PERGAMON

Atmospheric Environment 33 (1999) 401–407

ATMOSPHERIC ENVIRONMENT

Atmospheric volatilization of methyl bromide, 1,3-dichloropropene, and propargyl bromide through two plastic films: transfer coefficient and temperature effect

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Received 19 December 1997; accepted 8 May 1998

Abstract

Atmospheric emission of methyl bromide (MeBr) and its potential alternative chemicals such as 1,3-dichloropropene (1,3-D) and propargyl bromide (PrBr) can contribute to air pollution and ozone depletion (for MeBr). One of the main sources of these chemicals is from agricultural soil fumigation. To understand the volatilization dynamics, emission of MeBr, 1,3-D, and PrBr through a polyethylene-based high-barrier film (HBF) and a virtually impermeable film (VIF) was measured using an air flow and sampling system that produced $>90\%$ mass balance. The experiment was conducted outdoors and was subjected to ambient daily temperature variations. The HBF film was found to be very permeable to 1,3-D and PrBr, but somewhat less permeable to MeBr. The VIF film was very impermeable to 1,3-D, PrBr, or MeBr. Measured volatilization flux, in general, exhibited strong diurnal variations which were controlled by film temperature. Unlike the HBF film, a time lag (~ 12 h) was observed between high-temperatures and high-emission flux values for the VIF film. An impermeable film may be used as an effective means of controlling the atmospheric emission of MeBr and its alternative chemicals.

Keywords: Methyl bromide; 1,3-Dichloropropene; Propargyl bromide; Plastic film; Polyethylene; VIF; Permeability

1. Introduction

Atmospheric emission of volatile organic chemicals is a main source of air pollution, which is attributed significantly to arthropogenic activities (Singh et al., 1992). Methyl bromide (MeBr) is a very toxic volatile organic chemical which has been used in large quantities in soil fumigation. Conventional methods of agricultural fumigation can yield a 21–87% of the applied MeBr (Yagi et al., 1995; Yates et al., 1996a, 1997) being lost to atmospheric volatilization. Furthermore, MeBr has recently been put under scrutiny because of its large potential for depleting stratospheric ozone (Singh and Kanakidou, 1993; Butler, 1996). To meet the urgent demand of reducing MeBr input into the atmosphere, there is a need either for alternative management methods that are effective in reducing MeBr volatilization, or for alternative chemicals that could replace MeBr while maintaining effective pest control and without adverse environmental effects.

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PII: S1352-2310(98)00200-3

To protect ozone, chemicals that can be used in soil fumigation as alternatives to MeBr may include 1,3-Dichloropropene (1,3-D) and propargyl bromide (PrBr, C_3H_3Br), among others. It has been shown that 1,3-D is a very effective soil fumigant which controls many of the soil-born pests and plant pathogens that MeBr controls (Noling and Becker, 1994; Kotcon and Loria 1987). Recent research has shown that PrBr has the chemical properties similar to 1,3-D (Yates and Gan, 1998) although it is usually used as a chemical intermediate (Konig and Eilers, 1997). Agricultural use of 1,3-D in California is currently restricted to very low rates and under restricted conditions because of observed large emission losses. Chen et al. (1995) and Leistra and Frissel (1975) reported that about 25–50% of the applied 1,3-D could be lost to atmospheric volatilization.

To reduce volatilization loss, a simple alternative management method is to use a barrier film that is much less permeable to these volatile chemicals than the conventional polyethylene-based high-barrier film (HBF). It has been shown (Kolbezen and Abu-El-Haj, 1977; Houck and Mackey, 1989; Yates et al., 1996b) that materials such as Mylar, Saran, or aluminum foil are much less permeable to 1,3-D and MeBr than the HBF films. A new plastic film, the Hytibar (Klerk's Plastic, Belgium), has been developed by incorporating a EVOH (ethylene vinyl alcohol) barrier polymer between two layers of HBF and has been shown to have permeabilities to 1,3-D and MeBr that approach two orders of magnitude less than the HBF films (Daponte, 1995; Yates et al., 1996b). Once contained in the soil for a reasonable amount of time, degradation by soil water and organic materials will reduce these chemicals to less or non-toxic forms that will no longer endanger the environment. The potential effectiveness of a highly impermeable film or VIF, such as the Hytibar, would make its use a single most effective measure in controlling emissions of MeBr or its alternative chemicals into the atmosphere.

The permeability of HBF films to MeBr has been found to increase very significantly with increasing ambient air temperature (Kolbezen and Abu-El-Haj, 1977; Yates et al., 1996c). This is a primary cause for large MeBr emission fluxes near solar noon when the temperature is high and for the lower emission rate observed at night when the temperature is low (Wang et al., 1997a). Therefore, most emission loss is attributed to times when the temperature is high or the permeability of the HBF films is large. Since little information exists on the Hytibar film, it is important to determine whether its permeability is also temperature dependent and how it would be affected by diurnal temperature changes, as compared to the HBF films.

The purpose of this study was to quantify the volatilization dynamics of MeBr and its potential alternatives, i.e., 1,3-D and PrBr, through a HBF and the Hytibar film. To determine the temperature effect, the experiment was conducted outdoors to allow diurnal temperature variations. A volatilization transfer coefficient was used to characterize the permeability of the two plastic films to MeBr, 1,3-D, and PrBr.

2. Materials and methods

2.1. The emission measurement

Emissions of MeBr, 1,3-D, and PrBr were measured using an air flow and sampling system similar to that reported by Wang et al. (1997b). The system, as shown in Fig. 1, included a flow-through flux chamber, an enclosure that served as the chemical source, and an air flow system for sampling the fumigant (or VOC). The flux chamber was placed onto the source enclosure after a layer of HBF or Hytibar (both ~0.035 mm thick) was sealed over the opening of the enclosure. Air flow through the chamber and the sampling bypass flow was measured with electronic flow meters and automatically recorded with a datalogger. To measure chemical concentration in the source enclosure, a small Teflon tube was inserted through a port on the source enclosure and sealed to prevent leakage. The tube was also used to inject the chemicals into the enclosure. Thermocouples were installed in the chamber and inside the enclosure to measure air temperatures, which were used to estimate the mean temperature of the two plastic films for each sampling interval.

Emission samples were collected continuously and the sampling tubes were replaced at 3 h intervals. Air samples from inside the enclosure were taken at selected times after injection. Similar to procedures used by Gan et al. (1994), a Hewlett-Packard 5890 gas chromatograph (GC) with electron capture detector was used for the analysis of MeBr, 1,3-D, and PrBr content absorbed on each sampling tube. Injections were made from a Tekmar 7000 headspace autosampler (HSA) equipped with a 7050 sample carrousel.

The experiment was conducted at four different times with following combinations of plastic film and chemical selection: (1) MeBr with HBF; (2) MeBr with Hytibar; (3) 1,3-D and PrBr with HBF; and (4) 1,3-D and PrBr with

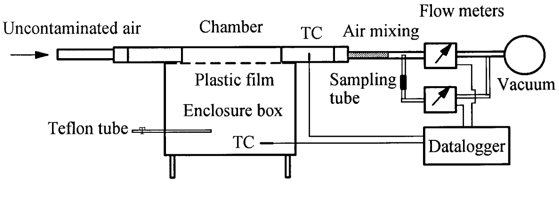

Fig. 1. Schematic of a closed air flow and sampling system for determining volatilization characteristics of volatile organic chemicals through different plastic films.

Hytibar. We tested 1,3-D and PrBr at the same time since they were found not to interact and could be easily separated on the GC.

2.2. Data analysis

Chemical emission from the source enclosure to the overlaying flux chamber can be described as (Jury et al., 1983)

$$J = h(C_{\mathrm{e}} - C_{\mathrm{a}}) \tag{1}$$

where J is the volatilization flux (M L^{-2} T^{-1}); h is a mass transfer coefficient (L T^{-1}); C_{e} is the chemical concentration inside the enclosure (M L^{-3}); and C_{a} is the chemical concentration in the air stream flowing through the chamber (M L^{-3}). According to Wang et al. (1997a), the mass transfer coefficient is attributed primarily to resistance from the plastic film and its dependency on temperature, which can be described with a form similar to the Arrhenius equation. Therefore, equation (1) can be rewritten as

$$J = h_{\mathrm{r}}^{\mathrm{Tr}} \exp\left(\frac{T_{\mathrm{r}} - T_{\mathrm{a}}}{R T_{\mathrm{a}} T_{\mathrm{r}}} \xi_{\mathrm{p}} E_{\mathrm{a}}^{\mathrm{hr}}\right)(C_{\mathrm{e}} - C_{\mathrm{a}}) \tag{2}$$

where $h_{\mathrm{r}}^{\mathrm{Tr}}$ is the reference mass transfer coefficient at a reference temperature (T_{r}); ξ_{p} is an empirical phase adjustment factor to account for any phase mismatch between the volatilization flux and film temperature induced from potential differences in thermal properties between different films; $E_{\mathrm{a}}^{\mathrm{hr}}$ is the activation energy for the reference mass transfer coefficient (J mol^{-1}); T_{a} is the air or film temperature (K); and R is the universal gas constant ($=8.314$ J mol^{-1} K^{-1}). Therefore, the task is to determine $h_{\mathrm{r}}^{\mathrm{Tr}}$, ξ_{p}, and $E_{\mathrm{a}}^{\mathrm{hr}}$ for combinations of MeBr, 1,3-D and PrBr with the HBF and Hytibar films. This was achieved by a nonlinear least-square fit using measured values of J, T_{a}, C_{e}, and C_{a}. The flux (J) was calculated from the flux chamber measurement: dividing the total emitted mass by the sampling area and the sampling interval. The air or film temperature (T_{a}) was estimated as the mean air temperature between that inside the enclosure and in the chamber during the same sampling interval as the emission measurement. Chemical concentration inside the enclosure (C_{e}) during the emission sampling interval was linearly interpolated between the two nearest sampling points taken from the enclosure. Chemical concentration in the flux chamber (C_{a}) was readily estimated from the flux samples averaged over each sampling interval (3 h).

3. Results and discussion

3.1. Emission dynamics

With the HBF film, measured emission flux density of PrBr reached a maximum (2440 µg m^{-2} s^{-1}) right after its injection in the enclosure box (Fig. 2A). Its emission remained large during the first 24 h (500–2440 µg m^{-2} s^{-1}), and decreased rapidly to <100 µg m^{-2} s^{-1} in about 36 h. This is consistent with the concentration measurement inside the enclosure, since over 95% of the chemical has been lost by emission (Fig. 2C). Emission flux density of MeBr is much smaller than PrBr. The maximum reached only 122 µg m^{-2} s^{-1}, which is comparable to field measurements with the flux chambers (118 µg m^{-2} s^{-1}, Yates et al., 1996c). Consequently, its concentration in the enclosure decreased at a slower rate than PrBr. It appears that the HBF film was very permeable to PrBr and somewhat less permeable to MeBr. Close examination indicates that the flux density of MeBr emission follows a diurnal pattern with large fluxes near solar noon when the temperature was high and low emission near midnight with low temperatures (Fig. 2A and B). This diurnal pattern is consistent with measurements from a large-scale field experiment (Yates et al., 1996a). For PrBr, however, the drastic decrease in

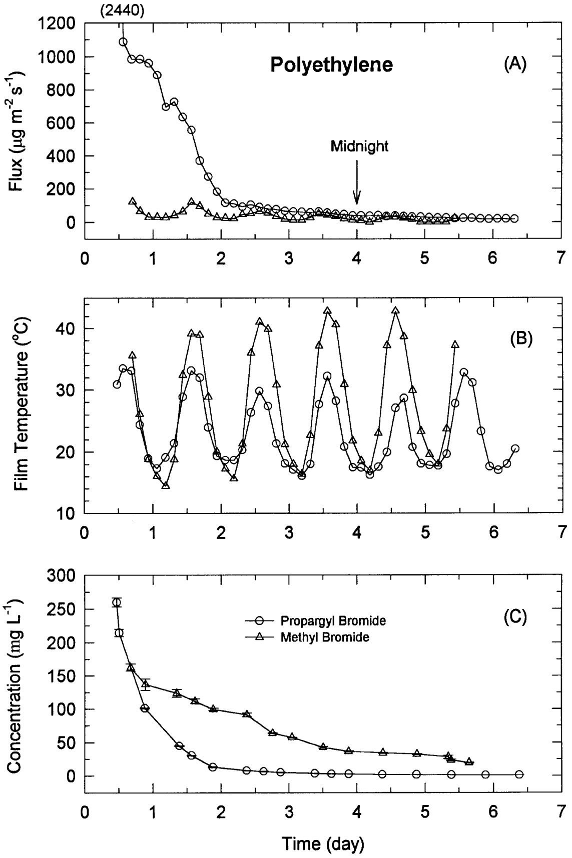

Fig. 2. Volatilization of propargyl bromide and methyl bromide through the polyethylene-based high barrier film: (A) volatilization flux density, (B) film temperature, and (C) concentration in the source enclosure, error bars = ± SE ($n = 3$).

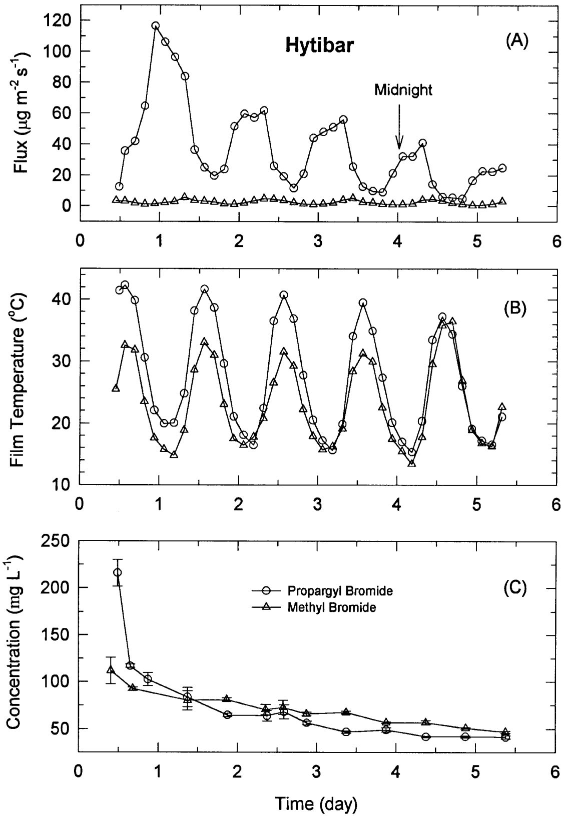

Fig. 3. Volatilization of propargyl bromide and methyl bromide through the Hytibar film: (A) volatilization flux density, (B) film temperature, and (C) concentration in the source enclosure, error bars = ± SE ($n = 3$).

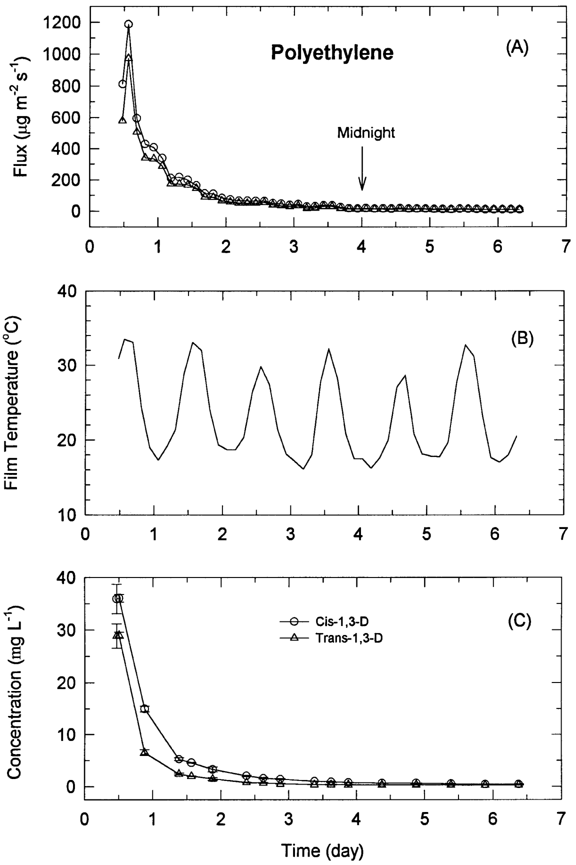

Fig. 4. Volatilization of cis- and trans- 1,3-Dichloropropene through the polyethylene-based high barrier film: (A) volatilization flux density, (B) film temperature, and (C) concentration in the source enclosure, error bars = ± SE ($n = 3$).

its emission flux density quickly depleted the chemical from the enclosure and its dependence on temperature is not apparent.

The emission was significantly reduced for both MeBr and PrBr with the Hytibar film (Fig. 3A). For PrBr, the maximum emission flux reached only 118 $\mu g\,m^{-2}\,s^{-1}$, as compared to 2440 $\mu g\,m^{-2}\,s^{-1}$ with the HBF film. This maximum flux occurred about 12 h after PrBr injection rather than immediately following its application in the enclosure. It is also very evident that its emission exhibits a strong cyclic behavior. For MeBr, the emission flux remained very low ($<10\,\mu g\,m^{-2}\,s^{-1}$) with small daily variations. It appears that, for both chemicals, a daily maximum occurred in the early morning and a minimum at late afternoon or before midnight. This pattern is the opposite of that for the HBF film where the maximum flux occurs in the afternoon and the minimum in early mornings following the diurnal temperature variations (Fig. 3B). The drastic reduction in emission rates is attributed primarily to the high barrier film, i.e., the Hytibar, since initial chemical concentrations in the enclosure were similar to that for the HBF film (Figs. 3C and 2C, 215 vs. 258 $mg\,\ell^{-1}$ for PrBr; 113 vs. 165 $mg\,\ell^{-1}$ for MeBr).

The maximum flux density reached, respectively, about 1200 and 1000 $\mu g\,m^{-2}\,s^{-1}$ for the cis- and trans-isomers of 1,3-D with the HBF film (Fig. 4A). This maximum was reached at 3 h after injection and the emission rate decreased rapidly to $<100\,\mu g\,m^{-2}\,s^{-1}$ in 24 h, following a trend similar to that for PrBr. Because of the rapid change in flux values, its dependence on temperature is difficult to characterize, compared to the Hytibar film (Fig. 4A and B). Source concentration of 1,3-D in the enclosure decreased, in a corresponding fashion to the flux density, from an initial 29–36 $mg\,\ell^{-1}$ to $<5\,mg\,\ell^{-1}$ in 24 h (Fig. 4C). The ineffectiveness of the HBF film in containing 1,3-D is consistent with field measurements by Schneider et al. (1996) who found that the presence or absence of a HBF film had very little effect on the magnitude of 1,3-D loss to the atmosphere.

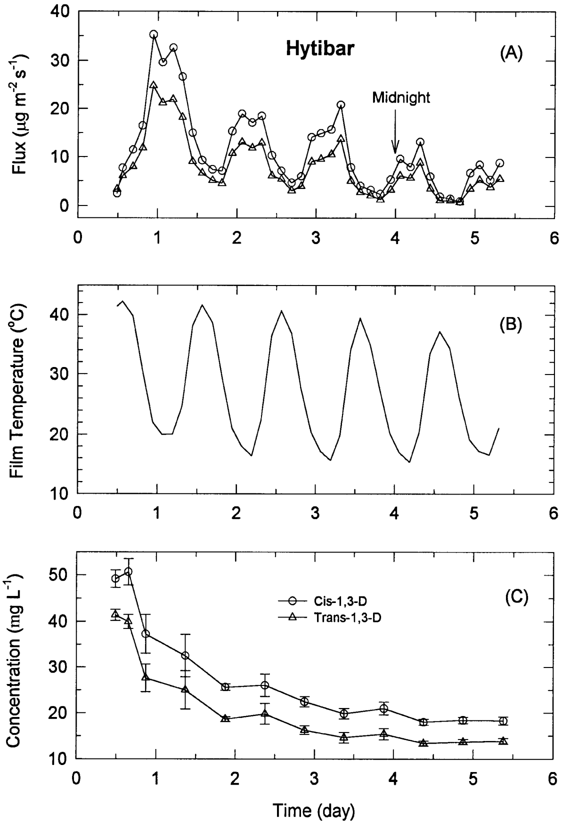

Fig. 5. Volatilization of cis- and trans- 1,3-Dichloropropene through the Hytibar film: (A) volatilization flux density, (B) film temperature, and (C) concentration in the source enclosure, error bars = ± SE ($n = 3$).

With the Hytibar film, 1,3-D emission was significantly reduced. About 12 h after injection, the emission flux density reached a maximum of only 35 and 25 $\mu g\ m^{-2}\ s^{-1}$ for the cis- and trans- isomers, respectively (Fig. 5A). Concentration in the source enclosure decreased at a slower rate than for the HBF film (Figs. 5C and 4C). The emission was also strongly cyclic following a diurnal cycle with a daily maximum occurring in early morning and a minimum in the late afternoon (Fig. 5A and B). Therefore, it appears that the Hytibar film not only significantly reduces the emission of the three chemicals, but delays the breakthrough of the chemicals for about 12 h. The time lag between the occurrences of daily maximum or minimum flux and temperature led us to investigate how the film permeability changes as a function of temperature. To quantify this effect, we constructed replicated double-chambers separated with the Hytibar film. MeBr was injected in one chamber and allowed to diffuse (through the film) to the second chamber for 48 h at 10, 20, 30, and 43°C constant temperatures, respectively. The ratio of the total mass applied in the source chamber to the amount of MeBr diffused into the second chamber increased with increasing temperature from 10 to 30°C, but decreased from 30 to 43°C. A more detailed study is currently underway to verify this effect. During the outdoor volatilization study, measured film temperature (as the mean of ambient air and that inside the enclosure) reached a daily maximum of 39–42°C between noon and late afternoon hours and a daily minimum of 15–17°C after midnight. However, the occurrences of the highest and lowest flux values were in the opposite phase of the diurnal temperature profile. It is likely that the EVOH polymer may have changed the thermal properties of the Hytibar film such as the heat capacity and conductivity (Birley and Couzens, 1974). This, in turn, would increase the residence time for the chemical molecules to diffuse through the film, creating a time lag in responding to the relatively rapid changes in air temperature.

3.2. Parameters for characterizing chemical emission through HBF and the Hytibar films

The closed air flow and sampling system worked well for the outdoor characterization of atmospheric volatilization dynamics of the three chemicals. A mass balance of 87.5–99.1% was obtained for different combinations of chemical and films (Table 1). The mass balance for HBF film (mean = 97.9%) was slightly closer to 100% than for the Hytibar film (mean = 91.1%). It is possible that the low permeability of the Hytibar film would keep the chemical in the source enclosure for a long time, thus increased the potential for leakage losses.

When not considering temperature effect, estimated mass transfer coefficients (i.e., h, from equation (1)) were very strongly cyclic on a diurnal bases, which indicates that other mechanisms such as temperature may be controlling this important parameter. With equation (2), the emission flux can be described very closely ($R^2 = 0.71$–0.99) by fitting the temperature-dependent reference mass transfer coefficient (h_r^{Tr}), the phase adjustment factor (ξ_p), and its activation energy (E_a^{hr}). For a particular combination of the plastic film and chemical, the fitted h_r^{Tr}, ξ_p, and E_a^{hr} values are listed in Table 1. These fitted parameters are reasonable because they agree with estimates from an independent laboratory measurement. For example, h for MeBr using a HBF was reported as 0.389 $\mu m\ s^{-1}$ when measured in the laboratory with $T_a = 20$–$23°C$ (Yates et al., 1996b), the value obtained in this study was 0.210 $\mu m\ s^{-1}$ for $T_a = 20°C$. The reference mass transfer coefficient was about 190 times smaller for Hytibar than for the HBF film for either 1,3-D or PrBr; and was about 240 times smaller for MeBr. The time lag in the Hytibar film in responding to temperature changes is reflected as negative ξ_p values, as compared to the HBF film.

To maintain or reduce current MeBr levels in the stratosphere (8.9 ppt, Singh et al., 1997) and meet the

Table 1
Fitted parameters for characterizing the permeability of polyethylene and Hytibar films to 1,3-Dichloropropene, propargyl bromide, and methyl bromide gas diffusion

Parameter[a]	Chemical[b]			
	cis-1,3-D	trans-1,3-D	PrBr	MeBr
Polyethylene Film ($\xi_p = +1$)				
Mass balance (%)	97.4	98.3	99.1	96.9
h_r^{Tr} ($\mu m\ s^{-1}$)	3.034	2.330	2.738	0.2096
E_a^{hr} ($J\ mol^{-1}$)	26.282	17.220	144.601	340.080
R^2	0.970	0.956	0.992	0.958
Hytibar Film ($\xi_p = -1$)				
Mass balance (%)	91.8	92.2	92.7	87.5
h_r^{Tr} ($\mu m\ s^{-1}$)	1.157×10^{-2}	2.005×10^{-2}	1.476×10^{-2}	8.769×10^{-4}
E_a^{hr} ($J\ mol^{-1}$)	222.193	243.860	292.360	256.572
R^2	0.750	0.708	0.726	0.798

[a] Parameters ξ_p, h_r^{Tr}, and E_a^{hr} are, respectively, the phase adjustment factor, the reference mass transfer coefficient at a reference temperature ($T_r = 20°C$), and the activation energy for h_r^{Tr}, as described in Eq (2).

[b] Cis- and trans- 1,3-D = cis- and trans-isomers of 1,3-dichloropropene; PrBr = propargyl bromide; MeBr = methyl bromide.

demand of MeBr phaseout as a soil fumigant, the potential alternative fumigants should be considered and used in a manner that minimizes their atmospheric emission. The replacement of conventional HBF films used in fumigation with a VIF film such as the Hytibar will reduce the atmospheric emission of MeBr, 1,3-D or PrBr to very low levels. This will significantly reduce their potential environmental risk and may promote the long-term use as a chemical alternative to MeBr without suffering from loss to additional chemicals.

Acknowledgements

The authors would like to thank Ms. Q.P. Zhang and R. LeMert for analyzing MeBr samples in the laboratory.

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