, and the high emission rates result partly from the use of application methods that are high in volatilization potential. In this study, we compared volatilization of fumigant 1,3-dichloropropene (1,3-D) from a sandy loam in 60-cm packed columns after applied by different methods. The most volatilization was found with injection into uncovered soil at a shallow depth (e.g., 56% loss for the 20-cm injection) and application via surface drip irrigation (>90%). Volatilization fluxes and cumulative losses, however, rapidly decreased as the injection depth was increased, and the total loss was only 27% for the 40-cm injection. The commonly used polyethylene plastic was ineffective in reducing the volatilization because of its high permeability to 1,3-D. Water application after the 20-cm injection resulted in substantially reduced volatilization, but the least loss (22%) was obtained when an emulsifiable formulation of 1,3-D, Telone SL, was drip-applied to the subsurface at 20 cm. Our results indicate that variables most influencing 1,3-D volatilization are injection depth and water management. By optimizing these variables, application methods with low emission potential can be developed.

SOILBORNE plant pathogens and parasitic nematodes can cause extensive damage to many crops, especially in intensive agriculture. Over the last few decades, soil disinfestation has been primarily through the use of fumigants (Noling and Becker, 1994; UNEP, 1995). As limited by their properties, however, many fumigants have had undesirable effects on the environment or humans, and the use of many has been, or will soon be, suspended. Recently, emissions of methyl bromide (MeBr) were implicated in stratospheric ozone depletion, and the production and importation of this most important fumigant are expected to be discontinued in the U.S. by 2001 (USEPA, 1993). Though nonchemical approaches are being explored as alternatives to MeBr, few of them appear to be widely applicable at this time (Noling and Becker, 1994; UNEP, 1995). Thus, preserving the few remaining fumigants by minimizing their negative environmental effects is of immediate importance.

The fumigant 1,3-dichloropropene (1,3-D) is considered as one of the most viable alternatives to MeBr (Noling and Becker, 1994). The high vapor pressure of 1,3-D, however, suggests that a great fraction of the applied chemical may volatilize into the air following soil applications. Model simulations and laboratory experiments estimated that 2 to 77% volatilization loss would occur after subsurface injection (McKenry and Thomason, 1974; Leistra and Frissel, 1975; Basile et al., 1986; Chen et al., 1995). High 1,3-D concentrations were found in the air near fumigated areas in 1990, which prompted a suspension of the use of 1,3-D in California between 1990 and 1994 (CDFA, 1990). High 1,3-D air concentrations were also reported in a few other studies (Albrecht, 1987; Brouwer et al., 1991; van Welie et al., 1991). Although intrinsically caused by its properties, the emission of 1,3-D is clearly also attributable to the utilization of efficacy-oriented application methods that are high in volatilization potential.

A variety of 1,3-D application methods have evolved over the past few decades. The most commonly used method is chisel injection at various depths into uncovered fields (Lembright, 1990). Though injection at >45 cm is now required in California, shallower injections are still used in other areas. To enhance efficacy, injection into plastic sheet covered soil, application via drip irrigation systems, and irrigation after injection (water sealing), have also been used (McKenry and Thomason, 1974; Albrecht, 1987; Lembright, 1990; Schneider et al., 1993, 1995a,b, 1997; Sipes and Schmitt, 1996; Becker and Mueller, 1997). Although 1,3-D concentrations in ambient air from fumigated fields were measured in a few recent studies (van den Berg, 1992; van den Berg et al., 1994; Baker et al., 1996), the influence of application methods on 1,3-D volatilization has not been adequately evaluated. Schneider et al. (1993, 1995a,b, 1997) monitored 1,3-D air concentrations above pineapple [*Ananas comosus* (L.) Merr.] fields in Hawaii after applying Telone by chisel injection, drip irrigation and chisel injection followed by plastic mulching, and noticed different concentration profiles among the treatments. Under field conditions, because large field sizes and buffer zones are required for measuring volatilization, it is difficult to simultaneously compare more than two treatments. Because many soil and climatic parameters may vary spatially or temporally, the effect of application method is difficult to infer from separately conducted field studies.

The primary objective of this study was to determine the effect of application method on 1,3-D volatilization from soil under controlled conditions. Variables considered were injection depth, surface covering, and water-aided application. Distribution of 1,3-D in soil was also monitored to predict the possible effect of application methods on the efficacy.

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Abbreviations: 1,3-D, 1,3-dichloropropene; HDPE, high density polyethylene.

MATERIALS AND METHODS

Chemicals and Soil

Two commercial formulations of 1,3-D, Telone II [0.63 g mL^{-1} (Z)-1,3-D and 0.54 g mL^{-1} (E)-1,3-D] and Telone SL [0.40 g mL^{-1} (Z)-1,3-D and 0.40 g mL^{-1} (E)-1,3-D], were provided by DowElanco (Indianapolis, IN). Telone II is the commonly used formulation for shank injection, while Telone SL is an emulsifiable formulation designed for drip application. 1,3-D standard [48% (Z)-1,3-D and 50% (E)-1,3-D] was purchased from Chem Service (Bellefonte, PA). The Greenfield sandy loam (coarse-loamy, mixed, thermic, Typic Haploxeralf) used in this study was taken from the 0- to 30-cm depths in a field at the University of California's Moreno Valley Agricultural Experimental Station. The organic matter content of the soil was 0.92%, and pH was 7.4. Before use, moist soil was passed through a 2-mm sieve, and the water content was adjusted to 12% (wt./wt.).

Enclosed Column System

The closed, packed soil column system used in this study has been described in detail elsewhere (Gan et al., 1996, 1997). This system, when combined with model extrapolation to correct for the effect of the closed lower boundary, has given reproducible estimates for MeBr volatilization losses (Gan et al., 1996, 1997). The columns were prepared by uniformly packing the Greenfield sandy loam into 60 cm high by 12.5 cm (i.d.), bottom-sealed glass columns to 1.60 g cm^{-3} bulk density, 0.19 volumetric water content, and 0.21 volumetric air content. A sampling chamber [4 cm high by 12.5 cm (i.d.), top sealed] with an inlet and an outlet was then sealed onto the top of the soil column. A constant air flow of 150 mL min^{-1} was established by connecting the outlet to a stable vacuum source. Sampling ports were installed at 10-cm increments along the column by inserting half-hole Thermogreen septa (Supelco Co., Bellefonte, PA) into 4-mm holes through the column wall. These sampling ports allowed the injection of fumigant at a selected depth, and the sampling of soil air for monitoring fumigant diffusion in the column. In such a system, it can be assumed that dissipation of 1,3-D from the column is caused only by degradation and volatilization.

Injection Depth

In the first experiment, volatilization of 1,3-D was measured from soil columns after 100 μL Telone II was injected at the 20-, 30-, or 40-cm depth. The soil surface was not covered, and two replicates were used for each treatment. The application rate was equivalent to 120 mg per column, or 98 kg ha^{-1}. Immediately after the application, sampling tubes containing 600 mg activated carbon (ORBO-32, Supelco) were placed between the outlet and the vacuum source to collect volatilized 1,3-D from each column. The sampling tubes were replaced every 2 h for the first 72 h and every 6 h thereafter. To analyze 1,3-D in the carbon sample tubes, the carbon granules were first transferred into 9-mL headspace vials, and after 1.0 mL benzyl alcohol was added, the headspace was analyzed on a Tekmar 7000 Headspace Autosampler (Tekmar Co., Cincinnati, OH) in tandem with an HP5890 GC equipped with an ECD (Hewlett Packard, Wilmington, DE) (Gan et al., 1994). Sampling was continued until 1,3-D level in the sample tubes fell below the detection limit (0.1 μg per tube). Volatilization fluxes in μg h^{-1} were obtained by dividing the recovered 1,3-D by the sampling interval, and cumulative volatilization losses was calculated by integrating volatilization fluxes over sampling time.

To follow the distribution of 1,3-D in soil, 0.5 mL of soil air was periodically withdrawn from the middle of the column through the sampling ports at various depths and transferred into headspace vials. The concentration of 1,3-D isomers in the vials was analyzed on the headspace-GC system.

As the column bottom was closed, downward diffusion of 1,3-D was stopped at 60 cm from the surface, which resulted in overestimated volatilization losses. The overestimation was greater for deep injections than for shallow injections. In our previous studies on MeBr, a model extrapolation procedure was developed that makes use of the measured soil air concentration profiles and predicts volatilization losses for the scenario of no lower boundary (Gan et al., 1996, 1997). The same approach was used in this study to extrapolate the measured volatilization losses to the more field-like infinite lower boundary condition.

Surface Cover

To determine the effect of surface cover on 1,3-D volatilization, the soil surface of two columns was covered with 1.4-mil (0.035-mm) high density polyethylene (HDPE) film (TriCal Co., Hollister, CA). In the covered columns, the fumigant had to diffuse through the plastic to enter the sampling chamber. Injection of Telone II (100 μL) was made at the 20-cm depth. Similar procedures were used for determining volatilization fluxes and soil air concentrations. Correction for the lower boundary effect was made by using the aforementioned method.

Application with Water

Volatilization of 1,3-D was also monitored in treatments that integrated 1,3-D application with *water sealing* or drip application. To simulate water sealing, after 100 μL Telone II was injected at 20 cm into an uncovered column, 184 mL (or 15 mm) of water was dripped onto the soil surface at 2 mL min^{-1}. To simulate drip application, Telone SL was first dissolved in water at 500 mg L^{-1}, and then 240 mL of the solution was delivered at 2 mL min^{-1} into an uncovered column either on the surface (surface drip) or at the 20-cm depth (subsurface drip) by using a syringe pump. The total delivery time was 2 h, and fumigant application was similar to that used in the other treatments. Delivery of fumigant solution or water was made through Teflon tubing preinstalled at a proper position, and volatilization was measured during and after the application. During water or solution application, sampling ports at the lower section of the columns were temporarily removed to prevent air from being trapped below the wet front, and they were reinstalled at the end of application. Since little 1,3-D was found to reach the lower boundary of the column, no model extrapolation was made to correct for the measured losses in these treatments.

RESULTS AND DISCUSSION

Effect of Injection Depth

Volatilization fluxes of (Z)-1,3-D for injection at different depths are shown in Fig. 1. Similar volatilization fluxes and cumulative losses were obtained for the same treatment repeated under the same conditions (data not shown). Volatilization of 1,3-D was rapid after subsoil injection, especially for applications at shallow depths (Fig. 1). For instance, after the 20-cm injection, the total loss measured over a 20-d period was 64% for (Z)-1,3-D and 60% for (E)-1,3-D (Table 1), and about

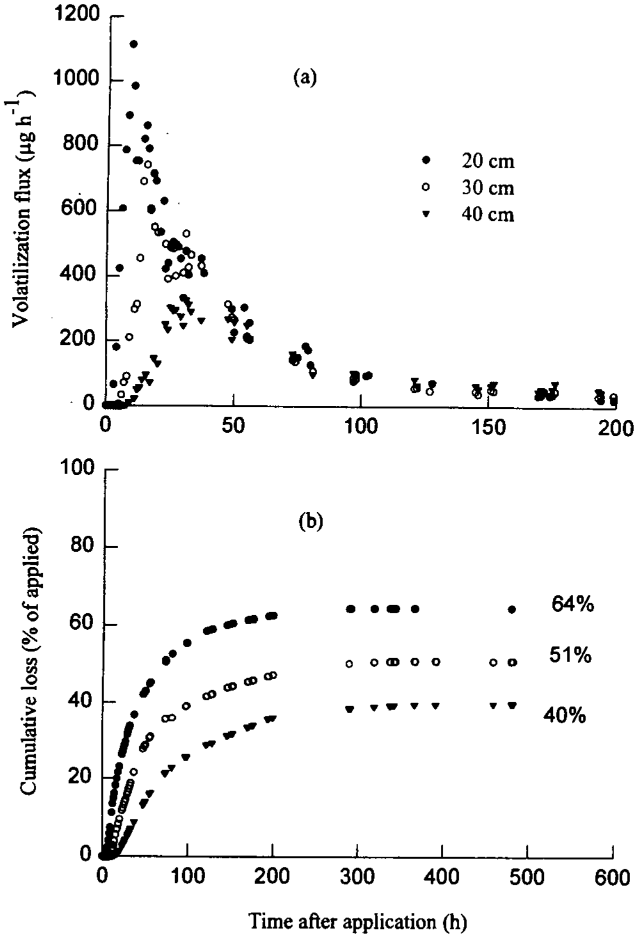

Fig. 1. Volatilization of (*Z*) 1,3-dichloropropene from uncovered soil columns after injection at different depths. (*a*) Volatilization fluxes in µg h^{-1} for the first 200 h; and (*b*) Cumulative losses calculated as % of applied.

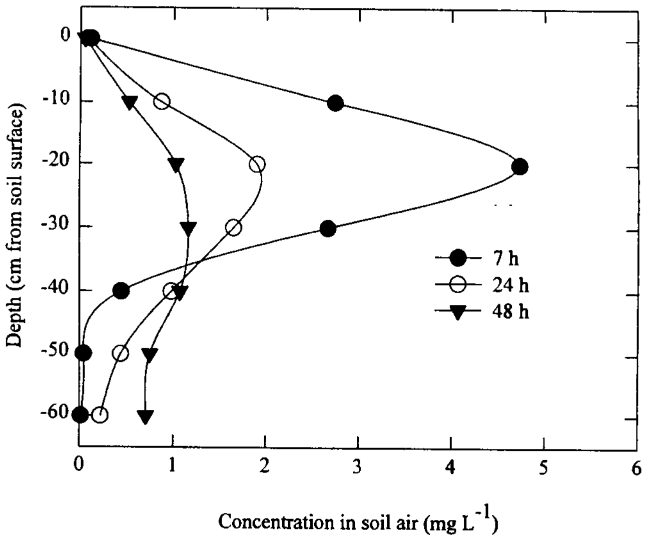

Fig. 2. Concentrations of (*Z*) 1,3-dichloropropene in soil air measured at different time intervals after injection at 20 cm in an uncovered soil column.

80% of the loss occurred during the first 4 d. The rapid volatilization may be attributed to the rapid diffusion and comparatively slow degradation of 1,3-D in the soil. The rapid transport of 1,3-D was seen from the change of its distribution pattern in the soil with time (Fig. 2). For instance, following the 20-cm injection, 1,3-D concentration at the application point (20 cm) was 10 times greater than that at 40 cm 7 h after the treatment, but the difference diminished quickly with time, and the two concentrations became equal at 48 h. The first-order half-life of 1,3-D degradation in the Greenfield sandy loam (20°C and 12% moisture content) was measured to be 5 d in a preliminary experiment. In contrast to the rapid diffusion, this degradation rate, though greater than that reported for many other xenobiotics, may be too slow. The disparity between diffusion and degradation rates may be expected to result in high 1,3-D emissions for shallow applications in an uncovered soil.

Table 1. Measured and extrapolated† (in parenthesis) total volatilization losses of 1,3-D isomers under various application conditions (in the percentage of applied).

Treatment	(*Z*) 1,3-D	(*E*) 1,3-D	Average‡
		%	
20 cm (uncovered)	64 (58)	60 (55)	62 (56)
30 cm (uncovered)	51 (43)	43 (37)	47 (40)
40 cm (uncovered)	40 (30)	31 (24)	36 (27)
20 cm (HDPE covered)	58 (52)	48 (44)	53 (48)
20 cm (irrigated)	35	34	34
Surface drip	90	97	92
Subsurface drip	24	20	22

† Volatilization losses extrapolated to infinite lower boundary condition by fitting measured 1,3-D concentrations in soil air into a vapor transport model.

‡ Loss averaged over the two isomers.

As the injection depth increased, the maximum fluxes became smaller, and the cumulative losses decreased (Fig. 1). The maximum detected flux was 1100, 740, and 330 µg h^{-1} for the 20-, 30-, and 40-cm injection, respectively. The measured total volatilization loss (averaged over the two isomers) was 36% for the 40-cm injection, which was 26% less than the 20-cm injection (Table 1). After correcting for the lower boundary effect, the calculated total loss from the 20-, 30-, and 40-cm injections was 56, 40, and 27%, respectively (Table 1), and the loss for the 40-cm injection was 29% less than that for the 20-cm injection.

The dependence of 1,3-D volatilization on injection depth supports the concept that deep injection is effective for minimizing fumigant volatilization (Reible, 1994; Schneider et al., 1995b; Jury et al., 1997). To reduce air pollution by 1,3-D, the amended regulations mandate that Telone II be applied at depths >45 cm in California (CDPR, 1994). Deep placement has also been found effective for reducing MeBr emissions (Gan et al., 1997; Yates et al., 1997b). In the field, only 21% of the applied MeBr was emitted following a 68-cm injection into bare soil, (Yates et al., 1997b), which was smaller than the loss reported in any other study that was conducted at a shallower depth (Yagi et al., 1993, 1995; Majewski et al., 1995; Yates et al., 1996a,b). The reduced volatilization following deep injections can be attributed to the greater distance and longer time that a chemical has to travel to reach the surface, which allows more degradation to occur (Reible, 1994; Jury et al., 1997). In practice, however, fumigant concentrations in the topsoil layers decrease as injection depth increases, which may lead to reduced effectiveness

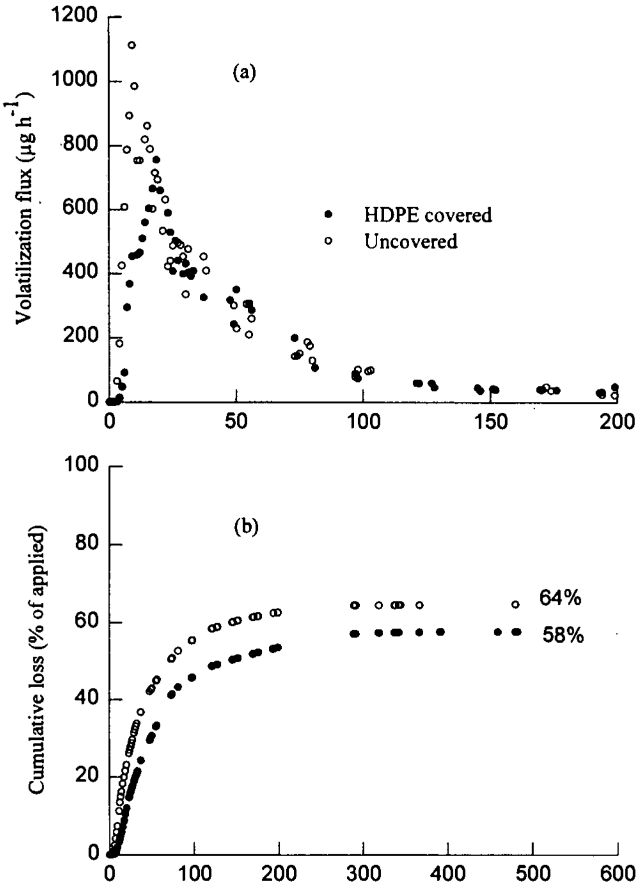

Fig. 3. Volatilization of (*Z*) 1,3-dichloropropene from polyethylene film-covered and uncovered soil columns after injection at 20 cm. (*a*) Volatilization fluxes in µg h^{-1} for the first 200 h after treatment; and (*b*) Cumulative losses calculated as % of applied.

against pests residing near the surface for deep injection applications (Sipes et al., 1993).

Volatilization of the two isomers of 1,3-D followed similar trends, but the loss of (*Z*)-1,3-D was consistently greater than that of (*E*)-1,3-D (Table 1). For instance, following the 30-cm injection, the maximum flux was 740 µg h^{-1} for the (*Z*) isomer, but only 325 µg h^{-1} for the (*E*) isomer; and the measured loss was 51% for the (*Z*) isomer, but only 43% for the (*E*) isomer. The differences may be explained by their different physical–chemical properties. In comparison to (*E*)-1,3-D, the (*Z*) isomer has a lower boiling point (104.1 vs. 112. 6°C) and solubility (2180 vs. 2320 mg L^{-1} at 25°C), but a higher vapor pressure (5700 vs. 4500 Pa at 25°C) and Henry's Law constant (0.0018 vs. 0.0011 m^3 atm mol^{-1} at 25°C). Similar differences were also observed by others under field conditions (van den Berg, 1992; van den Berg et al., 1994).

Effect of Surface Cover

Compared with the uncovered, 20-cm injection, the maximum fluxes and cumulative losses were only slightly smaller from the polyethylene film covered columns (Fig. 3 and Table 1). After correcting for the lower boundary effect, the total loss from the covered columns was only 8% less than that from the uncovered columns (Table 1). Covering fumigated fields with polyethylene sheets is a common procedure in MeBr application (Lembright, 1990), and is sometimes used in 1,3-D treatment to achieve a better efficacy (Albrecht, 1987; Schneider et al., 1995b, 1997). Our results indicate that surface covering with the tested polyethylene plastic is not very effective for reducing 1,3-D volatilization losses. This is in agreement with recent field observations (Schneider et al., 1995b), where the near-surface air concentrations were found to be similar between uncovered and polyethylene-covered treatments, and among plots that were covered with narrow or wide stripes of plastic sheets. Numerous studies also revealed that high MeBr emissions occur from polyethylene film-covered soil (Brown and Rolston, 1980; Yagi et al., 1993, 1995; Jin and Jury, 1995; Majewski et al., 1995; Yates et al., 1996a,b; Gan et al., 1997). The significant volatilization loss from covered soil is caused by the high permeability of fumigants through polyethylene films. Methyl bromide penetrated through polyethylene plastic at 7.4 mg m^{-2} h^{-1} under a 1000 mg L^{-1} concentration gradient, while 1,3-D isomers, at 75 mg m^{-2} h^{-1} under the same gradient, permeates even more rapidly through the same film than MeBr (Yates et al., 1997a).

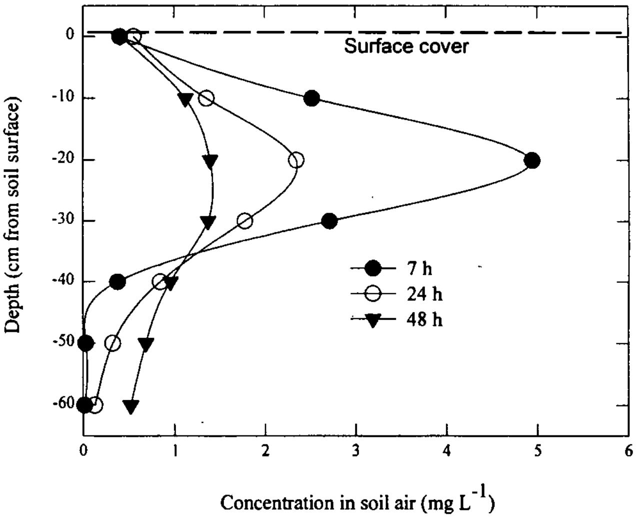

Fig. 4. Concentrations of (*Z*) 1,3-dichloropropene in soil air measured at different time intervals after injection at 20 cm in a polyethylene film-covered soil column.

The high permeability of 1,3-D isomers through the plastic was reflected in the very small build-up of 1,3-D concentration under the surface cover (Fig. 4). In the covered columns, 1,3-D distribution patterns were similar to those in the uncovered columns (Fig. 2). These results imply that unless some other mechanisms are operating (e.g., surface covering may cause soil solarization and alter soil water movement), any efficacy increase that may be derived from the use of a polyethylene surface cover will likely be small.

Integration of 1,3-D Application with Water Management

Applying 15 mm water as a water seal to the soil surface immediately after injecting 1,3-D at the 20-cm depth resulted in substantially reduced volatilization fluxes and cumulative losses (Fig. 5) as compared with the nonirrigated application at the same depth (Fig. 1, Table 1). The maximum fluxes of (Z)-1,3-D decreased from 1100 μg h^{-1} for the nonirrigated treatment to only 220 μg h^{-1} for the irrigated treatment (Fig. 5). The measured total loss of 34% from the water seal treatment was 28% less than that for the nonirrigated, 20-cm injection (Table 1).

Much higher volatilization of 1,3-D was observed during the simulated surface drip application of Telone SL. The maximum fluxes detected during the 2-h application, at 1.7×10^4 μg h^{-1} for (Z)-1,3-D and 1.3×10^4 μg h^{-1} for (E)-1,3-D, were >15 times that from the 20-cm injected, uncovered application, and 50 times that from the 40-cm application (Fig. 5). By the end of the 2-h application, 50% of the applied dosage had been lost via volatilization. In the next 24 h, an additional 25% of (Z)-1,3-D and 32% of (E)-1,3-D was emitted. Instantaneous volatilization losses following surface drip application indicate that little 1,3-D actually entered the soil; and this should lead to poor control over the pests residing in the subsurface soil layers. As the wind speed across an uncovered field may be many times greater than that in the used column system, even faster volatilization may occur under field conditions. Near-surface air concentrations of 1,3-D were measured after Telone SL was applied into polyethylene film-covered plots via drip lines buried at 0- to 5-cm depths (Schneider et al., 1997). Even though additional water was immediately applied to leach the applied fumigant into the soil, the initial concentrations were still substantially greater than those from an injection treatment.

In contrast to the surface drip treatment, volatilization of 1,3-D following a simulated subsurface drip application at the 20-cm depth was much lower (Fig. 5), and the total loss, at 24% for (Z)-1,3-D and 20% for (E)-1,3-D, was the smallest among all treatments (Fig. 5 and Table 1). The maximum flux was <100 μg h^{-1} for both isomers, and occurred nearly 3 d after the application (Fig. 5a). The overall volatilization was much more gradual than for the other treatments, with about 40 and 25% of the total loss occurring during the first 4 d for the (Z) and (E) isomer, respectively.

In the water seal treatment, distribution of 1,3-D (Fig. 6a) was more limited to the application depth (20 cm) compared with that in the nonirrigated columns (Fig. 2), and low 1,3-D concentrations were detected at the very shallow depths (2 and 5 cm) (Fig. 6a). Movement of 1,3-D in soil was even more restricted after subsurface drip application, and the chemical remained near the application point (20 cm) for a prolonged time (Fig. 6b). The retarded movement of 1,3-D in these treatments was apparently a result of the reduced vapor diffusion caused by the increased water content or decreased air

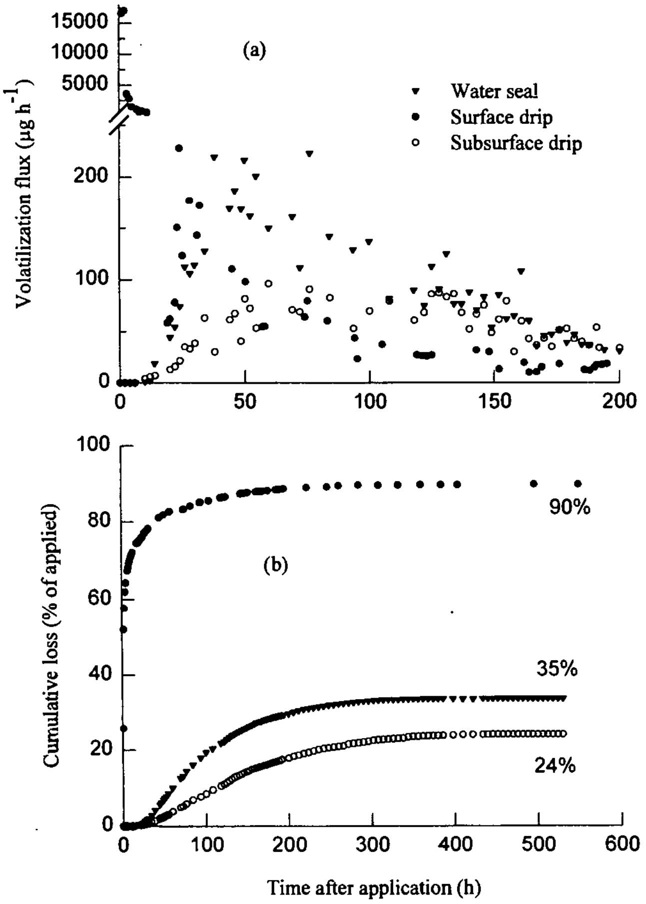

Fig. 5. Volatilization of (Z) 1,3-dichloropropene from uncovered soil columns after water-aided applications. (*a*) Volatilization fluxes in µg h^{-1} for the first 200 h after treatment; and (*b*) Cumulative losses calculated as % of applied.

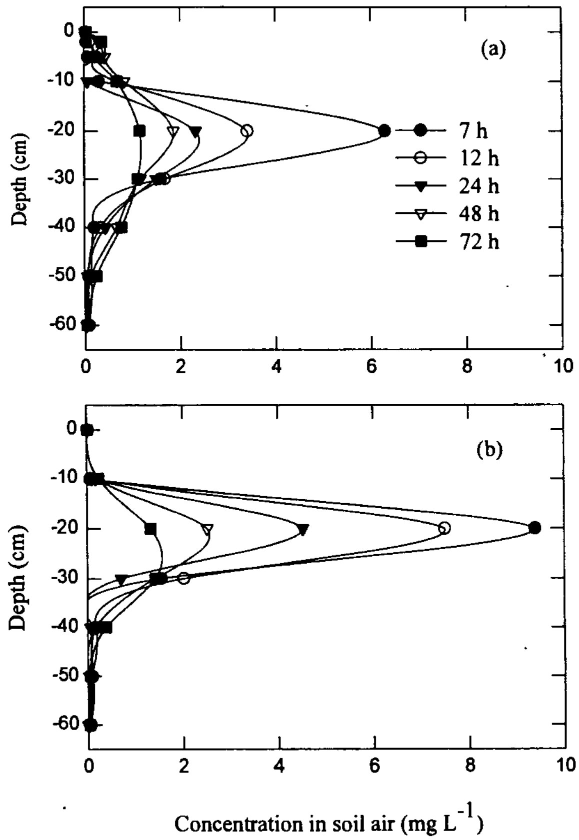

Fig. 6. Concentrations of (Z) 1,3-dichloropropene in soil air measured at different time intervals after water-aided fumigant application (*a*) 15 mm surface irrigation after the 20-cm injection (Telone II); and (*b*) Subsurface drip application at 20 cm (Telone SL).

porosity in the soil. The focused distribution suggests that if the point of application is inside the infested zone, subsurface drip application may produce better efficacy due to the increased concentration–time product. For the same reason, however, subsurface drip application may provide inadequate control over pests residing near the soil surface or in the deep layers below the drip lines.

CONCLUSIONS

The results from this study clearly show that application methods can have a significant effect on 1,3-D volatilization. Of the tested variables, injection depth and soil water regime were the most influencing. The most volatilization occurred after application by shallow injections or through surface drip irrigation in uncovered soil. The use of polyethylene film as a surface cover was ineffective in reducing 1,3-D emissions, and the rapid volatilization losses were due to the high permeability of the fumigant through the plastic membrane. Increasing injection depth, however, was found to be highly effective in reducing volatilization losses, and the total loss from a 40-cm injection was only about 50% of that from a 20-cm injection. Emissions were greatly reduced by combining subsoil injection with surface irrigation, and by applying an emulsifiable formulation through a subsurface drip irrigation system.

Some processes that may affect 1,3-D emissions under field conditions, however, were not considered in the current study. These include chisel traces created by injection, and soil temperature gradients and diurnal variations, among some others. Field studies should therefore be conducted to measure 1,3-D emissions and evaluate its efficacy using the low-emission methods identified in this study.

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