

HILGARDIA

A JOURNAL OF AGRICULTURAL SCIENCE PUBLISHED BY THE CALIFORNIA AGRICULTURAL EXPERIMENT STATION

539 E4 V.42 Wo.14 Volume 42, Number 14 · November, 1974

# Factors that Affect Deep Penetration of Field Soils by Methyl Bromide

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JAN 24 1975

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Effective control of the root rot fungus, Armillaria mellea, with methyl bromide requires diffusion of adequate dosages to depths as deep as the roots of the dead and diseased plants. Factors that govern diffusion to these depths are soil texture, porosity as it relates to moisture, amount and method of application, and the soil covers used. Diffusion patterns were determined by gas chromatographic analysis of soil atmospheres. A crop of sudangrass dried the soils sufficiently in one season to depths of 8 feet to enable methyl bromide to diffuse readily. Best downward movement was obtained by placing methyl bromide about 3 feet deep; downward movement was greater than lateral, and adequate dosages were obtained 10 to 12 feet deep. Increased dosage increased the range of effective fumigation. A plastic cover was required to give control at the surface and top 1 foot of soil. Polyethylene is a relatively poor cover compared to other plastics. The soils studied were coarse sandy loams and the fine-textured silt and sandy clay loams. Some results of these studies have been applied to commercial fumigations.

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### Factors that Affect Deep Penetration of Field Soils by Methyl Bromide<sup>1</sup>

### INTRODUCTION

FUMIGATION WITH METHYL BROMIDE (MB) has been used for many years to control certain plant pathogens in the upper few feet of soil. For example, *Rhizoctonia*, *Phytophthora*, *Pythium*, and nematodes can be successfully controlled by such treatment. Theoretical and practical aspects of soil fumigation are discussed in some excellent reviews by Goring (1962, 1967, 1970), Munnecke (1967, 1972), and Kreutzer (1960).

Few reports have been made, however, on the factors that affect movement of MB below those upper few feet and down to 10 to 12 feet. Deep penetration is of particular interest in California where Armillaria mellea, commonly called oak root fungus, often seriously affects deep-rooted perennial crops such as citrus, grapes, prunes, and many ornamentals. Armillaria mellea infection is a serious problem also on a wide variety of plants in temperate and tropical climates throughout the world (Raabe, 1962).

Armillaria mellea may be endemic in California mountains and foothills. Infested roots and trunk pieces are often carried by flood waters and buried in outwash plains. Many of these areas are being brought into agricultural production. The fungus may lie dormant for many years in these infested wood pieces but will resume active parasitism in roots of susceptible plants that contact it.

Further serious sources of infection are the infested root pieces remaining in the soil when dead or dying plants are removed from established agricultural lands.

Fumigation with adequate dosages of MB effectively controls *A. mellea*; thus it is important to know more about the movement of MB in the deeper horizons of the soil.

Usually, to fumigate for pathogens near the surface, the toxicant is applied by chisel injection with machine-drawn equipment. Some soil is compacted into the channel with presser wheels, and the soil is immediately covered with a polyethylene tarp approximately 1 mil thick. For deeper penetration of MB, chisel injection depth may be increased to 3 feet, but the power requirements are very great. Unfortunately, polyethylene is relatively porous to MB, the MB quickly vaporizes, moves via chisel channels to the soil surface, and escapes through the tarp. These factors make it difficult to obtain adequate concentrations of MB in the deeper soil horizons except in very porous soils.

<sup>&</sup>lt;sup>1</sup>Submitted for publication October 29, 1973.

This study was designed to measure the effects of the fixed soil characteristics (composition, bulk density, and available pore space) and the transient characteristic porosity which is directly related to moisture content. Various methods of application of MB also were evaluated. These experiments extended over five years and involved over 20 separate trials on four different lands with a range of soil types. Preliminary reports have been published (Kolbezen, Munnecke and Stolzy, 1969; Munnecke Kolbezen, and Stolzy, 1969; Munnecke, Wilbur, and Kolbezen, 1970; Rackham *et al.*, 1968).

### METHODS

The experiments reported here followed a basic pattern: selecting sites with desired soil characteristics, treating with MB, and determining diffusion patterns by gas chromatographic analysis of soil atmospheres. ments are identified and described in table 1 and are referred to as Hinkley, Moreno, UCR (Riverside), and SCFS (U.C. South Coast Field Station).

The soil physical parameters were determined by methods given by Black (1965) and Baver (1956). Soils porosity data were derived indirectly by two procedures: soil moisture was

### Soils

The field soils used in these experi-

TABLE 1

PHYSICAL CHARACTERISTICS AT VARIOUS DEPTHS OF THE FOUR SOILS USED IN THE DEEP PENETRATION EXPERIMENTS: THE SOIL BULK DENSITY ( $P_b$ ), PER CENT SOIL VOLUME NOT OCCUPIED BY SOLIDS ( $\Sigma$  E), AND THE PARTICLE SIZE DISTRIBUTION

	H (Bryn H san	inkley: Mawr, C anford dy loam	(al.)	(U.C	foreno: . Exp. S Moreno ilt loam	ta.)	UCR: (U.C. Riverside) Ramona sandy loam				SCFS: (So. Coast Field Sta.) Moreno sandy clay loam		
Depth	Рь	Σ	Е	Рь		ΣE	Рь		ΣΕ		Рь	İ	ΣΕ
in.	g/cc	per	cent	g/cc	pe	r cent	g/cc	g/cc		r cent	g/cc	p	er cent
12	1.56	4	1	1.71 36		36	1.3		43		1.45		45
24	1.49	4	3	1.48 44		1.4		49		1.48		44	
36	1.58	4	0	1.68 37		1.3		49		1.28		52	
48	1.52	4	3	1.45	45 45		1.3		52		1.31		51
60	1.57	4	1	1.48	44		1.3		49		1.37		48
72	1.64	3	8	1.74	34		1.5	1	44		1.42		46
96	1.65	3	8	1.90		28	1.6		42				_
	Per cent particle size:												
	Sand	Silt	Clay	Sand	Silt	Clay	Sand	s	ilt	Clay	Sand	Silt	Clay
6	. 65	26	9	12	65	23	74	2	0	6	_		_
12								.			61	21	18
18	67	23	10	11	60	29	64	2	8	8			
24								•	• •		65	23	12
30	. 56	33	11	10	55	35	58	3	4	9	•••		
36								·			40	32	28
42	54	34	12	15	48	37	63	2	8	9			
48			1.0	17				·			39	33	20
54	51	30	13	17	49	35	04	2	.9	°	50		
66		•••		20	50	30	64	·		8	50	20	1 22
72		•••			50			1.1			61	23	16
96				16	56	29					60	22	18
102							58	3	3	9			
			1	1		1	1	1					1

measured by neutron thermalization, and bulk density was obtained by gamma absorption. The data were verified, in some instances, by direct determinations on undisturbed soil cores. Supplementally, at the time of each experiment, samples for soil moisture analyses were obtained when holes were dug by auger for placement of the soil atmosphere sampling equipment.

Certain commonly used terms should be defined here:  $P_w$  is weight per cent soil moisture based on soil dry weight,  $P_v$  is per cent soil volume occupied by water,  $\Sigma E$  is per cent soil volume not occupied by solids, and  $\Sigma a$  is per cent soil volume occupied by gases. Then  $\Sigma E =$  $\Sigma a + P_v$ . CT is the accumulated dosage of MB in ppm days = the product of concentration (ppm) × time (days).

In most experiments, soil moisture was altered by planting either sudangrass, Sorghum sudanense Piper (Stopf.) var. Trudan, or safflower, Carthamus tinctorius L. At SCFS where summer temperatures rarely exceeded 38° C, and at Moreno and Bryn Mawr where temperatures frequently exceeded 38°C, sudangrass was most efficient for this purpose. The fields were irrigated in late winter, then sown, fertilized and irrigated again in the spring to obtain maximum growth. No water was added after mid-June, and there was no appreciable rainfall during the summer months. In several years, there was less than 0.5 inch rainfall through the growing season and on into the following January. In some cases, the grass was mowed at the flowering time to keep it growing vegetatively. The effect of mowing on rate of water withdrawal was examined, but no conclusive differences were detected if comparisons were made late in the year after the mowed area had regained full growth. Mowing, however, retarded soil desiccation for a few weeks. This, and localized variations in plant vigor in a planted area, enabled us to find plots with graded moisture contents within relatively small areas. Usually the crop was allowed to stand until just before the plots were used which may have been October, November, or even January.

When MB was applied by injection with chisels, the grass was mowed and removed, and the stubble and roots incorporated into the soil by disking. With deep placement fumigation, the plots were not disked.

### Application of fumigant

MB was applied either at the soil surface beneath a cover, or by machine chisel injection, or by hand injection to depths as great as 5 feet. Most of the results from applications made by machine will be reported separately.

### Surface application

MB was released as a gas through a tube under a cover, consisting of either polyethylene, Mylar, Saran, or Saranex synthetic films, usually  $20 \times 20$  or  $25 \times$ 25 feet. Usually the entire dosage was applied from a previously prepared small tank containing the correct amount of MB. The edges of the covers were buried in slit trenches 12 to 18 inches deep and sealed with firmly tamped soil. The covers were supported on a network of wires to insure even exposure of the surface. To avoid the confounding effects of excessive temperatures or sunlight and the attendant effect on the diffusivity of the films, the plots were shaded. Best results were obtained with covers made of styrofoam sheets 0.5 inch thick  $(4 \times 12 \text{ ft.})$  glued together with canvas strips and laid over the plots.

### Hand injections

MB was applied by hand at various depths in holes approximately 3 inches in diameter made with a soil auger. Cans containing one pound MB were chilled in a food freezer and transported to the field packed in dry ice. The can was gripped by a special tool, placed in the hole, and punctured in place through the top. As desired, from one to four cans were placed into a hole, the top can covered with a wad of burlap or coarse gravel, and the hole filled with soil and tightly tamped. This procedure avoided danger to operators from early release of the gas and also ensured that a definitely known quantity was applied. Readings from thermocouples attached to each of four stacked cans indicated that the gas was released slowly over a period of 6 to 11 hours for the different cans. The MB was introduced in various patterns: in a single hole, in a closely spaced cluster of two to four holes around a single point, or in a rectangular grid of many points covering several hundred square feet.

### Sampling and analysis of methyl bromide

Sampling points for soil atmospheres, arranged in vertical and horizontal patterns, were installed before MB application. For example, in one experiment, sampling stations were placed 2, 6, 10, and 14 feet horizontally from the point of application of the gas. At each station there were sampling points 1, 3, 6, and 9 feet deep. Thus, 16 points were sampled in a two-dimensional vertical grid extending from the point of injection. Often, a similar set was installed in the opposite direction from the injection point. In other cases, depending on the porosity of the soil, the horizontal distances were varied, and occasionally the vertical sampling was extended to 12 feet. When MB was released beneath the cover or injected into the soil by chisels behind a moving tractor, samples were obtained from similar stations at selected sites in the treated area.

Samples of soil atmosphere were withdrawn through  $\frac{1}{16}$ -inch outer di-

ameter stainless steel tubing. The sample inlet end, buried at the desired point, was protected by covering it with a 100-mesh wire screen, while at the aboveground end, a soldered brass ball served as a mount for a  $\frac{1}{4}$ -inch rubber septum. To sample, the septum was pierced by the needle of a 20-ml glass syringe, the first 20-ml sample discarded, and the next 20 ml taken to the laboratory for analysis. Leakage was avoided by coating the plunger of the syringe with a film of Triton X-100, then, after the sample was taken, embedding the needle of the syringe in a gas-impervious sponge. There was no detectable loss in concentration of methyl bromide by storage in the syringes for six hours; even after 22 hours the loss was only 5 to 7 per cent. Syringes containing samples of soil atmosphere were transported up to 50 miles to the laboratory in this manner. Usually, samples were analyzed within six hours after removing them from the field.

The sample in each syringe was injected in three portions into the gas sampling valve of a gas chromatograph; detection was by flame ionization. The column was 2 feet of 1/4-inch aluminum tubing packed with 20 per cent silicone Hi-vac grease on Chromosorb W. Column temperature was 100°C and the retention time, 35 seconds. Peak height was an accurate measure of methyl bromide concentration. The accuracy of the system was checked by comparing readings of a series of standard methyl bromide concentrations measured before and after each set of field samples were analyzed.

Generally, all points in a field were sampled the day following treatment with MB, every other day for the next week, and at increasing intervals thereafter until data were considered adequate.

The equipment and procedures for sampling and analysis are presented in detail elsewhere (Kolbezen and Abu-El-Haj, 1972).

#### Covers

Polyethylene sheeting is relatively permeable to MB (Waack *et al.*, 1955). However, because it is used routinely in commercial field fumigations, films

### Effect of sudangrass planting on soil moisture

The effect of planting sudangrass on the withdrawal of moisture from the fine-textured soils in the Moreno plots is shown in figure 1. The curves represent the 0 to 12 foot moisture profiles of three very closely spaced plots on the dates indicated. Winter rains had been heavy through February; later some irrigation was necessary, but water was withheld onward from late May. By this time the planting was well established and there was no rain during the summer months.

The June 2 measurement showed the moisture content to be very high in the 0 to 6-foot profile. Five weeks later on July 7, much water had been withdrawn, and by November 20, the soil of 1 (0.001 inch), 4, or 6 mils thick were used in some of these experiments. Mylar (2-mil), Saran (2-mil), and Saranex (2-mil), which are practically impervious to MB, were used in many experiments to improve confinement of the applied MB and reduce unknown losses through the cover.

### RESULTS

was relatively dry. There was little effect below 8 feet, presumably because the roots did not penetrate beyond that depth. Moisture in this figure is expressed as weight per cent, since these plots were used for diffusion experiments, and soil samples were available from auger-hole digging operations.

In contrast, the curves in figure 2 show the moisture profiles of an adjacent area with similar soil structure profiles, but this plot was kept fallow throughout the year. Immediately after the heavy winter rains (March 19), the moisture content was very high at all levels down to 8 feet. After four months (July 14) there was little



Fig. 1. Soil moisture profiles of three closely adjacent plots in the fine-textured silt loam at Moreno on three dates of the same year showing withdrawal of water by sudangrass.



Fig. 2. Soil moisture profiles of a Moreno plot similar to those in figure 1, but kept fallow throughout the year.

change in the upper strata, but below 5 feet there was considerable decrease. Considerable evaporative drying occurred at 0–1 foot by November 20, but there were only minor changes in the deeper regions between July and November. These data were obtained by use of a neutron probe, and the moisture is expressed as volume per cent  $(P_v)$ .

As a result of this and similar experiments, soil usually was dried by growing sudangrass for a summer and fumigating in the fall and early winter months before the rains began.

### MB penetration in dry vs. wet soil

Many experiments, made on three soil types over a period of five years, demonstrated the importance of having soil dry for maximum distribution of MB gas. Since results were similar, only representative data will be presented.

**Hinkley:** A dry and a wet plot were compared. The  $P_v$  of the two plots were: dry, 3.7, 4.6, and 4.3 per cent at 3, 5, and 8 foot depths; wet, 13.6, 13.5, and 14.0 per cent moisture at the same depths, respectively. MB was injected with a commercial fumigating machine



Fig. 3. Concentration of methyl bromide (MB) in soil atmosphere vs. time following a commercial application of 4 lb. MB/100 sq. ft. covered with 1-mil polyethelene tarp. Soil was Hinkley sandy loam with moisture content: dry, 3.7-4.6 per cent; wet, 13.5-14 per cent.

into the soil with chisels set 10 inches deep and 12 inches apart. The dose was 4 pounds per 100 square feet of soil surface. The plots were covered immediately with a 1-mil polyetheylene tarp. Methyl bromide in the soil atmosphere was analyzed daily for three weeks at two locations in each plot at depths of 0.5, 3, 5, and 8 feet.

The data are shown in figure 3. Higher concentration of MB were attained in all depths in the dry soil than were attained in the wet soil except at the 0.5-foot level, where they were approximately equal. This was most noticeable 8 feet deep, where concentrations lethal to A. mellea<sup>t</sup> were obtained within three days in the dry soil, but in the wet soil, the concentration-time dose (CT) never reached what probably was a lethal level.

South Coast Field Station: More refined experiments on the effect of soil moisture were made. Square plots totalling 500 square feet were used. Gas was applied through tubing into the air over the soil, which was covered with Saran and shaded with a gabled, well ventilated, black polyethylene canopy. The edges of the Saran were buried in trenches 18 inches deep. Samples were taken from the air at the surface under the covers, and from soil atmospheres at 3, 6, and 9 feet deep. Duplicate plots were prepared, and each plot contained four sampling stations. After the treatment, soil atmosphere samples were taken daily from all points until max-

<sup>2</sup> LD95 of MB to A. mellea is an exposure of infested roots to an MB concentration  $\times$  time dosage (CT) of 6000 ppm days where concentration is greater than 500-600 ppm. This CT, while not lethal *per se*, places sufficient stress on A. mellea so that it is killed by the biological complex of field soils (Munnecke, Wilbur, and Kolbezen, 1970).



Fig. 4. Comparison of the concentration of methyl bromide (MB) in deep soil atmospheres in a wet and a dry plot in the sandy clay loam at SCFS. MB applied at 1 lb. per 100 sq. ft. as a gas under impervious Saran cover. Weight per cent moisture at 0.5, 3.6, and 9 ft. deep respectively was: dry, 14.6, 18.7, 15.6, and 16.9; wet, 24.8, 31.2, 17.9, and 23.7.

imum concentrations were recorded at every depth (approximately two weeks); after that they were taken less frequently for 35 days. In this experiment, the fall of 1967, the dose applied was 1 pound per 100 square feet of surface.

The data are shown in figure 4. In the dry plot (fig. 4B) the gas quickly penetrated to 6 feet, reaching a concentration of 1000 ppm in five days. In contrast, by the same time, most of the gas in the wet plot (fig. 4A) was held in the upper 3 feet and under the cover, and a concentration of only 130 ppm was attained at the 6 foot depth. A dose lethal to A. mellea was reached in the dry plot at all depths, probably even including 9 feet, whereas in the wet plot it was reached only at the surface and at 3 feet. In the dry plot with open pore spaces, diffusion produced nearly uniform concentrations throughout after 15 days. These fumigations were conducted at near-optimum conditions. The treated plots were covered with Saran, shaded, and the edges carefully buried to a depth of 18 inches on all sides. However, these optimum conditions made it possible to control many variables attendant to such a fumigation and to more completely isolate the



Fig. 5. Concentration vs. time of methyl bromide (MB) in the same soil used in figure 4; MB applied at the higher dose of 4 lb./100 sq. ft. under impervious Saranex cover. Characteristics of wet and dry plots compared in figure 6.

variable of the effect of moisture content on diffusion.

When analysis of soil atmospheres showed that after 135 days, all traces of MB had disappeared, the experiment was repeated on these same plots in the spring. But this time the dose was increased to 4 pounds MB per 100 square feet, and the cover was Saranex which is equivalent to Saran in its ability to retain MB. The covers were shaded with styrofoam sheets.

The results are presented in figure 5. Maximum concentrations were reached at the 3-foot depth in both wet and dry plots in five days. At 6 feet, the maximum concentration was reached in six days in the dry plot (fig. 5B), and in 16 days in the wet plot (fig. 5A), while at 9 feet, the maxima were attained in 15 and 25 days, respectively. More striking is the cumulative dosage



Fig. 6. Per cent fine particle content (A), per cent open pore space (B), and volume per cent moisture (C) vs. depth of soil used to obtain diffusion data in figure 5.

(CT) at the various depths attained in 50 days. In the dry plots, the CT 370K (K = 1000), 240K, andwere 125K ppm days at depths of 3, 6, and 9 feet, while in the wet plot they were 350K, 135K, and 45K. These results, the much greater CT in the deeper horizons of the dry plot, can be reconciled by referring to the moisture content, the open pore spaces, and fine particle-content profiles in figure 6. The wet and dry plots were adjacent, and particle-size distributions were similar. The curve in figure 6A shows a layer above and below 3 feet with a high percentage of fine particles (less than 100  $\mu$ ). The high moisture-holding capacity and fine-pore diameters of such a laver provided an effective barrier to downward diffusion when it was wet. The dry plot had been dried with a planting of sudangrass. The  $P_w$  of the wet and dry plots are compared in figure 6C, and the per cent open pore space in figure 6B. From the diffusion data in figure 5, it is clear that even such fine-textured soils may be successfully fumigated if sufficiently dried.

In the experiment just described (figs. 5 and 6), information on lateral diffusion was obtained by placing sampling stations 1, 2, 3, and 4 feet outside the covered and treated area, and sampling points 1, 3, 6, and 9 feet deep at each station. The CT attained in 50 days at each distance and depth is shown in table 2. The CT were calculated by integration of curves similar to those in figure 5; calculations included only those days when concentration of MB was 600 ppm or greater (see footnote 2). In the dry plot, all areas sampled received CT lethal to A. mellea except at lateral distances of 3 and 4 feet at the 1 foot depth. In the wet plot, however, effective CT were obtained in a much smaller space outside the treated area.

**Moreno:** The effect of moisture content on diffusion in the fine-textured silt

	CT* (in thousands) of MB diffusing at following distances (ft.) from edges of wet or dry plots									
		1		2		3	4			
Depth (ft.)	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet		
1	61	23	32	10	†	†	t	†		
3	102	57	64	7	44	†	23	†		
6	117	21	68	†	48	†	40	†		
9	55	†	41	t	37	†	16	†		

TABLE 2 LATERAL DIFFUSION OF METHYL BROMIDE FROM SURFACE-TREATED, COVERED PLOTS (DRY AND WET SOIL)

\* CT is the product, concentration (ppm) times time (days). † CT is less than 6,000 ppm days.

loam soils at Moreno was studied; in these experiments, the MB was applied deep in the soil in augered holes. The land was planted to sudangrass, and as the soil dried during the year, four 1pound cans of MB (stack is 18 inches high) were applied in a hole 5 feet deep in June and July. In October and January, the MB was applied as a single 1-pound can into each of four tightly clustered holes 3 feet deep.

Sampling stations were spaced at 2, 5, and 8 feet in opposite directions from the injection site with sampling points at each station 1, 3, 6, 9, and 12 feet deep. No covers were used.

In a preliminary experiment on April 2, four 1-pound cans of MB were placed in a single hole 5 feet deep. The soil was so wet (March 19 moisture profile in fig. 2) that diffusion in all directions was totally restricted. The MB seemed to reside as a "lump" at the application site, with only the nearest sampling points showing measurable concentrations of MB.

In the June experiment (June 2 moisture profile fig. 1), upward diffusion was almost totally restricted, but downward diffusion was rapid, undoubtedly facilitated by the impervious cover formed by the wet upper layers. In July (July 7, fig. 1), the upper strata had dried somewhat, and significant quantities of MB were detected at the 3-foot levels; downward diffusion was again rapid.

By late October, the sudangrass was extensively wilted and probably not significantly changing the moisture levels in the soil (Nov. 20, fig. 1). In the Oct. 29 experiment (four 1-pound cans of MB placed 3 feet deep) downward diffusion was rapid, but concentrations lethal to A. mellea were found also at the 3-foot depth. By this time, the heavy clay soil had developed a network of deep cracks, as deep as 20 inches, and escape through these cracks accounted for the low levels of MB found at 1 foot.

In January, after very little rain and no significant changes in soil moisture, the October experiment was repeated, but this time the plot was covered with a  $20 \times 20$  foot, 4-mil thick, polyethylene tarp to prevent immediate escape of MB from the cracks. Within the 28 days of measuring MB concentration, all sampling points within the volume of this plot received CT lethal to A. mellea except at the 1-foot depth 8 feet laterally from the charge hole. Here again, the cracks may be the reason since this point was near the edge of the tarp.

The analytical results for the June and October experiments are shown in table 3. The data show the concentrations in ppm of MB detected on the days indicated at depths of 1 to 12 feet from the soil surface and distances of

### TABLE 3 CONCENTRATION OF METHYL BROMIDE (PPM) IN SOIL ATMOSPHERES OF MORENO SILT LOAM SOIL AT VARIOUS DEPTHS AND DISTANCES AT VARIOUS TIMES AFTER APPLICATION

М	applied Jun	ne 2, 1969†		MB (ppm) applied Oct. 29, 1969†						
Depth and Pw*	Days after applic.	Later applic	al distance ation point	from (ft.):	Depth and	Days after	Lateral distance from application point (ft.):			
		2	5	8	Pw*	applic.	2	5	8	
	1					3	264	147		
	2					5	253	234		
	3					7	157	249		
1 ft./19%	7				1 ft./10%	12	71	200		
	10					24	lost	lost		
	15					33	23	61		
	25									
						3	2,630	530		
	1					5	1,760	760	44	
	2					7	1,120	830	155	
	3	15			3 ft./12%	12	863	630	155	
3 ft./27%	7	119				24	310	270		
	10	251				33	230	160		
	15	327								
	25	12								
						3	22,200	4,420	88	
	1	56				5	15,000	6,190	370	
	2	1,590				7	22,200	4,230	220	
6 ft./23%	3	3,350	32			12	4,170	2,880	580	
	7	3,790	250		6 ft/13%	24	1,670	1,360	170	
	10	2,790	400	15		33	1,060	660		
	15	1,5 <b>6</b> 0	360	36		40	780			
	25	870	320	90						
						3	39,000	11,700	15	
	1	4,590				5	32,300	11,000	37	
	2	18,400	5,800	16		7	20,500	10,900	50	
	3	13,100	3,600	38		12	6,470	4,740	360	
9 ft./19%	7	5,320	1,700	112	9 ft./20%	24	3,400	2,720	340	
	10	3,600	1,400	215		33	2,440	1,710		
	15	2,700	1,300	215		40	1,760			
	25	1,500	930	400						
12 ft./24%						3	1,850	410	140	
	1	42,300	64	16		5	35,300	860	170	
	2	45,100	3,000	11		7	lost	2,200	190	
	3	31,500	4,900	123		12	8,500	4,310	400	
	7	14,000	4,160	750	12 ft./26%	24	5,170	3,400	1,220	
	10	10,000	3,890	950		33	3,800	2,500	680	
	15	5,400	2,280	1,030		40	2,500			
	25	2,500	1,360	930				1		

\*  $P_w$  is weight per cent moisture in the soil horizon. † All blanks in table: samples analyzed, concentration MB was nil or trace.

2 to 8 feet from the point of application. The  $P_w$  at each depth was also recorded. Blank spaces in the table indicate nil or trace MB at these points. A striking feature shown by the data is the very rapid downward movement of MB in both experiments, although it was somewhat less for October than for June. Because upward diffusion was significant in the October trial, concentration gradients downward were less steep. The high concentration attained at 12 feet in both trials indicates that very deep penetration is possible when the MB source is placed deep in the soil.

The experiments described, and numerous other observations, show that even though fine-textured soils are practically impervious to diffusion of MB when wet, they can be sufficiently dried to allow adequate gas penetration for successful fumigations.

Another experiment performed in the fine-textured sandy clay loam soil at SCFS demonstrated that when soil, initially so wet that it allowed only poor diffusion of MB, is dried, there is a transition range in moisture wherein diffusion increases rapidly with small decreases in moisture. This would indicate that in this range, continuous pores are freed of water and results in greatly increased rates of diffusion.

Four  $20 \times 20$  foot plots with varied moisture contents were located within an area of 100 feet square. This SCFS soil was well characterized (table 1), and moisture measurements were made just prior to the experiment. From the data it was possible to calculate the open-pore volume for each 6-inch increment in depth, and from this the average per cent pore volume free of water for each plot in the 0 to 6 foot horizon. The data are shown in figure 7 and the terms defined in the legend.

The plots were covered with Saranex covers and styrofoam shaders and treated at the surface under the covers with 4 pounds of MB per 100 square



Fig. 7. Cumulative dosage (48 days) at the 6 and 9 ft. depths in four plots in the sandy clay loam soil with graded moisture contents at SCFS. Methyl bromide applied at 4 lb. per 100 sq. ft. at surface under Saranex cover.

feet of surface. Soils atmospheres were analyzed at the surface and at 3, 6, and 9-foot depths for periods up to 48 days.

The CT's attained in the four plots at the 6- and 9-foot depths in relation to per cent open pore volume are shown in figure 7. At the 6-foot depth, CT increased from 80K to 240K when pore volume increased from 19.8 to 30 per cent of the soil volume. It is interesting to note the increasing steepness of the curve as it passes through the points denoting plots II to IV. It is in the moisture range found in plots III and IV of this experiment, and others (e.g., October experiment, table 3), that we found that even the finest textured soils we studied became sufficiently open with continuous pores to allow ready diffusion of MB.

At the 9-foot depth, the CT attained in plot IV was five times that in plot I. This was to be expected, since CT 6 feet deep was the result of downward movement of MB from the soil surface, and the 6-foot depth may then be considered the MB source for the 9-foot depth. The much higher CT in plot IV at 6 feet therefore resulted in higher CT at greater depths and emphasizes the desirability of adequate drying of the soil to achieve deep penetration.

In summary, drying the soil greatly facilitated downward diffusion when MB was applied at the surface, when injected relatively shallowly with commercial application machines, or when placed in holes 5 feet deep beneath the surface. The beneficial effects of drying were equally apparent in sandy loam, sandy clay loam, and in silt loam soil.

### Effect of depth and pattern placement

Effect of depth, single point. Three plots at SCFS were arranged in a triangle with respect to each other in an area  $76 \times 80$  feet to try to obtain a homogeneous soil. The weight per cent moisture contents were 7, 9, 6, and 10 per cent at the 0.5, 3, 6, and 9-foot depths, respectively. Paired sampling stations were placed in opposite directions 3, 6, and 9 feet horizontally from the point of MB application, and each station had sampling points 0.5, 3, 6, and 9 feet deep from the soil surface. No covers were used. Each plot was treated with four 1-pound cans of MB, 2 feet deep in one, 3 and 5 feet deep in the other two. Soil atmosphere samples for analysis were taken 1, 3, 5, 11, and 22 days after the MB was applied. The plot with 3-foot deep placement had to be abandoned early in the experiment because of encroachment of water into the plot.

The data in figure 8 show the CT (in thousands) for 22 days at each sampling point for the plots with MB placement 2 or 5 feet deep. The results were, in part, qualitatively predictable; treatment 2 feet deep resulted in higher CT



Fig. 8. Cumulative dosage of methyl bromide (MB) attained in 22 days at depths of 0.5, 3, 6, and 9 ft. from a point application of 4 lb. placed 2 or 5 ft. deep. Note difference in vertical scale for the 0.5 ft. depth.

at the 0.5 and 3-foot levels at all horizontal distances from the MB charge, while the 5-foot deep treatment resulted in higher CT at the 9-foot depth. However the data at 6 feet, where CT were almost identical, again demonstrated the ready downward diffusion of MB (compare table 3) when it was placed beneath the surface in an otherwise undisturbed soil. In this experiment, CT lethal to A. mellea were attained at 6 and 9-foot depths to a radius of nearly 9 feet for both the shallow and deep placements.

## Effect of multiple point deep placement

A plot at SCFS was used to study the diffusion pattern of MB resulting from multiple point deep placement of MB sources.  $P_w$  ranged from 8.5 to 14.4 per cent depending on depth and position in the field; thus the soil in the entire plot was relatively dry and would allow good diffusion of MB.



Fig. 9. Plot plan for study of multipoint deep placement of methyl bromide (MB) at 1 or 4 lb. per charge hole 5 ft. deep. Central parallelogram test plot is surrounded by supporting sources of MB; sampling stations are located at a, b, c, d.

Two closely adjacent plots with similar soil conditions were prepared for the multipoint pattern treatment; a diagram of a plot is shown in figure 9. The injection holes were bored in rows 10 feet apart at staggered intervals 10 feet apart along the rows. The central parallelogram of four injection sites (1 to 4 in fig. 9) was the test plot; the other ten sites served as border treatments, thus the test plot could be considered to be within a field treated in this manner. Sampling stations (a to d in fig. 9) were placed along the long diagonal of the parallelogram 1, 3, 6, and 9 feet from a corner injection site. Stations a and bwould be used to study the expanding MB diffusion pattern from injection site 1, station c was central to the triangle of sites surrounding it, and d was the center of the short diagonal. Thus the latter two were at points where MB from adjacent sites would first overlap. Sampling points at each station were 1, 3, 6, and 9 feet deep.

One plot received a single 1-pound can of MB placed 5 feet deep at each of the 14 sites. The other plot was treated with four 1-pound cans per hole at the same depth. No covers were used. Samples of soil atmospheres were analyzed one, three, five, and seven days after treatment, thereafter every three to five days for a total of 35 days.

The time-vs-MB-concentration plot for all the sampling points in the 1pound per hole treatment is shown in figure 10. The vertical line at each point in the figure represents the range of concentrations present at the four stations a to d at the specified time and depth. For example, on day 25, at the 3-foot depth all stations had concentrations within the short vertical line at that point. At points without this line, concentrations at the four stations were nearly identical. This shows that from about the fifth day onward, MB had diffused to a series of equal-concentration planes, that after the twelfth day, the 9-foot depth had the greatest and the 1-foot the least. Such a gradient can be expected from laws of diffusion in



Fig. 10. Concentration of methyl bromide in soil atmospheres vs. time for treatment of 1 lb. per hole according to pattern in figure 9. Vertical lines at each point include concentrations at all stations (a to d in fig. 9) for the given time and depth.

this porous soil with zero concentration at the surface. It is interesting to compare this situation with that in figure 4B where the use of impervious cover resulted in equal concentration throughout the sampled soil mass.

Depending on depth and location, the concentrations of MB varied greatly during the first five days. To avoid complicating figure 10, most data for this early period are omitted. Concentration of MB at every sampling point rose to above 1000 ppm by the second day, and all points reached a maximum by the fifth day except at the 9-foot depth at stations c and d where the maximum was reached in seven days.

The fumigation with 1 pound of MB

per 100 square feet would have been lethal to A. mellea at all points in this plot including the 1-foot depth, although it probably was not lethal at the zero to 1-foot depth. However, with the large underground reservoir of MB, if a cover were used, probably lethal concentrations would have been attained at the surface also.

### Effect of dose

The effect of varying the dose on distribution of MB in soils was investigated with surface applications, single point, and pattern injections. In one experiment at Hinkley, four sampling stations were installed in a square plot encompasing 300 square feet of dry



Fig. 11. Effect of applied dose (1 lb. or 4 lb.) of methyl bromide (MB) on the concentration in soil atmospheres vs. time in Hinkley sandy loam. MB was applied as a gas under mylar cover.

soil ( $P_w$ , 4 per cent at surface and down to 5 feet) with sampling points at each station at 2-foot intervals from 1 to 9 feet deep. The plot was covered with a 2-mil Mylar tarp suspended a few inches above the soil on a network of wires and buried in the soil around the edges.

MB at 1 pound per 100 square feet was applied as a gas to the surface beneath the tarp, and one month later when all traces of MB had gone, the plot was treated again with 4 pounds per 100 square feet in the same manner. The data for the analysis of MB concentrations for both dosages is shown in figure 11. Increasing the dose resulted in more rapid diffusion because of greater concentration gradients, and also greater penetration with high concentration of the gas into deep soils. The maximum concentration attained at each depth for the 1- and 4-pound per 100 square feet treatments are shown in figure 12. The divergence of the two curves with increase in depth indicates



Fig. 12. The data of figure 11 plotted as the maximum concentration of methyl bromide attained at various depths by treatment at the surface under mylar cover with 1 or 4 lb. per 100 sq. ft.

that increasing the dosage progressively increases depth of penetration when MB is applied at the surface of the soil. Calculation of CT for the two treatments shows that the 4-pound treatment would be lethal to A. mellea at all depths, while with the 1-pound treatment, the CT was inadequate at the 7 and 9-foot depths.

The effect of dose when applied at a single point by deep injection was investigated at SCFS in three closely adjacent plots with very similar moisture characteristics. The three plots, designated A, B, C, were treated with 1, 2, and 4 pounds, respectively, of MB applied in 1-pound cans; the 2 and 4pound treatments were placed, 1 pound per hole, into closely clustered adjacent holes augered to the desired depth and can be considered point sources. In plots A and B, 1 and 2 pounds, the charge of MB was placed 3 feet deep, while in plot C, 4 pounds, the charge was 2 feet deep. Water had entered into the original 4-pound plot with placement 3 feet deep, and it became unsuitable for comparison with the other two. Plot C was part of another experiment, but the effect of dose on deep penetration is so striking that this plot is included in this discussion.

The  $P_w$  of the three plots at the 0.5, 3, 6, and 9 foot depths, respectively, were: plot A: 8.5, 10.1, 8.9, and 12.4 per cent; plot B: 5.0, 8.0, 9.4, and 13.7 per cent; plot C: 5.4, 8.7, 5.7, and 12.7 per cent. At these  $\mathrm{P}_{\mathrm{w}},$  this soil was dry and porous, and the three plots were quite similar. Sampling points in each plot had been installed 0.5, 3, 6, and 9 feet deep at horizontal distances of 3, 6, and 9 feet from point of application. Soil atmospheres were analyzed 1, 3, 5, 11, and 22 days after application. The data in figure 13 show on a linear scale the concentrations of MB in soil atmospheres for the first 11 days of the experiment. Each set of curves represent data obtained from equivalent points



Fig. 13. Concentration vs. time for deep placement of methyl bromide (MB) at a point for 1, 2, and 4 lb. per hole. Sampling point locations are designated by H and D. Thus, 3H, .5D is a point 3 ft. horizontally from point of application and .5 ft. deep from surface of soil. Letter E indicates effective dosage for *Armillaria* control, M indicates marginal dosage. Note change in vertical scale by factors of five for each horizontal distance at the three lower depths.

in the three plots. Fairly good proportionality exists between the dose applied and the concentration of MB in the soil atmosphere at a given point and date. Note the change in the vertical scale by a factor of 5 for each horizontal distance for the three lower depths.

When the curves were integrated for CT (ppm  $\times$  days) at each sampling point in the plots, the CT was clearly effective (E on the curve in fig. 13), marginal (M), or ineffective by our criterion that CT of about 6,000 is lethal to *A. mellea*. Concentrations less than 600 ppm were not used in calculating the CT, since it was shown previously that dosage response was too uncertain at these concentrations to be validly applied for such purposes.

An examination of the figure emphasizes the very efficient downward diffusion of MB obtained when the source is placed deep into undisturbed soil. At 3 feet horizontally, even the 1-pound dose was effective 9 feet deep. Increasing the dose increased the effective lateral distance: the 1-pound dose was not effective at any sampled depth 6 feet horizontally, while the 4-pound dose was effective or nearly so to a distance of 9 feet at all depths below 3 feet. The very low concentration obtained at 0.5 feet deep is due to rapid escape of the gas to the surface, since no covers were used.

Comparison of the dry plots of figures 4 and 5, also the wet plots in the same figures, gives an additional quantitative illustration of the effect of dosage applied on the concentration of MB attained in deep soils.

### A final observation on the effect of dosage

The experiment, in which the MB was put into the soil in a pattern of points using 1 pound of MB per 5foot deep hole, was discussed earlier and illustrated with figures 9 and 10. In the duplicate experiment, where 4 pounds of MB per hole were used, analytical data resulted in MB concentration vs. time curves similar to those in figure 10. but concentrations at all points were much higher. Since the two experiments were identical except for the MB quantities applied, each of the 16 sampling points in one plot (depths of 1, 3, 6, and 9 feet at each of the four sampling station a, b, c, d in fig. 9) had a point, equivalent in location and depth, in the other plot. If the concentration of MB at each sampling point in the 4-pound plot was compared with the concentration at its equivalent point in the 1pound plot, one obtained a set of ratios, concentration 4 pounds/concentration 1 pound, for all equivalent points on a specific sampling date. When these ratios were plotted vs. time, figure 14 was obtained. The shaded band in the figure includes all 16 possible ratios; thus for the sampling 16 days after



Fig. 14. Ratio of the concentrations of methyl bromide (MB) in soil atmospheres (4 lb/1 lb. treatment) at comparable plot locations resulting from treatment with 4 or 1 lb. MB per hole according to plan in figure 9. Shaded band contains all ratios from equivalent sampling points and dates in the two plots.

treatment, all concentration ratios lie between 4 and 5.5. A few specially aberrant points are indicated by the shaded dot in the figure. Thus, when the applied MB dosage was quadrupled, the concentration ratios, and therefore the CT, were increased by factors of four to seven; these ratios were sustained from the third day after treatment to the thirty-fifth day. More important, high CT at greater depths would be attained; after 35 days, the concentration 9 feet deep was still 2000 ppm in the 4-pound treatment, while it declined to 360 ppm in the 1-pound treatment.

#### Effect of type and thickness of tarp

A single experiment will be described here; other observations and conclusions may be derived from data already presented and will be discussed later.

A commercial fumigation of a strawberry plot was studied for deep penetration. Parallel plots 33 feet wide and 220 feet long were treated with 360 pounds per acre of MB applied by shanks 0.5 foot deep, 1 foot apart. The treated area was covered immediately by the same machine with polyethylene tarps 1, 4, or 6 mils thick. The soil was sandy loam with coarse sand in the 3 to 5 foot depth. Moistures throughout the area were quite uniform, the  $P_w$  averaging 6.5, 10.4, 7.8 and 8.5 per cent at the 0.5, 1, 3, and 5 foot depths respectively. Atmospheres obtained from the surface under the tarp and from depths of 1, 3, and 5 feet were analyzed 2, 20, 44, and 120 hours after treatment.

The CT that accumulated in five days at the surface and at the depths indicated is shown in figure 15. In this case, CT was calculated if MB concentration exceeded 100 ppm instead of, as usually, 600 ppm. Otherwise, there would be lit-



Fig. 15. Effect of polyethylene tarp thickness on dosage of methyl bromide (MB) (ppm  $\times$ days) accumulated in five days vs. depth for three thicknesses. Treatment was 360 lb/A by tractor-drawn machine with chisels 6 in. deep 1 ft. apart and the cover laid immediately by same machine.

tle comparative data since concentrations exceeded 600 ppm at the 3-foot depth only under the 6-mil tarp and in no case at the 5-foot depth.

The CTs under the 6-mil tarp were roughly triple those under the 1-mil tarp at all depths measured, and about 1.7 times those under the 4-mil tarp. After five days, the concentration at the surface was 360 ppm under the 6mil tarp and 20 ppm under the 1-mil tarp. At the 5-foot depth, also after five days, the concentrations of MB were 350 and 100 ppm under the same respective tarps. Thus, the heavier tarp, that more effectively retarded escape through the cover, allowed greater downward diffusion. However, the effect of kind of tarp material is even greater as will be noted in the discussion.

### DISCUSSION

Of the several methods available for bringing biocides into contact with soil pathogens, namely thorough mixing with the soil, movement with soil water.

and diffusion in the vapor phase, the only practical method for *A. mellea* control is via vapor diffusion. The first two methods are practical for treatment near the surface; however, as noted before, *A. mellea* is found as deep as roots will grow, and dispersion in the vapor phase in sufficiently porous soil is the only reasonable way to bring toxicants into contact with deeply buried inoculum.

Many soil conditions and characteristics are interrelated in soil fumigations. Characteristics such as composition, texture, and total porosity are measurable but fixed in a given soil. Total porosity as determined by compaction or mechanical cultural practices may feasibly be altered only in the upper few feet. In this study we have concentrated on the factors that are controllable or manipulative in their effect on deep penetration, such as confinement by covers, amount and method of toxicant application, and porosity as governed by moisture content. It is generally recognized that diffusion is the most important factor in the movement of a fumigant, which, in turn, is governed by porosity and moisture content.

Methyl bromide was chosen for this work because it is a good general biocide —a highly diffusible gas, relatively unreactive in the low organic content mineral soils encountered in California, not strongly sorbed by damp soils (Chisholm and Koblitsky, 1943), with a Henry's law constant favorable to broad dispersion (3.8 at 20° C). More important, since effective dosages (CT) of MB for A. mellea have been determined (Munnecke, Wilbur, and Kolbezen, 1970), the extent of effective treatment is delimited by a study of the diffusion patterns.

Determination of diffusion patterns by analysis of soil atmospheres has advantages over a once commonly used technique of determining the effect on buried biological specimens (fungi, insects, nematodes). Some advantages of the technique we used are: (a) soil structure is not disturbed: (b) it is possible to follow the continuous change of the fumigant concentration in the soil air by analyses as frequent and for any time duration desired; (c) variabilities of biological testing are eliminated; and (d) once a diffusion pattern is known, its effective range is determined for any organism whose response can be determined independently.

Earlier workers have analyzed soil atmospheres by chemical methods—for carbon disulfide (Fleming and Baker, 1935), for ethylene dibromide (Call, 1957); while Siegel, Erickson, and Turk (1951) used radioactively-labelled halogens to study the diffusion of 1,3-dichloropropene and ethylene dibromide. The current availability of gas chromatographic techniques (GC) make such analyses extremely accurate and rapid, and GC is finding increasing applications in soil vapor studies.

### Effect of soil moisture

The water solubility and sorption to solids of MB are relatively low, and these factors are not as important in the diffusion of MB as is the availability of continuous pores. It will therefore diffuse readily in wet soils if such pores are present (Goring, 1962, p. 73). Some moisture is desirable in sandy soils to prevent too rapid dispersion of the MB and loss to the atmosphere. The frequently confirmed observation that soil pathogens are more susceptible to fumigants in wetter soils may not be of great importance with A. mellea. We have often observed that A. mellea-infested roots in fairly dry soils remain wet and thus can partition MB into the water phase which presumably is required to exert its fungitoxic action.

Saturated soils are not fumigable since all pores are blocked with water. Diffusion through the water phase is extremely slow; Hartley (1960) estimated that for the same volume concentration, the rate is 10 to 30 thousand times more rapid in the vapor state than in the dissolved phase. In our experiment in the saturated fine-textured silt loam at Moreno (soil moisture March 19 in fig. 2), diffusion of MB was totally restricted. Similar results were obtained in this same soil at field capacity (June 2 in fig. 1 and table 3), where practically no MB diffused into the upper 3 feet of soil. Another example is shown in figure 4, where the high moisture content in the upper 3 feet in this sandy clay loam severely restricted diffusion to lower depths.

These and other observations demonstrate the futility of fumigating very wet soils (especially fine-textured silt and clay soils) and expecting adequate diffusion. Many workers comment on the difficulty of treating heavy clay soils. McBeth (1954), for example, avoided fumigating clays with moisture equivalent greater than 30 per cent and recommended increased dosage if moisture was greater than 25 per cent of field capacity. Bliss (1951) found that a moist clay layer at the 6-foot depth was a barrier to the movement of carbon disulfide beyond that depth.

When the wet silt loam at Moreno was dried, however, by a crop of sudangrass to the permanent wilting point, the soil became permeable to MB (October 29 in table 3 and November in fig. 1). The  $P_w$  had now been reduced to 10 to 13 per cent in the upper 6 feet of this soil. The data in figure 7 show that when the moisture in the sandy clay loam at SCFS had been reduced to  $P_w$  of about 13 per cent (plots III and IV in fig. 7) the soil became sufficiently porous for ready diffusion of MB.

Heavy clay and silt clay soils were not available for our studies, but from observations on the Moreno silt loam, these should present extraordinary difficulties in attaining adequate porosity. As the  $P_w$  approached the wilting point ( $P_w$  10 to 13 per cent, table 3) the Moreno plot developed a network of large cracks as deep as 20 inches. But the clods were penetrated by MB when escape through the cracks was prevented by use of a heavy plastic cover, and each clod was surrounded by a high concentration of MB. Heavy clay soils may have  $P_w$  at permanent wilt to about 30 per cent and porosity consisting only of very fine pores. It would seem that not all undisturbed soils are fumigable by gaseous diffusion alone.

Loam and sandy loam soils are ideally suited to fumigation by diffusion. In the Ramona sandy loam at UCR, MB diffused readily in mid and late summer without the previous planting of a desiccating crop. In such soils, with field capacities of 10 to 20 per cent  $P_w$ , and wilting percentage of 4 to 6 per cent (Hausenbuiller, 1972, p. 123) fumigation is possible at  $P_w$  well above the wilting point. For example, in the sandy loam at Hinkley, downward diffusion of MB was compared in the wet  $(P_w 14 \text{ per cent})$  and dry  $(P_w 4 \text{ per})$ cent) plots (fig. 3). The dry plot was at the permanent wilting point of the sudangrass crop. While diffusion to the 8 foot depth was quite restricted in the wet plot, considerable CT was obtained 5 feet deep. This wet plot was very nearly porous to good diffusion, and from experience we know that it would have been adequately fumigated at all analyzed depths if the MB had been applied as multiple point deep injections in otherwise undisturbed soil.

It is difficult or almost impossible to generalize for all soil types a single moisture content beyond which diffusion would be restricted. Yet a broad generalization may be made. The finetextured soils have a greater field capacity, but a greater percentage of this must be removed to yield porosity sufficient for good diffusion. All the soils in these studies, from the sandy loam at UCR and Hinkley to the fine silt and clay loam at Moreno and SCFS, became sufficiently porous for good movement of MB when  $P_w$  was in the neighborhood of 10 to 15 per cent. Since  $P_w$  is probably the simplest of analyses on a soil, this moisture can be a general guide for the fumigability of such soils. Duffusion may be adequate or even rapid at higher  $P_w$  (note 9 and 12-ft. depths in table 3), but the 10 to 15 per cent would ensure adequate porosity in such soils.

Soils should be dried by a deep-rooted crop as evaporative drying or gravitational flow are not adequate for the purpose in fine soils (figs. 1 and 2, Rackham *et al.*, 1968). In all our work, sufficient porosity was attained with a planting of sudangrass that was allowed to go to full wilt. Heavy soils may have  $P_w$  much greater than 15 per cent at wilt, but such soils were not studied. Raski *et al.* (1972) have cited data on the ability of sudangrass to dry soil down to 8 feet.

### Diffusion pattern

Various authors present conflicting reports on the shape of the control pattern resulting from placement of a fumigant beneath the soil surface. Call (1957) found the diffusion pattern to be skewed upward with ethylene dibromide, and concentration below an injection point was lower than that at an equal distance above. He found no effect of downward gravity flow. Youngson, Baker, and Goring (1962) noted no gravity effect with MB and chloropicrin. Siegel, Erickson. and Turk (1951) on the other hand, noted the pattern to be skewed strongly downward with 1,3-dichloropropene and ethvlene dibromide, and Fleming and Baker (1935) described the pattern of carbon disulfide as cone-shaped with apex near the injection point. Bywaters and Pollard (1937) and Higgins and Pollard (1937) noted that carbon di-

sulfide patterns were skewed downward and surmised that there may be some gravitational effect in very coarse soils. Schmidt (1947) noted that chloropicrin and DD mixture moved downward and laterally in a cone-shaped pattern in dry and wet soils, but distribution was more uniform in all directions in medium-moisture soils.

In our work it was difficult to assess unequivocally whether a gravitational or mass flow process was involved in the movement of the rapidly diffusing MB because of a sudden discontinuity of normal diffusion gradients at the soil surface. If the soil was tarped, concentration at the surface was affected by loss through the tarp; if untarped, the surface concentration was zero. Neither situation provided normal gradients which would be present if MB were diffusing through additional volumes of soil. Deep placement, however, reduces the effect of this variable, and a few illustrations will show the strikingly strong downward movement of MB when it is placed several feet into undisturbed soil. Also, anisotropy can confound our kind of studies where measurements may involve 10 to 12 vertical feet of soil.

When MB was placed 5 feet deep (open circles in figure 8), sampling points below the 5 foot depth, at the sampling station 3 feet horizontally from the point of application, received higher CT than those above. Thus, the CT attained in 22 days were 3.5K, 60K, 195K, and 160K ppm days at 1, 3, 6, and 9 feet deep, respectively. This illustrates the good deep penetration possible; however, it does not indicate preferred diffusion downward, because escape from the surface necessarily results in lower dosages near the surface.

The diffusion pattern derived from table 3 is similar to the cone shape described for carbon disulfide by Fleming and Baker (1935). Another example of a cone-shape pattern may be seen in figure 13. The sampling points 3H,9D and 6H,6D are within inches equidistant from the point of application, yet the point (3H,9D) more directly below the injection point received maximum concentration, double that of the 6H,6D point for all three dosages of MB applied. These and other data show that from a single point, MB will diffuse downward and laterally, but downward movement is more rapid than lateral.

When MB was placed into the soil as a number of single point injections arranged in a regular pattern, diffusion resulted, after several days, in a series of isoconcentration planes (fig. 10) as was predicted by Hemwall's (1962) mathematical model.

### Method of application

Surface or shallow chisel application of MB in fumigable soils will give adequate CT in deep soils only if impervious covers are used (figs. 4, 5, 11). If polyethylene is used as a cover, deep penetration is possible only in very porous soils (fig. 3), since this cover is a relatively poor barrier for escape to the atmosphere.

A recent trend in commercial fumigation is toward deep shank application (24 to 30 in.) with chisels 5 to 6 feet apart; the soil may not be or may be cross-ripped beforehand to a depth of 36 inches in two or more directions. This method can be superior to the more shallow applications, but it has some limitations. The requirement for a dry, porous soil is even more important, or clods will not be penetrated. Although the unbroken lumps have a vastly greater surface area exposed to the gas, the soil now also contains a great number of channels for rapid escape to the surface. The surface should be sealed as tightly as possible, perhaps by cultipacking and rolling. But with volatile fumigants the upper foot may be poorly fumigated. It is better that the treated area be covered by a tarp, a practice followed by many commercial fumigators. Fumigants less volatile than MB could probably be effectively applied in this manner, if deep treatments are desired.

In our observations, the best way is to apply MB at a point or at separated points in a pattern with the access hole filled and tamped to at least the degree of compaction of the otherwise undisturbed soil. The curves in figure 10 show that adequate fumigation was obtained at all points and all depths, even at 1 foot, by placing a pattern of MB sources 5 feet deep. In figure 8, the data show that deep penetration was obtained by placement of a single charge of MB either 2 or 5 feet deep. Total dosages were nearly identical at the 6-foot depth, and, as expected, the 2foot placement produced better fumigation near the surface. But both depths of placement resulted in nearly adequate fumigation 6 and 9 feet deep to a radius of nearly 9 feet. Figure 13 also illustrates the excellent deep penetration by MB from point charges placed 2 or 3 feet deep; and table 3 shows that, while placement 3 feet deep resulted in significant amounts of MB near the surface, good fumigation was obtained to depths below 12 feet.

We concluded from these and other observations that placement into an access hole at the 5-foot depth originally investigated was not necessary, but that depths of about 3 feet were adequate to obtain diffusion to depths of 12 feet in moderate moisture soilswhile at the same time attaining reasonable concentration at the 1-foot depth. Greater depths could be used, for example, when inoculum is suspected deep in land-filled stream beds or old planting sites. Furthermore, both a deep and an additional shallow placement could be used to place a charge of MB above and below an impervious horizon, such as a clay lens, or to obtain

better control deep and near the surface.

If MB is applied near the surface, deep penetration is limited by poorly permeable horizons. The effect of such horizons is magnified in the deeper regions, since the MB entering each horizon is the source for the one below it. Deep placement then is, in effect, a "head start" for deep penetration. Furthermore, escape to the atmosphere is slowed by the thick soil cover.

As a result of these studies, commercial rigs assembled on farm tractors have been developed that can rapidly auger holes to a desired depth, inject a measured amount of MB, and compact the access hole leaving the surrounding soil otherwise undisturbed.

### Effect of covers

Perhaps the greatest boon to good fumigation practice would be the development of economical tarping materials less pervious to MB than polyethvlene. Such materials are available commercially, but at the present time, costs are prohibitive, and the 11 to 13 foot widths required for field application machinery are not manufactured. If such covers were used, less toxicant would be needed, and loss to the atmosphere would be reduced. A comparison of the effect of the use of impervious covers, or polyethylene, or no cover at all, can be illustrated with the following examples:

When MB was applied at the surface under impervious covers in the dryplot experiments illustrated by figure 4 (Saran cover), figure 5 (Saranex), and figure 11 (Mylar), the concentrations at the surface and at all depths in the plot converged, as time passed, toward equal concentrations. The concentrations at the surface and the upper regions remained high and generally higher than in the greater depths. Thus, in the dry plot in figure 5, concentration gradients were such

that MB was diffusing downward for the entire 50 days of testing, and the curves converge to a common concentration.

When the more pervious polyethylene was used as cover, the surface concentration decreased rapidly (dry plot in figure 3), and after about five days, the concentration gradients in the figure show that movement of MB was upward toward the surface. In the dry plot, the effect of loss through the cover is not as pronounced as it is in less porous soil (wet plot in fig. 3) since the underground reservoir of MB more readily diffuses upward to maintain concentration under the cover.

Increasing the thickness of polyethylene cover has a beneficial effect (fig. 15), but in this commercial fumigation at 360 pounds per acre with shanks 6 inches deep and 1 foot apart, a concentration of 600 ppm was attained 3 feet deep only under the 6-mil cover and in no case at 5 feet deep. The use of better tarp materials would have a much greater effect than increasing the thickness of polyethylene. Furthermore, disposal of waste tarp material is becoming an increasingly serious problem.

The use of polyethylene as cover is intermediate in effect to that when impervious cover or no cover at all is used. In the deep placement experiment where no cover was used, upward diffusing MB resulted in CT just adequate for A. mellea control at the 1foot depth. However, if concentration gradients from the 3-foot depth upward are compared under stabilized conditions (e.g., the fifteenth day, in fig. 10, and the dry plot, fig. 3), they are steeper when no cover was used. Concentration in the upper few inches is certainly negligibly low. Thus, with the highly volatile MB, while deep placement is ideal for deep penetration, little or no control can be expected in the upper foot without a cover. The decision to cover then depends on the need to control in this near-surface region.

Occasionally, irrigation or sprinkling of the soil surface has been recommended as a seal following fumigation. Lear (1951) attained good control of nematodes with Dowfume G when 0.5 to 1 quart per square foot (0.2-0.4 in.)of water was applied within 30 minutes of the fumigation. We found that basins filled with 1 to 4 inches of water formed a significant barrier to escape of MB. In our years of conducting these experiments, occasional rains of a few tenths to over an inch per day fell on uncovered plots. If this wetting formed an effective barrier, one would expect the concentration decay curves (e.g., 1-foot depth, fig. 10) to become more

horizontal during and directly after such rains. We have not seen this effect, and it seems that wetted soil is a barrier only if sufficient water to puddle it is added.

### Effect of dose

Most authors conclude correctly that increasing the amount of toxicant applied increases the range of control, while some conclude that increasing the dosage increases the persistence of high concentration within a volume of soil but does not increase the range. Figures 11 to 14 illustrate in several ways that increasing the amount of applied MB increases the concentration and persistence at a given point and also increases the range of adequate fumigation.

### CONCLUSION

Field soils can be successfully fumigated with deep penetrations of MB if the soil type and its moisture content is considered—along with the method of application and the careful use of cover.

It is most important that an A. mellea-infested area be completely delimited. Rackham et al. (1968) describe in detail such a procedure. Dead or diseased plants, as well as large roots near the trunk and surface, must be removed and the soil replaced and compacted.

Soil must be fairly dry. If the weight per cent moisture of soils, from the more coarse sandy loams to the finetextured clay and silt loam soils, is 10 to 15 per cent or less, these soils are permeable to MB. Sandy soils, even at field capacity, may have moisture content less than this, but they will be permeable. Soils can be dried over the summer with a desiccating crop such as sudangrass or safflower and fumigated in the fall and early winter months. California climate, with little summer rain, is very favorable to such treatment. The crop should be removed and the stubble disked to smooth the soil surface.

The MB can be applied by deep shank injection about 24 to 36 inches deep, with shanks 5 to 6 feet apart, and the shank channels tightly compacted. A better way is to apply the MB into a pattern of holes 10 to 12 feet apart in a square or diamond pattern about 3 feet deep and to tightly compact the access hole. Otherwise, the soil is undisturbed. A machine has been developed to apply MB by this latter method, but its use is still relatively limited.

Earlier attempts to apply liquid MB through the bore of a mechanically driven screw auger were not satisfactory, because the sudden high pressure developed when MB contacted the soil caused high losses before adequate compaction of the access hole was accomplished. Although improvements have been made for using liquid MB, the recent development of a thixotropic MB gel, which evolves MB more slowly, offers intriguing possibilities for applying MB in this manner. A serious problem, however, is that the gel may completely block the tubing in the application equipment.

A convenient method for treating small areas or isolated infected sites is by placing chilled cans of MB, punctured at the top, into holes drilled by auger, then filling the holes with compacted soil. This method can also be used to retreat spots in treated fields where infection recurs as it is wishful thinking to expect total eradication from an economically reasonable treatment. The high phytotoxicity of MB must always be considered, and deep spot placement of MB must be at least 15 feet removed from any living plant.

Neither method of deep placement will yield adequate dosages of MB in the top foot or so of the soil. The treated area should be tarped, and the tarp should be left in place for at least a week. There may be situations where the need for tarping may be circumvented, such as when a less volatile material is used for treatment near the surface together with deeply placed MB for deeper fumigation. Or, as is successful with certain nematodes, the upper foot of soil is thoroughly desiccated (lethal to some nematodes), and MB is applied deep for control from 1 foot downward. Good compaction of chisel tracks and the soil surface would

be required when no tarps were used.

The dosage should be 600 to 800 pounds per acre. When the MB is applied in augered holes at 10- to 12-foot spacing, this would be 2 pounds per hole. Data acquired thus far indicate that grape vineyards require at least the higher dosage.

The advantage of more impervious covers is so evident from data presented here, for both surface and deep treatments, that added cost would be warranted when one considers the relatively mediocre fumigations so often obtained with polyethylene. Better confinement, by whatever method of application, would require less MB and losses to the atmosphere would be reduced. Deep placement, as for A. mellea control, is ecologically and economically desirable, since several feet of soil forms a cover in addition to the polyethylene. Benefits of better plastic materials would be especially noticeable for surface treatments, such as for strawberry preplant fumigations.

Deep fumigation with methyl bromide is technically feasible and economically justified in areas where deep rooted crops are attacked by *A. mellea*. However, equipment is needed to rapidly and economically place MB deep in the soil without undue loss during the process, along with more impervious tarping materials than are now available.

### ACKNOWLEDGMENTS

The authors wish to thank the Dow Chemical Company, Great Lakes Chemical Corporation, and Tri-Cal Incorporated for financial support and donations of fumigant chemicals and tarping materials. We thank them also for their interest and cooperation in conducting field experiments.

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