Nylon Film Enclosures for Protection of Foods from Exposure to Sulfuryl Fluoride and Methyl Bromide during Structural Fumigation

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Sulfuryl fluoride (SF) and methyl bromide (MB) residues were quantified in four refrigerated (3 °C) and nine cupboard (22 °C) foods by gas chromatographic headspace analysis after fumigation and aeration. Food items were protected in double-sealed bags of two nylon films or polyethylene film before 20 h exposures to 6113, 780, or 93 mg·h/L SF or 707 mg·h/L MB (3 °C); or 6582, 742, or 105 mg·h/L SF or 735 mg·h/L MB (22 °C). Mean corrected residues for all foods and enclosure types ranged from 0 to 103 ppb w/w for SF and from 0 to 46 300 ppb w/w for MB. No SF residues were detected in any foods protected by nylon enclosures at the intermediate and low SF exposures. MB residues in foods were reduced up to >900-fold in nylon bags as compared to residues detected in foods enclosed in polyethylene film. Residues of both SF and MB were significantly reduced in foods protected by nylon film relative to foods enclosed in polyethylene bags regardless of exposure conditions.

Keywords: *Fumigant residues; headspace analysis; food protection; polyethylene film*

Structural fumigation with sulfuryl fluoride (SF) and methyl bromide (MB) has been practiced for decades (Hunt, 1949; Stewart, 1957) to control drywood termites, wood-boring beetles, and various miscellaneous structural and household pests not easily controlled by other, less demanding, methods. Fumigation is the treatment of choice over localized or "spot" treatments when infestations are extensive or difficult to access or delineate or when a residual treatment is undesirable. The resident pest population is eradicated because fumigation exposes the entire structure and all of its contents, including wood matrices (Scheffrahn et al., 1992c), to lethal concentrations of the gaseous toxicant. Over 200 000 structural fumigations are conducted annually in the United States, the majority for control of drywood termites in California, Florida, Hawaii, and other southeastern and western states (Scheffrahn et al., 1988; Scheffrahn, unpublished observation).

Label directions for SF (DowElanco, 1993) and MB (Great Lakes Chemical Corp, 1992) dictate that foods, feed, and medicines must either be properly protected from fumigant exposure (e.g., sealed metal or glass containers or, on older SF labels, polyethylene bags) or be removed from the structure because unprotected commodities are likely to harbor transient or permanent residues of either SF (Meikle and Stewart, 1962; Osbrink et al., 1988; Scheffrahn et al., 1989a,b) or MB (Daft, 1988, 1989; DeVries et al., 1985; Meikle and Stewart, 1962) after fumigation. Many consumer foods packaged in manufacturer-sealed containers of various materials and closure types are also susceptible to fumigant exposure and residue formation (Scheffrahn et al., 1992b), and often the packaging does not constitute adequate protection.

The removal of foods from a house or commercial building before fumigation of the structure is often impractical or even unfeasible. Scheffrahn et al. (1990) demonstrated that nylon polymer film was more refractive to fumigant permeation than standard polyethylene film. Empty double nylon bags partially filled with air allowed the entry of only 0.003 and 0.02% of external time-weighted concentrations of SF and MB, respectively (Scheffrahn et al., 1990). It was hypothesized that differences between within-bag concentrations of fumigant and external exposure concentrations of this magnitude would greatly minimize or preclude residue formation of foods enclosed in such bags. This would allow for a convenient method of *in situ* protection and storage of food items during fumigation.

The current study was conducted to validate the performance of nylon bags for the reduction or prevention of fumigant residues in foods. Following fumigation, we quantified SF and MB residues in various cupboard and refrigerated commodities sealed in nylon and polyethylene bags.

MATERIALS AND METHODS

Note: Sulfuryl fluoride and methyl bromide are restricted-use pesticides. These gases are colorless and odorless at concentrations harmful or lethal to humans and must be handled with extreme caution by certified personnel.

Foods and Enclosures. Thirteen food items, procured from a local retail outlet, were selected to represent common proteinaceous, fatty, carbohydrate, and raw vegetable food commodities found in typical households. Refrigerated (3 °C) foods included ground beef, ca. 100 g (27–29% fat), wrapped in butcher paper; Tropicana orange juice in an opened, half-full ≤473 mL paper carton; iceberg lettuce, unwrapped, quartered head; and whole milk in an opened, half-full ≤473 mL paper carton. Cupboard (22 °C) foods included single whole Red Delicious apples; Nabisco Shredded Wheat breakfast cereal in two unopened, unlined 23.6 g cardboard boxes; Ken-L-Ration Kibbles 'n Bits dry dog food, ca. 100 g, in a no. 1 kraft paper bag; unbleached wheat flour, ca. 100 g, in a no. 1 kraft paper bag; Carnation nonfat dry milk, ca. 100 g, in a no. 1 kraft paper bag; Crisco vegetable oil in an opened and reclosed, half-full ≤473 mL polyethylene terephthalate bottle; a single whole Russet potato; a single whole Hostess Twinkies

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Table 1. Sulfuryl Fluoride and Methyl Bromide Residues[a] in Refrigerated Foods Fumigated in Nylon and Polyethylene Film Enclosures for 20 h at 3 °C and Aerated before Analysis

food and enclosure material	postfumigation aeration time (h)	mean fumigant exposure (mg·h/L ± SD)			
		sulfuryl fluoride			methyl bromide
		93.3 ± 0	780 ± 26.3	6113 ± 669	707 ± 78.6
ground beef					
nylon C	2		0 ± 0	11.6 ± 6.3	49.9 ± 29.1
	6		0 ± 0	1.7 ± 3.5	42.0 ± 32.8
nylon D	2		0 ± 0	7.7 ± 10.0	91.3 ± 66.8
	6		0 ± 0	2.4 ± 4.9	71.2 ± 62.3
polyethylene	2	0 ± 0	4.2 ± 1.1	24.9 ± 22.3	$46.2 \times 10^3 \pm 14.8 \times 10^3$
	6	0 ± 0	1.7 ± 1.3	7.0 ± 6.6	$22.3 \times 10^3 \pm 6.21 \times 10^3$
orange juice					
nylon C	2		0 ± 0	8.8 ± 10.5	14.5 ± 6.3
	6		0 ± 0	5.3 ± 3.4	25.9 ± 17.3
nylon D	2		0 ± 0	14.4 ± 9.6	28.0 ± 10.4
	6		0 ± 0	10.0 ± 4.5	31.5 ± 20.0
polyethylene	2	0 ± 0	13.6 ± 8.4	24.2 ± 16.0	$6.27 \times 10^3 \pm 1.97 \times 10^3$
	6	0 ± 0	7.4 ± 4.6	20.1 ± 11.1	$7.06 \times 10^3 \pm 2.63 \times 10^3$
iceberg lettuce					
nylon C	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
nylon D	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
polyethylene	2			0 ± 0	26.8 ± 22.5
	6			0 ± 0	0 ± 0
whole milk					
nylon C	2		0 ± 0	0 ± 0	54.5 ± 32.0
	6		0 ± 0	0 ± 0	16.3 ± 18.9
nylon D	2		0 ± 0	0 ± 0	69.7 ± 28.3
	6		0 ± 0	0 ± 0	62.8 ± 39.8
polyethylene	2	0 ± 0	5.3 ± 3.1	43.1 ± 17.8	$16.3 \times 10^3 \pm 6.92 \times 10^3$
	6	0 ± 0	4.3 ± 2.9	20.9 ± 10.9	$12.3 \times 10^3 \pm 3.23 \times 10^3$

[a] ppb w/w. Means ± SD. Means, $n = 4$, calculated from exposure, standard, and recovery (see regression equations, Table 4) corrected values. Limit of detection = 1 ppb w/w.

snack cake in the manufacturer's wrapper; and McNeilab Tylenol acetaminophen 325 mg caplets in an opened and reclosed, half-full snap-cap high-density polyethylene bottle (cotton plug removed).

The following three bag types were used as experimental protective enclosures for the above food items: type C large nylon oven cooking bags (Reynolds Metals Co.) (Scheffrahn et al., 1990), 36 × 51 cm, 0.019 mm thick, 1 ply, nylon 6 and 66 polymers); type D large nylon Fumebags (Soil Chemicals Corp. Products) (Scheffrahn et al., 1990), 61 × 93 cm, 0.025 mm thick, proprietary nylon polymer; and polyethylene bags (Cole-Parmer no. N-06502-95), 46 × 61 cm, 0.051 mm (2 mil) thick, 1 ply, low-density polyethylene.

Enclosure of Foods in Bags and Fumigation. A single unit of each food item was individually enclosed in two bags before fumigation. The food item was placed in the first bag and the bag was filled with air inside a rigid container to 1.33 ± 0.2 cm^3 of air/cm^2 of bag surface area. The open end of the bag was twisted thrice, bent 180° on itself, and secured with a wire twist-tie to complete the seal (Scheffrahn et al., 1990). The sealed bag was then placed in a second bag of the same film type. The airspace between the two bags was filled to 0.33 ± 0.1 cm^3/cm^2 (outer bag area), and the second bag was sealed as above. The double-bagged food samples were fumigated with SF and MB in a 4.2-m^3 chamber (Scheffrahn et al., 1987b). Food items were exposed to up to three target concentrations of SF (Vikane, DowElanco, 99%), 360, 36, and 3.6 mg/L (i.e., theoretical time-weighted exposures of 7200, 720, and 72 mg·h/L in this study, respectively), and to MB (Meth-O-Gas, Great Lakes Chemical Corp., 100%) at 36 mg/L. Measured exposure concentrations varied slightly from target concentrations because of slight chamber sorption and/or leakage. For fumigations at 360 mg/L SF and 36 mg/L MB, treatments included all food types from both classes (refrigerator and cupboard) in the three different double-bag enclosures (nylon types C and D and polyethylene) and were replicated four times each. Because of labor and equipment limitations, each fumigation included 12 samples consisting of two replicates of foods in the same class in each of the three enclosure types. Only treatments yielding detectable residues at 360 mg/L SF were fumigated at the next lower SF exposure.

Cupboard food samples were randomly placed on open shelves inside the chamber. Chamber air temperature was set at 22 ± 1 °C. The chamber was sealed, the fumigant introduced, and the chamber concentration verified from triplicate samples taken at 0.25 and 19.75 h from an external chamber sampling port according to the method of Scheffrahn et al. (1990). For refrigerated foods, samples were randomly distributed on shelves inside an upright refrigerator held at 3 ± 1 °C. The refrigerator was housed inside the fumigation chamber, which was maintained at 22 ± 1 °C. In addition to chamber verification samples as taken above, refrigerator verification samples were also taken in triplicate at 2 and 19.75 h via two Tygon sampling lines which recirculated air within the refrigerator food compartment. The sampling lines were connected from a second external chamber sampling port to two 6.4 mm o.d. copper tubes propped between the flexible magnetic door seal and the refrigerator body. To ensure fumigant entry under worst-case assumptions and still maintain an internal temperature of 3 °C, two additional 3 cm pieces of 9.5 mm o.d. copper tubing were placed between the seal and body on the opposite side of the door from where sample lines entered.

The actual time-weighted fumigant exposure values (mg·h/L) of cupboard food samples were determined by calculating mean fumigant concentration of the 0.25 and 19.75 h chamber verification values and multiplying by 20 h. For refrigerated foods, the mean of 2 and 19.75 h refrigerator concentration values were multiplied by 19 h to give an accumulated dosage reflecting fumigant diffusion lag into the refrigerator. All accumulated dosage values assume linear gas diffusion and/or loss.

At 20 h, each fumigation was terminated. The chamber was aerated by opening two external gate valves for 0.1 h, after which time the chamber door was opened and the bagged samples were removed. The bags were immediately opened and food samples removed. Cupboard foods were transferred to shelves in each of three incubators, one for each bag type,

Table 2. Sulfuryl Fluoride and Methyl Bromide Residues[a] in Cupboard Foods Fumigated in Nylon and Polyethylene Film Enclosures for 20 h at 22 °C and Aerated before Analysis

food and enclosure material	postfumigation aeration time (h)	mean fumigant exposure (mg·h/L ± SD)			
		sulfuryl fluoride			methyl bromide
		105 ± 0	742 ± 10.8	6582 ± 210	735 ± 91.8
fresh apple					
nylon C	2			0 ± 0	447 ± 286
	6			0 ± 0	235 ± 49.6
nylon D	2			4.3 ± 5.1	112 ± 40.0
	6			0 ± 0	51.7 ± 42.9
polyethylene	2		0 ± 0	41.5 ± 19.6	$25.5 \times 10^3 \pm 4.71 \times 10^3$
	6		0 ± 0	0 ± 0	$17.4 \times 10^3 \pm 3.73 \times 10^3$
dry cold cereal					
nylon C	2			0 ± 0	538 ± 139
	6			0 ± 0	148 ± 15.2
nylon D	2			0 ± 0	290 ± 78.1
	6			0 ± 0	44.7 ± 46.2
polyethylene	2		0 ± 0	36.7 ± 46.2	$27.2 \times 10^3 \pm 5.34 \times 10^3$
	6		0 ± 0	9.7 ± 12.0	$9.19 \times 10^3 \pm 1.39 \times 10^3$
dry dog food					
nylon C	2		0 ± 0	3.8 ± 2.5	$1.10 \times 10^3 \pm 246$
	6		0 ± 0	0 ± 0	329 ± 101
nylon D	2		0 ± 0	5.0 ± 4.0	144 ± 62.6
	6		0 ± 0	0.8 ± 0.9	40.9 ± 57.2
polyethylene	2	0 ± 0	7.5 ± 1.3	103 ± 11.5	$44.1 \times 10^3 \pm 8.24 \times 10^3$
	6	0 ± 0	0.5 ± 0.5	18.1 ± 4.3	$25.0 \times 10^3 \pm 7.01 \times 10^3$
wheat flour					
nylon C	2			0 ± 0	342 ± 426
	6			0 ± 0	91.8 ± 121
nylon D	2			0.3 ± 0.8	68.6 ± 96.6
	6			0 ± 0	3.4 ± 6.8
polyethylene	2		0 ± 0	3.8 ± 2.7	$6.68 \times 10^3 \pm 6.82 \times 10^3$
	6		0 ± 0	0.8 ± 1.7	$2.41 \times 10^3 \pm 2.27 \times 10^3$
nonfat dry milk					
nylon C	2			0 ± 0	26.9 ± 23.2
	6			0 ± 0	0 ± 0
nylon D	2			0 ± 0	11.3 ± 22.7
	6			0 ± 0	0 ± 0
polyethylene	2		0 ± 0	0 ± 0	$1.42 \times 10^3 \pm 680$
	6		0 ± 0	0 ± 0	308 ± 117
vegetable oil					
nylon C	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
nylon D	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
polyethylene	2		0 ± 0	0 ± 0	0 ± 0
	6		0 ± 0	0 ± 0	84.5 ± 21.0
fresh potato					
nylon C	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
nylon D	2			0 ± 0	0 ± 0
	6			0 ± 0	0 ± 0
polyethylene	2		0 ± 0	0 ± 0	847 ± 62.9
	6		0 ± 0	0 ± 0	471 ± 63.1
snack cake					
nylon C	2			0 ± 0	76.5 ± 90.1
	6			0 ± 0	30.3 ± 36.5
nylon D	2			0 ± 0	57.0 ± 66.0
	6			0 ± 0	10.6 ± 17.3
polyethylene	2	0 ± 0	2.5 ± 1.0	7.5 ± 6.8	$6.17 \times 10^3 \pm 6.58 \times 10^3$
	6	0 ± 0	0 ± 0	1.0 ± 1.3	$1.82 \times 10^3 \pm 1.86 \times 10^3$
acetaminophen					
nylon C	2			0 ± 0	71.5 ± 36.3
	6			0 ± 0	43.3 ± 17.0
nylon D	2			0 ± 0	11.5 ± 5.4
	6			0 ± 0	3.5 ± 3.2
polyethylene	2		0 ± 0	0.8 ± 1.2	$4.97 \times 10^3 \pm 1.14 \times 10^3$
	6		0 ± 0	0 ± 0	$3.16 \times 10^3 \pm 1.19 \times 10^3$

[a] ppb w/w. Means ± SD. Means, $n = 4$ except as noted in Table 3, calculated from exposure, standard, and recovery (see regression equations, Table 4) corrected values. Limit of detection = 1 ppb w/w.

set at 22 ± 1 °C. Refrigerated foods were placed in three incubators held at 3 ± 1 °C.

Postfumigation Handling and Residue Analysis. At 1–2 h postfumigation, 12 10 g subsamples of each food/bag combination were removed from incubators and transferred into headspace vials (120 mL serum bottles; Scheffrahn et al., 1987a). The lettuce, apple, and potato were subdivided by knife before loading into vials. At 2 h postfumigation, sample vials and an empty air (background contamination) vial were sealed with Fisher 20 mm TFE-lined septa. At 5–6 h postfumigation, the above procedure was repeated except that vials were sealed at 6 h postfumigation. Fumigant distribution between sample matrix and vial headspace was allowed to equilibrate for 24 ± 2 h at 22 ± 1 °C prior to GC analysis of SF and MB by the gas chromatographic method of Scheffrahn et al. (1990). A calibration plot of gas chromatograph electron-

Table 3. Mean Residues[a] of All Food Samples (n) per Enclosure Type within a Given Aeration and Fumigant Exposure Class

refrigerated foods, aeration time and enclosure material	fumigant exposure (mg·h/L) SF 780	SF 6113	MB 797
2 h aeration			
nylon C	0.0a (12)[b]	5.1a (16)	30a (16)
nylon D	0.0a (12)	5.6a (16)	47a (16)
polyethylene	7.7b (12)	23.1b (16)	17220b (16)
6 h aeration			
nylon C	0.0a (12)	1.8a (16)	21a (16)
nylon D	0.0a (12)	3.1a (16)	41a (16)
polyethylene	4.5b (12)	12.0b (16)	10416b (16)

cupboard foods, aeration time and enclosure type	fumigant exposure (mg·h/L) SF 742	SF 6582[c]	MB 735
2 h aeration			
nylon C	0.0a (4)	0.4a (36)	77a (36)
nylon D	0.0a (4)	1.0a (38)	289a (36)
polyethylene	1.1b (36)	20.3b (40)	12979b (36)
6 h aeration			
nylon C	0.0a (4)	0.0a (36)	17a (36)
nylon D	0.0a (4)	0.1a (38)	97a (36)
polyethylene	0.1a (36)	3.2b (40)	6645b (36)

[a] ppb. Means within each class followed by the same letter are not significantly different (Student–Newman–Kuels test, $P > 0.05$). [b] Numbers in parentheses = n. [c] Additional replicates included were one potato, one cake, and two acetaminophen for polyethylene and one potato and one flour for nylon D.

capture detector response was generated with three- or five-level standard dilutions of fumigant in empty headspace vials. An external standard was quantified before and after the analysis of each 12-sample series. Background laboratory air samples were also run after each headspace series. The minimum level of detection for both fumigants was 1.0 ppb w/w (Tables 1–3). Residues were evaluated in commodities exposed to the highest SF concentration first. If no residues were detected in a given commodity, that commodity was deleted from the next lower exposure level. In a few instances, additional replicates were included for commodities that yielded residues near detection limits (see footnotes, Table 3). The bag-type variable for each aeration and fumigant exposure class was evaluated with one-way ANOVA (complete randomized design) for fumigant treatments (Table 3). Significant differences ($P > 0.05$) among residue means for each aeration and exposure class were separated by Student–Newman–Keuls test using the PROC GLM procedure (SAS Institute, 1988).

Recovery efficiency of SF and MB was determined by fortifying untreated commodities in vials at five levels approximating those found in fumigated samples (Table 4). Standard air dilutions of SF and MB were injected through headspace vial septa 24 h prior to GC analysis to allow for analyte equilibrium in the food matrices. Regression of natural logs of fortification concentrations on natural logs of headspace concentrations was calculated by PROC REG (SAS Institute, 1988) for each commodity. Samples at each fortification level were replicated four times. Regression equations of recoveries were used to correct residue concentrations in headspace samples.

RESULTS AND DISCUSSION

Mean accumulated exposures of all the bagged refrigerated foods were 6113, 780, and 93 mg·h/L for SF and 709 mg·h/L for MB. Typical structural fumigation rates are ca. 140 mg·h/L SF and ca. 207 mg·h/L MB for drywood termite control (Scheffrahn et al., 1992a). Corrected fumigant residues detected in the refrigerated foods are listed in Table 1. Neither SF nor MB was detected in any background laboratory air samples. At the highest SF exposure (6113 mg·h/L, >40 times the drywood termite rate), residues between 2 and 24 ppb were detected with all bag types in ground beef and orange juice and at 43 ppb in whole milk protected by polyethylene bags. Lettuce and whole milk in nylon bags yielded no detectable SF residues. At 780 mg·h/L SF (ca. 6 times the drywood termite rate), no residues were detected in either nylon enclosure type for refrigerated commodities, although, in polyethylene bags, low residues were detected at both 2 and 6 h of aeration in ground beef, orange juice, and whole milk.

MB residues were detected in ground beef, orange juice, and milk regardless of bag types (Table 1). However, refrigerated foods in nylon bags yielded MB residues 200–>900-fold less than those found in polyethylene bags. Residues in lettuce were limited to 2 h aeration samples in polyethylene enclosures only.

Bag-protected cupboard foods were exposed to mean accumulations of 6582, 742, and 105 mg·h/L for SF and 735 mg·h/L for MB resulting in residues (Table 2). Of these nine foods, only the apple, dog food, and flour yielded SF residues after 2 h of aeration in nylon bag samples at the 6582 mg·h/L exposure. Except for 0.8 ppb of SF found in nylon D protected dog food, no residues were detected in any other nylon bag samples

Table 4. Log–Log Regression Equations of Recoveries of Sulfuryl Fluoride (SF) and Methyl Bromide (MB) from Unfumigated Foods in Tables 1 and 2 Spiked at Five Fortification Levels[a]

commodity	sulfuryl fluoride regression eq	R^2	methyl bromide regression eq	R^2
	Refrigerated Foods			
ground beef	$C_r = 0.9977C_f - 0.2805$	0.999	$C_r = 0.9940C_f - 1.1653$	0.993
orange juice	$C_r = 1.0424C_f - 0.2765$	0.997	$C_r = 1.0703C_f - 0.9169$	0.989
lettuce	$C_r = 1.6117C_f - 2.5141$	0.937	$C_r = 1.0483C_f - 0.9438$	0.996
fresh milk	$C_r = 1.0185C_f - 0.2428$	0.993	$C_r = 1.1318C_f - 1.5006$	0.992
	Cupboard Foods			
fresh apple	$C_r = 0.8993C_f - 0.6233$	0.958	$C_r = 1.0071C_f - 0.5035$	0.998
dry cereal	$C_r = 0.7784C_f + 0.0417$	0.986	$C_r = 1.0145C_f - 1.0239$	0.990
dry dog food	$C_r = 0.8168C_f + 0.1620$	0.980	$C_r = 1.0155C_f - 1.1815$	0.996
wheat flour	$C_r = 1.0648C_f - 0.5090$	0.998	$C_r = 1.1565C_f - 1.9744$	0.991
dry milk	$C_r = 0.9700C_f - 0.4429$	0.958	$C_r = 1.0299C_f - 1.0547$	0.994
vegetable oil	$C_r = 0.9140C_f - 0.1989$	0.995	$C_r = 0.9949C_f - 1.9826$	0.995
fresh potato	$C_r = 0.9993C_f - 2.4903$	0.442	$C_r = 1.0391C_f - 1.8808$	0.991
snack cake	$C_r = 0.8660C_f - 0.0943$	0.974	$C_r = 0.9785C_f - 0.4017$	0.994
acetaminophen	$C_r = 0.9311C_f - 0.0300$	0.980	$C_r = 0.9786C_f + 0.0842$	0.998

[a] SF fortification levels: 2.5, 7.5, 22.5, 67.5, and 202.5 ppb w/w for refrigerated foods and 2.5, 5, 10, 20, and 40 ppb w/w for cupboard foods. MB fortification levels: 4.65, 46.5, 465, 4651, and 23256 ppb w/w for all foods. [b] C_r = natural log of recovered headspace concentration in ppb; C_f = natural log of fortification concentration.

at this SF exposure when samples were aerated for 6 h. Polyethylene bag samples yielded residues in apple, cereal, dog food, flour, snack cake, and acetaminophen. At 742 mg·h/L, no SF residues were detected in nylon-protected samples and only low-level residues were found in dog food and snack cake protected by polyethylene.

As with refrigerated foods, residues in cupboard samples fumigated with MB were consistently higher than residues of comparable (742 mg·h/L) SF-exposed samples (Table 2). As expected, MB residues were highest among the higher fat or more sorptive dry commodities with the exception of vegetable oil. The oil was protected in polyethylene terephthalate bottles with snap-cap closures in which some latent MB breakthrough (i.e., commodity sorption originating from packaging during aeration) was detected in 6 h samples.

Mean separation analysis of residue values using bag type as the tested variable within each aeration and fumigant exposure class is given in Table 3. No significant differences ($P > 0.05$) were observed between the mean residues of foods protected in either nylon enclosures. However, significant differences were detected in all but one exposure class (742 mg·h/L SF, 6 h cupboard foods) between residues in nylon-enclosed foods and residues detected in polyethylene-protected foods.

Log–log regression equations of recoveries from fortified commodities are listed in Table 4. As reported previously (Osbrink et al., 1988), percent recovery of SF is generally high, even at parts per billion level fortifications. MB recoveries in this study were found to be considerably lower with many commodities, especially fatty ones, as has been previously noted (Daft, 1988, 1989; Scheffrahn et al., 1992b). Poor recoveries may be due to MB dealkylation in commodities (Meikle and Stewart, 1962), nonoptimum equilibration time (DeVries et al., 1985), and high fat affinity (Daft, 1988, 1989).

The results of this study demonstrate that, under normal field conditions, the enclosure of foods in nylon film before fumigation will preclude detectable SF residues in foods and restrict MB residues to well below 1 ppm. Osbrink et al. (1988) and DeVries et al. (1985) respectively demonstrated that the desorption of SF and MB from commodities is rather rapid during the first 1–2 days after exposure. This would further lessen the potential for residue exposure to humans if foods are not immediately consumed after fumigation. Currently there are no food residue tolerances established for SF (E. M. Thoms, DowElanco, personal communication), and tolerances for MB are pending (J. E. Sargent, Great Lakes Chemical Corp., personal communication). For comparison with residue values in this study, the established inhalation exposure limits (threshold limit values) for SF and MB are 5000 and 3000 ppb (v/v), respectively (DowElanco, 1993; Great Lakes Chemical Corp., 1992).

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LITERATURE CITED

Daft, J. L. Rapid determination of fumigant and industrial chemical residues in food. *J. Assoc. Off. Anal. Chem.* **1988**, *71*, 748–760.

Daft, J. L. Determination of fumigants and related chemicals in fatty and nonfatty foods. *J. Agric. Food Chem.* **1989**, *37*, 560–564.

DeVries, J. W.; Broge, J. M.; Schroeder, J. P.; Bowers, R. H.; Larson, P. A.; Burns, N. M. Headspace gas chromatographic method for determination of methyl bromide in food ingredients. *J. Assoc. Off. Anal. Chem.* **1985**, *68*, 1112–1117.

DowElanco. Vikane specialty gas fumigant specimen label; Indianapolis, IN, 1993.

Great Lakes Chemical Corp. Directions for use of the products Brom-O-Gas (GLK 160G); West Lafayette, IN, 1992.

Hunt, R. W. The common dry-wood termite as a pest. *J. Econ. Entomol.* **1949**, *42*, 959–962.

Meikle, R. W.; Stewart, D. The residue potential of sulfuryl fluoride, methyl bromide and methanesulfonyl fluoride in structural fumigations. *J. Agric. Food Chem.* **1962**, *10*, 393–397.

Osbrink, W. L. A.; Scheffrahn, R. H.; Hsu, R.-C.; Su, N.-Y. Sulfuryl fluoride residues of fumigated foods protected by polyethylene film. *J. Agric. Food Chem.* **1988**, *36*, 853–855.

SAS Institute. *SAS/STAT User's Guide*, release 6.03; SAS Institute: Cary, NC, 1988.

Scheffrahn, R. H.; Osbrink, W. L. A.; Hsu, R.-C.; Su, N.-Y. Desorption of residual sulfuryl fluoride from structural and household commodities by headspace analysis using gas chromatography. *Bull. Environ. Contam. Toxicol.* **1987a**, *39*, 769–775.

Scheffrahn, R. H.; Su, N.-Y.; Osbrink, W. L. A. Precise dosage delivery method for sulfuryl fluoride in small-chamber fumigations. *J. Econ. Entomol.* **1987b**, *80*, 705–707.

Scheffrahn, R. H.; Mangold, J. R.; Su, N.-Y. A survey of structure-infesting termites of peninsular Florida. *Fla. Entomol.* **1988**, *71*, 615–630.

Scheffrahn, R. H.; Hsu, R.-C.; Osbrink, W. L. A.; Su, N.-Y. Fluoride and sulfate residues in foods fumigated with sulfuryl fluoride. *J. Agric. Food Chem.* **1989a**, *37*, 203–206.

Scheffrahn, R. H.; Hsu, R.-C.; Su, N.-Y. Fluoride residues in frozen foods fumigated with sulfuryl fluoride. *Bull. Environ. Contam. Toxicol.* **1989b**, *43*, 899–903.

Scheffrahn, R. H.; Hsu R.-C.; Su, N.-Y. Evaluation of polymer film enclosures as protective barriers for commodities from exposure to structural fumigants. *J. Agric. Food Chem.* **1990**, *38*, 904–908.

Scheffrahn, R. H.; Bloomcamp, C. L.; Su, N.-Y. Indoor airborne residues of methyl bromide and sulfuryl fluoride following aeration of fumigated houses. *Indoor Air* **1992a**, *2*, 78–83.

Scheffrahn, R. H.; Bodalbhai, L.; Su, N.-Y. Residues of methyl bromide and sulfuryl fluoride in manufacturer-packaged household foods following fumigation. *Bull. Environ. Contam. Toxicol.* **1992b**, *48*, 821–827.

Scheffrahn, R. H.; Su, N.-Y.; Hsu, R.-C. Diffusion of methyl bromide and sulfuryl fluoride through selected structural wood matrices during fumigation. *Mater. Org.* **1992c**, *27*, 147–155.

Stewart, D. Sulfuryl fluoride—a new fumigant for control of the drywood termite, *Kalotermes minor* Hagen. *J. Econ. Entomol.* **1957**, *50*, 7–11.

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