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# Comparison of leakage rates of methyl bromide and sulfuryl fluoride during structural fumigations

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## Abstract

In structural fumigations, half-loss time (HLT) is the most frequently used indicator for comparing fumigant leakage rates. In practical situations where gas leakage rates during structural fumigations are compared, environmental conditions generally are not analyzed in detail and sealing quality is assumed to be constant or fixed. This gives a false impression that a certain gas fumigant might be contained in a structure better than another fumigant. During commercial structural fumigations at the Hal Ross Flour Mill, Department of Grain Science and Industry, Kansas State University, Manhattan, Kansas, USA, leakage characteristics of Methyl bromide (MB) and Sulfuryl fluoride (SF) were compared by taking internal and external environmental conditions into consideration.

Two sets of one 24-h MB and one 24-h SF fumigation experiments were conducted in May and August 2009. Mill sealing and fumigations were conducted by two separate commercial fumigators. After sealing, sealing quality prior to a fumigation was verified by a building pressurization test. The mill was subjected to different pressure levels generated by a specially made fan. At each pressure level, the air flow rate through a calibrated fan was measured. The observed air flow rate plotted as a function of pressure quantified leakage characteristics of the mill. In two MB and SF fumigations, gas concentrations were continuously monitored during the entire fumigation period. A weather station was installed on the roof of the mill to monitor outside barometric pressure, wind speed and direction, temperature, and relative humidity. Inside the mill, a temperature and relative humidity data logger was placed on each of the five floors of the mill.

Results of this study provided a quantitative side-by-side comparison between MB and SF in the same facility. The pressurization test showed that sealing effectiveness can be quantitatively determined ahead of fumigation. It also confirmed the sealing quality for all fumigations was essentially similar. MB and SF showed similar gas distribution and leakage characteristics. Although the observed HLTs of the fumigations were different, those differences could be explained by the differences in environmental conditions, primarily wind speed, and to a certain extent mill temperature, rather than inherent properties of MB and SF gases.

Keywords: Structural fumigation, Half-loss time, Grain-processing facility, Sulfuryl fluoride, Methyl bromide

# 1. Introduction

In the United States, the primary fumigant used for structural fumigation in food-processing facilities (e.g., flour mills) had been Methyl bromide (MB) until Sulfuryl fluoride (SF) was introduced. SF was registered in the United States for post-harvest use in January 2004 under the trade name ProFume<sup>®</sup> by Dow AgroSciences LLC, Indianapolis, Indiana, USA. MB was phased out in the United States as of 2005, but it continues to be available to certain end users through the critical use exemption (CUE) process. Two reasons cited for allowing CUE were that the alternatives, in this case SF, should be as cost effective as MB, and the end users needed transition time to embrace SF and other MB alternatives.

Most studies in which fumigation experiments are conducted in commercial food processing facilities primarily focus on efficacy against insects and/or on insect population rebound (Drinkall et al., 2003; Reichmuth et al., 2003; Drinkall et al., 2004; Small, 2007). In a structural fumigation, half-loss time (HLT) is the most frequently used indicator to characterize fumigant leakage rates. Cryer (2008) used

computational fluid dynamics simulations to compare leakage characteristics between MB and SF from a flour mill subjected to various fixed wind speeds, and found that under the same conditions the HLTs for MB and SF were nearly identical. Another computer simulation study by Chayaprasert et al. (2009) supported this view. However, in typical discussions where gas leakage rates during structural fumigations are compared, environmental conditions are not analyzed in detail and sealing quality is not characterized. Therefore, it makes it difficult to interpret the effectiveness of a structural treatment absent such a characterization.

Chayaprasert (2007) reported on data collected from five SF and one MB fumigation experiments in three flour mills which included fumigant concentrations, indoor temperature and relative humidity, and outside weather conditions (i.e., temperature, relative humidity, barometric pressure, solar radiation, and wind speed and direction). Analysis of the data resulted in concluding that sealing quality and environmental factors should be considered when comparing structural fumigants. However, sealing quality effectiveness is generally not quantified whenever a fumigation is done. Therefore, the effectiveness of a fumigant should be based on how the gas behaves and how environmental factors affect how a gas behaves within a structure, and consequently how that affects fumigation efficacy.

The objective of the current study was to experimentally compare leakage characteristics of MB and SF while maintaining all the other influencing parameters as close to identical as possible.

#### 2. Materials and methods

#### 2.1 Fumigation procedure

The Hal Ross Flour Mill has five floors with a total volume of approximately 9,628 m<sup>3</sup> (340,000 ft<sup>3</sup>). Figure 1 shows a photo of the mill and its generic floor plan which is the same for every floor. All five mill floors during fumigation were interconnected through stairwell doors and air supply vents, in addition to openings between certain floors to accommodate equipment. Two replications of one 24-h MB and one 24-h SF fumigation experiments were conducted in this mill (Table 1). In each set, the two fumigations were carried out within a three-week time span. All fumigations were done by professional fumigators following label directions. Preparation for all fumigations was similar. Prior to each fumigation, the mill was cleaned and sealed. Sealing quality was verified by building pressurization tests (described in Section 2.2 below). Two 20-inch fans were placed on each floor to facilitate gas distribution. These fans were operating during the entire exposure period. One fumigant introduction point was selected at approximately the same location on every floor (Fig. 1b). The amount of fumigant (i.e., weight) being introduced on each floor and the corresponding introduction time for each fumigation are listed in Table 1. The amount of fumigant used for the August fumigations was less than during May fumigations because of higher inside mill temperatures.



**Figure 1** (a) The Hal Ross Flour Mill and (b) its floor plan.

	1 5	8 8				
	MB1	SF1	MB2	SF2		
	Gas introduction time					
	6:40P 6 May 2009	6:00PM 27 May,2009	2:50PM 11 August 2009	2:45PM 19 August 2009		
Floor	Introduced amount (kg)					
1 <sup>st</sup> floor	22.7+22.7 <sup>a</sup>	113.6	22.7	113.6		
2 <sup>nd</sup> floor	22.7	113.6	22.7	56.8		
3 <sup>rd</sup> floor	22.7	113.6	22.7	113.6		
4 <sup>th</sup> floor	45.4	113.6	45.4	113.6		
5 <sup>th</sup> floor	45.4	113.6	45.4	113.6		
Total	181.6	568.0	158.9	511.2		

<b>Table 1</b> The quantity of MB and SF fumigants used and ga	s introduction times.
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<sup>a</sup>Top-up release at 9:50AM 7 May 2009

#### 2.2 Pressurization test

At approximately two hours before the gas introduction of each fumigation, the building sealing quality was quantitatively evaluated by a pressurization test. The pressurization test was conducted using the E3 blower door fan (Infiltec, Waynesboro, Virginia). This fan is capable of delivering a maximum airflow rate of  $2.57 \text{ m}^3$ /s (5,450 cfm). As the speed of the fan varies, it pressurizes the tested building with different level of airflow rates. The fan was attached to one of the exit doors. During each pressurization test, the building was subjected to different pressure levels between 20 and 130 Pa. At each pressure level, the flow rate through the fan and the static pressure difference across the blower door were measured by the DM4 micro-manometer (Infiltec, Waynesboro, Virginia). The gas-tightness characteristic of the mill was determined by plotting the pressure-flow rate relationship.

#### 2.3 Fumigation monitoring

Six monitoring lines were evenly distributed on each floor of the mill. Fumigant concentrations were continuously monitored automatically by the Spectros Instruments Single Point Monitor (Spectros Instruments, Hopedale, Massachusetts) at 10 locations. The other 20 locations were monitored using either the Spectros Instruments Single Point Monitor or Fumiscope (Key Chemical and Equipment, Clearwater, Florida) manually throughout the 24-h exposure time. In addition to fumigant concentrations, environmental conditions were monitored. A HOBO<sup>®</sup> U30 weather station (Onset Computer Corporation, Bourne, Massachusetts, USA) was installed on the roof of the mill. The weather station recorded barometric pressure, wind speed and direction, temperature, and relative humidity every minute. Temperature and relative humidity inside the mill were monitored by HOBO<sup>®</sup> H8 data loggers (Onset Computer Corporation, Bourne, Massachusetts, USA) every three minutes (one logger per floor).

# 3. Results and discussion

A plot of the pressure-airflow rate curves representing the sealing effectiveness of all fumigation experiments is shown in Figure 2. The data points for the MB1, SF1 and MB2 fumigations were on an almost identical curve (i.e., dashed line in Fig. 2), indicating similar gas-tightness levels. The result of the pressurization test for the SF2 fumigation was adversely affected by strong prevailing wind around the mill experienced during the test, resulting in notably more scattered data points. However, the lower bound of the scattered data points, which is the highest building gas-tightness possible for the SF2 fumigation, coincides with the pressure-airflow rate curves for the other three fumigations. Assuming that this lower bound represented the sealing quality for the SF2 fumigation, the plot in Figure 2 in general confirms that the sealing quality of all four fumigations in this study was nearly identical. In other words, if the pressure differences across the flour mill building envelope were the same, the fumigant leakage rates observed from these four fumigations would be the same as well. Thus, any differences in the leakage rates (i.e., HLTs) between the fumigations were caused by the fumigation-to-fumigation variations in environmental conditions.

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Figure 2 Results of the pressurization tests.

Figures 3a, 3b and 3c show barometric pressures, outside temperatures, and outside relative humidity, respectively, during the four fumigations. Note that the barometric pressure reduction due to the difference in height between the weather station and ground level has been compensated for in the barometric pressure curves in Figure 3a. Substantial variations in these environmental conditions were observed both within each fumigation and between fumigations. Within each fumigation, the differences between the highest and lowest values of barometric pressure, temperature, and relative humidity, were approximately 3-9 mbar, 9-12°C, and 40-50%, respectively. Nevertheless, the inside temperature and relative humidity were relatively stable during the entire exposure periods for all fumigations. Within each floor, the inside temperature and relative humidity varied less than 1°C and 5%, respectively, and the differences in the inside temperature and relative humidity between floors were less than 4°C and 20%, respectively (data not shown). This implied that the heat transfer rate between the inside and outside, and the heat generation and accumulation rates within the mill were balanced. Similar observations can be expected for other buildings with the same gas-tightness level.



Figure 3 Plots of (a) barometric pressures, (b) outside temperatures, and (c) outside relative humidity during the four fumigations.

The fumigant concentrations at all monitoring locations during the MB1, SF1, MB2 and SF2 fumigations are plotted as curves with markers in Figures 4a, 4b, 4c and 4d, respectively. Both MB and SF showed similar gas characteristics. Initially, the fumigant concentrations increased rapidly. For the first three fumigations, even gas distribution was established throughout the mill within the first four hours. For the SF2 fumigation, however, it took 10 hours. During the first three fumigations, all the stairwell doors were opened, while during the SF2 fumigation only the stairwell doors on the 3<sup>rd</sup>, 4<sup>th</sup> and 5<sup>th</sup> floors were opened. Partitioning of the 1<sup>st</sup> and 2<sup>nd</sup> floors caused slower gas movement from these two floors to the other floors. However, in some buildings partitioning very leaky areas as separate fumigated volumes will be beneficial in preventing excessive fumigant loss. The sudden peak in gas concentration at the 15<sup>th</sup> hour of the MB1 fumigation was due to adding an additional 22.7 kg of MB. Generally, once the fumigant was well mixed, it remained evenly distributed throughout the building and the concentrations gradually decreased over time. The differences in concentrations within the mill were between 2 and 7 g/m<sup>3</sup> most of the time.



Figure 4 Fumigant concentrations at all monitoring locations and outside wind speeds during the (a) MB1, (b) SF1, (c) MB2 and (d) SF2 fumigations.

After even gas distribution was achieved, the SF1 and MB2 fumigations showed relatively constant HLTs, while the HLTs for the MB1 and SF2 fumigations changed as the fumigations progressed. By observation, the concentration curves for the four fumigations were divided into sections and the average HLT for each section was calculated. The average HLTs and the corresponding elapsed time periods in which the HLTs were calculated are summarized in Table 2. The HLTs for the MB1, SF1, MB2 and SF2 fumigations were 10.2-111, 19.7, 26.0 and 9.9-26.1 hours, respectively. The wind speed data collected during the fumigations are superimposed on the concentration plots in Figure 4 (transparent lines). Although the effect of rapid wind fluctuations could not be seen in the gas concentration curves, a strong correlation between HLT and wind speed could be observed. The average wind speeds corresponding to the HLTs of the same elapsed time periods are listed in Table 2. Regardless of the type of fumigant, when the average wind speed was less than 2 m/s, between 2 and 3 m/s, between 3 and 4 m/s, and greater than 4 m/s, the HLT was much greater than 26 h, approximately 26 h, between 20 and 15 h, and approximately 10 h, respectively. This indicates no noticeable difference in the leakage characteristics of MB and SF. Banks and Annis (1984) analytically showed that the overall ventilation rate ( $d^{-1}$ ), which is defined as the total volume of the enclosure divided by the volumetric gas loss rate, during fumigation in grain storages is a summation of individual ventilation rate associated with atmospheric pressure, buoyancy and wind forces. However, the effects of other environmental conditions, specifically buoyancy (i.e., inside-outside temperature difference) and barometric pressure pumping forces, were not observable in the concentration data, and thus no other correlations with the HLTs could be established. It is likely that these forces were overshadowed by wind.

Table 2	Half-loss times (HLT), average wind speeds, and corresponding elapsed time periods in which thes
	two values are calculated.

Fumigation	Elapsed time period (h)	HLT (h)	Avg wind speed (m/s)
	5 <sup>th</sup> -10 <sup>th</sup>	111.0	1.65
MB1	$10^{\text{th}}$ -15 <sup>th</sup>	16.4	3.52
	17 <sup>th</sup> -24 <sup>th</sup>	10.2	7.12
SF1	5 <sup>th</sup> -24 <sup>th</sup>	19.7	3.67
MB2	5 <sup>th</sup> -24 <sup>th</sup>	26.0	2.16
953	11 <sup>th</sup> -21 <sup>st</sup>	26.1	3.00
SF2	21 <sup>st</sup> -24 <sup>th</sup>	9.9	6.90

## 4. Conclusions

Results of this study provided a quantitative side-by-side comparison between MB and SF under nearly identical conditions in the same facility. The pressurization test showed that sealing effectiveness can be quantitatively determined ahead of fumigation. It also confirmed the sealing quality of all fumigations to be similar. SF and MB showed similar gas distribution and leakage characteristics. Although the observed HLTs of the fumigations were different, those differences could be explained by the differences in environmental conditions, primarily wind speed, and to a certain extent mill temperature, rather than inherent gas properties of MB and SF.

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