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Case Studies

Health Hazard Evaluation of Methyl Bromide Soil Fumigations

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Plant Protection and Quarantine (PPQ) officers of the U.S. Department of Agriculture's Animal and Plant Health Inspection Service apply a variety of soil fumigants and post-emergence herbicides in their attempts to eradicate witchweed from the United States. They use methyl bromide to spot fumigate small, isolated areas of soil contaminated by witchweed.

Witchweed (*Striga asiateca*) is a parasitic annual that can severely damage corn, sorghum, sugar cane, dryland rice, and more than 60 other grass species.[1,2] After its seeds germinate, witchweed penetrates the roots of host plants, robbing them of water and nutrients.[2] Though witchweed is one of the most serious crop pests hindering cerealcrop production in Africa, the Middle East, and Far East, eastern parts of North Carolina and South Carolina are the only places in the Western Hemisphere where it has been found.[1]

A PPQ spot fumigation involves puncturing a pressurized can of 98 percent methyl bromide and 2 percent chloropicrin under a plastic covering or tarpaulin. The first steps of a spot fumigation are clearing away all debris and, if possible, tilling the soil. Tilling is thought to increase the likelihood that methyl bromide will penetrate the soil and contact buried witchweed seeds. When tilling is not done, a spot-fumigated area may include knocked down corn stalks, corn stubble, or heavy grass cover.

A narrow trench is dug around the perimeter of a 10-foot by 15-foot area to be fumigated, and a 4-foot section of 4-inch diameter polyvinyl chloride pipe having three sections cut from it is placed near the center. Next, a 1.5-pound can of Brom-o-gas® (Great Lakes Chemical Corporation, West Lafayette, Indiana) is placed in the pipe on top of a block of wood having a nail sticking through it. A sheet of clear 6-mil plastic having a thickness of 0.15 millimeters is then laid over the area, and its edges are covered with dirt to make a secure enclosure (Figure 1). The can punctures when pressed against the nail, and the pipe acts like a trough, holding the released methyl bromide and chloropicrin until they evaporate completely. A PPQ officer leaves the site immediately afterward.

Though done on a smaller scale, a PPQ spot fumigation is similar to other tarpaulin methods used to apply methyl bromide to soil before planting crops (e.g., tomatoes, strawberries, tobacco, and peppers).[3–5] Releasing a 1.5-pound can of Brom-o-gas under a tarpaulin covering 150 square feet equals an application rate of 436 pounds of methyl bromide per acre (490 kilograms/hectare), which is similar to the rates recommended for larger tarpaulin methods.[5,6]

The agricultural use requirements of methyl bromide prescribe that workers must wait at least 48 hours before removing a tarpaulin. Because of concern for the health risks associated with overexposure to methyl bromide, the director of the witchweed eradication program requested a NIOSH health hazard evaluation of their spot-fumigation process. The primary purpose was to evaluate the adequacy of the 48-hour waiting period by estimating the time needed for methyl bromide

FIGURE I
Preparation of a 10-foot by 15-foot spot-fumigation site on tilled soil.

and chloropicrin to decay to "safe levels." Air sampling methods, findings, and recommended improvements to the process are described in this case study.

Methyl Bromide

Methyl bromide (CAS number 74-83-9) is a broad-spectrum, restricted-use pesticide used to control insects, nematodes, weed seeds, and rodents.[7] Approximately 27,000 tons of methyl bromide are used annually in the United States for soil fumigation (87%), commodity and quarantine treatment (8%), and structural fumigation (5%).[3]

Human Health Effects

Methyl bromide is a colorless, nonflammable gas that is odorless and tasteless at air concentrations considered unsafe.[8,9] Odor thresholds reported for methyl bromide range from 20 to 1000 ppm.[10] Methyl bromide is a pulmonary irritant and neurotoxin. Short-term inhalation exposure may cause headache, dizziness, nausea, vomiting, blurred vision, slurred speech, convulsions, and death. Short-term inhalation exposure to high concentrations may cause lung irritation resulting in congestion with coughing, chest pain, and shortness of breath. The onset of lung effects may be delayed.[11]

Prolonged or repeated exposures to methyl bromide may cause a variety of central nervous system symptoms including visual disturbances, slurred speech, numbness of the arms and legs, confusion, shaking, and unconsciousness.[11] Neurological signs and symptoms may be delayed for several hours to a few days after an exposure has ended.[12]

Methyl bromide may be absorbed through the skin and cause systemic toxicity.[5,8] Skin contact with liquid methyl bromide may cause irritation.[7,9] Prolonged skin contact may cause burns and blistering.[5] To avoid prolonged skin contact, applicators are advised not to wear tight clothing, jewelry, gloves, or boots.[7]

Liquid methyl bromide may cause severe corneal burns, but its vapors do not appear to be irritating to the eyes.[9]

Occupational Exposures

NIOSH considers methyl bromide a potential occupational carcinogen.[13,14] According to an earlier policy, this meant occupational exposures to potential carcinogens should be controlled to the lowest feasible level. Based on the limit of quantitation of the NIOSH analytical method used at the time of this policy, the lowest feasible level was 4.7 ppm.[13] The NIOSH carcinogen policy was changed in 1995. According to the revised policy, NIOSH RELs will be adopted for potential occupational carcinogens, but one has yet to be adopted for methyl bromide.

Based on results from an inhalation study done using laboratory rats, the ACGIH® TLV® for methyl bromide was reduced in 1997, to a TWA concentration of 1 ppm with a skin notation.[15] The previous TLV was 5 ppm. The OSHA PEL for methyl bromide is a ceiling limit of 20 ppm with a skin notation.[16] The NIOSH immediately dangerous to life or health (IDLH) level is 250 ppm.[14]

Most published reports of human health effects following methyl bromide exposure concern its use as an agricultural fumigant.[6,17–21] One author reviewed reports published between 1953 and 1981 describing 60 fatalities and 301 cases of systemic poisoning related to fumigant uses of methyl bromide.[18] A report was also published containing descriptions of six severe intoxications and four fatalities that occurred in California between 1957 and 1966 in the food processing industry. The products handled were nuts, fruits, and grains.[19] An air concentration of 100 ppm was estimated from reconstructed conditions at two of the work sites.

A report was published describing the acute respiratory and neurological symptoms of four unprotected workers who removed plastic tarpaulins after a soil-injection application of methyl bromide.[6] The fumigant, 98 percent methyl bromide and 2 percent chloropicrin, had been applied ten days earlier to six acres at a rate of 350 pounds per acre. Though chloropicrin is meant to be a warning agent, none of the workers reported sensing immediate irritation or an odor. The author concluded that 2 percent chloropicrin could not be relied on as a warning agent and recommended that a self-contained breathing apparatus (SCBA) be worn when methyl bromide exposures exceed 5 ppm.

Neurobehavioral function was studied among soil fumigators exposed to methyl bromide at an average air concentration of 2.3 ppm.[21] The workers reported a significantly higher prevalence of 18 symptoms consistent with methyl bromide toxicity than non-exposed workers. Also, the fumigators did less well than non-exposed workers on 23 of 27 behavioral tests. They also did significantly less well on a finger sensitivity test and one of cognitive performance. The authors concluded that low-level methyl bromide exposures may produce slight neurotoxic effects.

Chloropicrin

Chloropicrin (CAS number 76-06-2) is a restricted-use pesticide used as a soil and enclosure fumigant.[7] It is a colorless, oily liquid that is a severe irritant of the eyes, mucous membranes, skin, and lungs.[22] Chloropicrin causes eye irritation beginning at 0.3 to 0.4 ppm, and its odor threshold is approximately 1 ppm.[8,23,24] An air concentration as low as 1.3 ppm may cause respiratory irritation.[24]

Because chloropicrin is an irritant at low levels, it is added as a warning agent to odorless fumigants like methyl bromide. However, "experience has shown that chloropicrin vapor may disappear before methyl bromide vapor and therefore the warning properties are lost."[9]

The NIOSH, OSHA, and ACGIH exposure limits for chloropicrin are a TWA of 0.1 ppm.[13–16] The EPA recommends that workers wear respiratory protection when chloropicrin exposures are 0.1 ppm or greater.[7] When this level is exceeded at any time, an exposed worker must wear an air-purifying, organic-vapor respirator, a SCBA, or a combination supplied-air respirator with auxiliary SCBA.

The NIOSH IDLH level of 2 ppm is based on acute inhalation toxicity data in workers and animals.[14,25] IDLH documentation for chloropicrin references a 1931 article in which the authors stated "a few seconds exposure to 4 ppm renders a man unfit for action."[25]

Methods

The EPA requires that workers wear respiratory protection when methyl bromide exposures exceed 5 ppm.[7] When this level is exceeded at any time, an exposed worker must wear a SCBA or a combination supplied-air respirator with auxiliary SCBA. Thus, for this study, a safe waiting period before removing a spot-fumigation tarpaulin was specified as the time needed for the methyl bromide level under the tarpaulin to reach 5 ppm or less.

A brief pilot study of two spot fumigations was conducted first, and air samples were collected only for methyl bromide. The pilot study's purposes were to gain insight to the methyl bromide levels under tarpaulins and to estimate a sampling duration that would not result in overloading the sampling media. Low-flow sampling pumps ran at 0.02 liters per minute (L/min) for one hour. Air samples were collected at 27 hours and 47 hours after the spot fumigations were started. The soil was not tilled.

A few weeks after the pilot study, six spot-fumigation sites were started simultaneously on freshly tilled, sandy soil, and both methyl bromide and chloropicrin measurements were taken. Measurements were taken first above and below the tarpaulins at 15 minutes after starting. Air samples were collected above each tarp to learn whether 6-mil plastic prevented methyl bromide from leaking through. Methyl bromide and chloropicrin measurements were taken under all six tarpaulins after six hours and daily for five days after the start, and under three tarpaulins after two weeks. To measure methyl bromide, sampling pumps ran at 0.02 L/min for durations ranging from 5 minutes, soon after the start, to 60 minutes on the last sampling day. To measure chloropicrin, sampling pumps ran at 0.1 L/min for durations ranging from 12 to 60 minutes.

To sample under a tarpaulin, a slit smaller than the diameter of a sampling tube was cut in the plastic. The tip of each tube was carefully inserted through the slit so that the plastic sealed around it. The inlet of each tube was then positioned in the space between the surface of the tarpaulin and the soil. After each sampling period, a sampling hole was patched using tape.

Five days after starting, one tarpaulin was removed, and two others were cut for aeration. One tarpaulin was cut once in the center along its entire length and perpendicular to the wind's direction. The other was cut at three equidistant locations along its width and in the same direction as the wind. Twenty-four hours later, methyl bromide and chloropicrin measurements were taken above the soil where the first tarpaulin had been and under sections of the two cut tarpaulins.

Methyl bromide air samples were collected and analyzed according to the revised version of NIOSH Method 2520.[26] (Problems with the first version of Method 2520 included reduced adsorption capacity at high humidity, difficult-to-prepare standard solutions, sample instability, decreasing recovery as loading decreased, and an insufficiently low quantitation limit.[27] Each sampling train consisted of three tubes in series, a drying tube holding 9 grams of sodium sulfate, a 400-milligram charcoal tube, and a 200-milligram charcoal tube. Sampling trains were connected by tubing to low-flow sampling pumps. Direct-reading detector tubes were also used. Dräger tube 3/a for methyl bromide (part number 6728211) measures methyl bromide concentrations from 3 to 100 ppm.[28]

The revised version of NIOSH Method 2520 did not solve completely all the problems of its predecessor. Thus, steps were taken to samples cold before analysis and keep the time as short as possible between sample collection and analysis. Immediately after a sampling period, all sampling tubes were capped and put in an insulated, 1-quart cooler containing cold packs. When the NIOSH researcher arrived at his motel room, sampling tubes were removed from the cooler and put in a refrigerator's freezer. Methyl bromide samples and cold packs were put back in a cooler just before being mailed overnight to the NIOSH laboratory.

Air samples for chloropicrin were collected and analyzed using a 1991 OSHA stopgap method for which recovery and storage stability studies were completed. Each sampling train consisted of two XAD-4 tubes connected by tubing to a low-flow sampling pump. Samples were analyzed using a gas chromatograph having an electron capture detector.

Results and Discussion

During the pilot study, temperatures under the two tarpaulins ranged from 17°C (62°F) to 23°C (74°F). Light rain started on the evening of the first day and ended the next morning. Despite the rain, the soil under both tarpaulins remained dry.

Methyl bromide air concentrations under pilot-study tarpaulins were approximately 23,000 ppm and 26,000 ppm after 27 hours and 16,000 ppm and 18,000 ppm after 47 hours. All of the methyl bromide was collected on the first charcoal tube of each sampling train; none broke through to any of the backup tubes.

On most days of the six-tarpaulin study, the weather was sunny and hot; daytime temperatures were in the 90s. However, a 2-hour thunderstorm brought a half-inch of rain on the evening of the second day, and an inch of rain fell during the ninth and tenth days of the study.

Because daytime temperatures under the tarpaulins ranged from 49°C (120°F) to 60°C (140°F), air sampling results were corrected to a standard temperature of 25°C.[14,15] As with the air samples of the pilot study, all methyl bromide was collected on the first charcoal tube of each sampling train.

Table I shows the sampling results for the six-tarpaulin study. Average methyl bromide air concentrations ranged from 15,000 ppm, measured 24 hours after the start, to 50 ppm after five days.

Average chloropicrin air concentrations ranged from 340 ppm, measured 15 minutes after the start, to 0.6 ppm after six days. Chloropicrin levels measured on the first four days of the study exceeded its IDLH level of 2 ppm.[25] An air concentration of 0.6 ppm may be sufficient to cause eye irritation and warn of possible methyl bromide exposure if a tarpaulin is removed too soon.[8,23,24]

Data in Table I show steadily declining chloropicrin levels under intact tarpaulins. However, methyl bromide levels do not share this tendency. For example, the average methyl bromide air concentration after six hours (3600 ppm) is less than the average concentration after 15 minutes (13,400 ppm), but the average concentration after 24 hours (15,000 ppm) is greater than either of these concentrations.

One possible reason for the apparent discrepancy in the methyl bromide data concerns problems encountered with the shipment of the first set of samples. Methyl bromide samples collected on the first three days of the six-tarp study were collected on a Thursday, Friday, and Saturday. They were mailed together that Saturday evening for Sunday delivery. Unfortunately, they did not arrive at the NIOSH laboratory until Monday. When the cooler was opened, several tubes were found to have warmed to room temperature. The problem caused by the delay in receiving the cooler was compounded by a leaking cold pack. The cooler's spout had torn the cold pack. These problems emphasize the importance of keeping methyl bromide samples cool and analyzing them as soon as possible after collection.

Because of the sample shipment problems, methyl bromide levels reported for 15 minutes, 6 hours, 24 hours, and 2 days after starting may be underestimates. By comparing the methyl bromide levels of the pilot study, underestimation may be the greatest for samples collected at 15 minutes and 6 hours.

Methyl bromide levels under three intact tarpaulins at six days after starting are greater than any of the levels measured during the three previous days. Methyl bromide air samples taken at 5 and 6 days were analyzed together. The unexpectedly high concentrations may have resulted from a laboratory calculation error or an error in standard preparation.

Fifteen minutes after starting the six-tarpaulin study, 30-minute methyl bromide measurements taken directly above each tarpaulin ranged from none detected (one air sample) to 21 ppm. Like other methyl bromide measurements made on the study's first day, these air concentrations may also be underestimated. The limit of detection was 2 ppm.

After large-scale tarpaulin applications of methyl bromide, California Environmental Protection Agency (EPA) requires that unprotected workers not enter an application area for a minimum of five days. After five days, workers are allowed to aerate a treated area by mechanically cutting tarps using a tractor-mounted cutting wheel. Twenty-four hours later, unprotected workers are allowed to remove the tarpaulins.[29]

The day after removing a tarpaulin, methyl bromide and chloropicrin were not detected directly above the soil. The limits of detection were 6 ppm for methyl bromide and 0.0002 ppm for chloropicrin. However, 24 hours after cutting two spot-fumigation tarps, methyl bromide air concentrations of 80 ppm and 170 ppm were measured under sections of plastic. (Because of a problem with the methyl bromide samples collected six days after the start, these air concentrations may be overestimated.) Chloropicrin air concentrations under these tarps were 0.002 ppm and 0.003 ppm. These levels are below those expected to warn of possible methyl bromide exposure.[8,23,24]

TABLE I

Methyl Bromide (CH_3Br) and chloropicrin (CCl_3NO_2) air concentrations (ppm) under the tarpaulins of six spot-fumigation sites

Time after starting	Site 1		Site 2		Site 3		Site 4		Site 5		Site 6		Average	
	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2	CH_3Br	CCl_3NO_2
15 Minutes	14,800	270	17,100	360	8200	—	11,800	210	13,500	220	15,200	660	13,400	340
6 Hours	2900	84	4000	120	3400	57	4300	60	3900	62	3300	26	3600	68
24 Hours	13,900	24	16,400	26	16,500	21	15,800	18	14,700	26	12,800	14	15,000	22
2 Days	8700	4.6	8800	5.0	10,800	7.2	9500	6.0	8500	7.5	10,000	3.6	9400	5.6
3 Days	190	5.2	170	3.2	160	6.4	160	6.0	190	8.5	130	3.3	170	5.4
4 Days	70	3.9	100	3.0	90	3.8	100	3.2	130	4.8	130	1.8	100	3.4
5 Days	60	1.1	40	0.6	40	0.6	50	0.6	60	0.7	40	0.5	50	0.7
6 Days	ND[A]	ND[A]	800	1.0	80[B]	0.002[B]	640	0.9	170[B]	0.003[B]	610	0.05	680[C]	0.6[C]
2 Weeks	—	—	ND[D]	ND	—	—	ND[D]	ND	—	—	ND[D]	ND	ND	ND

ND means none detected.

[A]Tarpaulin removed completely 24 hours earlier. Samples taken directly above soil.

[B]Tarpaulin cut for aeration 24 hours earlier.

[C]Average of three values.

[D]Limit of detection for charcoal-tube sampling method was 7 ppm. Detector-tube sampling showed methyl bromide present at less than 5 ppm.

Two weeks after starting the spot fumigations, methyl bromide and chloropicrin were not detected under three intact tarpaulins. The limits of detection for these laboratory-analyzed samples were 7 ppm for methyl bromide and 0.0001 ppm for chloropicrin. However, detector-tube sampling showed that methyl bromide was present at levels less than 5 ppm.

Conclusions

Methyl bromide levels under spot-fumigation tarpaulins that were measured at the end of the 48-hour waiting period far exceeded the study's 5 ppm *safe level*. Chloropicrin levels exceeding its NIOSH IDLH level were measured for four days after the start.

Some methyl bromide levels may have been affected by sample shipment or analysis problems. Consequently, care is needed when doing air sampling for methyl bromide to ensure that NIOSH Method 2520 is followed completely.

Methyl bromide measurements taken directly above tarpaulins showed that the gas leaked through 6-mil plastic. This is not an important health risk for PPQ officers, because they leave immediately after puncturing a can of methyl bromide. However, methyl bromide leakage may be important to workers involved in large-scale tarpaulin applications.

Residual air concentrations of methyl bromide were measured under tarpaulins cut 24 hours earlier, but chloropicrin levels were too low to provide warning. Thus, whatever the size of the area treated, exposures to pockets of methyl bromide remaining under poorly aerated section of cut tarpaulins should be considered a health risk for which there may be no warning.

Recommendations

The following changes were recommended for improving the health and safety aspects of PPQ spot-fumigations:

- The waiting period before removing spot-fumigation tarpaulins should be extended to longer than 48 hours. Because soil and environmental conditions vary, a definite waiting period cannot be defined. However, a week or longer may be necessary.
- Regardless of the time that has passed since starting a spot fumigation, measurements should be made before removing a tarpaulin using detector tubes or some other direct-reading method to ensure methyl bromide levels are less than 5 ppm.
- Chloropicrin should be relied upon as a warning agent for methyl bromide only at the start of a spot fumigation. This recommendation agrees with those of other researchers who have reported that chloropicrin may not be detected or be present though toxic levels of methyl bromide are present.[6,9,18]

Many atmospheric scientists believe that methyl bromide is an important ozone-depleting chemical. Thus, at the Ninth Meeting of the Parties to the Montreal Protocol in 1997, decisions were made that developed countries should stop using methyl bromide in 2005 and that developing countries should stop all use in 2015.[3] The Montreal Protocol is an international treaty developed in the late 1980s to control world production and use of ozone-depleting chemicals. Under the Clean Air Act, the U.S. EPA took steps to phase out methyl bromide use in the United States. The EPA froze U.S. production and importation of methyl bromide at 1991 levels. Beginning in 1999, methyl bromide production and importation will be reduced gradually until 100 percent reduction occurs on January 1, 2005.[3] Quarantine, preshipment, and critical agricultural uses of methyl bromide are exempt from any control measures.

The approaching ban on methyl bromide has caused evaluation of replacement soil fumigants. The EPA has reported that methyl iodide equals methyl bromide in controlling plant pathogens and weeds, and it will not adversely affect atmospheric ozone levels.[3] Though methyl iodide may benefit atmospheric ozone levels, the health risk to pesticide handlers may not change. Like methyl bromide, methyl iodide has poor warning properties.[8] Also, occupational exposure limits are lower for methyl iodide. Both the NIOSH REL and the ACGIH® TLV®-TWA are 2 ppm with a skin notation.[13–15] NIOSH considers methyl iodide a potential occupational carcinogen.[13,14] The NIOSH immediately dangerous to life or health level is 100 ppm.[14] The OSHA PEL for methyl iodide is 5 ppm with a skin notation.[16]

When evaluating other soil fumigants, an important consideration should be the health risk associated with worker exposures to potential replacements and any warning agents. The EPA has promised that an evaluation of methyl iodide's toxicity will be added to its methyl bromide phaseout web site when the information is available.[3]

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