CONCENTRATIONS AND SOURCES OF FORMALDEHYDE AND VOLATILE ORGANIC COMPOUNDS IN FOUR NEW MANUFACTURED HOUSES

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ABSTRACT

The concentrations of formaldehyde, individual volatile organic compounds (VOCs) and total VOCs (TVOC) were measured in four new manufactured houses on three occasions over nine months following construction. Ventilation rates were also measured. A mass-balance model was used to calculate area-specific emission rates of the target analytes. Formaldehyde concentrations were all less than a guideline value of 50 ppb. One-half of the 58 target VOCs had median concentrations at or below 1 ppb. The most abundant VOCs were terpene hydrocarbons, ethylene glycol, hexanal, 2-butanone and acetic acid. Concentrations of hexanal, other aldehydes and acetic acid often exceeded their odor thresholds. The median TVOC concentration was 1.6 mg m^{-3}. In general, there were no large decreases in the emission rates of individual VOCs or TVOC over the course of the study. The data suggested that wood products were a dominant source of VOCs in all houses.

INTRODUCTION

Indoor sources of volatile organic compounds (VOCs) and building ventilation are important determinants of indoor air quality (IAQ) in houses. Many materials used to construct and finish houses emit VOCs and some emit formaldehyde. The trend in new house construction is to make building envelopes tighter. Consequently, ventilation rates are often relatively low. Elevated sources of indoor contaminants in combination with low ventilation rates create a potential for degraded IAQ that may affect occupant health and comfort. Prior to this study, there were no published VOC data for contemporary manufactured houses in the USA.

The objectives of the study were to: 1) quantify concentrations of total VOCs (TVOC), individual VOCs, and formaldehyde in four new manufactured houses over nine months following construction; 2) compare the concentrations to typical values and to odor thresholds; 3) document temporal changes in emission rates; and 4) evaluate the effectiveness of several ventilation and source modifications for reducing concentrations of VOCs.

METHODS

The four, two-section houses were constructed at a single manufacturing plant in Florida, USA. They were produced and set up at an adjacent model center during July and August 1997. All houses were equipped with a heating, ventilating and air conditioning (HVAC) system and a central occupant-controlled exhaust fan. The HVAC systems were operated daily. One of the houses had supplemental mechanical ventilation. Material substitutions were made in several of the houses either during the manufacturing or set up phases. The houses were decorated and fully furnished, but unoccupied. Air samples for the analysis of VOCs and formaldehyde were obtained in September and November 1997 and May 1998.

On each occasion, the samples were collected at a central location in each house and at a nearby outdoor location. Formaldehyde samples were collected on cartridges treated with 2,4-dinitrophenylhydrazine. Samples for TVOC and VOCs were collected on Tenax®-TA sorbent tubes. Ventilation rates were measured concurrently with the collection of air samples by tracer-gas decay using sulfur hexafluoride as the tracer gas.

Formaldehyde samples were analyzed by high-performance liquid chromatography following U.S. EPA Method TO-11. Sorbent tubes were analyzed for TVOC and individual VOCs by thermal desorption gas chromatography/mass spectrometry (GC/MS) using a modification of U.S. EPA Method TO-1. For the analysis of TVOC, a GC/MS total-ion-current chromatogram was integrated over a retention-time range bounded by n-heptane and n-heptadecane. The area response was calibrated with a mixture of ten common alkane and aromatic hydrocarbons. 58 target VOCs were selected for analysis. Some of the compounds are indicative of specific indoor sources. Others have low odor thresholds or are strong sensory irritants. 42 of the compounds are among the 63 target VOCs recommended to be included in an analysis of TVOC [1]. The concentration data for the target VOCs were also summarized as ΣVOC (*i.e.*, the sum of the individually measured VOC concentrations).

A steady-state mass-balance model was used to calculate area-specific emission rates with indoor and outdoor concentrations, ventilation rates and house volumes and areas as inputs.

RESULTS

The house specifications are given in Table 1. House M2 had supplemental mechanical ventilation provided by an outside air duct connected to the HVAC return duct and operated intermittently by a FanRecycler™ control device [2]. Low-VOC paints were used in House M2, and a low-emitting carpet assembly was installed in Houses M2 and M4.

Table 1. Specifications for the four houses.

Parameter	M1	M2	M3	M4
Floor area, m^2	112	169	141	131
Volume, m^3	273	412	344	320
Supplemental vent.	No	Yes	No	No
Ventilation rate, h^{-1}	0.57 - 0.78	0.53 - 0.71	0.35 - 0.36	0.35 - 0.50
Carpet area, m^2	72	128	94	96
Sheet vinyl area, m^2	0	29	29	21
Low-VOC paint	No	Yes	No	No
Low emitting carpet	No	Yes	No	Yes

The concentrations of TVOC and ΣVOC in the four houses are shown in Figures 1 and 2, respectively. TVOC concentrations ranged from 0.81-3.0 mg m^{-3} with a median value of 1.6 mg m^{-3}. TVOC concentrations in Houses M2 and M4 were relatively constant over time. For House M3 only, there was a substantial decrease in TVOC concentration from the 1st to the 2nd sampling period. For House M1, the TVOC concentration in the final sampling period was distinctly lower than concentrations in the 1st and 2nd periods. ΣVOC concentrations ranged from 44-100% of the TVOC concentrations. Typically, the individual VOCs accounted for ~70% of the TVOC values. The median ΣVOC concentration was 1.2 mg m^{-3}.

Five target VOCs, n-propylbenzene, isopropyl acetate, trichloroethene, tetrachoroethene and 1,4-dichlorobenzene, were not detected. 14 target VOCs had concentrations that were consistently at or below 1 ppb in all houses and sampling periods. These were benzene,

ethylbenzene, 1,3,5-trimethylbenzene, naphthalene, 4-phenylcyclohexene, n-pentadecane, n-hexadecane, propylcyclohexane, butylated hydroxytoluene, methyl isobutyl ketone, 1-phenylethanone, ethyl acetate, butyl acetate, and benzothiazole. About one-half of the target VOCs had median concentrations at or below 1 ppb. The concentrations (ppb) of 23 selected VOCs are listed in Table 2. These were either the dominant or most odorous compounds. The uncertainty for sampling and analysis of most VOCs was about 10% relative standard deviation. The relative uncertainty for the glycol ethers and acetic acid was 35% or more. Seven VOCs were among the most-abundant compounds in all four houses. These were α-pinene, β-pinene, 3-carene, ethylene glycol, hexanal, 2-butanone, and acetic acid. Acetic acid was the dominant compound.

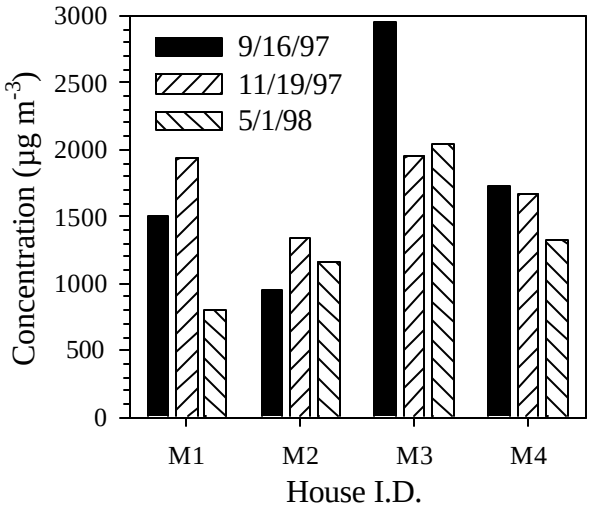

Figure 1. TVOC concentrations.

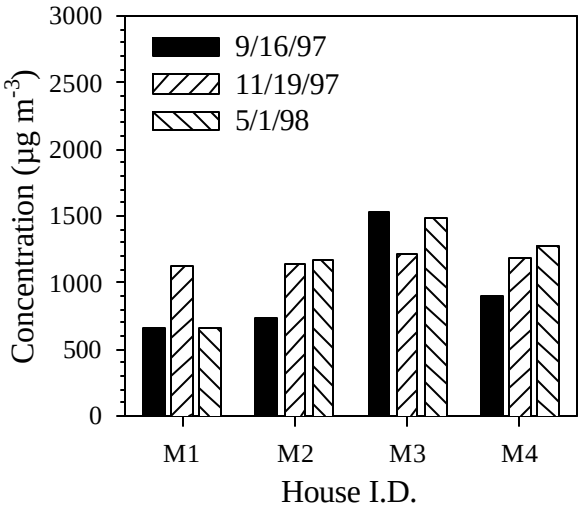

Figure 2. ΣVOC concentrations.

The formaldehyde concentrations are shown in Figure 3. The median formaldehyde concentration was 37 ppb, and all concentrations were less than 50 ppb.

The ranges of ventilation rates measured in the houses are shown in Table 1. Houses M1 and M2 had the highest ventilation rates, while House M3 had consistently lower rates. All of the values meet or exceeded the ASHRAE minimum ventilation standard of 0.35 h^{-1}.

The area-specific emission rates of TVOC are shown in Figure 4. The median TVOC emission rate was 1.6 mg m^{-2} h^{-1}. There was no obvious trend of TVOC (or ΣVOC) emission rates decreasing with time throughout the study. However, for Houses M1, M3 and M4, the TVOC emission rate in the final sampling period was less than the emission rate in the 1st sampling period. The TVOC emission rate in House M2 was constant over time. In the final sampling period, acetic acid accounted for 44-56% of the ΣVOC (mg m^{-2} h^{-1}) emission rates in the four houses.

There was also no general trend in the temporal profiles of the area-specific emission rates of the target VOCs. Among the dominant VOCs reported in Table 2, the emission rates of only n-decane, n-undecane, α-pinene, and 2-butanone decreased from the 1st to the final sampling period in all four houses. On the other hand, the emission rates of 1-octanol, 2-butoxyethanol, nonanal and acetic acid increased from the 1st to the final sampling period in all houses. The acetic acid emission rate increased by a factor of two to ten.

Table 2. Concentrations (ppb) of selected individual VOCs in the four houses on three sampling dates.

Compound	Chemical Class	M1			M2			M3			M4			Med.* Conc.
		9/97	11/97	5/98	9/97	11/97	5/98	9/97	11/97	5/98	9/97	11/97	5/98	
Toluene	AromaticHC	2	3	1	6	3	2	4	2	2	3	3	2	2
n-Decane	AlkaneHC	16	14	1	1	1	<1	8	4	2	2	1	1	1
n-Undecane	AlkaneHC	7	7	2	<1	<1	<1	3	2	1	1	1	<1	1
n-Dodecane	AlkaneHC	1	1	1	2	3	2	10	8	6	5	4	3	3
n-Tridecane	AlkaneHC	1	1	1	6	7	6	21	17	12	12	10	7	7
n-Tetradecane	AlkaneHC	1	1	1	4	4	5	13	10	11	6	6	5	5
α-Pinene	TerpeneHC	13	19	5	9	31	10	35	32	15	23	25	9	17
β-Pinene	TerpeneHC	3	6	2	2	7	3	11	8	5	7	6	3	5
3-Carene	TerpeneHC	3	6	2	1	5	2	15	11	7	9	10	4	5
d-Limonene	TerpeneHC	2	3	1	1	3	2	7	5	4	5	5	3	3
1-Butanol	Alcohol	2	6	3	3	4	1	4	4	5	3	4	4	4
1-Octanol	Alcohol	1	1	1	1	1	2	2	1	3	1	1	3	1
Phenol	Alcohol	1	1	1	2	2	4	4	4	6	3	3	4	3
Ethylene glycol	Glycol	32	39	12	41	44	17	<11	<10	21	<9	44	21	21
1,2-Propanediol	Glycol	<3	12	4	<3	5	<3	<4	<3	<3	<3	4	3	<3
2-Butoxyethanol	Glycol	1	2	1	3	5	5	2	2	3	3	6	6	3
Hexanal	Aldehyde	8	20	8	10	19	12	26	22	17	25	25	16	18
Heptanal	Aldehyde	1	2	1	1	1	1	2	2	3	2	2	2	2
Octanal	Aldehyde	1	2	2	2	2	2	3	3	4	3	3	3	3
Nonanal	Aldehyde	2	2	2	2	3	3	3	3	4	4	3	4	3
2-Butanone	Ketone	6	8	2	16	28	6	7	4	4	8	7	5	6
Acetic acid	Acid	25	102	142	80	122	264	123	120	267	53	126	275	122
Texanol®	Ester	2	7	3	2	2	2	2	1	2	2	2	2	2

*Median Concentration.

The area-specific emission rates of formaldehyde ranged from 24-68 μg m^{-2} h^{-1} with a median value of 41 μg m^{-2} h^{-1}. The values for each house varied by less than a factor of two, and there was no consistent trend with time. The hexanal emission rates ranged from 45-137 μg m^{-2} h^{-1} with a median value of 85 μg m^{-2} h^{-1}. For Houses M1, M2 and M4, the highest hexanal emission rates occurred in the 2nd sampling period. In the final sampling period, the rates were in a narrow range of 56-65 μg m^{-2} h^{-1}. The formaldehyde and hexanal emission rates were not correlated with each other (r = 0.54).

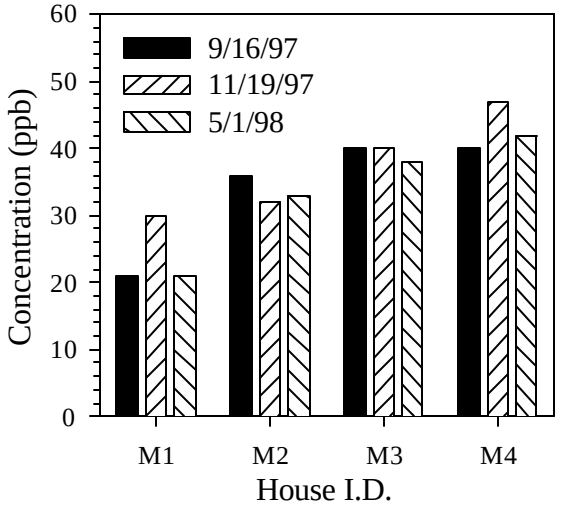

Figure 3. Formaldehyde concentrations.

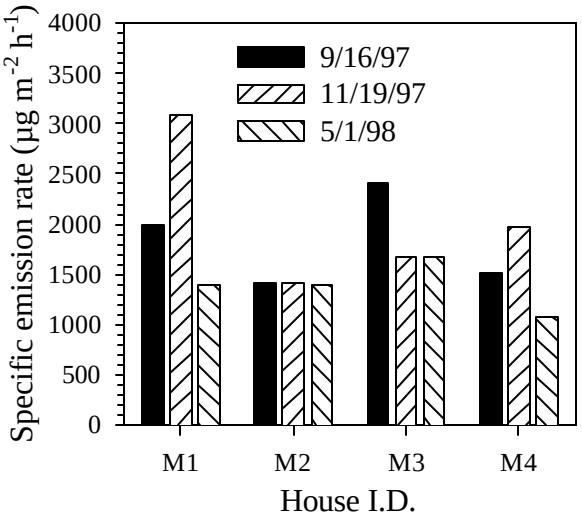

Figure 4. TVOC specific emission rates.

DISCUSSION

TVOC concentrations in various indoor environments are frequently about 1,000 μg m^{-3}, or lower [3]. One study of a probability-based sample of ~200 occupied houses of all ages recorded a median TVOC concentration of 700 μg m^{-3} [4]. The TVOC concentration in House M3 during the 1st sampling period was about four times higher than this value while the median TVOC concentration in the four houses over the course of the study was only about twice this value. Supplemental mechanical ventilation may have contributed to the consistent, and generally low, TVOC concentrations in House M2.

In the past, formaldehyde concentrations in manufactured houses were frequently in excess of 100 ppb [5]. Guidelines for formaldehyde concentrations in buildings are now established at relatively low levels. The formaldehyde concentrations in the four houses were all lower than the most restrictive guideline in the USA of 50 ppb [6]. The low formaldehyde concentrations are probably due to the exclusive use of gypsum board panels for walls and ceilings and the relatively low emissions of formaldehyde from current wood products.

Indoor air concentration data for a number of VOCs have been summarized from the literature [3]. Compounds that had notably elevated concentrations in the study houses relative to these data included the alkane and terpene hydrocarbons, n-decane, n-undecane, n-dodecane, n-tetradecane, α-pinene and β-pinene. Other compounds in the houses with elevated concentrations relative to the literature were 1-butanol, hexanal, nonanal, and 2-butanone.

A number of alcohols, aldehydes and carboxylic acids produce objectionable odors at low concentration [7]. The hexanal concentrations in the houses frequently were near or exceeded

the hexanal odor threshold of 14 ppb. The odor thresholds for octanal and nonanal are 1 and 2 ppb, respectively. The concentrations of octanal and nonanal consistently were at or slightly exceeded these values. The odor threshold for acetic acid of 140 ppb was approached in all houses during the 2nd sampling period and was exceeded in all houses during the final period. Thus, while the concentrations of TVOC and VOCs in the houses were, in general, not abnormally elevated, it is expected that some occupants would be able to detect objectionable odors due to elevated concentrations of aldehydes and acetic acid.

The data suggest that IAQ in the houses was primarily impacted by a few dominant VOC sources. Phenol and at least a portion of the normal alkane hydrocarbons, such as n-tridecane and n-tetradecane, may have originated from the sheet vinyl flooring used only in Houses M2, M3 and M4. Wood and engineered wood products emit terpene hydrocarbons, such as α-pinene, β-pinene and d-limonene. Engineered wood products also emit hexanal and other aldehydes including heptanal, octanal and nonanal. It is likely that the terpene hydrocarbons and aldehydes originated from engineered wood products used in the construction and finishing of the houses. The possible sources include cabinetry, plywood floor decking and oriented-strand-board roof decking. The primary source of acetic acid is uncertain.

The effects of the two source substitution treatments were difficult to evaluate. House M2 with the low-VOC paints had some of the highest concentrations of ethylene glycol that is used as a solvent in many latex paints. It is possible that conventional latex paints were used in the house during the decorating phase or for touch up. The concentrations of specific VOCs typically emitted by carpets and carpet cushions were low in all four houses, suggesting that contemporary carpet materials are a relatively minor source of VOCs.

ACKNOWLEDGEMENT

Sponsored by the U.S. Depart. Of Energy, Assist. Secretary for Energy Efficiency and Renewable Energy, Office of Building Technology, State and Community Programs.

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