Sources of Formaldehyde, Other Aldehydes and Terpenes in a New Manufactured House

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Abstract
Formaldehyde, less-volatile aldehydes, and terpene hydrocarbons are generally the predominant air contaminants in new manufactured and site-built houses. This study was conducted to identify the major sources of these compounds in a typically constructed, new manufactured house. Specimens of materials used within the house envelope were collected from the production facility. These were individually preconditioned for 19 ± 4 days and tested for emissions of formaldehyde and the other target compounds using small-scale chambers. Several cabinetry materials, passage doors and the plywood subfloor were the predominant sources of formaldehyde and other aldehydes. The plywood subfloor was the predominant terpene source. Whole-house emission rates for combined materials were predicted based on the emission factors and the corresponding material quantities. These predicted rates were compared to whole-house emission rates calculated from measurements made at the house three months after its installation. For 11 of 14 target compounds including formaldehyde, the predicted and calculated rates were within a factor of two. This generally good agreement indicates that the predominant sources were correctly accounted for. Based on these results, practices are proposed for reducing the concentrations of the target compounds in newly constructed houses.

Key words Houses; Formaldehyde; Aldehydes; Wood products; Emissions; Source reduction.

Practical Implications
Formaldehyde may cause sensory irritation among occupants of new houses and less-volatile aldehydes can result in persistent odors. This study demonstrates that the sources of these compounds in new houses are predominantly engineered wood products used for cabinetry, passage doors and the subfloor. The identification of the contaminant sources provides the basis for the development and evaluation of effective mitigation practices. Several practices are proposed that should be relatively simple to implement at modest cost.

Introduction

The concentrations and emission rates of volatile organic compounds (VOCs) were shown to be similar among 11 new manufactured and site-built houses in four different locations (Hodgson et al., 2000). This was attributed to strong similarities in materials and building practices. Formaldehyde, other aldehydes and terpene hydrocarbons (HCs) were generally the predominant compounds. Exposures to formaldehyde are of concern because formaldehyde is a potent sensory irritant and is classified as a probable human carcinogen (Lui et al., 1991; U.S. EPA, 1994). Higher molecular weight aldehydes can produce objectionable odors at low concentrations. The odor thresholds for hexanal and other aldehydes are often exceeded in new houses and may remain elevated for months after construction (Lindstrom et al., 1995; Hodgson et al., 2000). Terpene HCs are of potential concern because they react with ozone to produce ultrafine particles (Weschler and Shields, 1997). Animal studies also indicate that strong sensory irritants are formed by terpene-ozone reactions (Wolkoff et al., 2000). Wood and engineered wood products are the likely major sources of aldehydes and terpene HCs in new houses.

This study was conducted to identify and verify the major sources of aldehydes and terpene HCs in a new manufactured house. Laboratory emission tests were conducted with a number of wood and engineered wood products and measurements were made in the house. Although only a single house was studied, the information on sources is anticipated to have broad application to residential construction due to the widespread use of similar materials and building practices.

Material and Methods

The manufactured house was typical of better quality two-section houses produced in Florida. It was completed in November 1999. Within three weeks, it was installed at a nearby site. The house was used daily as a sales model. It was decorated, fully furnished, but unoccupied. There were three bedrooms and two bathrooms. The floor area was 160 m^2; the volume was 414 m^3. 19% of the floor was ceramic tile; 69% of the floor was carpeted. The manufacturer supplied a detailed list of materials used in the house. Between December 1999 and January 2000, we collected ~30 specimens of the major materials from the production facility. These were cataloged, packaged in aluminum foil, and shipped to the laboratory by airfreight. The specimens were stored at room conditions in their original packages until they were tested. Most materials were tested within three months of collection.

Air sampling in the house and outdoors was conducted in March 2000. The house ventilation rate was quantified concurrently by tracer gas decay. The ventilation rate measurement and the VOC air sampling and analytical methods for field and chamber work have been described previously (Hodgson et al., 2000). The only change was that the VOC sorbent samplers (P/N 16251; Chrompack, Bergen of Zoom, The Netherlands) were modified by the addition of a 15-mm section of Carbosieve S-III 60/80 mesh (P/N 10184, Supleco Inc., Bellefonte, PA, USA) toward the outlet end (an equivalent length of Tenax-TA was removed). VOCs collected on sorbent samplers were quantitatively analyzed by thermal desorption gas chromatography/mass spectrometry. Formaldehyde and acetaldehyde were collected on treated silica cartridges and analyzed by high-performance liquid chromatography.

Specimens for chamber testing were cut from larger pieces of the materials. The exposed surface area of a specimen was typically 0.02 m^2. Cut edges and backs of specimens were covered with stainless steel plates and sealed with low-emitting aluminum foil tape. All specimens were conditioned for 19 ± 4 days prior to emission testing. Salthammer (1997) utilized the same preconditioning period for furniture in order to obtain data useful for estimating long-term exposures. Conditioning was conducted in individual 19-L containers supplied with charcoal filtered air at ~4 L min^{-1}. The temperature and humidity of the conditioning chambers were maintained at 20 ± 3^o C and 45 ± 15% relative humidity (RH). At the end of the conditioning period, a test specimen was transferred to a 10.5-L stainless-steel chamber maintained at 23 ± 1^o C and 50 ± 10% RH. Nitrogen was introduced at 1.0 ± 0.05 L min^{-1}. For the 0.02 m^2 sample size, the chamber loading ratio (L) was 1.9 m^2 m^{-3}. At the 5.7 h^{-1} ventilation rate (N), the L/N value was 0.33 h m^{-1}. The specimen was maintained at this condition for 48 hours. Then, gas samples for VOCs were collected from the chamber exhaust.

Emission rates (mass per time) and emission factors (mass per area-time) of VOCs were calculated for the house and the chamber tests using the steady-state form of the mass-balance model for well-mixed chambers (ASTM, 1997).

Results

The quantities of the predominant wood and engineered wood products contained within the house envelope are listed in Table 1. The cabinetry in the kitchen, baths and utility area was identically constructed and was composed of seven major materials. The majority of these

materials had a vinyl coating, either white or woodprint, applied to one or more surfaces. The top surface of the particleboard (PB) countertop was finished with decorative laminate. There were five materials with ~44 m^2 of bare, unfinished surfaces. The other predominant wood products contained within the house were the passage doors (molded high-density fiberboard with a white acrylic factory finish) and the plywood subfloor under the carpeted areas.

The emission factors for individual VOCs had a lower limit of quantitation of ~3 $\mu g\ m^{-2}\ h^{-1}$. Only three materials had significant emissions of terpene HCs. These were the PB counter top, cabinet frame lumber and plywood subfloor (Table 2). The predominant terpene HCs were α-pinene, β-pinene and d-limonene. Other detected terpenes included Δ^3-carene, camphor and borneol. The plywood subfloor had the highest terpene HC emission factors and predicted whole-house emission rates.

Six materials were significant sources of formaldehyde and other aldehydes. The emission factors shown in Table 3 are for bare surfaces, i.e., the underside of the PB counter top, the uncoated side of the PB used for the cabinet cases and the exposed medium density fiberboard (MDF) surface of the cabinet stiles. The emissions of aldehydes from the vinyl-coated surfaces of the cabinetry materials were very low, frequently $<3\ \mu g\ m^{-2}\ h^{-1}$. The PB case had the highest formaldehyde emission factor but did not emit measurable amounts of the other aldehydes. The MDF surface of the stiles had a relatively high formaldehyde emission factor. The PB countertop and the stiles generally had the highest emission factors for the other aldehydes.

The aldehyde emission factors for the six materials were multiplied by their respective surface areas in Table 1 to produce estimates of whole-house emission rates (Table 4). The PB case and the passage doors had the highest formaldehyde emission rates. The fractional contributions of the formaldehyde sources are illustrated in Figure 1. The plywood subfloor had the highest predicted emission rates for hexanal and the other aldehydes.

The indoor and outdoor concentrations of terpene HCs and aldehydes for the house approximately three months after the house was installed are presented in Table 5.

The ventilation rate at the time of sample collection was 0.28 h^{-1} ($r^2 = 0.997$). The whole-house VOC emission rates were calculated assuming that 95% of the house volume was ventilated at the measured rate. Uncertainties in the emission rates were estimated by propagating the relative standard deviation for the VOC concentrations of 7% (Hodgson, 1999) and conservative 10% uncertainties for the ventilation rate and the ventilated house volume. The

calculated emission rates are compared to the sums of the predicted rates for the individual indoor sources in Table 5. For 11 of the 14 compounds, the predicted rates were within a factor of 2 ± 0.1 of the calculated rates. For all compounds except d-limonene and formaldehyde, the predicted rates were less than the calculated rates.

Tests were conducted to evaluate the effectiveness of various barriers for reducing the emissions of terpene HCs and aldehydes from the plywood subfloor. Plywood that had been stored in the laboratory for 6-8 months was utilized for these tests. Plywood specimens were prepared, conditioned and tested for emissions as described above. Then, various treatments were applied, and all assembly edges were sealed. The assemblies were returned to the chambers and tested for emissions after five or seven days. The first of five treatments consisted of a standard bonded polyurethane carpet cushion and a residential carpet installed over the plywood. The next two treatments utilized different brands of spill barrier carpet cushion combined with the same carpet. Cushion A was bonded polyurethane; cushion B was prime polyurethane. The fourth treatment consisted of a reinforced aluminum foil radiant barrier with small uniformly spaced perforations. The fifth treatment consisted of a non-woven fiber, exterior weatherization membrane. The treatment materials were separately tested for VOC emissions. None of these materials were sources of the target compounds.

The emission factors of terpene HCs and aldehydes for the treatments are compared to the emission factors for the bare plywood in Table 6. The emission factors of the three terpene HCs and many aldehydes for the plywood varied substantially among the test specimens. The installation of the standard carpet cushion and carpet over the plywood had no distinguishable impact on the emission factors of all compounds. The other treatments generally resulted in lower emission factors. However, in most cases it is difficult to evaluate the significance of these reductions due to the relatively high uncertainties in the emission factors for plywood. There were consistently 20-40% reductions for the treatments in the emission factors for hexanal, the predominant compound. The perforated aluminum barrier and the non-woven fiber membrane effectively reduced the formaldehyde emission factor.

Vinyl coated passage doors in an identical style are commercially available. A newly manufactured vinyl coated door was obtained. Both the finished surface of the door skin and the door assembly were prepared, conditioned and tested for emissions as described above. These had a formaldehyde emission factor of 5 $\mu g\ m^{-2}\ h^{-1}$ or less.

Discussion

The indoor concentrations of the three terpene HCs were at the low ends of the concentration ranges reported previously for 11 new manufactured and site-built houses; the formaldehyde concentration was slightly higher than the range of reported values; and the concentrations of the other aldehydes were within the reported ranges (Hodgson et al., 2000). These comparisons imply that the study house had typical sources of terpene HCs and aldehydes. The generally good agreement between the calculated and predicted emission rates of formaldehyde and other aldehydes in the study house suggests that the predominant sources of these compounds were correctly accounted for.

The formaldehyde emission factors for five of the indoor sources were generally consistent with the results of a study of formaldehyde emissions from contemporary materials used in California houses (Kelly et al., 1999). In that study, the materials were conditioned for 2-7 days prior to 24-h small-scale chamber testing at typical and exaggerated room conditions. Six industrial particleboards (IPs) were tested; the range of formaldehyde emission factors at typical conditions was 104–237 $\mu g\ m^{-2}\ h^{-1}$. Formaldehyde emission factors for three vinyl-coated IPs were <20 $\mu g\ m^{-2}\ h^{-1}$. Three MDFs were tested; the range of emission factors was 258–364 $\mu g\ m^{-2}\ h^{-1}$. No high-density fiberboard product equivalent to the passage doors was tested. Emission factors for phenol-formaldehyde wood products, such as hardboard and softwood plywoods, were <10 $\mu g\ m^{-2}\ h^{-1}$.

Only a few studies have measured the emissions of VOCs other than formaldehyde from engineered wood products. Bauman et al. (1999) studied the emissions of terpene HCs from PB and MDF specimens produced in North America. Among the 37 PB specimens, those composed of softwood species had the highest terpene HC emission factors. The predominant compounds were pinenes, camphene, Δ^3-carene, p-cymene, limonene and borneol. Emission factors were measured at 48 hours with no preconditioning of materials using small-scale chambers. The median emission factors for α-pinene, β-pinene and limonene were 152, 60 and 23 $\mu g\ m^{-2}\ h^{-1}$, respectively. The 18 MDF specimens (both softwood and hardwood) were not significant sources of terpene HCs. This was attributed to the use of elevated temperature and steam pressure for processing MDF. Of the two PBs in the current study, only the PB counter top was a measurable source of terpene HCs. The plywood subfloor, composed of softwood species, had

terpene HC emission factors that were generally comparable to the emission factors for softwood PBs.

Almost all of the PB and MDF specimens studied by Bauman et al. (1999) were found to emit aldehydes ranging from formaldehyde through nonanal; however, these emissions were not quantified. Su et al. (1999) found that pentanal and hexanal comprised 28 and 12%, respectively, of the total VOCs released during the drying of hardwood flakes for oriented strand board (OSB). They stated that these and other aldehydes are oxidation products of wood components formed during wood drying operations. Barry and Corneau (1999) measured the emissions of aldehydes from OSB panels in a large-scale chamber. The predominant compound was hexanal; pentanal was also a significant constituent. They also reported that pentanal and hexanal dominated the emissions of VOCs from plywoods. In the current study, hexanal was the predominant compound in addition to formaldehyde that was released by the three wood products with substantial aldehyde emissions.

Because formaldehyde is a potent irritant, implementation of practices to reduce formaldehyde concentrations may reduce the potential for sensory irritation among occupants of new houses. Wherever possible, wood products with a urea-formaldehyde resin system should be avoided as they have substantially higher emissions of formaldehyde (Kelly et al., 1999). However, in many cases there may be no practical substitutes. In this study, the predominant sources of formaldehyde were the high-density fiberboard passage doors and the bare PBs and MDF. These were presumably made with a urea-formaldehyde resin system, although the finish on the doors also may have been a source. Vinyl was found to be an effective coating for substantially reducing the emissions of formaldehyde and other aldehydes from the PB case and the MDF cabinet doors. This suggests that the cabinet cases should be constructed with fully coated materials, such as PB with vinyl on both sides. The use of frameless cabinets would eliminate the stiles. Vinyl coated passage doors were shown to have substantially lower formaldehyde emissions than the standard doors. Applied finish materials for cabinets and doors other than vinyl may also be effective. For example, laminate was shown to be an effective emission barrier (Kelly et al., 1999). Thus, a laminate backing sheet applied the undersides of the PB counter tops, a common construction practice, should effectively reduce the formaldehyde emissions from this source.

The concentrations of pentanal, hexanal, heptanal, octanal, 2-octenal and nonanal were all elevated over their respective odor thresholds in the study house (Table 5). The data indicate that the bare plywood subfloor under standard carpet systems is probably the major source of the emissions of these compounds. Thus, control practices directed at this source may help to reduce the occurrence and persistence of objectionable odors. This study shows that spill barrier carpet cushions, a relatively new product category, may be partially effective.

Conclusions

Emission factors and predicted whole-house emission rates of formaldehyde, other aldehydes and terpene HCs were quantified for the predominant wood and wood product sources used in the study house. The general agreement between the laboratory derived whole-house emission rates and the calculated whole-house emission rates determined from the field study of the house suggests that we accounted for the predominant sources of these compounds. It is probable that elevated concentrations of these compounds and the associated potential for sensory irritation and objectionable odors can be reduced in new house construction by several relatively simple practices directed at these sources. These practices are the use of vinyl coated or alternative passage doors, the construction of cabinet cases with fully coated PB, the use of frameless cabinets, the application of a backing sheet to the underside of PB countertops, and the installation of a carpet cushion with a integral spill barrier. The one-time investment required for these modifications is anticipated to be modest.

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Table 1 Quantities of wood and engineered wood products within the house envelope

Material Description	Volume (m^3)	Density (g cm^{-3})	Mass (kg)	Surface Area (m^2) Bare	Coated
Cabinetry Materials					
PB[a] Counter Top	0.192	0.81	155	12.1	---
PB Case, Vinyl 1 Side	0.117	0.64	74	9.2	9.2
PB Case, Vinyl 2 Sides	0.087	0.64	55	---	13.7
Hardboard, Vinyl 1 Side	0.037	1.05	39	11.7	11.7
Stile[b], Vinyl 3 Sides	0.093	0.78	72	4.9	7.4
Door[b], Vinyl coated	0.168	0.77	129	---	17.6
Frame Lumber	0.127	0.47	60	6.6	---
Cabinetry Totals	0.82		584	44	60
Passage Door				---	25
Plywood Subfloor	3.0	0.60	1,830	111[c]	49

[a]PB = Particleboard
[b]Material is medium-density fiberboard (MDF)
[c]Area of plywood subfloor overlain by carpet; remainder not exposed

Table 2 Emission factors (EFs) and predicted whole-house emission rates (ERs) of terpene hydrocarbons for indoor sources

	PB Counter Top		Cabinet Frame Lumber		Plywood Subfloor	
Compound	EF (μg m^{-2} h^{-1})	ER (mg h^{-1})	EF (μg m^{-2} h^{-1})	ER (mg h^{-1})	EF (μg m^{-2} h^{-1})	ER (mg h^{-1})
α-Pinene	23	0.27	14	0.09	196	22
β-Pinene	7	0.09	17	0.11	49	5.4
d-Limonene	6	0.07	<3	<0.02	71	7.9

Table 3 Aldehyde emission factors for indoor sources

	Emission Factor ($\mu g\ m^{-2}\ h^{-1}$)					
	Cabinetry Materials, Bare Surface				Passage	Plywood
Compound	PB Top	PB Case	Hardbd.	Stile	Door	Subfloor
Formaldehyde	87	470	10	330	153	10
Acetaldehyde	40			20	11	15
Pentanal	47			36	8	27
Hexanal	220			260	42	165
2-Furaldehyde	6		72	7		
Heptanal	11			7		3
2-Heptenal	7			9		5
Benzaldehyde	16			42	3	5
Octanal	20			28		8
2-Octenal	16			29		12
Nonanal	18			40		22

Table 4 Predicted whole-house emission rates of aldehydes for indoor sources

	Whole-House Emission Rate ($mg\ h^{-1}$)					
	Cabinetry Materials, Bare Surface				Passage	Plywood
Compound	PB Top	PB Case	Hardbd.	Stile	Door	Subfloor
Formaldehyde	1.05	4.3	0.12	1.64	3.8	1.06
Acetaldehyde	0.47			0.10	0.29	1.66
Pentanal	0.56			0.17	0.19	3.0
Hexanal	2.7			1.25	1.06	18.3
2-Furaldehyde	0.07		0.84	0.03		
Heptanal	0.13			0.03		0.40
2-Heptenal	0.08			0.04		0.54
Benzaldehyde	0.19			0.21	0.07	0.59
Octanal	0.24			0.14		0.86
2-Octenal	0.19			0.14		1.28
Nonanal	0.22			0.19	0.06	2.4

Table 5 Concentrations and whole-house emission rates of terpene hydrocarbons and aldehydes in study house

Compound	Outdoor Conc. ($\mu g\ m^{-3}$)	Indoor Conc. ($\mu g\ m^{-3}$)	Indoor Conc. (ppb)	Odor Threshold[a] (ppb)	Calculated ER[b] ($mg\ h^{-1}$)	Predicted ER[c] ($mg\ h^{-1}$)	Pred/Calc w/in 2 x? Y/N[d]
α-Pinene	1.4	232	41.6	692	25 ± 4	22	Y
β-Pinene	<0.5	73.9	13.3	Nd[e]	8.1 ± 1.3	5.6	Y
d-Limonene	<0.5	40.3	7.2	437	4.4 ± 0.7	8.0	Y
Formaldehyde	4.7	94.9	77.4	871	9.9 ±1.6	12.0	Y
Acetaldehyde	4.6	42.5	23.6	186	4.2 ±0.7	2.5	Y
Pentanal	<0.5	72.9	20.7	6.0	8.0 ± 1.3	3.9	Y
Hexanal	1.5	267	65.3	13.8	29 ± 5	22	Y
2-Furaldehyde	<0.5	15.4	3.9	776	1.69 ± 0.27	0.94	Y
Heptanal	0.7	25.3	5.4	4.8	2.7 ± 0.4	0.56	N
2-Heptenal	<0.5	13.7	3.0	13.5	1.51 ± 0.24	0.66	N
Benzaldehyde	5.5	18.8	4.3	41.7	1.46 ± 0.23	1.05	Y
Octanal	1.5	44.3	8.5	1.3	4.7 ± 0.7	1.24	N
2-Octenal	<0.5	19.3	3.7	2.0	2.1 ± 0.3	1.60	Y
Nonanal	5.8	46.6	8.0	2.2	4.5 ± 0.7	2.9	Y

[a]Odor thresholds are from Devos et al. (1990)
[b]Emission rates (ERs) ± 1 stdev. are calculated from measured concentrations and house parameters
[c]Predicted whole-house ERs are the sums of the ERs for indoor wood and wood product sources
[d]Y/N = Predicted ER is within factor of 2 ± 0.1 of calculated ER, Yes or No?
[e]Nd = No odor threshold data available

Table 6 Emission factors of terpene hydrocarbons and aldehydes for plywood subfloor alone, assemblies of subfloor, carpet cushions and carpet, and subfloor with barrier materials

	Emission Factor ($\mu g\ m^{-2}\ h^{-1}$)					
Compound	Plywood Subfloor[a]	Assembly[b] w/ Std. Cushion	Assembly w/ Barrier Cushion A	Assembly w/ Barrier Cushion B	Subfloor w/ Perf. Al Barrier[c]	Subfloor w/ Fiber Barrier[d]
α-Pinene	34 ± 14	38	14.3	23	18.2	20
β-Pinene	5.4 ± 2.4	8.5	<3	3.2	3.5	<3
d-Limonene	25 ± 18	23	9.2	9.4	16.7	33
Formaldehyde	8.6 ± 2.4	6.4	3.2	4.9	<3	<3
Acetaldehyde	2.6 ± 1.1	<3	<3	<3	<3	<3
Pentanal	40 ± 6	32	27	23	35	27
Hexanal	172 ± 13	165	105	116	135	126
2-Furaldehyde	3.6 ± 0.9	3.6	<3	<3	<3	<3
Heptanal	10.0 ± 0.7	8.9	5.5	6.0	6.4	7.1
2-Heptenal	4.2 ± 1.8	3.6	<3	<3	<1	<3
Benzaldehyde	3.4 ± 2.1	3.6	<3	<3	<3	<3
Octanal	24 ± 8	18.4	10.6	15.7	11.0	12.2
2-Octenal	14.0 ± 9.1	9.7	3.2	6.0	<3	<3
Nonanal	29 ± 14	18.3	8.2	16.1	10.1	10.9

[a]Mean ± 1 stdev. for 5 replicate specimens
[b]Assemblies tested with subfloor, carpet cushion and carpet
[c]Perforated and reinforced aluminum foil radiant barrier
[d]Non-woven fiber, exterior weatherization membrane

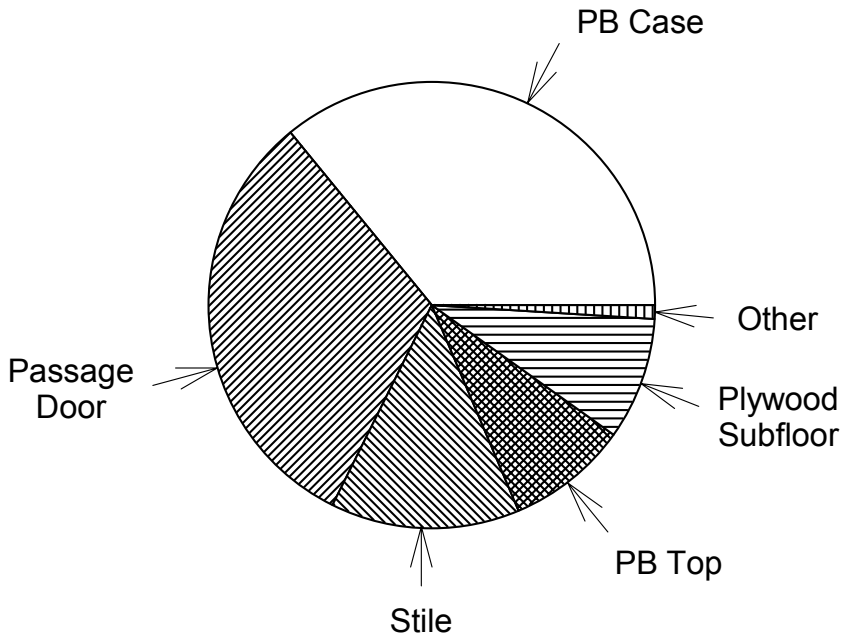

Fig. 1 Fractional contributions of indoor sources to whole-house formaldehyde emission rates