Methyl Bromide Emission Reduction with Field Management Practices

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Current methods of soil fumigation may allow up to 65% of the applied methyl bromide (MeBr) to escape to the atmosphere. To protect stratospheric ozone and prevent economic losses from MeBr phaseout, there is an urgent need to find alternative fumigation techniques that can reduce MeBr emission. A field experiment was conducted to study and compare the effect of different management methods on MeBr emission reduction. Tested parameters included injection depth (0.25 and 0.6 m), use of polyethylene or a high-barrier plastic, bare soil, and irrigation. MeBr emission was estimated by sampling for the increase in soil bromide ion. Deep injection increased MeBr degradation and reduced total emission. Compared to bare soil, covering with plastic tarp significantly reduced MeBr emission. MeBr emission was reduced to <15% with the high barrier plastic. Irrigation and tarping enhanced MeBr containment and degradation. Effective treatment of citrus nematodes, fungi, and yellow nutsedge seeds was achieved for shallow injection with tarp or deep injection with the high-barrier plastic. The optimal method for pest control and MeBr emission reduction appears to be a combined use of the high-barrier tarp, irrigation, and deep injection.

Introduction

Methyl bromide (MeBr) is an effective soil fumigant for controlling weeds, soil-born disease pathogens, plant-parasitic nematodes and fungi. Recent research has indicated that agricultural emission of MeBr can contribute to the depletion of stratospheric ozone (*1*, *2*), and a complete phaseout of MeBr use by the year 2001 has been scheduled by the U.S. Congress. Previous and current research has not shown the existence of any other single soil fumigant that can be used to replace MeBr while maintaining the efficacy for effective and broad-spectrum pest control. The cost resulting from the suspension of MeBr for soil fumigation alone would exceed $1 billion annually (*3*). The scheduled phaseout of MeBr use was based on the emission rate from current management practices. Unless much lower emission can be achieved, it is very unlikely that MeBr use will be permitted in the U.S. after 2001. Therefore, it is necessary to evaluate the current fumigation practices and test potential management alternatives to reduce MeBr emission.

For soil fumigation purposes in California, MeBr is traditionally injected as a liquid at 0.2−0.3 m for shallow injection or about 0.6 m for deep application. The treated soil is often covered immediately with a layer of plastic tarpaulin (tarp) to retard MeBr emission into the atmosphere and enhance control efficacy. The conventional polyethylene (PE) tarp, however, is not effective in inhibiting MeBr emission, because under warm conditions and shallow injection, up to 87% (*4*) and 65% (*5*) of the applied MeBr can escape to the atmosphere. Injecting MeBr as a gas (called "hot-gas" injection) has often been used in greenhouse nurseries and for very localized turf treatments. While chemical injection through drip irrigation systems is commonly performed (*6*), injecting MeBr gas into buried drip lines has recently become popular because it often uses existing drip irrigation systems required for crop production. The hot-gas injection method has been widely used for field crops such as pepper, melons, and strawberries. MeBr application into preinstalled drip systems would have less chance for MeBr loss than shank application because the field can be covered with plastic tarps prior to application.

Preliminary work has been carried out (*7*) in searching for high-barrier films that are less permeable to MeBr than the widely used PE films. A new plastic film (Hytibar, Klerk's Plastics, Inc.) manufactured by incorporating a barrier polymer (EVOH) in the center of two layers of PE is claimed to have very low permeability to MeBr (*8*). Preliminary tests in the laboratory have shown that this new plastic is about 70 times less permeable to MeBr than the conventional PE films (*9*). The replacement of the PE films with less permeable barrier tarps, such as the Hytibar, could be a very efficient way of reducing MeBr emission, since it requires the least change in current management practices.

Because the transport of MeBr in soil fumigation is mostly through gas-phase diffusion, increasing injection depth would delay and reduce the surface emission according to the diffusion theory. This is due to an increased path length from the MeBr source to the soil surface and increased residence time for degradation. Since gas-phase diffusion is directly related to soil porosity, the emission can also be reduced or retarded if the air-filled porosity of surface soil is reduced. In a laboratory column study Jin and Jury (*10*) demonstrated that increasing the water content of surface soils can effectively reduce the emission rate, and the percent of emission reduction is proportional to the amount of water applied. No field experiment has been conducted to determine the effectiveness of surface irrigation on reducing MeBr emission.

Field quantification of MeBr emission has been made with many approaches including meteorological (*5*, *11*), chamber (*4*, *12*), and bromide ion (Br^-) appearance method (*13*). The meteorological-based measurements require extensive instrumentation, air concentration measurements, and a large open field. The chamber method may alter local environmental conditions and corrections for temperature and pressure may be required (*12*, *14*, *15*). The Br^- appearance method offers a simple and accurate indirect estimation of MeBr emission because MeBr degrades to the simple mineralized form of Br^- that can be easily and accurately analyzed in the laboratory. This method is especially suitable for small plot studies where soil variability is less of a problem and direct flux measurement with chambers would be prohibitively expensive.

The overall goal of this study was to experimentally test the effectiveness of new fumigation management methods on MeBr emission reduction. Controlled parameters included the use of (a) a new plastic film (the Hytibar) as compared to the conventional PE film, (b) MeBr application at 0.25 and 0.6 m depths, and (c) irrigation over the soil surface before MeBr application.

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TABLE 1. Summary on Methyl Bromide (MeBr) Application, Degradation or Potential Volatilization, and Pest Control

Treatment[a]	MeBr application (g)	MeBr application (g m^{-2})	MeBr degradation (g)	potential volatilization (%)	citrus nematode[b] (counts)	fungus[b] (counts)	yellow nutsedge[b] (counts)
Deep-PE	703	36.8	600	15	0, 0, 0	10, 10, 10	13, 13, 13
Deep-HP	695	50.8	590	<15	0, 0, 0	0, 0, 0	0, 0, 0
Shallow-PE+Irrig	639	36.3	368	<42	0, 0, 0	0, 0, 1	0, 0, 0
Shallow-PE	590	32.4	241	59	0, 0, 0	2, 0, 0	0, 0, 0
Deep-Bare	708	42.7	283	60	56, 72, 41	10, 10, 10	13, 11, 14
Shallow-Bare	605	34.5	80	87	0, 0, 0	10, 10, 10	13, 11, 14
Control-PE	0	0	0	0	498, 534, 462	10, 10, 10	13, 13, 12
Control-Bare	0	0	0	0	642, 1050, 1776	10, 10, 10	14, 14, 11

[a] Deep = 0.60 m injection; Shallow = 0.25 m injection; PE = polyethylene tarp; HP = the Hytibar plastic. [b] Survived citrus nematode (*Tylenchulus semipenetrans*), fungus (*Rhizoctonia solani*), and Yellow nutsedge (*Cyperus esculentus*) seed.

Experimental Section

Plot Construction. Six experimental plots were constructed in a field located on a University of California Agricultural Experimental Station near the Riverside campus. Each plot was about 3.4 × 4.9 m in size and at least 4.6 m apart from the next treatment. The soil in this area is an Arlington fine sandy loam (coarse-loamy, mixed, thermic, Haplic Durixeralf) and consists of approximately 64% sand, 29% silt, and 7% clay. To eliminate lateral diffusion of MeBr away from the plots, four layers of continuous plastic tarp were buried to 3 m depth along the sides of each plot. The four layers of plastic consisted of two layers of the Hytibar which were sandwiched between two layers of 0.10 and 0.15 mm (4- and 6-mil) PE tarps. The Hytibar plastic served as the impermeable barrier, and the PE tarps protected the Hytibar from punctures during the construction of the plots. In addition, the two layers of PE provided an additional diffusion barrier. A backhoe was used to excavate and backfill 0.46 m wide and 3 m deep trenches on the perimeter of each plot for the installation of these side-wall plastics.

To simulate shallow and deep injection, drip tapes (RO-DRIP, Roberts Irrigation Products, Inc.) used in conventional drip irrigation for row crops were installed at 0.25 and 0.6 m depth, respectively. A trencher was used to excavate 76 mm wide and 0.25 and 0.6 m deep trenches for the placement of these drip tapes. For the 0.25 m application, ten tapes were installed at a 0.31 m spacing whereas only two tapes were used for the 0.6 m injection. All the drip tapes were buried in the long (i.e., 4.9 m) direction and connected to a manifold with a common riser to the soil surface. We doubled each drip tape and offset the 0.2 m spaced openings to have a 0.1 m spacing to achieve a better application uniformity along the tapes.

Plot Treatments and MeBr Application. After installing the side-wall plastics and drip tapes and backfilling all the open trenches, we irrigated the experimental site with a sprinkler system to settle the soil from disturbances created during the construction and installation processes. We did not start the experiment until the volumetric soil water content was reduced (by drainage and evaporation) to about 18% at the injection depths. Before covering the plots with plastic films, the surface 0.15 m soil was rototilled to create a homogeneous layer which simulates a commercial field application.

Prior to MeBr application, we covered a 0.25 m application plot (Shallow-PE) and a 0.6 m plot (Deep-PE) with the PE film and sealed the four edges to the side-wall plastics. Following a similar procedure, a 0.6 m plot was covered with the Hytibar plastic (Deep-HP). Immediately before MeBr injection, we applied 263 L of water (or 14.9 mm depth which was the maximum amount before runoff occurred) to a 0.25 m plot and then covered it with the PE film (Shallow-PE+Irrig). The remaining 0.25 and 0.6 m application plots were not covered and left with the rototilled bare surface (named Shallow-Bare and Deep-Bare, respectively). To obtain background soil information and provide a control for MeBr efficacy assessment, we selected two areas, each about the same size as the MeBr plots, as the control plots. One of the control plots was covered with the PE film (Control-PE), and the other was left with the rototilled bare soil surface (Control-Bare).

Pure MeBr (CH_3Br) gas was applied on September 21, 1995, between 1020 and 1245 h by Tri-Cal following the hot-gas injection procedures (Tri-Cal Inc., 1029 Railroad Street, Corona, CA 91720). We recorded the amount of MeBr applied to each plot with a scale accurate to 1.0 g (Table 1). The time required to apply these amounts of MeBr to each plot, while maintaining a pressure of about 55 kPa, averaged 14 min for the three deep plots and about 5 min for the three shallow injection plots. It took more time for the deep plots because they had only $^1/_5$ the drip tape of the shallow plots.

Data Collection and Analysis. Because a large fraction of MeBr can be partitioned into the water phase and degraded by hydrolysis, it is important to determine the water content of soils fumigated with MeBr. Although no precipitation occurred during the experiment, we monitored soil water content with time domain reflectometry (TRASE, Soilmoisture Equipment Co.) equipped with probes installed at various depths to 1 m in the two control plots (Control-PE and Control-Bare) and to 0.3 m in the irrigated plot. Measurements were made at selected times after MeBr application. To compare temperature changes under tarp and bare conditions, we also measured soil temperature at variable depths from the two control plots. Soil bulk density was determined for different depths to 3 m using undisturbed samples collected from an excavation adjacent to the plots. They ranged from 1.30 (g cm^{-3}) near the surface to about 1.71 (g cm^{-3}) at deeper depths.

Soil-air profile samplers were installed for measuring MeBr gas concentration distribution. For the three deep injection plots, a profile sampler to 3 m was installed in the center of each plot with sampling ports located at 0.05, 0.25, 0.5, 0.6, 0.7, 0.85, 1, 1.5, 2, and 3 m depths. An additional 2 m sampler was installed respectively in Deep-PE and Deep-Hytibar next to one of the two injection lines to capture the early-time MeBr lateral diffusion. A 2 m sampler was installed in the center of each of the three shallow injection plots with sampling ports located at 0, 0.05, 0.15, 0.25, 0.35, 0.5, 0.75, 1, 1.5, and 2 m depths. Single port soil air samplers were installed at 0.25, 1, 2, and 3 m depths on the outside of the four layers of plastic containment walls to determine any potential MeBr breakthrough from the confinement. Coconut charcoal tubes were used for MeBr gas sampling from the soil air and, MeBr concentrations were analyzed on a gas chromatograph with an electron capture detector (*16*).

To determine pest control efficacy under these management treatments, three replicated samples of a common citrus nematode (*Tylenchulus semipenetrans*), a fungus (*Rhizoctonia solani*), and a common weed the Yellow nutsedge (*Cyperus esculentus*) seed were placed in the center of each plot (including Control-PE and Control-Bare) to 0.1 m depth. They

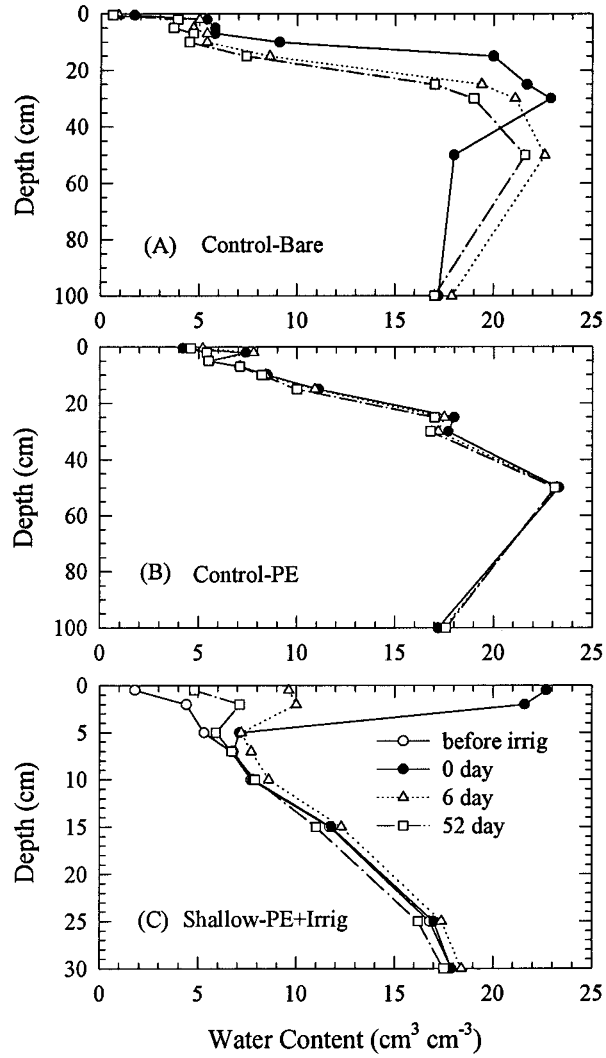

FIGURE 1. Volumetric soil water content in the control bare (A), control polyethylene or PE (B), and PE with irrigation (C) plots at 0, 6, and 52 days after MeBr application.

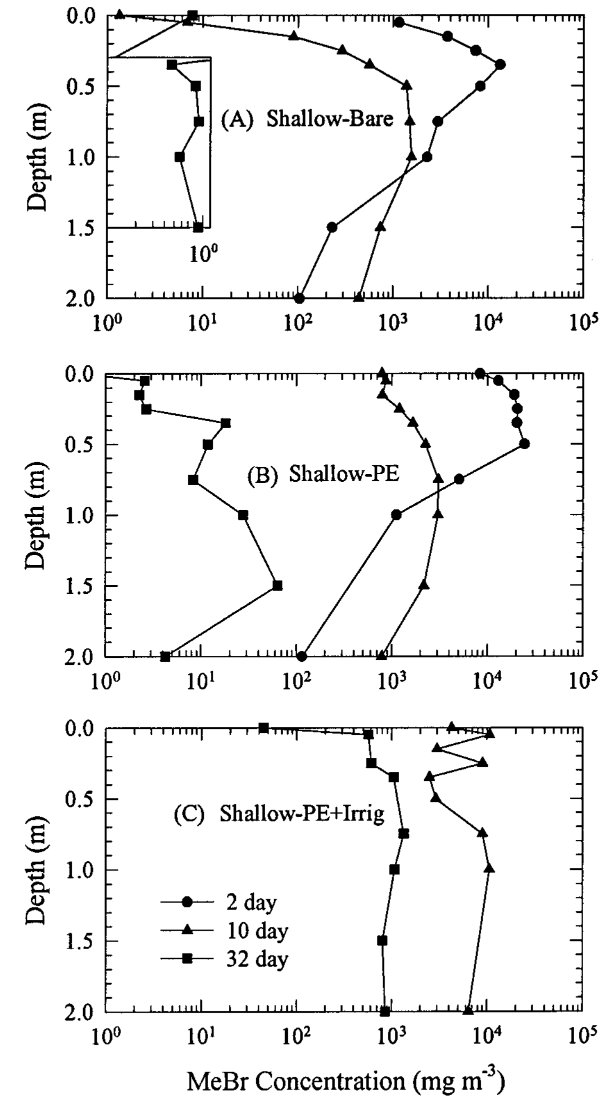

FIGURE 2. Methyl bromide (MeBr) concentration in soil air under bare (A), polyethylene or PE (B), and PE with irrigation (C) treatments at 2, 10, and 32 days after shallow injection at 0.25 m.

were removed 5 days after fumigation because current regulation requires the field be covered for 5 days after MeBr application. Surviving specimens from each plot were determined and compared to the controls. When removing the samples from the covered plots, we isolated an area about 0.3 m in diameter to prevent or reduce MeBr loss from the tarp containment.

For analysis of Br^- addition and potential emission estimates, soil core samples were taken before and about 133 days after MeBr application. A total of nine soil cores were taken from each of the six fumigated plots with three cores taken near the center of the plots to 3 m depth and the remaining six cores taken near the edges with three cores to 1.5 m, and three to 0.5 m depth, respectively. Sampling increments were between the following depths: 0, 0.05, 0.1, 0.2, 0.3, 0.5, 1, 1.5, 2, 2.5, and 3 m for shallow injection and 0, 0.1, 0.2, 0.3, 0.5, 0.7, 1, 1.5, 2, 2.5, and 3 m for deep injection. The frequency of soil samples was increased near the surface. This was to provide a more representative measurement of the total mass of degraded MeBr, since the injection depth or center mass should be near the surface and to 0.25 and 0.6 m depths. To determine potential MeBr movement beyond 3 m depth, we extended the three deep cores to 5 m depth for the Shallow-PE plot. Br^- concentrations in these soil samples were analyzed with a colorimetric QuikChem AE automated ion analyzer (Lachat Instruments).

Results and Discussion

Soil Water Content and MeBr Concentration. Soil water content decreased significantly in the upper 0.3 m of the Control-Bare plot (Figure 1A). The water content near the surface was only about 1% for the bare plot compared to about 4% for the covered plots. Plot covered with the PE tarp showed little or no change in soil water content (Figure 1B) due to the elimination of surface evaporation by the water impermeable plastic. The surface soil water content (~5 cm depth) increased from 1.7 to 22.6% and remained high (between 22.6 and 9.6%) in the irrigated plot (Figure 1C) during the first 6 days after MeBr injection. Because of drainage, an average of 0.6% increase in water content was observed between 7 and 30 cm depth 6 days after irrigation.

Similar MeBr concentration profiles were measured for the Shallow-Bare and Shallow-PE plots at 2 and 10 days after injection (Figure 2, panels A and B). However, the bare plot had much smaller concentrations (1.2×10^3 and 1.4 mg m^{-3} for 2 and 10 days after application, respectively) near the surface than the covered plot (8.5×10^3 and 8×10^2 mg m^{-3}) because of the absence of the PE tarp. The PE tarp kept the MeBr concentration near the surface at a relatively higher level during the first 10 days after application. The shape of these profiles represents very nicely the gas diffusion away from the source (i.e., at 0.25 m). The average concentration

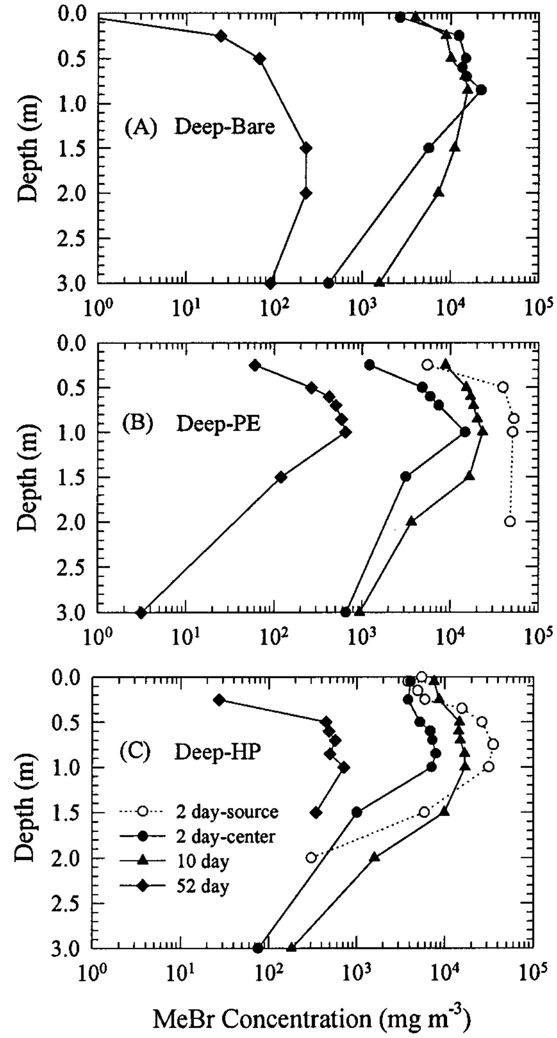

FIGURE 3. Methyl bromide (MeBr) concentration in soil air under bare (A), polyethylene or PE (B), and Hytibar plastic or HP (C) treatments at 2, 10, and 52 days after deep injection at 0.6 m.

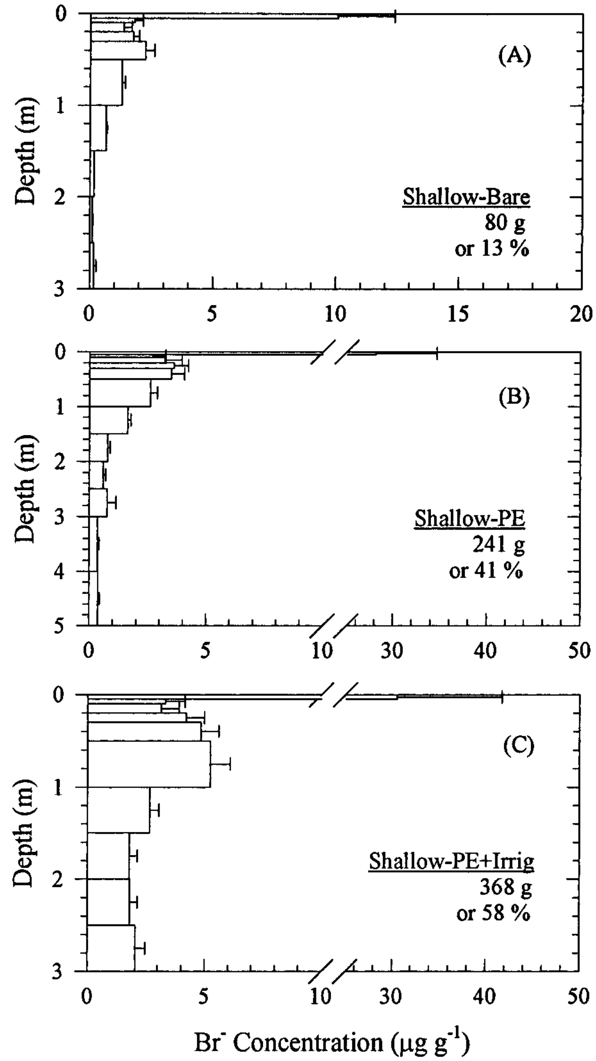

FIGURE 4. Soil bromide ion (Br^-) concentration distribution under bare (A), polyethylene or PE (B), and PE with irrigation (C) treatments at 130−137 days after shallow injection at 0.25 m. Error bars = +SD (*n* = 3−9).

in the upper 2 m soil reduced to about 14.1 mg m^{-3} for the Shallow-PE plot as compared to only about 0.65 mg m^{-3} for the Shallow-Bare plot at 32 days after MeBr application. In the irrigated plot, however, the concentration distributed evenly in the upper 2 m and remained at about 800 mg m^{-3} after 32 days (Figure 2C). This is about 60 times increase from the Shallow-PE plot. The average concentration was also higher (=6481 mg m^{-3}) than the Shallow-PE plot (=1668 mg m^{-3}) 10 days after application. The day 2 concentration data for the irrigated plot was lost because of an experimental error. MeBr distribution in the irrigated plot was more uniform than in the unirrigated plot in the upper 2 m soil and is attributed to water application and deep percolation of water which increased MeBr transport to deeper depths. Reduced effective soil porosity (due to irrigation) at the soil surface would create a sealing effect retarding MeBr diffusion loss into the atmosphere and keeping the concentration at a relatively high level for a prolonged time. Irrigation at the soil surface followed by applying a tarp offers a means of containing MeBr from emission into the atmosphere.

For the three deep injection plots, similar concentration profile shapes were observed at 2 and 10 days after injection (Figure 3), except in the Hytibar plot (Deep-HP) where the concentration at 3 m was about an order of magnitude smaller than observed in either the bare or the PE plot. The retarded downward movement was probably caused by a hard pan occurring at ~1.5 m which we observed during soil sampling. Early-time lateral diffusion was observed from the concentration profiles at 2 days after application (Figure 3, panels B and C) where the concentration was about an order of magnitude larger near the source than found between the two sources at 0.6 m depth. The difference disappeared after 10 days of redistribution. Unlike the shallow injection, the three deep plots had a similar average concentration over the 3 m depth at 52 days after application. The Deep-Bare plot had a much smaller concentration near the surface than the two covered plots. Both the PE and the Hytibar tarp kept the MeBr concentration near the surface at a relatively higher level. The concentration at deeper depths was higher in the bare than in the tarped plot at 52 days after application. This was caused by the reduced soil water content from evaporation, hence increased diffusion rate in the bare plot. Compared to shallow injection, applying MeBr at 0.6 m kept the concentration at a high level for a long time, even under bare conditions.

Only a trace amount of MeBr was detected outside the four layers of side-wall plastic containment. Therefore, there was no significant unaccounted MeBr loss during this experiment.

MeBr Degradation and Potential Emission. Because the background soil Br^- concentration remained at about 0.1 μg g^{-1} before and after the experiment, we believe the increased soil Br^- content was induced from MeBr degradation. For shallow injection, measured soil Br^- concentration was

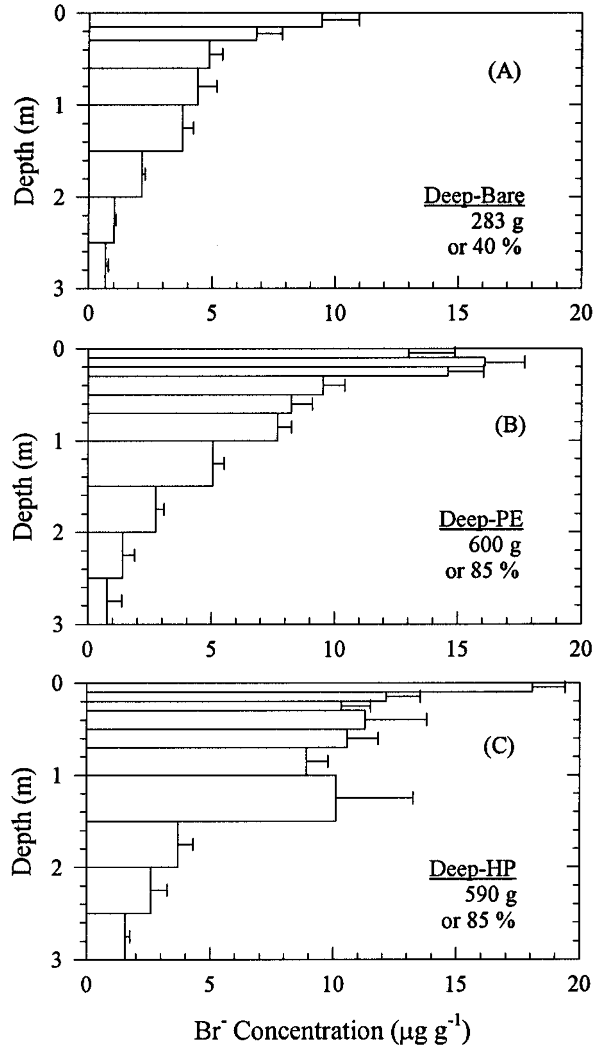

FIGURE 5. Soil bromide ion (Br^-) concentration distribution under bare (A), polyethylene or PE (B), and Hytibar plastic or HP (C) treatments at 130−137 days after deep injection at 0.6 m. Error bars = +SD (*n* = 3−9).

generally small with a maximum ranging from 2 to 5 $\mu g\ g^{-1}$ in the subsurface soil (Figure 4). The concentration in the surface 5 cm layer, however, was about an order of magnitude greater than the average over the 3 m profiles. This was caused by increased soil organic matter content occurring near the surface that had accelerated MeBr degradation into Br^-. MeBr degradation increased significantly throughout the profile when applying 14.5 mm water (Figure 4C). The increased soil water content would make MeBr hydrolysis more efficient because of an increase in MeBr resident time in the soil water phase. The amount of MeBr that can be converted from the measured Br^- was, respectively, 80, 241, and 368 g for the Shallow-Bare, -PE, and -PE+Irrig plot. These amounts would correspond, respectively, to 13, 41, and 58% of the total applied mass to these plots. Since no other losses for the applied MeBr occurred, the difference between applied and degraded would be lost from surface volatilization. The potential volatilization for the three shallow plots was then 87, 59, and 42%, respectively. The estimated 87 and 59% volatilization rates for the Shallow-Bare and -PE plots are in good agreement with studies (see refs 11 and 5 for bare and PE tarped treatment, respectively) from large-field experiments and using meteorological methods for emission estimation.

Taking soil samples to 3 m provided reasonable estimates of the total degradation in the shallow bare and PE plots because the average Br^- concentration between 1.5 and 3 m for the bare or 3 and 5 m for the PE plot was very close to the background concentrations (Figure 4, panels A and B). For the irrigated plot, however, the measured Br^- concentration at 3 m remained significantly higher than the background (Figure 4C). Therefore, additional degradation might have occurred below 3 m. The estimated total degradation would consequently be more than 58% or the potential emission be less than 42% for the irrigated plot.

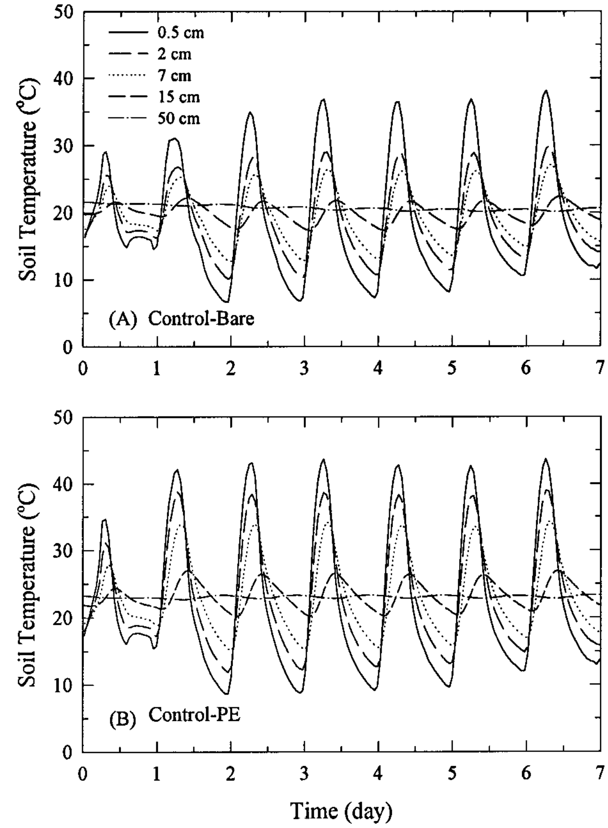

FIGURE 6. Measured soil temperatures from the bare (A) and tarp covered (B) control plots at 0.005, 0.02, 0.07, 0.15, and 0.5 m depths during the first 7 days of the experiment.

Applying MeBr at a deeper depth (0.6 m) increased soil Br^- concentration in the profile (Figure 5), compared to the 0.25 m shallow injection. The maximum concentration ranged from about 10 $\mu g\ g^{-1}$ for the Deep-Bare plot to 16 or 18 $\mu g\ g^{-1}$ for the two tarped plots. The enhanced degradation was primarily due to the increased MeBr residence time in the soil, because of longer traveling distance to the surface. Covering plots with either PE or the Hytibar tarp more than doubled MeBr degradation (85%) compared to the Deep-Bare plot (40%). Therefore, tarp cover is also effective in reducing emission for deep (0.6 m) injection. Although a similar degradation fraction was obtained (Figure 5, panels B and C), more MeBr degradation per unit soil volume had occurred in the Hytibar covered than in the PE plot because (a) the size of the Hytibar plot was smaller than the PE plot due to variations in the plot construction, hence more MeBr mass per unit volume, and (b) the measured Br^- concentration at 3 m remained significantly higher in the Hytibar plot than the background, thus, additional degradation might have occurred below 3 m. The degradation fraction in the deep Hytibar plot could be larger than the measured 85%. The presence of hard pan in the deep Hytibar plot may impede MeBr movement to lower depths or making it more susceptible for surface emission losses. This is probably why the less permeable Hytibar tarp had about the same emission losses as the deep PE plot. On the basis of the measured degradation ratio, potential volatilization was 60, 15, and <15% for the Deep-Bare, PE, and Hytibar plot, respectively.

Pest Control. Sufficient pest control (100% for *T. semipenetrans* and *C. esculentus*; 97% for *R. solani*) was achieved at 0.1 m depth with shallow injection and covering with the PE tarp either with or without irrigation (Table 1). Deep injection with the Hytibar tarp also reached 100% efficacy for all three targeted species. Covering the plot with PE in deep injection or no cover resulted insufficient pest control. The data also indicates that covering the soil surface with PE or Hytibar alone (without MeBr) can create some mortality in *T. semipenetrans* and *C. esculentus*. The presence of a plastic cover created a greenhouse or solarization effect where the soil temperature was significantly higher than the bare soil. Measured soil temperature profiles from the two control plots (Figure 6) indicated a 5−10 °C increase in the upper 0.15 m of soil for the covered plot. The increased temperature would increase the effectiveness of MeBr to the targeted pests (*17*).

This field research, conducted under environmental conditions similar to many commercial fumigation practices, has provided a very important comparison of MeBr potential volatilization estimates under different management techniques. The standard treatment, i.e., shallow injection with PE cover, produced an emission estimate that was very comparable to findings from recent field experiments on MeBr volatilization assessment. We also found that the new Hytibar plastic appeared to be effective in limiting MeBr from emission loss. Combined use of irrigation and the conventional PE tarp or the new Hytibar may prove to be an economical method for MeBr fumigation with reduced emission. Injecting MeBr to a deeper depth can also reduce the emission loss into the atmosphere.

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