

# Minimizing Emissions from Soil Fumigants

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## **Interpretive Summary:**

Reducing emissions from soil fumigation becomes mandatory due more stringent regulations for improvement of air quality. This research tested various potential surface seal and soil treatments to minimize emissions of 1,3-dichloropropene (1,3-D) and chloropicrin (CP) from shank-injection of Telone C35 (61% 1,3-D and 35% CP) in a field trial conducted in October 2006 at Parlier in the San Joaquin Valley of California. Treatments included non-irrigated or treated control, pre-irrigation (irrigation with sprinklers four days prior to fumigation), post-fumigation water seals (applied through irrigation with sprinklers following fumigant application) with or without chemical amendment as potassium thiosulfate (KTS), and combinations of standard high density polyethylene (HDPE) tarp and amended surface soil with either composted manure or KTS. Air emissions and concentrations of fumigants in soil gas were monitored for two weeks. The intermittent KTS applications following fumigation is much more effective to reduce fumigant emissions compared to the intermittent water seals but the post fumigation treatments are not favorable regarding to workers' safety due to potential exposure risks to fumigants. The pre-irrigation was more effective to reduce emissions than the intermittent water seals although less effective than the KTS treatments. Fumigant distribution in soil gas was not affected in the pre-irrigated soils indicating fumigation efficacy might not be affected. The pre-irrigation offers a favorable option in field practice because of its easy management and no worker exposure risks to fumigants involved. The application of composted manure to soil followed by HDPE tarp after fumigation resulted in equal or even higher emissions than the control, which was suspected by the high soil temperature under that tarp causing fumigant desorption and subsequently high emission rates during day time. Further study is needed for better understanding on using organic amendment to minimize emissions from soil fumigation.

## **Study Background**

Telone or 1,3-dichloropropene (1,3-D) and chloropicrin (CP) have been increasingly used as alternative soil fumigants to methyl bromide, which has been phased out because of its contribution to ozone depletion in the stratosphere. These alternative fumigants, however, are volatile organic compounds (VOCs) and their emissions from soil fumigation may result in formation of ground-level ozone harmful to humans and the environment (Segawa, 2005). More stringent regulations are under development in California to improve air quality especially in nonattainment areas (CDPR, 2007).

Research has identified various methods including plastic tarp, water seals, and soil amendment with organic materials or chemicals to minimize fumigant emissions from soil fumigation. The most commonly adopted practice, i.e., applying standard high density polyethylene (HDPE) tarp over the soil surface after fumigation, has been found to not effectively control 1,3-D emissions (Wang et al., 1999; Papiernik and Yates, 2002) unless the soil was pre-irrigated (Gao and Trout, 2007). Water seals (irrigation with sprinklers following fumigation) have been tested for effectiveness to minimize methyl isothiocyanate (MITC) and Telone product emissions (Sullivan et al., 2004; Gao and Trout, 2007). Treatment of surface soils with chemicals such as ammonium or potassium thiosulfate (ATS or KTS) that can react with fumigants to form nonvolatile compounds (Gan et al., 1998a; Zheng et al., 2006) or organic materials that adsorb or enhance degradation of fumigants in soils have been found to effectively reduce fumigant emissions (Gan et al., 1998b; Dungan et al., 2001; Xu et al., 2003; McDonald et al., 2006). A moist soil profile or irrigation prior to fumigation were also found to effectively minimize fumigant emissions (Gan et al., 1998c; Thomas et al., 2003; Gao and Trout, 2007; Gao et al., in press). Among all the options to minimize emissions, economic, effective, and environmentally safe methods for field application have not been clearly defined. This research is designed to test and compare several surface seal and soil treatment methods in field conditions.

### **Objectives:**

Our research goal is to develop feasible, i.e., efficient, cost-effective, and environmentally safe field management practices to minimize fumigant emissions from soil fumigation. The specific objective of this study was to determine the effectiveness of several surface seals and soil treatments methods including pre-irrigation, post-fumigation water seals with or without chemical amendment as KTS, and soil amendment with either KTS or composted manure in combination with standard plastic tarp.

### **Materials and Methods:**

A field trial was conducted in October 2006 at the USDA-ARS San Joaquin Valley Agricultural Sciences Center, Parlier, CA. The soil is a Hanford sandy loam (coarse-loamy, mixed, superactive, nonacid, thermic Typic Xerorthents) and properties of the soil were reported in earlier studies (Gao and Trout, 2007). During the field trial, the maximum and minimum air temperature ranged from 13–27 and 3–9°C, respectively.

#### **Soil preparation, fumigation and treatments**

A field strip (150 m long and 9 m wide) was prepared for applying fumigants. Treatments were applied perpendicular to the field length. The soil was cultivated to 76 cm depth two weeks prior to fumigation. Because the surface 15 cm of soil was dry, the field was pre-irrigated with hand-move sprinklers. Irrigation was stopped when the wetting front reached about 10 cm depth. The soil profile moisture condition was determined prior to fumigation (Fig. 6).

Half of the strip area (150 m long and 4.5 m wide) was fumigated on Oct. 17 by shank injection of Telone C35 to a depth of 45 cm below soil surface. The other half was not fumigated and served as a comparison to the fumigated area for efficacy studies. The fumigation was applied by TriCal using a Telone rig with 51 cm (20") spacing between the 8 shanks. Fumigation was at 0900 h and completed in one pass across the field. The total amount of Telone C35 applied was 26 kg (58 lbs) from weighing the fumigant tank before and after application. This is equivalent to 499 kg ha<sup>-1</sup> (445 lb ac<sup>-1</sup>), which is 82% of the targeted rate of 606 kg/ha (540 lb ac<sup>-1</sup>). Following fumigation, the field surface was cultivated by a spring tooth harrow followed by a ring roller in a one pass operation to close any shank traces and pack the soil surface. A similar operation was done for non-fumigated area the day before fumigation.

Six surface seal or soil treatments were applied in three blocks where treatments were randomized in each block. These treatments included irrigation prior to fumigation, water seals with or without KTS after fumigation, and amendment of surface soils with KTS or composted steer manure covered by HDPE tarp. Previous research indicated these treatments might reduce fumigant emissions. One of the main purposes in this field trial was to do side-by-side simultaneous comparisons of these treatments on emission reductions as well as on soil pest controls under field conditions. Efficacy against selected nematodes, pathogens and weeds were tested in this field trial but the data are not reported in this paper. The abbreviations and detailed information for these treatments are provided below:

1. Control (soil profile with ~30% field capacity [FC])
2. Manure + HDPE (composted steer manure incorporated to soil surface at 12,350 kg/ha (5 ton/ac), then covered by HDPE tarp)
3. KTS + HDPE (2:1 KTS/fumigant ratio, sprayed onto the soil surface in 1 mm water, then covered by HDPE tarp)
4. Pre-irrigation (applied 34 mm water with sprinklers 4 days prior to fumigation)
5. Intermittent water seals (applied 13 mm water with sprinklers to moisten the top 8 cm of soil to FC immediately following fumigation, plus an additional 4 mm at 12 h, 24 h, and 48 h)
6. Intermittent KTS applications (applied 2:1 KTS/fumigant ratio immediately following fumigation, and 1:1 KTS at 12, 24, and 48 h using the same amount of water with same application method as Treatment #5).

For Treatment 2 (Manure + HDPE), composted steer manure was spread over the soil surface at 5 tons/acre. The manure was incorporated into the surface soil using a tractor mounted rototiller. After fumigation was completed, the spring tooth harrow and a ring roller carried some surface soils away, additional amount (i.e., at 5 tons ac<sup>-1</sup> rate) of composted manure was spread over the soil surface but not incorporated. Then HDPE tarp was applied by TriCal.

For treatment 3 (KTS + HDPE), KTS was applied in 1 mm of water using a 3-m wide spray bar. The HDPE tarp was hand-applied to avoid the compaction of the wet surface soils.

For Treatment 4 (pre-irrigation), 34 mm water was applied four days prior to fumigation using 0.91 m x 0.91 m (30'x30') sprinkler irrigation system. This amount of water can wet the surface 20 cm soil layer to FC or was expected to achieve 60% of FC for top 30 cm soils by the time of fumigation. The soil moisture data measured the day before fumigation are reported in Figure 6.

For Treatment 5 (intermittent water seals), initial water seal following fumigation was applied to each plot using the same sprinkler-irrigation system as in the pre-irrigation treatment. A total 13 mm of water was applied and intended to moisten the top 8 cm of soil to FC. This amount of water was applied in about 2 h immediately following fumigation. Subsequent water seals at 12, 24, and 48 h, 4 mm of water were applied in about 0.5 h each time.

For treatment 6 (intermittent KTS applications), the application schedule and the amount of water used in delivering KTS to soil surface were the same as Treatment 5. At the initial application following fumigation, a 2:1 KTS/fumigant (w/w) ratio was used and the KTS solution was applied in the last 30 min of the irrigation. For the subsequent applications, i.e., at 12, 24 and 48 h, KTS was dissolved in 4 mm water and applied to soil surface in about 25 min.

## **Sampling and Measurement**

Sampling for air emissions and soil gas phase distribution of applied fumigants was conducted for about two weeks following fumigant application. Soil samples were taken at the end of the sampling period for residual fumigants in the soil and soil water content. For the control and pre-irrigated plots, soil water content was also determined on the day before fumigation and 4 days after fumigant application. Soil temperature at 10-cm depth was measured during one day of the monitoring period.

Sampling, sample processing and analysis followed similar procedures previously reported in Gao and Trout (2007) with some modifications indicated in the following. Briefly, emissions were measured using static (passive) flux chambers. The chambers were placed on soil or tarp for 15 min and then a 100-mL gas sample from inside the chamber was withdrawn using a gas-tight syringe through a sampling port at a flow rate  $\sim 100 \text{ mL min}^{-1}$ , and through an ORBO™ 613, XAD 4 80/40mg (Supelco, Bellefonte, PA) tube for trapping both 1,3-D and CP. The sampling tubes were immediately capped, stored on dry-ice in the field and transferred into an ultra-freezer ( $-80^{\circ}\text{C}$ ) in the laboratory. The tubes were extracted for fumigant analysis using a gas chromatography with a micro electron capture detector (GC- $\mu$ ECD). Emission sampling was done every 3 h for the first 48 h and every 4 h thereafter during the day. No sampling was done at night after the first two nights (1700 to 0800 h). Average flux or emission rates were calculated during the 15-min capture time based on fumigant concentration measured and soil surface area the chamber covered. Cumulative emissions of 1,3-D and CP were estimated by summing the products of the average of two consecutive emission flux values and the time interval between the two measurements over the time span of the study. This field sampling procedure was designed primarily to measure relative emission differences between treatments and may not accurately quantify absolute emission losses.

Fumigant concentration in the soil-gas phase were sampled using stainless steel sample probes installed at depths of 10, 30, 50, 70, and 90 cm below the soil surface and at two locations in each plot (a, adjacent to shank or fumigant injections lines; b, between fumigant injections lines). Measurements were done in one replicate of the treatments at 6, 12, 24, 36, 48, 72, 120, 168, 240, and 312 h following fumigation. Processing of the sampling tubes was the same as the emission samples.

At the end of the field trial, soil samples were taken at 20-cm depth intervals to 100 cm to determine residual fumigants in the soil. Samples were collected with an auger, mixed immediately and a portion placed in a screw-top glass jar that was stored on dry ice in the field, and in a freezer ( $-80^{\circ}\text{C}$ ) in the laboratory until extractions. The samples were extracted with ethyl acetate and analyzed using a GC- $\mu$ ECD system.

## **Results and Discussion:**

### **Emission Rates**

Figure 1 shows emission rates (flux) measured from six treatments for both 1,3-D and CP. The control which was irrigated two wks before fumigation along with the whole field gave the earliest high emissions that also lasted for 24 h. The emission peaks ( $24 \mu\text{g m}^{-2} \text{s}^{-1}$  for 1,3-D and  $13 \mu\text{g m}^{-2} \text{s}^{-1}$  for CP) for the control were much lower than soils without this irrigation as observed in earlier studies ( $80 \mu\text{g m}^{-2} \text{s}^{-1}$  for 1,3-D and  $50 \mu\text{g m}^{-2} \text{s}^{-1}$  for CP) (Gao and Trout, 2007; Gao et al., in press). The amendment of manure plus HDPE tarp resulted in surprisingly high emission rates, especially during day time (up to  $34 \mu\text{g m}^{-2} \text{s}^{-1}$  for 1,3-D and  $15 \mu\text{g m}^{-2} \text{s}^{-1}$  for CP) and had large diurnal variations. This contradicts earlier findings, where manure application to the soil surface reduced emissions, and indicates that soil or manure conditions or application methods need to be better understood to achieve emission reductions. The KTS application plus HDPE tarp substantially reduced emissions (emission peaks were  $11 \mu\text{g m}^{-2} \text{s}^{-1}$  for 1,3-D and  $< 2 \mu\text{g m}^{-2} \text{s}^{-1}$  for CP). Pre-irrigation (irrigated 4 days prior to fumigation) had similar emission peaks for 1,3-D as the KTS+HDPE tarp treatment but a much higher CP emission peak ( $5 \mu\text{g m}^{-2} \text{s}^{-1}$ ). The intermittent water seals after fumigation resulted in low emissions for the first 48 h when water seals were applied but emissions were high from 48-192 h. The intermittent water seals with KTS addition resulted in very low emissions for 1,3-D for the first 4 days and for CP through the whole monitoring period. The results showed that KTS more effectively for CP than 1,3-D, which also observed in the KTS application plus tarp treatment.

In general, the fumigant emission showed a diurnal pattern: a higher emission occurred during day time and lower emission occurred during night. This pattern was distinct in the manure incorporated treatment with HDPE tarp. The relatively higher soil temperature under the tarp and temperature-dependent adsorption and desorption processes of fumigants on the organic materials may contribute to the phenomenon observed.

### **Cumulative Emissions**

The cumulative and total emission losses of 1,3-D and CP as a percent of applied fumigant over a 2-wk monitoring period are shown in Fig. 2 and Table 2, respectively. The cumulative emission losses corresponded to the flux measurements. The control resulted in the earliest and highest 1,3-D emission losses in the first few days. After a few days, however, emissions of 1,3-D from the HDPE tarp over manure amendment exceeded the control mainly due the high daytime emissions. This resulted in the highest 1,3-D total emission loss (32% of applied) from Manure + HDPE tarp treatment although the CP total loss (17%) was slightly lower than the control (20%). In the treatment of KTS application plus HDPE tarp, much lower emissions of 1,3-D and extremely low emissions of CP were measured. The intermittent water seals did not reduce 1,3-D total emissions, which was mainly due to the emission increase after water seals were stopped, and produced slightly lower CP emissions compared the control. Intermittent KTS applications with sprinklers resulted in the lowest total emissions for both 1,3-D and CP. The pre-irrigation treatment from this field test resulted in lower emissions for both 1,3-D and CP than the control, manure + HDPE, and intermittent water seal treatments but higher than the KTS treatments. Thus the KTS applications show the most effectiveness on emission reductions, especially for CP. It should be noted that there are risks of fumigant exposure to workers who enter the field to carry out water or chemical applications after fumigant application. This risk may be reduced to some extent if irrigation lines on the field can be placed before fumigation and removal of the equipment can be done until the field is safe for reentry. There are still some requirements of equipment installation around the field following fumigation and for even spacing between the sprinkler laterals for uniform fumigant

application throughout the fields etc. Therefore, the pre-irrigation to minimize emissions has an advantage over post fumigation treatments. This treatment has also been tested in previous trials (Gao and Trout, 2007; Gao et al., in press). It is unlikely to reduce efficacy because fumigant concentration and distributions were not reduced in soil gas (also see below).

### **Fumigants in Soil Air**

Figure 3 depicts the 1,3-D concentrations in the soil-gas phase in soil profiles. Higher concentrations were observed at locations adjacent to shank-injection lines (Location a, top 6 graphs) compared to locations between shank-injection lines (Location b, bottom 6 graphs). The highest fumigant concentration observed for each treatment was observed at 6 or 12 h after fumigant injection for Locations a and 12 or 24 h for Locations b. This reflects the time required for fumigant distribution through the soil. Excluding larger variations at earlier sampling times, the fumigant concentrations at 12 h for Locations a ranged from 19 to 23 mg L<sup>-1</sup> and for Location b from 9 to 14 mg L<sup>-1</sup>. The lower fumigant concentrations in soil air appeared from KTS + HDPE tarp treatment or the intermittent water seal treatments for Locations a, although there were small differences at Locations b. The pre-irrigation treatment did not show any reduced fumigant concentrations compared to other treatments indicating that the higher soil water content is unlikely to affect fumigation efficacy. After two weeks of fumigant application, there were still a small amount of fumigants detected in the soil gas at all soil depths for all treatments (up to 1.2 mg L<sup>-1</sup> for 1,3-D and 0.7 mg L<sup>-1</sup> for CP). Distribution patterns of CP were similar to 1,3-D with generally lower concentrations than 1,3-D (data not shown).

### **Residual Fumigant in Soil**

Residual 1,3-D and CP extracted from soil samples taken at the end of the field trial are shown in Fig. 4. The data represent fumigants in the solid and liquid phase after emission losses and degradation. For 1,3-D, most treatments showed concentrations below 2 mg kg<sup>-1</sup>. Higher concentrations were observed in upper soil layers. The CP concentrations (mostly below 0.2 mg kg<sup>-1</sup>) were too low to exhibit a pattern. The highest 1,3-D concentration was from the Manure + HDPE treatment near the soil surface (up to 6 mg kg<sup>-1</sup>). The results indicate that with manure amendment, the soil retention capacity for fumigants might have increased. When temperature was high and HDPE tarp permeability increased, fumigants may be subject to desorption and contribute to emissions which may explain the high emissions observed during the day time. The tarp treatment showed much higher temperature than bare soils (Fig. 5). The results indicate that the mechanisms or interaction between organic materials and fumigants need to be further studied in order to use organic material amendment to reduce fumigant emissions.

### **Soil Water Content**

Soil moisture condition varied among treatments and over time within a treatment, especially for those with irrigations. Figure 6 shows soil water content measured the day before fumigation, 4 days after fumigation and at the end of field trial for the control and pre-irrigation treatments. The control had a uniform soil water distribution the day before fumigation (8%, v/v) which is about 30% of the FC (26%). The pre-irrigated soil had much higher soil water content (21%) at soil surface on the day before fumigation (~ 80% of FC), which was reduced to about 50% of FC 4 days later. This indicates that water infiltration through the soil and evaporation from the soil surface was fairly quick and it is important to apply fumigants within a limited time frame after a pre-irrigation. By the end of the field trial (2 wks later), the surface soil water content with the pre-irrigation reduced to about 35-40% of FC. As indicated above, the fumigant concentration in the soil profile was not affected by the pre-irrigation treatment with the amount of water applied and the soil water content throughout

the profile substantially below FC except initially at the soil surface, which can effectively control 1,3-D and CP emissions from shank injected Telone C35.

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**Table 1.** Total emission loss of 1,3-dichloropropene (1,3-D) and chloropicrin (CP) measured over 2 weeks after fumigation. Values in parenthesis are standard deviations of three replicates.

| Treatment               | Total Emissions (% of applied) |        |
|-------------------------|--------------------------------|--------|
|                         | 1,3-D                          | CP     |
| Control                 | 24 (3)                         | 20 (1) |
| Manure + HDPE           | 32 (7)                         | 17 (4) |
| KTS + HDPE              | 18 (4)                         | 3 (1)  |
| Pre-irrigation          | 19 (2)                         | 9 (5)  |
| Intermittent water seal | 24 (3)                         | 14 (3) |
| Intermittent KTS appl.  | 13 (3)                         | 3 (1)  |

Manure, composted steer manure; HDPE, high density polyethylene; KTS, potassium thiosulfate.



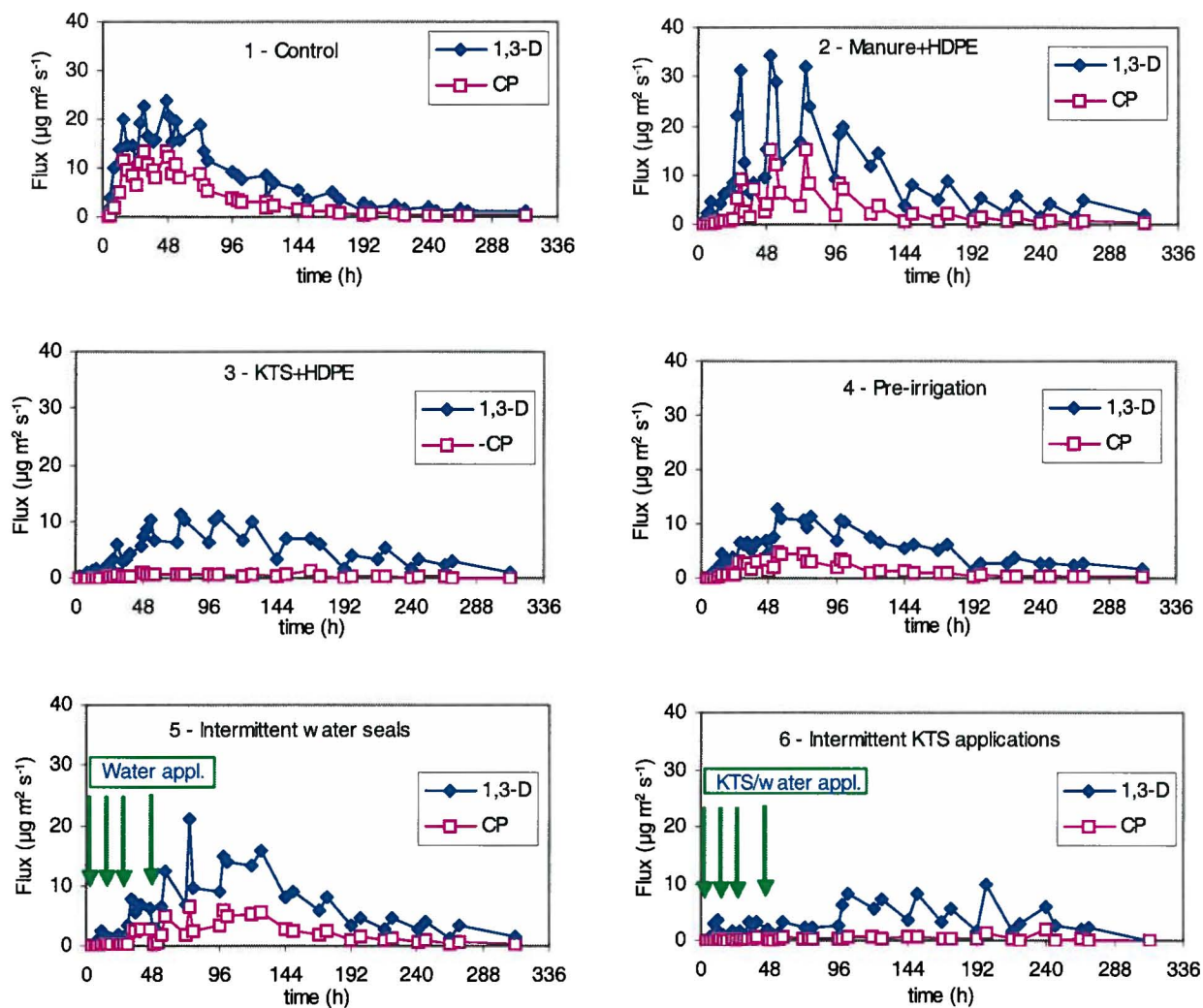


Figure 1. Effects of surface seal and soil treatments on emission flux of 1,3-dichloropropene (1,3-D) and chloropicrin (CP) from shank-injection of Telone C35. Plotted are averages of three replicates. Manure, composted steer manure; KTS, potassium thiosulfate.

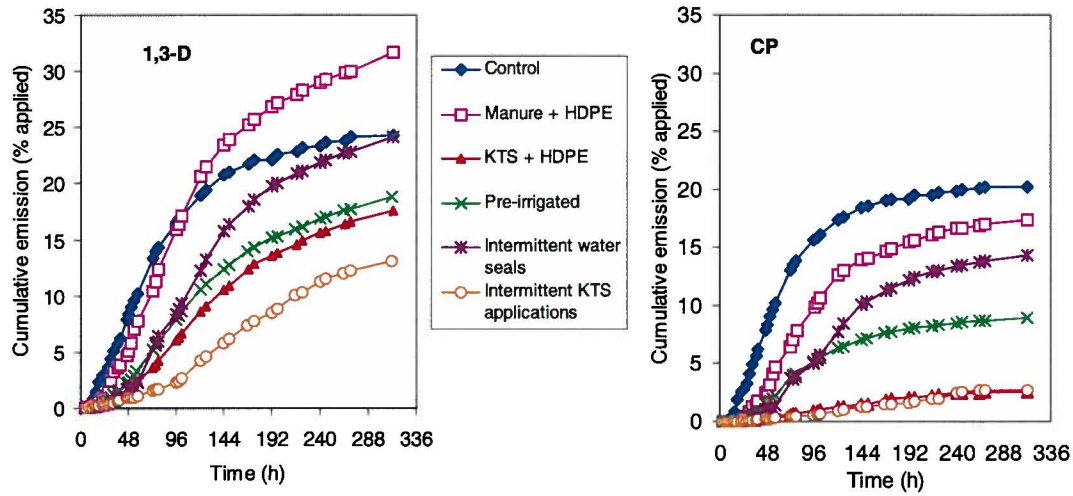


Figure 2. Cumulative emission losses of 1,3-dichloropropene (1,3-D) and chloropicrin (CP) from surface seal and soil treatments. Plotted data are averages of three replicates. Manure, composted steer manure; KTS, potassium thiosulfate.

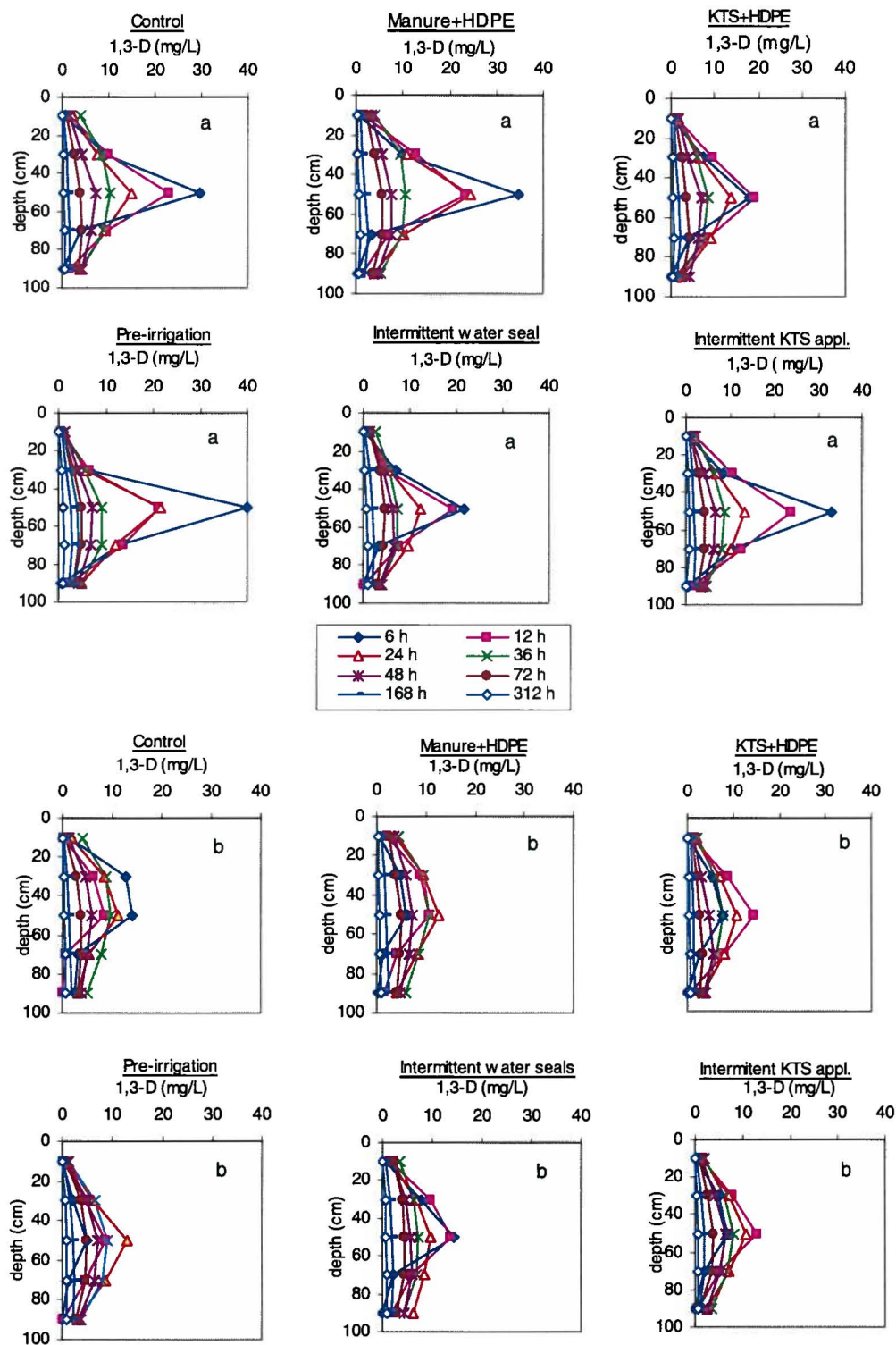


Figure 3. 1,3-dichloropropine (1,3-D) distribution in soil-gas phase at locations a=adjacent to fumigant injection lines (top six graphs), and b=between injection lines (bottom six graphs) under various surface treatments.

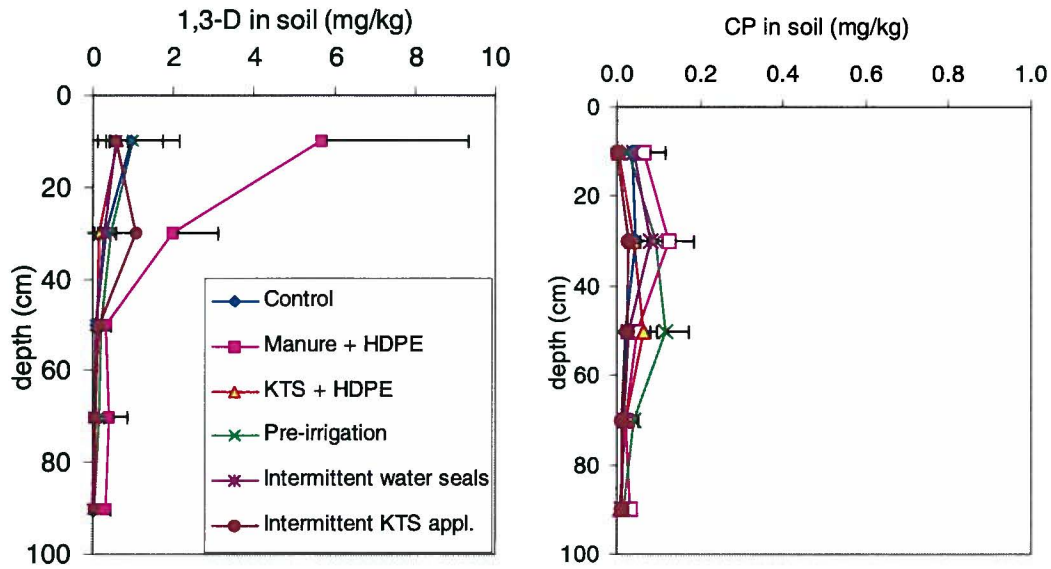


Figure 4. Residual 1,3-dichloropropine (1,3-D) and chloropicrin (CP) extracted from soil samples after 14 days of fumigations. Error bars are standard deviations of the mean (n=3).

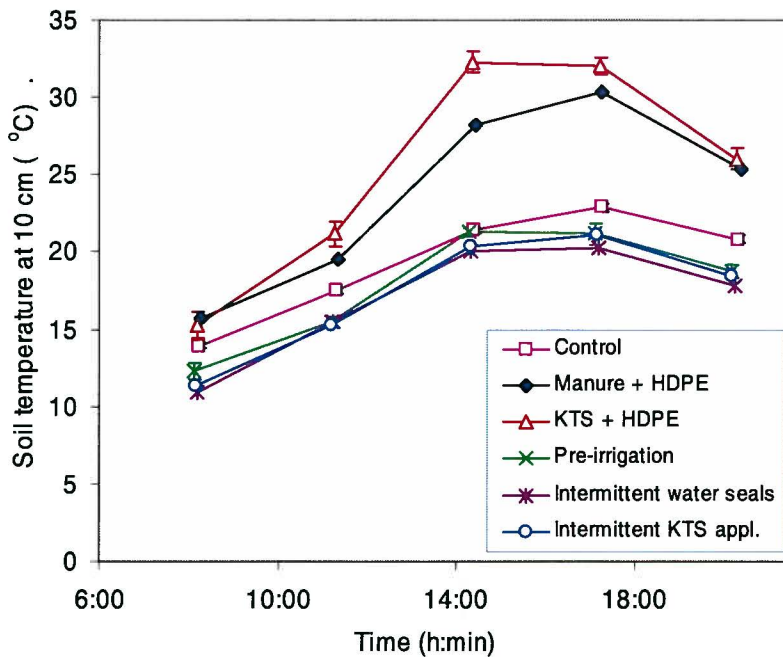


Figure 5. One-day soil temperature measurements at 10-cm soil depth during fumigant emission monitoring period. Error bars are the standard deviations of the mean (n=3).

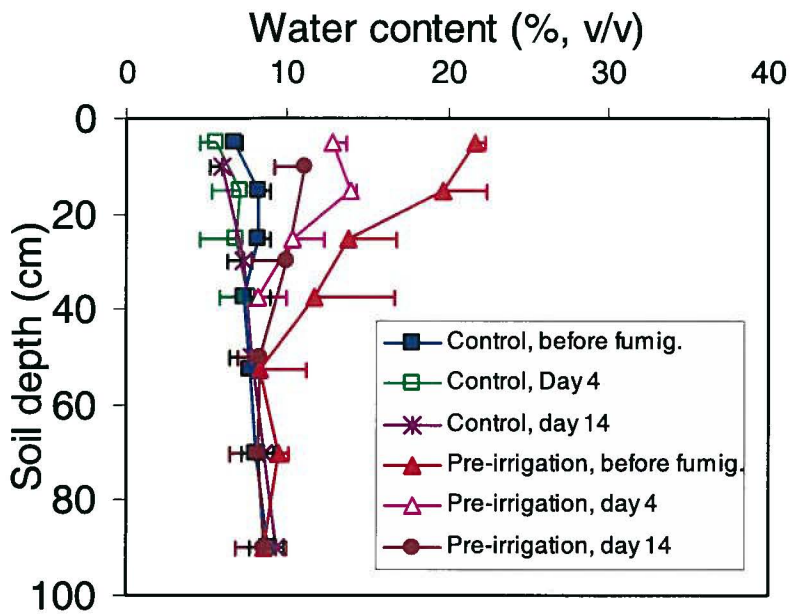


Figure 6. Soil water content measured the day before fumigation and 4 days after fumigation under various surface treatments. Horizontal bars are the standard deviations of the mean (n=3).

