Gas leakage and distribution characteristics of methyl bromide and sulfuryl fluoride during fumigations in a pilot flour mill

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Highlights

- The half-loss times (HLTs) during three methyl bromide (MB) and three sulfuryl fluoride (SF) fumigations were monitored.

- Concentrations of both fumigants within the mill ranged from 2 to 7 g/m^3.

- The observed HLTs for the MB and SF fumigations were in the range of 3.61 to 28.64 h and 9.97 to 31.65 h, respectively.

- HLTs were inversely related only to wind speeds.

1   Revised version

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8   **Gas leakage and distribution characteristics of methyl bromide and sulfuryl**

9   **fluoride during fumigations in a pilot flour mill**

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23

24 **Abstract**

25   The half-loss time (HLT) is used as an indicator to quantify gas leakage rates

26 during methyl bromide (MB) and sulfuryl fluoride (SF) fumigations. Comparisons of

27 HLTs between three MB and three SF fumigations were quantified in the Hal Ross pilot

28 flour mill, Department of Grain Science and Industry, Kansas State University, USA.

29 The sealing quality or gas tightness of the mill before each fumigation was verified by a

30 pressurization test. Fumigant concentrations during the six fumigations were monitored

31 continuously at 30 locations among the five mill floors during the 24 h fumigation period.

32 A weather station on the mill roof monitored barometric pressure, wind speed and

33 direction, temperature, and relative humidity. A data logger on each mill floor recorded

34 temperature and relative humidity. The pressurization test showed that the relationship

35 between airflow rate and building static pressure varied among the fumigations despite

36 the same areas being sealed by two separate fumigation service providers due to

37 environmental conditions not being identical among the fumigations. Concentrations of

38 both fumigants within the mill ranged from 2 to 7 g/m^3. The observed HLTs for the MB

39 and SF fumigations were in the range of 3.61 to 28.64 h and 9.97 to 31.65 h,

40 respectively, and were inversely related only to wind speeds during fumigation and not

41 any other environmental conditions recorded. In our study, the fumigant leakage rate

42 was found to be predominantly a function of wind speed rather than inherent gas

43 characteristics of MB and SF.

44

45 *Keywords:* Structural fumigation, Sealing quality, Leakage rates, Gas dynamics, Half-

46 loss time, Wind speed

47

1. Introduction

A structural fumigation is considered successful when the target dosage for an effective kill of all insect life stages is achieved. The dosage is a cumulative product of the fumigant concentration (C) over the exposure time (t), and is referred as the Ct product (Kenaga, 1961; Gandy and Chanter, 1976; Annis, 1999; Bell et al., 1999). The Ct product is a function of the amount of released fumigant, exposure time, and fumigant leakage rate. Fumigant leakage rate is quantified by the half-loss time (HLT), which is the time taken in hours (h) for 50% loss of the total fumigant concentration from the structure being fumigated. The gas leakage rate and HLTs are inversely related. In commercial fumigations, an ideal HLT should be >15 h, but realistic HLTs may range from 5 to 22 h (Chayaprasert, 2007).

Methyl bromide (MB) has been the primary fumigant used for structural fumigation in food-processing facilities such as flour mills (Taylor, 1994). Sulfuryl fluoride (SF) was registered in the United States for use in food-processing facilities in January 2004 under the trade name ProFume[®] by Dow AgroSciences LLC, Indianapolis, Indiana, USA. It is a viable replacement for MB, which was phased out in the United States in 2005 due to its adverse effects on stratospheric ozone, but continues to be available through the critical use exemption (CUE) process (US-EPA, 2010).

A majority of fumigation experiments conducted in commercial food-processing facilities focused on efficacy against insects and/or on insect population rebounds following the treatment (Drinkall et al., 2003; Reichmuth et al., 2003; Bell et al., 2004; Campbell and Arbogast, 2004; Drinkall et al., 2004; Small, 2007). Chayaprasert (2007)

3

71 reported that fumigant concentrations, indoor temperature and relative humidity, and

72 outside weather conditions alone cannot explain fumigant leakage rates without taking

73 sealing quality into consideration. Chayaprasert and Maier (2010) used experimental

74 building pressurization tests and computational fluid dynamics (CFD) model simulations

75 to evaluate the effect of building sealing quality or gas-tightness and weather conditions

76 on SF leakage rates. They concluded that sealing quality and environmental factors

77 should be considered when comparing structural fumigants. Cryer (2008) used CFD

78 simulations to compare leakage characteristics between MB and SF from two flour mills

79 subjected to various hypothetical fixed wind speeds, and found that under similar

80 environmental conditions the HLTs for MB and SF were nearly identical. A computer

81 simulation study by Chayaprasert et al. (2009) also supported this view.

82 Typically when HLTs during commercial structural fumigations are compared,

83 environmental conditions are not taken into consideration by fumigators. Additionally,

84 sealing quality effectiveness is rarely quantified whenever fumigation is done making it

85 difficult to interpret effectiveness of practical structural treatments. Therefore, the

86 current study objectives were to validate computer simulation results with empirical

87 measurements of gas leakage and distribution in a pilot flour mill subjected to MB and

88 SF fumigations and to relate gas leakage rates to environmental conditions.

89 **2. Materials and methods**

90 *2.1. Mill fumigation treatments*

91 The state-of-the art Hal Ross pilot flour mill belonging to the Department of Grain

92 Science and Industry, Kansas State University, Manhattan, Kansas, USA, was used for

93   the present study. The mill has five floors that occupy a total volume of ~9,628 m^3, and

94   Fig. 1 shows the mill exterior and generic floor plan which is essentially similar across

95   the floors. All mill floors have interconnected air supply vents, in addition to openings

96   between floors to accommodate equipment. Three MB and three SF fumigations were

97   conducted during 2009 and 2010. Each pair of MB and SF fumigations was carried out

98   within a three-week time span to ensure comparisons under approximately similar

99   environmental conditions. The fumigations were split between two separate professional

100   fumigation service providers following label directions and safety precautions. The mill

101   was cleaned and sealed prior to all fumigations. We did not compare the sealing

102   material and sealants used by these two service providers. Two 0.51-m diameter fans

103   were placed on each floor to facilitate gas distribution. These fans were in operation

104   during the entire 24 h exposure period. One fumigant introduction point was selected on

105   every floor. All of the stairwell doors were open with some exceptions. The first and

106   second floor doors were closed during the second SF fumigation, and the doors on

107   every floor were closed during the third SF fumigation to reduce fumigant leakage.

108   These decisions were made by the fumigators. The date of fumigation, amount

109   introduced on each floor, and the introduction time are shown in Table 1.

110

111   Six gas monitoring lines of different colors, made of nylon tubes with 4.3-mm

112   internal diameter, were placed on each mill floor. One line was placed on the mill floor at

113   the southwest corner and another line was placed near the ceiling at the northeast

114   corner. The other four monitoring lines were evenly distributed throughout each floor

115   both inside and outside of milling equipment, where there were bioassay boxes with

116 different life stages of the red flour beetle. Two or three of these lines were inserted into

117 different machines where bioassay boxes were located. The bioassay results are being

118 reported elsewhere and are not relevant to the objectives of this paper. The equipment

119 was closed after placement of the monitoring lines. Fumigant concentrations at 10

120 locations (2 per floor) were monitored automatically every 20 minutes by the Spectros

121 Single Point Monitor (Spectros Instruments, Hopedale, Massachusetts, USA). The

122 remaining 20 locations was monitored manually on an hourly basis by using either the

123 Spectros Instruments Single Point Monitor or Fumiscope (Key Chemical and

124 Equipment, Clearwater, Florid, USA) throughout the 24 h exposure period.

125   The environmental conditions during each fumigation were monitored using a

126 HOBO® U30 weather station (Onset Computer Corporation, Bourne, Massachusetts,

127 USA), which was installed on the mill roof to record barometric pressure, wind speed

128 and direction, temperature, and relative humidity at one-minuet intervals. A HOBO® H8

129 data logger (Onset Computer Corporation) on each mill monitored temperature and

130 relative humidity at one-minute intervals. During the third MB fumigation the weather

131 station failed to record wind speed, and wind speed data for this particular fumigation

132 were obtained from the weather station installed on the ground at the Agronomy Farm

133 located about 500 m to the west of the mill.

134   *2.2. Pressurization test*

135   One to two hours before each fumigation, the building sealing quality or gas

136 tightness was quantitatively evaluated by a pressurization test. The pressurization test

137 was conducted using the E3 blower door fan (Infiltec, Waynesboro, Virginia, USA). The

138 fan is capable of delivering a maximum airflow rate of 2.57 m^3/s. The fan was attached

139 to one of the exit doors on either the east or west side. During each pressurization test,

140 the building was subjected to different pressure levels between 10 and 140 Pa by

141 increasing the fan airflow rate. At each pressure level, the flow rate through the fan and

142 the static pressure difference across the blower door were measured by the DM4 micro-

143 manometer (Infiltec, Waynesboro, Virginia, USA).

144   *2.3. Data analysis*

145   The gas-tightness characteristic of the mill was determined by fitting a nonlinear

146 regression model (Equation 1) to the relationship between the flow rate across the

147 pressurization fan (Q, m^3/s) and the static pressure difference across the blower door

148 (p, Pa) (ASHRAE, 2001):

149
$$Q = bp^n \tag{1}$$

150 where, b is the flow coefficient (m^3/s-Pa^n) and n is a dimensionless pressure exponent.

151 All possible pair-wise combinations based on three pressurization tests for MB and

152 three for SF fumigations were compared by testing the deviation of individual models

153 (Equation 1) fit to the flow rate and pressure data to a pooled model (Draper and Smith,

154 1981). A significant difference ($P < 0.05$) between pooled and individual models

155 indicated that the relationship between flow rate and pressure was significantly different

156 between the two pressurization tests being compared. The six fumigations between MB

157 and SF resulted in 15 pair-wise comparisons.

158   The HLTs observed from the fumigations were estimated by a first-order kinetic

159 equation (Equation 2) of gas concentration readings over time (Banks et al., 1983;

160 Chayaprasert et al., 2008; Cryer, 2008):

161
$$C_t = \frac{C_i}{2^{\frac{t}{HLT}}}$$
(2)

162   where, C_t is the current concentration (g/m^3) at the elapsed time t (h) and C_i is the initial

163   concentration (g/m^3).

164   A direct comparison of the resulting HLTs between the MB and SF fumigations

165   could not be made without taking into account all of the weather conditions. Banks and

166   Annis (1984) showed that the overall ventilation rate (d^{-1}), which is defined as the total

167   volume of the enclosure divided by the volumetric gas loss rate during fumigation in

168   grain storages, is a summation of individual ventilation rates associated with barometric

169   pressure, buoyancy, and wind forces. One common method used to calculate air

170   infiltration rates, q (m^3/s), in buildings is the superposition method (Equation 3) in which

171   the wind and stack effects are determined separately and then combined together

172   based on a predefined correlation (ASHRAE, 2001):

173
$$q = \frac{A_L}{1000} \sqrt{c_s \Delta T + c_w U^2}$$
(3)

174   where, A_L is the effective leakage area (cm^2), c_s is the stack coefficient ((L/s)^2/cm^4-K), c_w

175   is the wind coefficient ((L/s)^2/cm^4-(m/s)^2), ΔT is the average indoor-outdoor temperature

176   difference (K), and U is the average local wind speed (m/s). HLT and q are related as

177   shown in Equation 4 (Banks et al., 1983; Chayaprasert, 2007):

178
$$HLT = \frac{V}{q} \frac{\ln(2)}{3600}$$
(4)

179   where, V is the volume of the fumigated building (m^3). Equations 3 and 4 were used to

180   establish any correlations between the HLTs calculated from Equation 2 and the

181   measured indoor-outdoor temperature differences and prevailing wind speeds.

182   Barometric pressure was not taken into account in the superposition calculations

183 because of lack of relationship between fumigant concentration and barometric

184 pressure. Wind direction can affect the fumigant leakage rate when tall structures are

185 neighboring fumigated structures (e.g., grain silos nearby a fumigated flour mill) or when

186 areas of leakage within fumigated structures are not evenly distributed on all sides

187 (Cryer, 2008; Chayaprasert et al., 2009). At the Hal Ross flour mill there were no

188 structures within a 200-m radius taller than half of the mill's height to alter wind direction

189 and influence gas leakage rates. Wind direction was, therefore, neglected in the

190 analysis of gas leakage rates.

191 **3.   Results and discussion**

192   The plots of the pressure-airflow rate curves representing sealing effectiveness

193 of all fumigation experiments are shown in Figure 2. Equation 1 satisfactorily described

194 the pressure and airflow data (r^2 = 0.819 to 0.995) (Table 2). The coefficients b ranged

195 from 0.098 to 0.279 while the coefficient n ranged from 0.445 to 0.655. The gas-

196 tightness was similar only for the first MB and first SF fumigations (F = 1.06; df = 2, 68;

197 P = 0.351). The gas-tightness was significantly different for the remaining 14 pair-wise

198 comparisons (F, range = 8.49 – 273.63; df, range = 2, 68 - 2, 145; P < 0.0005). This

199 could be attributed to differences in environmental conditions (see below) during each of

200 the fumigations, because data used in Equation 1 could not be corrected for differences

201 in environmental conditions. The result of the pressurization test for the second SF

202 fumigation was adversely affected by strong prevailing winds (6 to 8 m/s) during the test

203 resulting in more scattered data points. However, the lower boundary of the scattered

204 data points, which indicates the highest building gas-tightness, coincided with similar

205   pressure-airflow rate curves for the five other fumigations. In general, the pressurization

206   test results suggested that the differences in the HLTs were not caused by variations in

207   sealing quality but by the outside environmental conditions.

208   Substantial variations in barometric pressure, outside temperature, and outside

209   relative humidity were observed among fumigations (Figure 3A-C). The barometric

210   pressure curves in Figure 3A were adjusted for the barometric pressure reduction due

211   to the difference in height between the weather station on the mill roof and the ground.

212   The average values of barometric pressure, outside temperature, and relative humidity

213   between the fumigations ranged from 971 to 984 mbar, 13 to 26°C, and 63 to 84%,

214   respectively. Within each fumigation the differences between the highest and lowest

215   values of barometric pressure, outside temperature, and relative humidity were

216   approximately 3 to 9 mbar, 5 to 15°C and 30 to 60%, respectively. The inside

217   temperature and relative humidity were, however, stable during the fumigations (Table

218   3). On each floor the inside temperature and relative humidity generally varied by less

219   than 1°C and 10%, respectively, and the differences in the inside temperature and

220   relative humidity among floors were less than 4°C and 20%, respectively. The inside

221   temperatures were either equal to or higher than the outside temperatures with a

222   maximum difference of at least 10°C, except for the first and second MB fumigations,

223   where for a few hours, the opposite occurred. These findings suggested that at the gas-

224   tightness level achieved in this study air infiltration did not have an effect on the thermal

225   changes inside the flour mill. In addition to preventing rapid gas loss, good sealing

226   quality helps increase fumigation efficacy against insects and helps maintain stable

227   temperatures inside a fumigated building irrespective of outside temperature changes.

10

228   The fumigant concentrations over time near the ceiling across the five mill floors

229   for each of the fumigations are illustrated in Figure 4. For the MB and SF fumigations,

230   differences in fumigant concentrations within each floor were less than 3 and 5 g/m^3,

231   respectively (data not shown). Initially, the fumigant concentrations increased rapidly

232   and distributed well among the mill floors, after which the concentrations gradually

233   decreased over time. However, gas concentrations at one monitoring location in an

234   ingredient mixing drum on the third floor was an exception to this general observation.

235   During the first MB and all SF fumigations, the gas concentrations inside the mixing

236   drum did not decrease as fast as the other locations because of restricted gas

237   movement. The sudden peaks in gas concentrations 15 h after the initial fumigant

238   introduction in the first and third MB fumigations were due to adding more gas (Table 1).

239   SF gas was also added during the third fumigation at 14.5 h into the fumigation, but gas

240   monitoring data did not show any sudden peaks. The concentration differences within

241   the entire mill were between 2 and 7 g/m^3. Even gas distribution was established

242   throughout the mill within the first 4 h, except for the second and third SF fumigations in

243   which it took at least 10 h. The longer time for gas to equilibrate within the structure may

244   be due to the stairwell doors being closed during these two fumigations, making it more

245   difficult for the fumigant to circulate quickly among mill floors. In some structures,

246   partitioning very leaky areas as separate fumigated volumes can be beneficial in

247   preventing excessive fumigant loss.

248   In this particular study the observed HLTs correlated well with the outside wind

249   speeds regardless of whether or not the stairwell doors were closed. The even gas

250   distributions observed with MB and SF fumigations showed that these two fumigants

251 have similar gas distribution characteristics. In structures where commodities are

252 present distribution of MB and SF gases could be different due to different rates of

253 sorption by the commodities. However, this effect was nonexistent because the mill was

254 free of any stored commodity.

255 The hourly-average outside wind speeds during the fumigations were

256 superimposed on the corresponding concentration plots in Figure 4. While wind speeds

257 varied mostly within a range of 0 to 5 m/s, the rapid hour-by-hour wind fluctuations were

258 not reflected in the gas concentration curves. Except for the third MB fumigation, HLTs

259 for each fumigation shown in Figure 4 were calculated by dividing the gas concentration

260 curves over time into sections in which wind speeds were either above or below 5 m/s.

261 During the third MB fumigation at 8 h the gas concentration curves indicated a sudden

262 drop (Figure 4E), and thus the concentration curves after this time were divided

263 separately. For each divided section, the five concentration curves were first averaged

264 and Equation 2 was fitted to the average concentration over time data. The exposure

265 periods immediately after fumigant releases when concentration differences were

266 greater than 5 g/m^3 were excluded from the HLT calculations. The average estimated

267 HLTs (and SE), average wind speeds, average absolute inside-outside temperature

268 differences, and corresponding elapsed exposure periods are summarized in Table 4.

269 The HLTs for the MB and SF fumigations were in range of 3.61 to 28.64 h and 9.97 to

270 31.65 h, respectively. Williams et al. (2000) suggested HLTs above 24 h as desirable

271 and any values below 10 h as undesirable for structural fumigations. They reported

272 HLTs of 8 to 15 h to be common in food-processing facilities subjected to fumigation.

273 The range of HLTs observed reflects variation among structures in gas tightness

12

274   despite effective sealing, since all of the building gaps cannot be accurately identified or

275   sealed. Based on the pressurization test, the Hal Ross flour mill had nearly identical

276   sealing quality based on visual inspection, but the differences in HLTs were observed

277   across the six fumigations. Of all the weather variables observed, only wind speeds

278   predominantly affected HLTs, and HLTs were inversely related to wind speeds (Figure

279   5A).

280   Except for the last two HLTs of the third MB fumigation, when the average wind

281   speeds were not greater than 5 m/s, the HLTs were longer than 10 h, regardless of the

282   type of fumigant used. The last two HLTs of the third MB fumigation were 3.61 and 9.71

283   h while the corresponding average wind speeds were less than 5 m/s. These two

284   unexpectedly short HLTs were observed after the sudden drop in the fumigant

285   concentration during the third MB fumigation probably due to some seal damage which

286   we could not firmly identify. From Equations 3 and 4, if the stack effect was neglected, it

287   can be seen that:

288
$$HLT = \frac{x_1}{U} \qquad\qquad (5)$$

289   where, x_1 is a constant. Discarding the last two short HLTs of the third MB fumigation,

290   fitting Equation 5 to the data in Figure 5A resulted in the mean ± SE (no. observations =

291   8) x_1 value of 68.52 ± 2.85 and a r^2 value of 0.922. Similarly, combining Equations 3 and

292   4 with the wind effect neglected yields Equation 6:

293
$$HLT = \frac{x_2}{\sqrt{\Delta T}} \qquad\qquad (6)$$

294   where x_2 is a constant. However, such correlation in Equation 6 could not be

295   established as indicated by the scattered data points of the HLTs plotted against the

296   square roots of the average absolute inside-outside temperature differences in Figure

13

297   5B. This was likely attributed to the strong wind effect overshadowing the buoyancy

298   force. Chayaprasert and Maier (2010) found that as the wind speed doubled the HLT

299   decreased by half (Equation 5). Cryer (2008) neglected stack effect in his simulated

300   fumigations and the results indicated that the HLTs for MB and SF were

301   interchangeable. This finding was corroborated by a similar simulation study by

302   Chayaprasert et al. (2009) in which both the wind and stack effects were included in the

303   simulations. The high r^2 value of the curve fitting result (Equation 5) in the present study

304   indicated a strong correlation between the HLTs and wind speeds rather than the type

305   of fumigant used. In addition, when wind is the dominant force of gas leakage, HLT

306   were inversely proportional to the prevailing wind speed. These empirical findings

307   provide a quantitative basis to support the fact that HLTs are influenced by

308   environmental conditions, which should be taken into consideration during structural

309   fumigations.

310   **4. Conclusions**

311   This study provided a quantitative side-by-side comparison between MB and SF

312   fumigations in the same flour mill. The pressurization test showed that sealing

313   effectiveness can be quantitatively determined ahead of a fumigation to quantify gas

314   tightness of a structure. The concentrations of both fumigants varied within a range of 2

315   to 7 g/m^3, which implied similar gas distributions with the mill. The observed HLTs

316   decreased with increasing wind speeds regardless of the type of fumigant used. Our

317   results suggest that for a given level of gas tightness of a structure, fumigant leakage

318   rate is a function of the driving forces such as wind speeds rather than inherent gas

319   characteristics of MB and SF.

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320

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415

Figure Captions

417 Figure 1. Hal Ross flour mill and a generic mill floor plan. Note that only one gas

418 introduction point was selected from one of the two points shown in the figure. Only the

419 southwest and northeast gas monitoring locations are represented in the figure out of

420 the six locations.

421 Figure 2. Results of the building pressurization test for each of the six fumigations.

422 Figure 3. Barometric pressures (A), temperatures (B), and relative humidities (C)

423 recorded by the weather station on the mill roof during each of the six fumigations.

424 Figure 4. Fumigant concentrations over time (solid lines) near the ceiling among all five

425 mill floors and hourly-average outside wind speeds outside the mill (open circles) during

426 the first MB (A) and SF (B), second MB (C) and SF (D), and third MB (E) and SF (F)

427 fumigations.

428 Figure 5. Relationship between HLT values (Table 4) and average wind speeds (A) and

429 HLT values and the square roots of the average absolute inside and outside

430 temperature differences (B). The data points for MB and SF fumigations were plotted as

431 closed circles and closed squares, respectively. The dashed line in A shows Equation 5

432 fitted to the data. Note that the last two HLT values of the third MB fumigation (open

433 circles) were not included in the curve-fitting calculations (see text for details).

434 Table 1. Quantities of MB and SF fumigants used and gas introduction times.

| Fumigation | Fumigant introduction | | Exposure period (h) | Introduced amount (kg) on mill floor | | | | | |
	Date	Time		First	Second	Third	Fourth	Fifth	Total
MB1	6 May 2009	6:40 pm	24	22.7 +22.7[a]	22.7	22.7	45.4	45.4	181.6
SF1	27 May 2009	6:00 pm	24.5	113.6	113.6	113.6	113.6	113.6	568.0
MB2	11 Aug 2009	2:50 pm	24	22.7	22.7	22.7	45.4	45.4	158.9
SF2	19 Aug 2009	2:45 pm	24	113.6	56.8	113.6	113.6	113.6	511.2
MB3	11 May 2010	5:00 pm	24.3	+22.7[b] +18.1[c]	22.7	22.7	45.4	45.4	199.6
SF3	25 May 2010	5:10 pm	25	113.6	113.6	113.6	113.6 +28.3[d]	113.6 +28.3[d]	623.7

435 [a]Top-up (additional gas) release at 9:50 am on 7 May 2009.

436 [b]Top-up release at 8:15 am on 12 May 2010.

437 [c]Top-up release at 9:45 am on 12 May 2010.

438 [d]Top-up release at 7:50 am on 26 May 2010.

439

440   Table 2. Coefficients (mean ± SE) from Equation 1 fitted to pressure-airflow rate data.

Fumigation	No. observations	b	n	r^2
MB1	34	0.102 ± 0.007	0.655 ± 0.017	0.982
SF1	38	0.112 ± 0.007	0.630 ± 0.016	0.978
MB2	38	0.105 ± 0.004	0.639 ± 0.009	0.993
SF2	70	0.279 ± 0.031	0.445 ± 0.027	0.819
MB3	72	0.105 ± 0.009	0.603 ± 0.021	0.916
SF3	77	0.098 ± 0.002	0.634 ± 0.005	0.995

441

442

443   Table 3. Mean ± SE values for temperature and relative humidity observed inside the flour mill during fumigations.

Fumigation	Floor				
	First	Second	Third	Fourth	Fifth
Temperature (°C)					
MB1	21.9 ± 0.009	22.2 ± 0.010	22.3 ± 0.014	23.0 ± 0.014	23.0 ± 0.008
SF1	23.3 ± 0.006	24.4 ± 0.004	25.2 ± 0.003	25.7 ± 0.009	25.6 ± 0.000
MB2	26.7 ± 0.013	28.6 ± 0.011	30.0 ± 0.010	30.9 ± 0.010	31.1 ± 0.005
SF2	27.9 ± 0.005	29.7 ± 0.009	31.1 ± 0.002	31.9 ± 0.001	31.1 ± 0.000
MB3	23.6 ± 0.007	23.8 ± 0.008	24.4 ± 0.000	24.7 ± 0.007	25.4 ± 0.009
SF3	27.6 ± 0.009	28.3 ± 0.010	28.4 ± 0.015	28.9 ± 0.014	29.3 ± 0.009
Relative humidity (%)					
MB1	46.3 ± 0.097	45.2 ± 0.088	44.3 ± 0.056	42.7 ± 0.064	40.9 ± 0.080
SF1	43.2 ± 0.030	40.1 ± 0.028	37.6 ± 0.025	36.7 ± 0.025	34.8 ± 0.028
MB2	57.6 ± 0.049	50.6 ± 0.043	46.0 ± 0.031	43.5 ± 0.029	41.3 ± 0.018
SF2	54.2 ± 0.027	46.5 ± 0.064	43.0 ± 0.023	41.1 ± 0.031	41.1 ± 0.031
MB3	34.7 ± 0.043	33.4 ± 0.035	32.1 ± 0.026	31.1 ± 0.021	29.2 ± 0.022
SF3	49.8 ± 0.122	46.5 ± 0.047	46.0 ± 0.036	43.1 ± 0.037	42.1 ± 0.066

444

445

446 Table 4. Mean and SE estimated half-loss times (HLT) and average wind speeds and corresponding elapsed time periods

447 in which these two values were calculated.

Fumigation	Elapsed exposure period (h)	Absolute average temperature difference (°C)	Average wind speed (m/s)	HLT (h)			
				No. observations	Mean	SE	r^{2a}
MB1	5-15	5.96	2.45	28	28.76	0.001	0.962
	17-24	4.03	7.12	19	9.65	0.002	0.971
SF1	5-24	8.00	3.67	55	19.75	0.000	0.992
MB2	5-24	5.35	2.16	41	28.64	0.000	0.976
SF2	11-21	12.25	3.00	24	28.29	0.000	0.986
	21-24	6.01	6.90	8	9.97	0.002	0.981
MB3	4-8	10.95	5.04	9	10.80	0.003	0.917
	8-15	12.50	4.93	18	3.61	0.004	0.983
	20-24	10.67	3.11	10	9.71	0.003	0.967
SF3	13-25	4.36	2.10	26	31.65	0.001	0.955

448

449 [a]The r^2 values were based on linear regression of hourly fumigant concentration (y) versus elapsed time (x).

450 In an hour, there were 2 to 3 points of average fumigant concentration data. The curve generated from

451 Equation 2 intercepts y-axis at $y = C_i$ (i.e., the average concentration at the beginning of each fumigant concentration

25

452   curve section). Plotting Equation 2 on a semi-log scale gives a straight line, the slope of which is

453   essentially the HLT.

454

455

Figure 1

1. Work area
2. Air ventilation shaft
3. Stair well
4. Service room
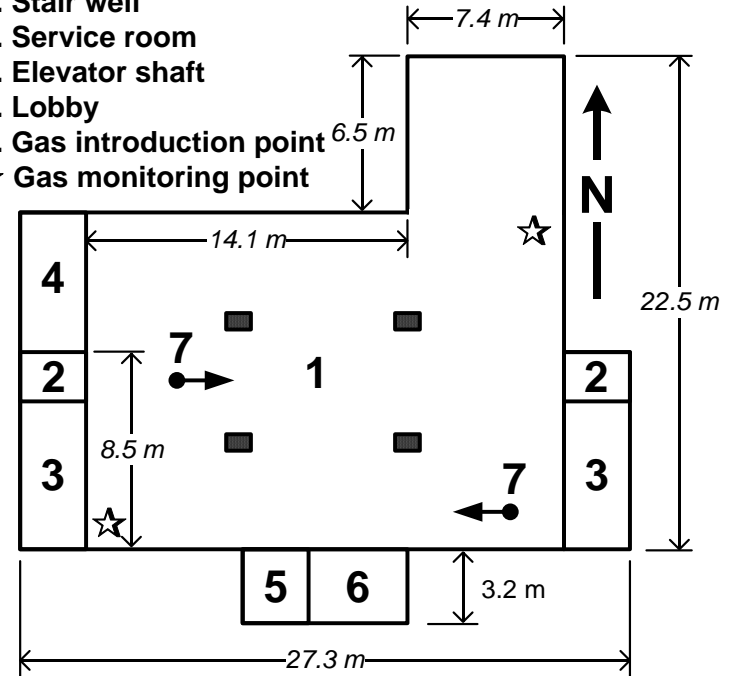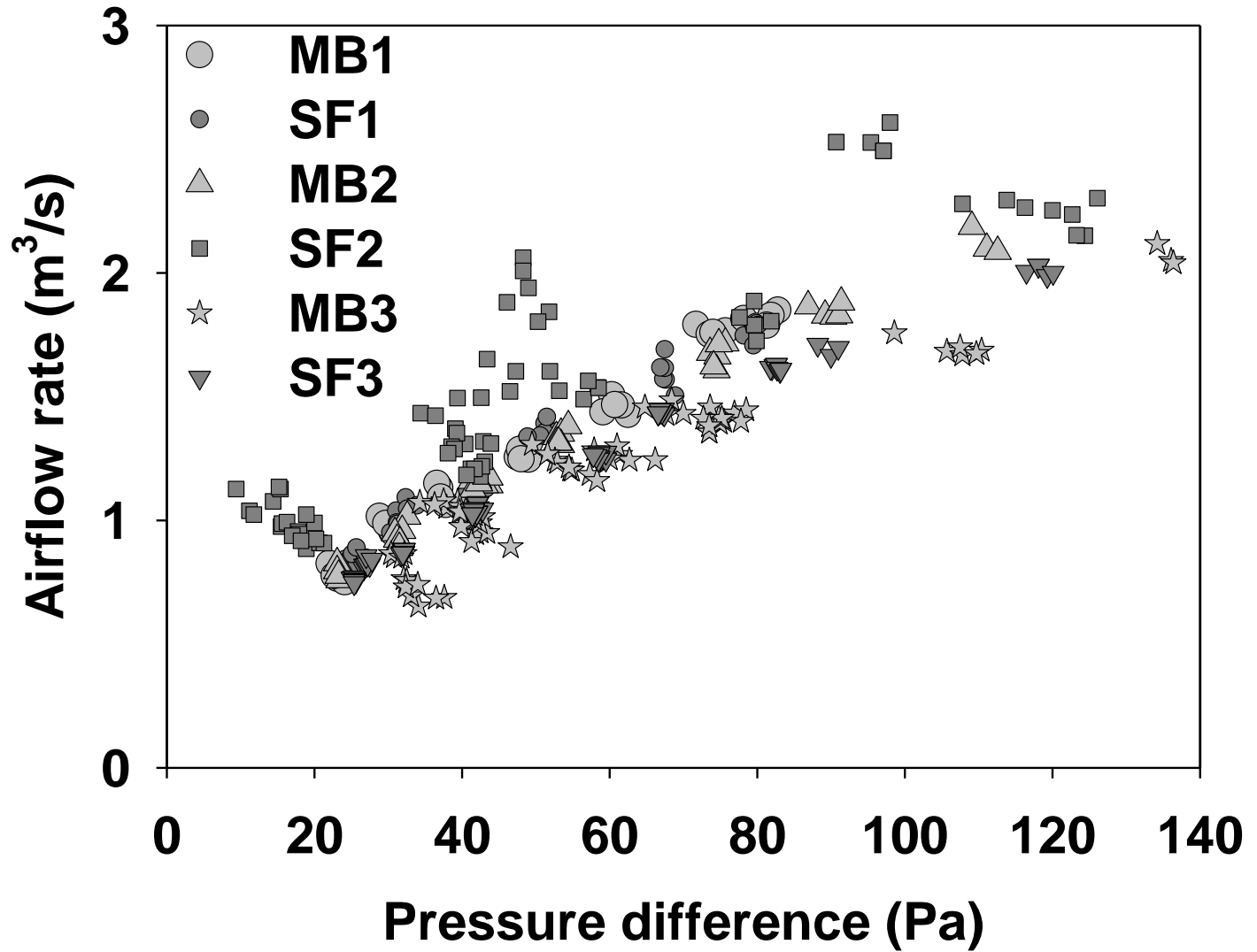5. Elevator shaft
6. Lobby
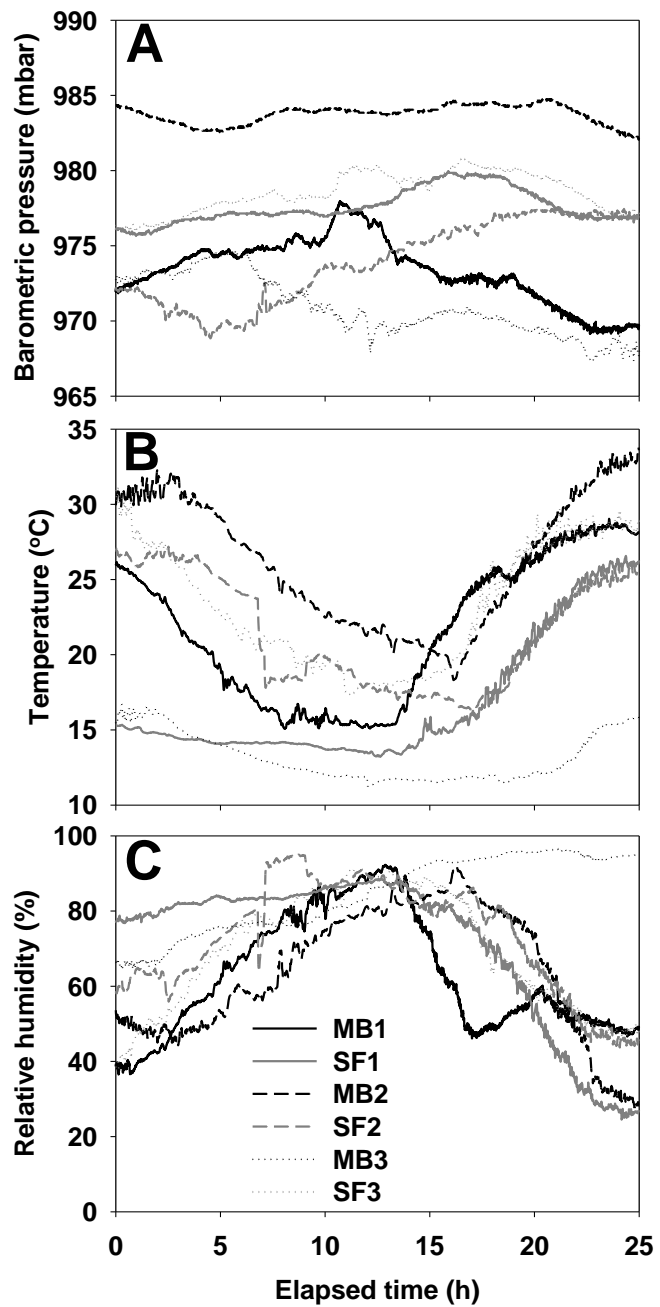7. Gas introduction point
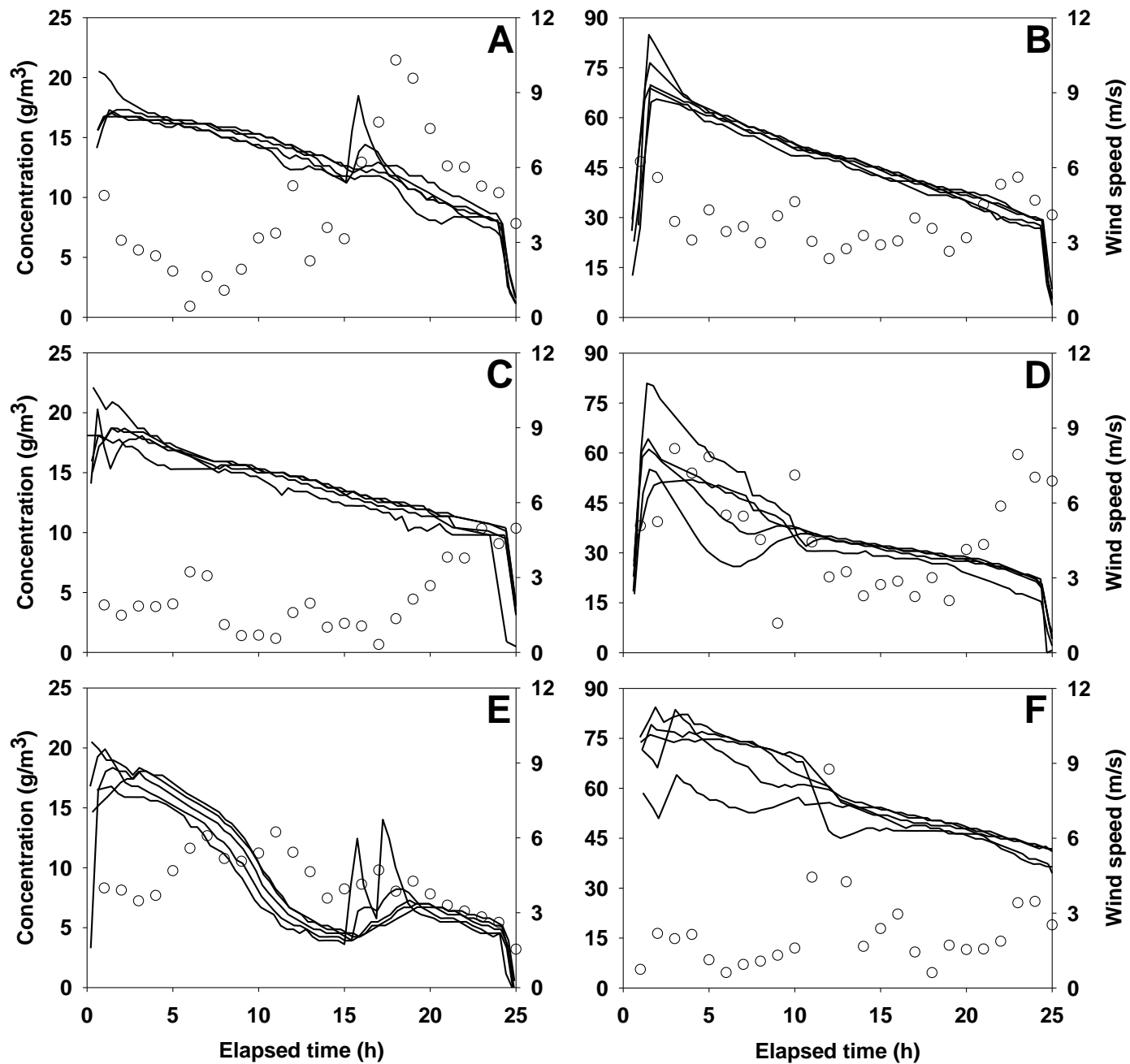☆ Gas monitoring point

Figure 2

Figure 3

Figure 4

Figure 5