

Methyl Bromide Emissions from a Covered Field: III. Correcting Chamber Flux for Temperature

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ABSTRACT

An experiment was conducted to investigate the environmental fate and transport of methyl bromide (MeBr) in agricultural systems. Part of this experiment involved the use of three flow-through chambers to estimate the MeBr flux through a sheet of clear polyethylene plastic covering the field. Using the chamber data, the total mass lost to the atmosphere was estimated to be 96% of the applied mass, and the results were highly variable between chambers (i.e., standard deviation of 298 kg or 35%). The air temperature inside the chamber was found to be much higher than the air temperature outside and was highly correlated with the diurnal variation in incoming solar radiation. Since the diffusion through polyethylene film was found to be strongly dependent on the temperature, a method was developed to correct the chamber flux density data for enhanced diffusion caused by increases in the temperature inside the chamber. After correcting for temperature, the estimated total MeBr emission was reduced to approximately 59% (21% standard deviation) of the applied amount, which is about 5% less than was measured using other methods. When chambers are used to measure volatilization of MeBr or other fumigants from fields covered with a sheet of polyethylene plastic, the chambers should be designed to minimize internal heating or some method should be used to correct the volatilization rate for the effects of temperature.

M^{ETHYL BROMIDE} (MeBr) has been used as a soil fumigant to control nematodes and soil fungi for >60 yr. It is currently estimated that approximately 65 to 85% (UNEP, 1992) of MeBr occurring in the environment is from natural sources, such as oceanic emissions and biomass burning. Other sources that contribute to the remaining 35 to 15 % include both urban and agricultural uses and deforestation. Recent evidence indicates that MeBr may cause significant damage to stratospheric ozone. The largest effects from ozone-depleting gases have been observed in the southern hemisphere. In more recent surveys, there have been noted changes in atmospheric ozone in the northern hemisphere. Due to provisions of the U.S. Clean Air Act, the use of compounds that significantly destroy stratospheric ozone will not be permitted. Therefore, the commercial use of MeBr is scheduled to be discontinued by the year 2001.

The U.S. Department of Agriculture-Cooperative State Research Service (USDA-CSRS), National Agricultural Pesticide Impact Assessment Program (NAPIAP) recently completed an assessment of MeBr and has determined that its scheduled cancellation will cause severe economic impacts on the agricultural community (NAPIAP, 1993). These effects will be most strongly felt in the states of California and Florida, which are the principal users of MeBr in agriculture. Because of the impact

of MeBr on agriculture, there is a great deal of interest in determining the environmental fate and transport of MeBr and potential alternatives that are used in agricultural systems.

Several major areas of uncertainty in the environmental fate and transport of MeBr have been identified in the "Methyl Bromide Science and Technology and Economic Synthesis Report" (UNEP, 1992). One area of particular concern is uncertainty in the MeBr mass emitted into the atmosphere from agricultural operations. This has led USDA-CSRS to initiate a project to investigate MeBr environmental fate and transport and to determine the fraction of MeBr that enters the atmosphere.

There have been several recent studies to obtain this information. Using passive chambers, Yagi et al. (1993) measured the MeBr emission from a southern California field after applying MeBr at approximately a 25-cm depth and covering the soil surface with plastic. Based on the measured flux, the authors concluded that 87% of the total MeBr applied to the field escaped into the atmosphere. The authors indicated that this value was higher than expected, given other recent estimates (UNEP, 1992; Singh and Kanakidou, 1993) that were based on mathematical models, but was similar in magnitude to the losses observed in glass-house studies (de Heer et al., 1984). Because of this unexpected result, additional information was collected to provide a mass balance estimate (Yagi et al., 1995). In addition, Yagi et al. (1995), conducted a second experiment using similar procedures and found that only 34 % of the applied MeBr escaped to the atmosphere. The uncertainties in the reported losses are large since only 10 to 15 samples of the volatilization rate were obtained during each 7-d experiment, only a few soil samples were taken to measure Br⁻ concentrations, the soil Br⁻ concentration after fumigation was measured to only 90 cm and, for the first experiment, initial Br⁻ concentration was available only at depth of 3.0 cm.

Although the flux-chamber method (Hollingsworth, 1980; Clendening et al., 1990; Livingston and Hutchinson, 1994) is probably one of the simplest methods for measuring pesticide flux, it suffers from several disadvantages- the method only measures the flux over a small area, which can cause the estimated flux rate to be highly variable, the flux estimates are sensitive to the placement of the chambers relative to the position where MeBr is injected (i.e., closeness to the source), and the presence of chamber can affect the area sampled, especially the local temperature and relative humidity. This can have a tremendous effect on experimental uncertainty.

In a study on strawberries (*Fragaria X ananassa* Duch.), Majewski et al. (1995) found that 32% of the

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Abbreviations: NAPIAP, National Agricultural Pesticide Impact Assessment Program; LDPE, low-density polyethylene; HDPE, high-density polyethylene.

applied MeBr was emitted into the atmosphere during the first 6 d following application. The MeBr application rate for this experiment was 392 kg/ha and the flux density was measured using the aerodynamic method (Parmele et al., 1972). The reported loss rate fell into the 30 to 60% range noted in the Montreal protocol, but a mass balance was not conducted.

The purpose of this paper is to show a recent estimate of MeBr emissions from a field treated with approximately 240 kg/ha MeBr and covered by a 0.025mm (1-mil) sheet of high-density polyethylene plastic. Like the experiments described by Yagi et al. (1993, 1995) and Majewski et al. (1995), the experimental conditions included a shallow injection of MeBr under plastic. Several methods were used to measure the MeBr emission rate and include flow-through flux chambers. The experimental design also included a mass balance, so that additional information concerning the degradation of MeBr to Br was obtained. From this experiment, we found it necessary to correct the chamber flux data for changes in the local temperature caused by the presence of the chamber on the plastic material. The corrected chamber flux data provides measurements that more closely match other methods for estimating the flux and provides a total emission rate which is in line with the mass balance information.

MATERIALS AND METHODS

The experiment was conducted in Field G7 located at the University of California's Moreno Valley Field Station between 26 Aug. 1993 and 13 Sept. 1993. The soil is a Greenfield sandy loam (a coarse-loamy, mixed, thermic, Typic Haploxeralf). Methyl bromide was applied to the field by a commercial applicator at a depth of approximately 25 cm. The MeBr was applied as a mixture containing 99.5 % methyl bromide (CH_3Br) and 0.5 % chloropicrin (CCl_3NO_2) and at a rate of approximately 240 kg/ha. The total mass of MeBr applied was 843 kg. Three flow-through flux chambers were used to estimate the MeBr flux from the field. The flux chambers were placed on the plastic material after application and remained in place throughout the first 5 d of the experiment. Then the plastic was removed from the field and the flux chambers were installed on bare soil and the measurements continued until the end of the experiment. The chambers were sampled at 2-h intervals during the daytime, and at 4 h between 2200 and 0600 h. A complete description of the experiment-including weather conditions, atmospheric soil-gas concentrations, MeBr degradation in soil, and an estimate of total emission to the atmosphere using MeBr degradation-is given by (Yates et al., 1996a). The volatilization rate from several methods that use atmospheric data including the aerodynamic, theoretical profile shape and integrated horizontal flux methods is given by (Yates et al., 1996b) and can be used for comparison with the flux-chamber data.

Flow-through flux chambers were used to calculate the pesticide flux density. Once the flow rate, surface area, and the difference in pesticide concentration at the inlet and outlet of the flux-chambers are known, the flux density can be estimated using

$$\text{Flux} = \frac{\text{Flow rate} (C_{\text{out}} - C_{\text{in}})}{\text{Sampled area}} \quad [1]$$

To obtain a flux value, 20 L/min of air was passed through the chambers to ensure that the concentration of MeBr would

not build up inside the flux chambers. The concentration in the chamber effluent tube was sampled using glass tubes containing 600 mg of coconut charcoal. The sampling rate through the charcoal tubes was 100 mL/min. After the sampling interval, the glass tubes were stored in a freezer until taken to the laboratory for analysis of the MeBr concentration. A complete description of the sampling methodology and laboratory analysis is given by Gan et al. (1995a,b). The inlet air was piped from an adjacent field to provide clean air to the chamber so that C_{in} would be zero. The flux calculations were obtained using the area of the surface sampled (0.31 m^2), the flow rate into the flux chamber (20 L/min), and the concentration that results during the sampling interval [mass on charcoal in (μg)/ $0.1 \text{ (L/min)/time interval (min)}$]. This sampling protocol was used throughout the experiment.

To obtain a mass balance, estimates are needed for the amount of MeBr degraded to the Br-, remaining in the vapor phase, sorbed to the solid phase, and dissolved into the liquid phase, and provides a means for checking the total MeBr emission into the atmosphere. The total mass emitted from the field must be less than or equal to the mass applied minus the amount degraded, that is

$$A \int_0^t f_z(0, t) dt \leq \text{Mass applied} \\ - \text{Mass degraded} - \text{Mass remaining} \quad [2]$$

where $f_z(0, t)$ is the areal-averaged vertical flux rate of MeBr from the soil surface into the atmosphere and A is the area of the field.

Temperature-Dependent Diffusion through Films

A study was conducted to quantify the changes in the MeBr diffusion rate through polyethylene films in response to increases in the ambient air temperature. An apparatus similar to Kolbezen and Abu-El-Haj (1977) was constructed to measure the diffusion through the polyethylene plastic used during this experiment. Two glass cylinders (0.125 m by 0.11 m height) each having one closed end were obtained and the internal volume measured. The edges of the glass at the open end of each cylinder were lined with latex tubing so that when placed together would provide a gas-tight container. A piece of high-density polyethylene film was inspected for holes and other damage and was placed between the two glass cylinders, creating two separate chambers. The device was held together using two plywood squares placed on the top and bottom of the glass cylinders and connected together with long screws and wing nuts.

Five milliliters of MeBr gas ($\sim 18 \text{ mg}$ mass) was introduced into the bottom chamber via a pre-installed septum using a gas-tight syringe. The system was transferred to an incubator that was set at several predetermined temperatures. After a 2-h equilibrium period, 5 mL of the air in the lower chamber was withdrawn using a gas-tight syringe and spiked into an ORBO-32 charcoal tube. The charcoal tubes containing MeBr were then analyzed using a headspace gas chromatograph method reported by Gan et al. (1995b). Four replicate samples were taken at 24, 38, 45, 55, and 65°C. The diffusion coefficient is reported as micrograms of MeBr diffusing through a square meter of the polyethylene film per second while maintaining a 1000 mg/kg (ppm) concentration difference across the film; allowing comparison with the results from Kolbezen and Abu-El-Haj (1977). The concentration difference was calculated from the initial amount of MeBr added to the lower chamber divided by the known volume. It was assumed that minimal leakage occurred during the 2-h equilibration period.

Construction of a Passive Chamber

A passive chamber was constructed following the specifications described by Yagi et al. (1993). The chamber was made from polyvinyl chloride pipe with approximately a 30 cm inside diameter. Its main body consisted of a 19 cm long section and had a removable 5 cm long rim with tedlar membrane sealed to one end to form a lid. A rubber gasket was glued onto the edge of the 19-cm section of pipe, and three spring latches were mounted between the rim and the main section of the chamber. This provided a tight fit between these parts of the chamber. A 1.9 cm diameter hole was drilled 9.5 cm from the chamber bottom so that a 76.2- μm diameter (0.003-inch) Type E (chromel-constantan) fine-wire thermocouple could be inserted into the chamber. The small gap between the chamber and the thermocouple wire was sealed with duct tape. The air temperature outside the chamber was measured with another fine wire thermocouple also located 9.5 cm from the soil surface and was positioned approximately 15 cm from the side of the chamber. To correlate the differential temperature with the intensity of solar radiation, a pyranometer was installed approximately 2 m from the soil surface and 1.5 m from the chamber. The thermocouples and pyranometer were measured using a data logger every 5 s and averaged over a 5-min interval. The measurements were made over 4 consecutive days, which experienced a range from completely overcast to clear-sky conditions. To observe the rate of temperature increase inside the chamber, the lid was placed on the chamber for 25 to 30 min, then removed, and the chamber remained open for 1 to 2 h before restarting the measurement cycle.

RESULTS AND DISCUSSION

Multiplying the flux density measurement by the specific time interval and integrating over the area of the field allows the total MeBr mass emitted from the soil surface to be determined. This is shown in Fig. 1A, as the flux density ($\mu\text{g}/\text{m}^2\text{s}$) emitted from the field during the first 7 d of the experiment. Each filled-circle is the average value for the three flux chambers located on the

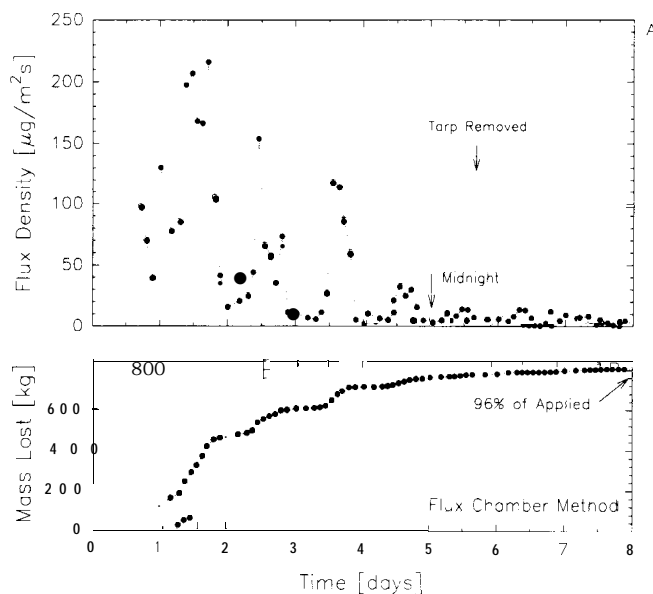


Fig. 1. Methyl bromide flux density (A) and cumulative mass emission (B) from the field during the first 8 d of the experiment. Values were determined using flux chambers. The integer values of times shown on the abscissa indicate midnight.

plastic. The integer values of time on the abscissa (i.e., 1, 2, 3, 4, etc.) indicate midnight of that particular day. Shown in Fig. 1B is the MeBr mass lost from the field and is obtained from integrating the chamber flux data shown in Fig. 1A. The amount of MeBr lost during the first 24 h after application is about 365 kg or 45% of the total mass volatilized from the field. During the next 24 h, an additional 202 kg was lost to the atmosphere, which is 25% of the total emissions.

When the MeBr mass lost to the atmosphere is estimated using atmospheric gradient-based flux methods, the slope of the mass lost vs. time is steeper at early time (Yates et al., 1996b). This is indicative of higher MeBr emissions with time. Using the aerodynamic method, 59% of the total mass emitted into the atmosphere was lost during the first 24 h and 88% by 48 h. For the theoretical profile shape and integrated horizontal flux methods, respectively, the fractional losses are 79 and 68% at the end of 24 h, and 97 and 86% by 48 h. These differences in temporal behavior suggest that the chambers may be measuring the MeBr volatilization rate for different environmental conditions than the atmospheric-based flux measurements. The chamber response is similar to the work of Yagi et al. (1993), who observed approximately 60 to 70% of the total volatilization losses occurring during the first 48 h. Other differences include the timing of the peak flux values. For the aerodynamic method, the peaks generally occur during the late morning to midday. For the flux chambers, they tend to occur after midday when temperatures are highest and diffusion through the plastic is the greatest. Like the aerodynamic method, the minimum flux values measured with the flux chambers occurred near midnight or early in the morning, although the magnitude for the chambers is generally greater during this time.

A serious problem occurs when the flux chamber data are used to estimate the total emission rate. When the flux values were integrated over space and time, the total mass emitted from the field is estimated to be 811 kg, which is about 96% of the 843 kg that was applied to the field and is similar in magnitude to the 87% value reported by Yagi et al. (1993). From the Br- information, which indicates the total MeBr degraded (325 kg or 39%), the total mass volatilized from the field should be approximately 518 kg (± 164 kg) or 61% ($\pm 19\%$) of the total applied. The amount of MeBr remaining in the vapor, solid and liquid phases at the time of sampling was $<0.05\%$ of the applied mass and, therefore, negligible (Yates et al., 1996a). The mass balance calculated from the flux chamber data is 135%. This suggests that flux chamber data, if not corrected for temperature, may overestimate the MeBr flux under these experimental conditions and that as an absolute measure, flux chambers alone may be inadequate for determining the MeBr emission rate.

Further evidence for this can be found from the atmospheric-based estimates of the fraction of the total applied MeBr lost to the atmosphere reported by Yates et al. (1996b). Several methods were used including: the discrete ($62\% \pm 11\%$) and profile ($67\% \pm 6\%$) aerodynamic methods, theoretical profile shape ($60\% \pm 3\%$),

Table 1. Total amount of methyl bromide volatilized during the experiment and mass balance.

Flux method used	Mass lost while soil covered	Percent lost while soil covered	Mass lost after plastic removed	Percent lost after plastic removed	Measured - applied mass†	Mass balance
	kg	%	kg	%	kg	%
Flux chamber, data not corrected for temperature	779 (298)§	92.3	32 (5)§	3.8	292	134.7
Flux chamber, corrected for temperature	464 (170)	55.0	32 (5)	3.8	- 22	91.3
Aerodynamic, discrete method‡	510 (88)	60.5	15 (3)	1.8	7	100.8
Aerodynamic, profile method‡	558 (46)	66.2	9 (1)	1.1	50	105.9
Theoretical profile shape‡	501 (26)	60.1	6 (2)	0.7	- 12	98.6
Integrated horizontal flux‡	579 (21)	68.7	9 (0)	1.1	70	108.3

† Mass applied 843 kg; measured mass remaining 0.26 kg; measured mass degraded 325 kg (SE \pm 164 kg) from Yates et al. (1996a).

‡ Data from Yates et al. (1996b).

§ Values in parentheses are standard deviations.

and integrated horizontal flux methods (70% \pm 3 %). These methods provide an average value of 64 % (\pm 10 %) for the fractional loss. A summary of the mass balance information for each method is given in Table 1. This discrepancy has caused a reexamination of the flux chamber measurements to find an explanation for the overestimate.

Temperature Correction for Enhanced Diffusion through Films

It is known that MeBr diffusion through polyethylene films increases with increasing ambient temperature (Kolbezen and Abu-El-Haj, 1977). Therefore, any increase in the temperature caused by the presence of the chamber on the plastic could cause the measured MeBr flux to be overestimated and not be representative of the rest of the field.

Kolbezen and Abu-El-Haj (1977) conducted a study that investigated the changes in diffusion rate through various films resulting from a change in ambient temperature. The experiment consisted of a chamber covered by the polyethylene film. Inside the chamber a concentration of 1000 mg/kg (ppm) was maintained, while outside the chamber clean air was moved across the film to maintain approximately a zero concentration. The tem-

perature range extended from approximately room temperature to 60°C. The data for 0.025 mm (1 mil) low-density polyethylene (LDPE) and high-density polyethylene (HDPE) films from Table I of Kolbezen and Abu-El-Haj (1977) are shown in Fig. 2A. In this figure, the differences in the rate of escape (i.e., flux density) at various ambient temperatures are plotted as open and filled circles and triangles to indicate the different films. Measurements for the film used in the field experiment are shown as open boxes in Fig. 2A. It is clear from this figure that there is a strong dependence between the rate of MeBr loss through the film and increases in ambient temperature. If the data for each experiment are normalized by dividing each rate of escape by its value at 23 or 24°C, the data constitutes a single relationship, which can be described, approximately, by a linear model shown in Fig. 2B as a solid line, that is

$$\text{Flux enhancement} = 1.03 + 0.067 AT$$

$$r^2 = 0.998 \quad [3]$$

Given the data presented, this relationship holds regardless of whether the film was a 0.025mm (1-mil) low- or high-density polyethylene. The relationship can be considered a flux enhancement factor, where the flux through the film increases by a factor of approximately

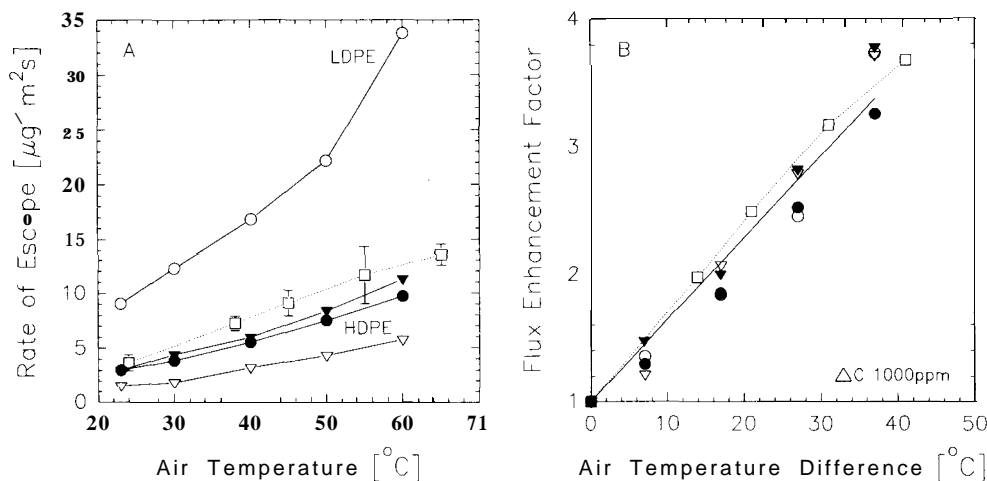


Fig. 2. (A) Rate of methyl bromide escape and (B) flux enhancement factor as a function of ambient temperature during the laboratory experiment. Portions of the data in (A) were obtained from Table 1 of Kolbezen and Abu-El-Haj (1977). In (B) the data were normalized by dividing each datum in (A) by its lowest value (i.e., at 23 or 24°C). The open boxes are laboratory results from the plastic film used in this experiment. The linear regression equation (thick solid line) is Flux enhancement factor = 1.03 + 0.067 AT.

two for each 20°C increase in ambient temperature. Therefore, a 40°C temperature increase would produce approximately 3.5 to 4 times more diffusion through a film compared with a film at a lower temperature. Although this relationship can be described by a linear model, at least to the first approximation, closer inspection suggests that the relationship is slightly curvilinear. The curvilinear nature would cause an increasing diffusion rate through the film at higher temperatures compared with the linear model. For the purposes of correcting the chamber flux data, however, a linear model is probably adequate and the chamber response can be adjusted for temperature effects using the model shown in this figure.

To use this information to correct the flux chamber data, the temperature difference inside and outside the chamber must be known. Once this is known, the flux enhancement factor can be used to correct the flux data. This approach assumes that the presence of the flux chamber on the plastic causes an increased temperature over the portion of the plastic where the flux measurement is taken. Recent measurements of temperature inside and outside the chambers used during this experiment validate this assumption. Since the plastic not covered by the flux chamber is at a lower temperature, the flux chamber produces a higher flux value, which is not representative of the whole field. It is also assumed that a representative value for the flux would occur if the temperature inside the chamber were identical to the outside temperature. That is, there is no inherent bias in flux measurements obtained from chambers. It is further assumed that the air temperature inside the chamber is very nearly the same as the temperature of the plastic. Given that factors and processes that affect measurements are highly variable at the field scale and that very little information is available relating diffusion through polyethylene plastic to temperature, this is probably a reasonable assumption.

During the experiment, the flux chambers did not have a permanently installed temperature sensor inside. Therefore, a direct measurement of the inside temperature is not available and it was necessary to find an alternative method for estimating the internal chamber temperatures. To arrive at a method, an experiment was conducted where a relative humidity and temperature sensor was placed inside and outside one of the chambers used during the experiment. A high correlation was found between incoming solar radiation measured with a pyranometer (LI-200S, LI-COR) and the internal chamber temperature. Since the incoming solar radiation was measured during both experiments, this information makes it possible to estimate the temperature differential. An example of this is shown in Fig. 3, where the measured differential air temperature is plotted along with a differential temperature estimated from the intensity of the incoming solar radiation, R_{in} (W/m^2). An equation describing this was obtained using linear regression

$$\Delta T_{est} [^{\circ}C] = (T_{inside} - T_{outside}) = 0.98 + 0.029 R_{in}$$

$$r^2 = 0.903 \quad [4]$$

The temperature response of a chamber is fairly compli-

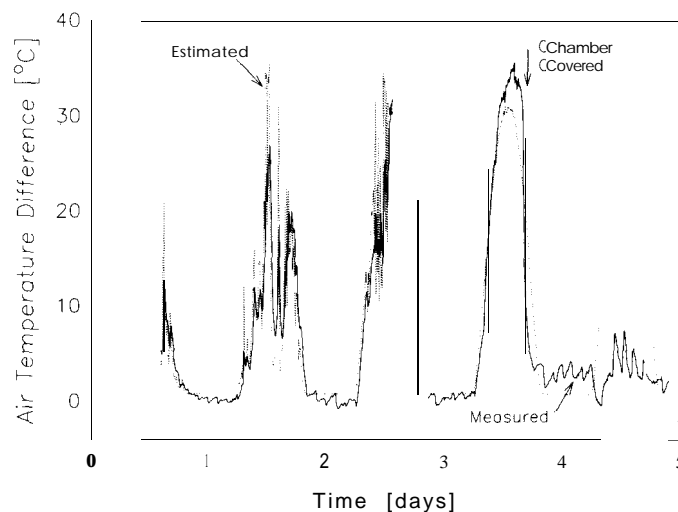


Fig. 3. Comparison of the measured temperature difference inside and outside the flux chamber with a temperature estimated from the intensity of the incoming solar radiation, R_{in} . The regression relationship is $T_{est} [^{\circ}C] = 0.98 + 0.029 R_{in} [W/m^2]$.

cated and depends on the shape, the materials used in construction, the flow rate through the chamber and possibly many other factors. For example, it is anticipated that a flux chamber made from transparent or translucent materials would have higher interior temperatures than those that are opaque, since they allow more sunlight to penetrate the chamber, trapping energy, which increases the temperature. Therefore, Eq. [4] is probably only useful for chambers of this particular design and a specific set of experimental conditions.

During the first 4 d of the experiment, there is high correlation between the incoming solar radiation and the differential temperature (see Fig. 3). After the third day, in an attempt to minimize the large difference in temperature inside and outside the chamber, aluminum foil was placed over the top of the chamber and the pipes carrying clean air to the inlet of the chambers were covered with straw. At this point there is a significant drop in the differential temperature during the day, but at night the differential temperature remains higher than previously observed.

Using incoming solar radiation, the temperature differential inside the flux chamber can be estimated. Coupling this to the flux enhancement factor, a corrected flux measurement can be obtained and is shown in Fig. 4. Comparing Fig. 1 and 4 shows that the timing of the peak daily emission has not changed. The most significant difference is that the total mass emitted from the field after correcting for temperature is about 496 (± 175) kg as opposed to 811 (± 303) kg. The loss represents about 59% of the total applied mass, which more closely follows the results from the other studies. During the first 24 h after application, approximately 227 kg (27% of applied) of MeBr was lost, which is 46% of the total emissions. During the next 24 h, an additional 117 kg (14% of applied) was lost. The corrected measurements underestimate the total mass lost by about 3 to 10% compared with the other methods. For the corrected

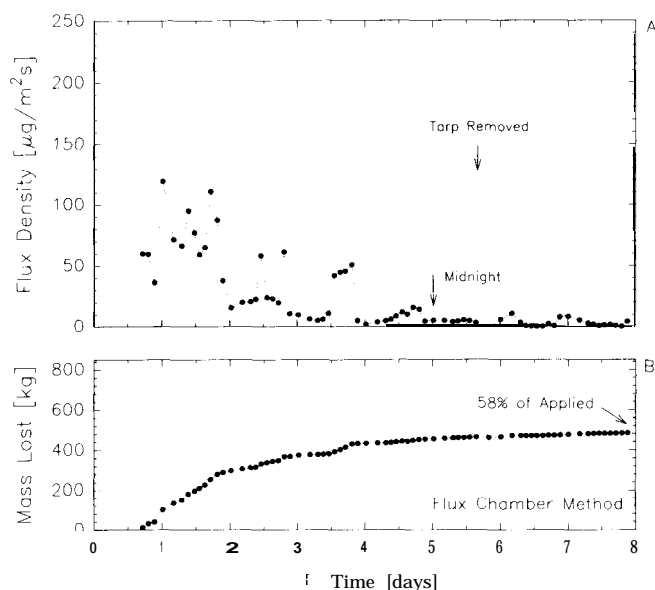


Fig. 4. (A) Methyl bromide flux density and (B) cumulative mass emission from the field during the first 8 d of the experiment after the temperature correction is applied to the data shown in Fig. 1. The integer values of times shown on the abscissa indicate midnight.

measurements, a mass balance of 97 % was obtained and is shown in Table 1.

A similar effect may have occurred during the experiments described by Yagi et al. (1993, 1995). Shown in Fig. 5 are differential air temperature response curves as a function of incoming solar radiation for a chamber built to the specifications given by Yagi et al. (1993) for times 0, 5, 10, and 20 min after the chamber cover is put in place. During periods of low incoming solar radiation, the air temperature inside the chamber is approximately 5°C higher than the air temperature outside the chamber. During early afternoon when the skies are clear, the air temperature inside the chamber can be 15°C higher than outside after 10 to 15 min. Using Eq. [3] (i.e., the solid line in Fig. 2) suggests that the MeBr flux density measured by the chamber may be from 1.4 to 2.1 times higher than the portions of plastic not covered by a chamber. Since most of the samples were collected during midday when solar radiation would be greatest, the estimated fraction of MeBr lost is probably greater by nearly a factor of 2. Accounting for the temperature effect may reduce their estimated total MeBr loss to around 30 to 50%, which would be in agreement with the results from other experiments and estimates used in the Montreal Protocol (UNEP, 1992).

These results also suggest that the reported mass balances of Yagi et al. (1995) (i.e., 106 and 104%) might have resulted from offsetting errors. For example, if the measured emission rate was overestimated and the measured Br- was underestimated, a mass balance of approximately 100% could result. Since the Br- concentration was measured to only 0.9 m depth and considering that MeBr can penetrate several meters deep in the field (Yates et al., 1996a), it seems plausible that an underestimate of the Br mass occurred. Further, the method used to estimate the initial Br- concentration for the

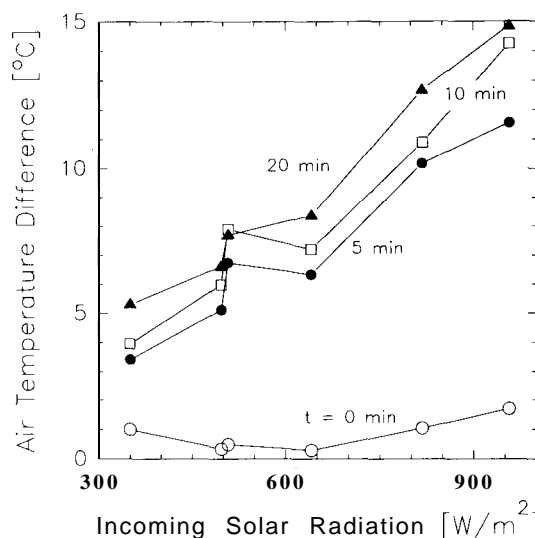


Fig. 5. Air temperature difference (inside - outside air temperatures) from a chamber similar to the one described by Yagi et al. (1993). The air temperature response to various levels of incoming solar radiation is reported at times 0, 5, 10, and 20 min after the chamber cover was put in place.

experiment described by Yagi et al. (1993) may have overestimated the initial mass, thereby underestimating the amount degraded.

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