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Sources and Concentrations of Formaldehyde and Other Volatile Organic Compounds in the Indoor Air of Four Newly Built Unoccupied Test Houses

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Key Words

VOCs
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Sources
Materials
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Abstract

Concentrations of formaldehyde and other volatile organic compounds (VOCs) were monitored over a 2-year period in the main rooms of four unoccupied test houses and in the outdoor air. During the construction and furnishing of the buildings 30 samples of materials were collected and subsequently tested using environmental test chambers to determine amounts of formaldehyde and other VOCs emitted. Concentrations of VOCs in the building were initially high and declined quickly during the first 6 months after construction. Formaldehyde concentrations showed a slower rate of decline with strong seasonal effects resulting in highest concentrations during summer months. The most important source of benzene and toluene was the outdoor air whereas almost all materials were potential sources of the wide range of VOCs found in the indoor air. Some materials such as paints and flooring are identified as the most important sources of the major VOCs occurring in the indoor air.
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Introduction

Volatile organic compounds (VOCs) occur in the indoor air because of their entry from outdoors and their emission from building, furnishing and consumer products. The resulting indoor concentrations depend upon the rate of emission and the rate of removal from the air by ventilation or adsorption onto surfaces. The factors controlling the emission and removal are dynamic and dependent upon temperature, humidity, wind speed and occupant behaviour including opening of windows and use of products [1].

The relative importance of the various sources of indoor pollutants is different for individual compounds

and locations. In a study of 173 homes in the UK, benzene in the outdoor air was found to be the major source of benzene in the indoor air although smoking of tobacco was also a significant source in some homes [2]. Toluene had both important indoor and outdoor sources whereas for undecane and formaldehyde only indoor sources were significant. The total VOC (TVOC) concentration in the indoor air was typically ten times higher than outdoors [3]. The TVOC indoors was due to a complex mixture of compounds, the mean number being 85 detectable peaks.

Studies in many other countries report that a complex mixture of VOCs occurs in the indoor air [4]. The materials and consumer products that emit these compounds are numerous [5, 6] and the nature of the emission from simi-

lar product types such as flooring materials can vary considerably [7]. Environmental chambers are increasingly being applied to determine the emission of VOCs from products under controlled environmental conditions [8]. Models are being applied to link the data on emission rates in a chamber to the concentration of VOCs that would result from the use of the product in a room or house [9]. This approach has been applied to the labelling of building materials in Denmark based upon the predicted impact of the product on indoor air quality [10].

The present paper complements previous studies of VOCs in occupied UK homes, by investigating the sources in four unoccupied test houses. In these houses the sources are the outdoor air and building and furnishing materials, and not products used by the occupants. The concentrations of formaldehyde and other VOCs were monitored for a period of 2 years immediately after construction. The sources of pollutants were investigated by undertaking emission testing of materials used in the construction and furnishing of the buildings and by monitoring the VOCs and formaldehyde in the outdoor air. The relationships between sources and the indoor concentrations are discussed.

Materials and Methods

The BRE Low-Energy Test Houses

The monitoring of the formaldehyde and other VOC concentrations in two matched pairs of test houses began when the construction and furnishing was completed in July 1992. The test houses were built at the Garston site of the Building Research Establishment (BRE), primarily for the investigation of the energy efficiency of homes constructed to a higher insulation standard than that required by the current UK Building Regulations. One pair of detached houses were built to meet the energy requirements and insulation standards used in Scandinavian countries and have a timber-framed construction externally clad with brick and tiling (houses T₁ and T₂). Mineral wool is used for insulation of the walls, timber-suspended floors and roof space. The other pair (houses M₃ and M₄) have double masonry cavity walls but are of the same size and have the same arrangement of rooms as T₁ and T₂. The walls and roof space are insulated with mineral wool and the suspended timber floor incorporates extruded polystyrene board.

The houses are unoccupied, though activities that consume energy in a normal home are simulated. Houses T₁ and T₂ have better thermal insulation and a more airtight structure than houses M₃ and M₄, and have mechanical ventilation and heat recovery. Ventilation in the M₃ and M₄ houses was provided by trickle ventilators and extract fans installed in the kitchens and bathrooms, complying with the UK Building Regulation requirements. In addition, blockable passive stack ventilators were installed in the kitchen and bathroom for separate examination of their effectiveness, and these are only used occasionally. Air temperature and humidity were monitored continuously in all rooms in each house.

The rooms in both types of houses have 15-mm plywood partition walls covered with 12.5-mm plasterboard and coated with emulsion paint. The houses are carpeted throughout, except the bathroom, kitchen and airing cupboard. The bathroom and kitchen have vinyl floor coverings. All the internal doors are painted with white gloss. The exterior doors and windows are wood stained. Medium-weight curtains with motorised cord pulls are fitted to the lounge/diner and bedroom windows. All the ceilings are coated with emulsion paint.

All the bedrooms are furnished with a bed, a chest of drawers and a bedside table. There is a built-in wardrobe in the main bedroom of each house. In the lounge/diner there is a dining table and four chairs, a coffee table, two low wall cabinets and a settee made of soft foam. All the furniture in the houses is old and made of hardwood. In the kitchen, there is a sink unit, melamine-coated chipboard work-tops and cupboards, an electric cooker and a gas boiler.

Materials Tested for Emission

Thirty materials were collected during the building and furnishing stage of the house construction. These were materials classed as either flooring, adhesives and sealants, walls and ceilings, windows and doors, timber supports or other polymeric. All the board products and timber pieces were wrapped in aluminium foil when collected; the vinyl and carpet materials were rolled up and enclosed in individual stainless steel cupboards. The paints, woodstain, caulk, putty and adhesives were sealed in their own containers and were stored at room temperature. The other materials were stored uncovered in steel cupboards.

Environmental Test Chambers

Each material was tested using either a 1-m³ environmental chamber or a 2.4-litre microchamber [11]. The 1-m³ chamber was operated at 23°C (±0.5°C) and 45 ± 3% RH with an air exchange rate of 1 h⁻¹ and an internal air velocity of 0.3 m·s⁻¹. Each sample was enclosed in the chamber, and the air leaving the chamber was monitored to determine the concentration of VOCs. The microchambers were located in a 23°C constant-temperature room and supplied with 'zero' grade air humidified to 45% RH. The air flow rate was 40 ml·min⁻¹ giving an air exchange rate of 1 h⁻¹ and the air leaving the microchamber was sampled at 20 ml·min⁻¹ using an adsorbent tube and pump. Different loading factors were used for the various materials. For the 1-m³ chamber, ratios of product surface area to chamber volume were used that were appropriate to a room situation (e.g. 0.4 m²·m⁻³ for flooring and 0.5 m²·m⁻³ for emulsion paints). For some materials that are not in direct contact with indoor air, such as supporting timbers, it is difficult to select a meaningful loading factor appropriate to a room. For these materials and others placed in the microchambers high loading factors (5–15 m²·m⁻³) were used to increase the sensitivity of the sampling and analytical procedure. All liquid products were applied by brushing onto glass and then this was placed immediately into the chamber or microchamber. Other materials were clamped to stainless steel supports.

Sampling and Analytical Methods

The sampling of the VOCs in the indoor air was carried out over a 2-year period using diffusive samplers exposed for periods of 28 days. The technique has been used previously by BRE to monitor concentrations of individual VOCs and the TVOC concentrations in occupied homes [12]. The sampler was the Perkin Elmer type adsorbent tube packed with Tenax TA. Samplers were sited and exposed in the lounge/diner, the kitchen, the bedrooms and the bathroom in

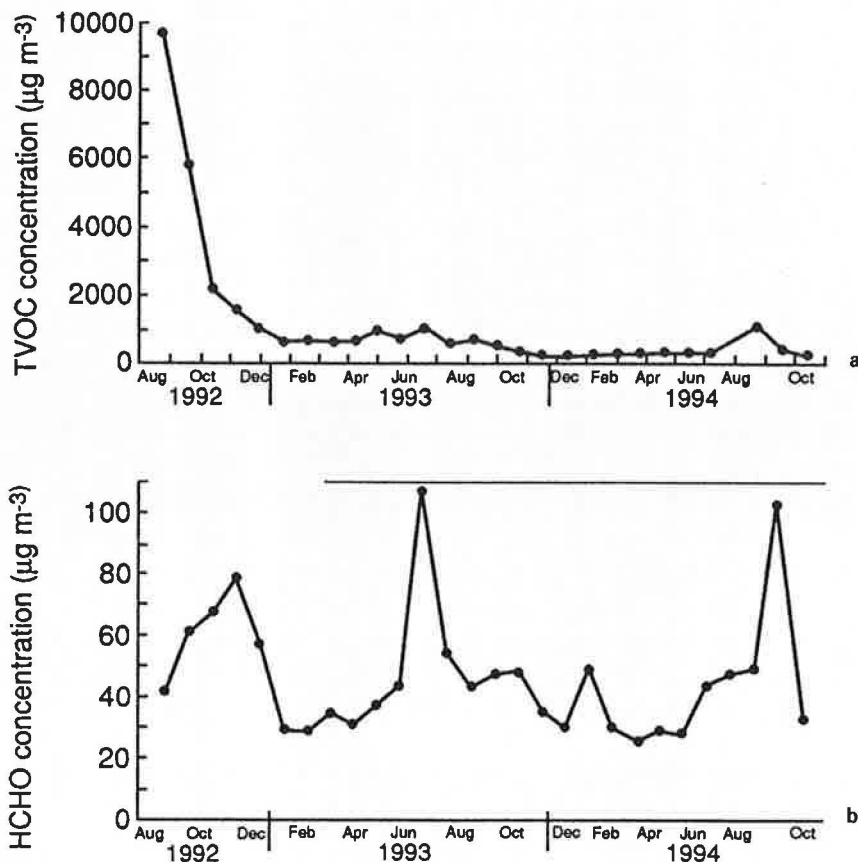


Fig. 1. **a** Changes of TVOC concentration over a 2-year period in the living-room of house T₁ (August 1992–September 1994). **b** Changes of formaldehyde concentrations over a 2-year period in the living-room of house T₁ (August 1992–September 1994).

each of the test houses. The tubes were suspended at near head height from central lighting units fixed to the ceiling. After sampling, the tubes were sealed with a stainless steel cap. For quantitative work, the tubes were analysed using thermal desorption/gas chromatography (TD/GC) with flame ionisation detection (FID). To assist with the identification of these and other major compounds found in the indoor air, some additional sampling was carried out in the 1st and 9th month of the 2-year monitoring period. These adsorbent tubes were analysed using TD/GC with ion-trap detection (ITD). Full details of the analytical method have been described elsewhere [3]. The detection limit of the method using a 28-day sampling period is 1–2 µg·m⁻³ depending upon the net diffusive uptake rate and the FID response factor for the particular compound.

The formaldehyde concentration was also monitored over the 2-year period at the same locations in the rooms in the test houses as for the VOC sampling. Passive samplers containing 2,4-DNPH (dinitrophenylhydrazine) coated on filter paper were used. The samplers were exposed to the room atmosphere for 3 days, giving a 3-day average concentration, and the sampling was repeated every 28 days. This method of diffusive sampling of formaldehyde has been described [13], together with the analytical method which involves the use of high-performance liquid chromatography. The detection limit for the method using a 3-day sampling period is 1 µg·m⁻³.

The concentration of formaldehyde and other VOCs in the outdoor air at ground level and 5 m from the front of the house T₂ were also monitored using the diffusive samplers. Monitoring of VOCs during the emission chamber experiments was undertaken using active sampling methods with the same type of Tenax adsorbent tube test used for diffusive sampling [7]. The measurement of formaldehyde emission was undertaken by the acetylacetone method using impingers [11].

Results

Test Houses

During the winter months, the temperature in the homes was more or less constant at 20°C, but in summer it fluctuated between 20–30°C. The humidity was about 33% during colder months, increasing to over 40% in June–September. The average air exchange rate was about 0.3 h⁻¹ in houses M₃ and M₄ and 0.5 h⁻¹ in houses T₁ and T₂. Figure 1 shows the changes in TVOC and formaldehyde concentrations in the living room of house

Table 1. Summary of the whole house concentration ($\mu\text{g}\cdot\text{m}^{-3}$) of TVOCs, benzene, toluene, xylene, undecane and formaldehyde (HCHO) during the first and second year

House		Year 1 (3/8/92–3/8/93)			Year 2 (3/8/93–3/8/94)		Summer	Winter
		mean	max	min	mean	RSD %	(16/3/93– 28/9/93) mean	(28/9/93– 15/3/94) mean
T ₁	TVOC	1,938	8,270	635	362	75	808	200
	benzene	27	149	3	4	24	4	4
	toluene	19	88	3	6	40	6	6
	xylenes	81	289	59	25	59	42	16
	undecane	75	261	32	18	61	34	10
	HCHO	59	122	29	42	33	63	37
T ₂	TVOC	1,603	6,472	648	315	57	500	203
	benzene	5	9	3	5	22	3	4
	toluene	12	38	4	6	42	6	6
	xylenes	64	227	32	21	48	36	15
	undecane	70	276	32	17	54	33	11
	HCHO	51	91	28	37	48	52	28
M ₃	TVOC	1,429	5,179	531	363	67	939	272
	benzene	8	35	3	7	36	4	5
	toluene	10	31	3	36	290	4	6
	xylenes	30	94	14	20	136	19	8
	undecane	57	176	26	16	59	39	10
	HCHO	41	106	17	40	43	53	30
M ₄	TVOC	1,954	5,660	863	896	61	1,578	802
	benzene	8	39	3	4	32	5	4
	toluene	12	32	5	7	42	9	7
	xylenes	33	85	16	45	183	25	26
	undecane	82	202	41	37	61	70	31
	HCHO	43	111	15	45	45	59	35
Out- side	TVOC	71	150	7	37	40	31	43
	benzene	6	11	3	5	33	4	4
	toluene	9	18	3	6	53	6	6
	xylenes	7	11	4	4	40	4	5
	undecane	4	24	6	1	20	2	1
	HCHO	3	5	2	2	63	3	2

RSD = Relative standard deviation (i.e. standard deviation expressed as percentage of mean).

T₁ during the 2-year period of monitoring. The TVOC concentration declined markedly during the first 6 months after completion of the building, from 9,700 to about $500 \mu\text{g}\cdot\text{m}^{-3}$, then a more gradual decline occurred with some fluctuations, to about $200 \mu\text{g}\cdot\text{m}^{-3}$ after 16 months. For formaldehyde, the pattern was different with peaks in June 1993 and September 1994 superimposed on a gradual decline in concentrations. The relationship between the formaldehyde concentration and temperature in the test houses has been discussed in more detail elsewhere [11].

Table 1 shows the whole house concentrations of formaldehyde, TVOC and four of the many individual VOCs found in the indoor air. These values were calculated by averaging the concentrations measured in the rooms for each month of the study. The four VOCs selected were benzene which is predominantly of outdoor origin, toluene and xylene which have significant indoor and outdoor sources and undecane which has predominantly indoor sources [12]. Student's t test was used to test for significant differences between whole house annual mean concentrations for each test house during years 1 and 2

and to test for differences in concentrations between rooms. The results of tests were regarded as statistically significant if the probability of the observed difference occurring by chance was less than 1 in 20 (i.e. 5% level).

The two types of houses have similar mean annual concentrations for each compound. The slightly higher concentration of formaldehyde in houses T₁ and T₂ in year 1 compared with houses M₃ and M₄ was not statistically significant. During year 2 the TVOC, xylene and undecane concentrations in house M₄ were higher than in the other 3 houses and this was due to the painting of a bedroom door in June 1994. For TVOC, benzene, toluene, xylene and undecane there was no statistically significant difference between the mean concentration in the rooms in each house for all three periods tested; year 1, year 2 and the mean of year 1 + year 2. There was also no significant difference between the formaldehyde concentrations measured in the different rooms in the test houses during the first year period. This indicates that the air within each house was well mixed and local sources did not have a strong influence on the annual mean concentration in a room. During the second year, the formaldehyde concentration in the kitchens was significantly lower than the concentrations measured in the bedrooms and bathrooms except for house M₃. The reason for this has not been identified.

Indoor concentrations of formaldehyde, TVOCs, xylenes and undecane were considerably higher than those measured outdoors. For example, the mean TVOC and formaldehyde concentrations indoors during year 1 were 20–27 and 14–20 times higher, respectively, than those outdoors. This shows that the construction and furnishing material used for the houses are major sources of these compounds in the indoor air. Concentrations of toluene and benzene in the indoor air were very similar to those outdoors although benzene was initially elevated in house T₁. Hence, the source of benzene and toluene in the indoor air of the test houses is predominantly from motor vehicle emissions that pollute the outdoor air.

The same passive sampling methods used in the present study were also applied in the BRE Indoor Environment Study which involved monitoring of formaldehyde and VOC concentrations in 174 homes in the Avon region of England [3, 14]. No measurements of rates of ventilation were made in these occupied homes. The mean year 1 TVOC and undecane concentrations in the test houses were greater than the 95 percentile of mean values found in Avon and declined to within the range given by the 50- and 75-percentile values in year 2. This was due to the strong influence of emissions from materi-

als and furnishings on the concentration of these compounds, especially during the first 6 months after construction. In both years formaldehyde concentrations were between the 75- and 95-percentile values measured in Avon. These relatively high values are in keeping with the study of occupied homes which found highest concentrations in newer homes although possible differences in ventilation rates between occupied homes and the unoccupied test houses may also be a factor.

The benzene and toluene concentrations are generally below the median values found in Avon homes and for toluene a number of readings are near the 10-percentile value. For benzene this is consistent with the strong influence of the outdoor concentration found in the Avon study. Higher concentrations indoors were caused by tobacco smoke and possibly other indoor sources. The mean toluene concentration indoors in Avon was 3 times the outdoor concentration. The relatively low values in the unoccupied test houses suggests that emissions associated with human activities, including the use of consumer products, may be particularly important sources of toluene in homes compared with construction materials and furnishings.

The seasonal variation in formaldehyde levels in the test houses is similar to that found in the occupied homes with higher levels during the summer months. TVOC concentrations in the test houses are also higher in summer whereas the study of occupied homes showed significantly higher levels in winter. This suggests that the winter peak in occupied homes is due to the influence of occupant activities affecting source strength and perhaps ventilation rate, and this outweighs any influence that temperature has on the emission rate of VOCs from materials and furnishings.

Figure 2 shows examples of chromatograms given by ATD/GC/FID analysis in the 1st, 9th and 24th sampling events in the living rooms of two of the test houses. These illustrate the decline in both the number and size of detectable peaks over the 2-year monitoring period. The pattern of peaks in the different houses is similar, but not identical in the four houses. The total number of individual VOCs detected ranged from 41 to 254 and the largest number occurred at the start of the monitoring period. During the last 6 months of the study, the total number of detectable peaks was typically about 50 in the indoor air, which is less than the mean value of 85 found in occupied homes [3].

Table 2 shows the compounds that were dominant on at least one occasion in the GC chromatogram from the analysis of diffusive samplers exposed in the main bed-

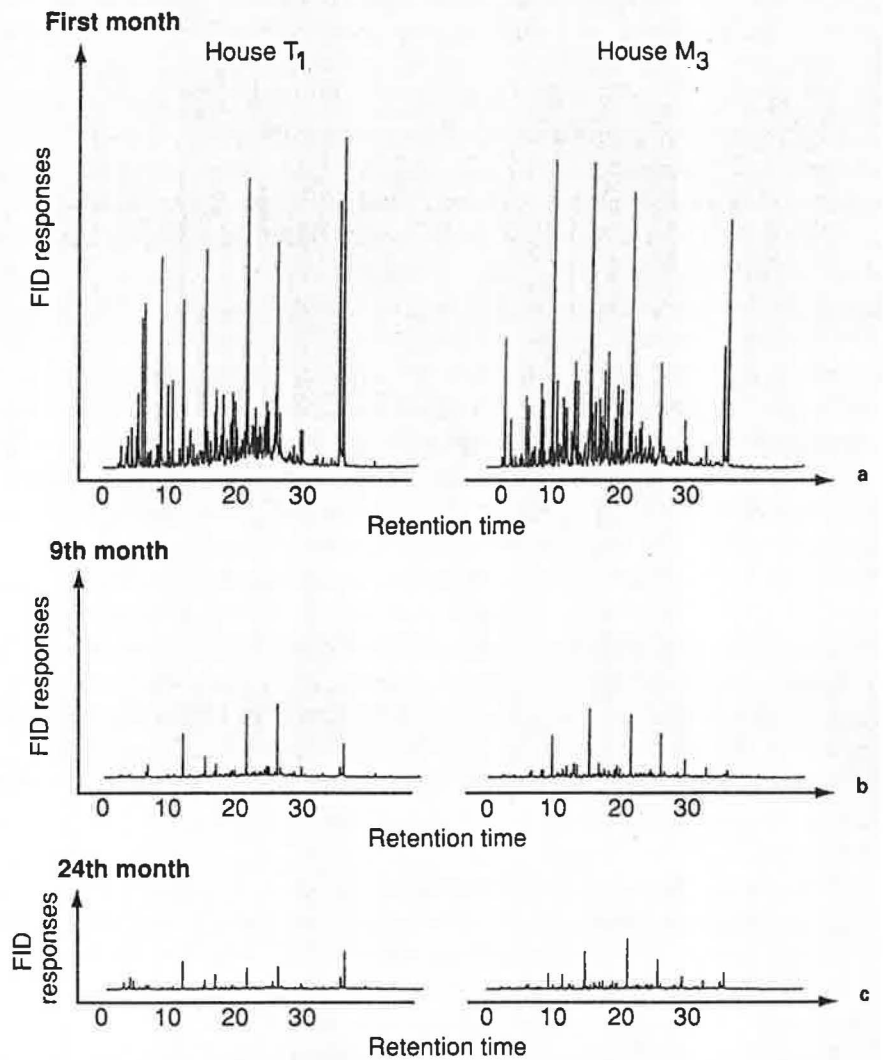


Fig. 2. FID chromatograms of VOCs emitted in living-rooms of two test houses. **a** First month. **b** Ninth month. **c** Twenty-fourth month.

room of each house. Undecane was most frequently the dominant compound in each house. In houses M₃ and M₄, decane was also dominant for a considerable proportion of sampling events, which was not the case for the houses T₁ and T₂. Texanol (2,2,4-trimethyl-1,3-pentanediol monoisobutyrate) was more often an important component of the VOC mixture in T₁ and T₂ than in M₃ and M₄. These results differ from those found in occupied homes in several respects. Toluene was most frequently the dominant compound in occupied homes, but was never dominant in the test houses. Also notable was the importance of limonene and some very volatile VOCs (VVOCs) which were not major compounds in the air in the test houses. It is likely therefore that toluene, limonene and

the VVOCs are released during activities by occupants and the building and furnishing materials are not major sources of these compounds. However, it is also possible that other materials not used in test houses are sources of these compounds in other buildings.

Emissions from Materials

While not every material present in the houses could be examined, the majority likely to emit VOCs into the indoor air were tested. Uncertainty exists about how representative the various off-cuts and duplicate samples provided by the builders are of the much larger amounts of materials used in the houses. Also the period of storage before testing of some solid materials means it is less clear

what contribution they may have made to the VOC content of the indoor air when the houses were first completed.

Table 3 lists the 5 major compounds found by TD/GC/FID analysis of an air sample taken during the first day a material was placed in the emission test chamber. Vinyl flooring, carpet, chipboard flooring, wood stain, wood primer, undercoat, gloss paint and three types of emulsion paints were tested in the 1-m³ environmental chamber. All other materials were tested in the 2.4-litre micro-chambers. Emission rates of individual compounds released from each material were recorded during the chamber tests but are not presented here. This is because work is ongoing to compare emission rate values obtained using 1 m³ and micro-chambers.

The results of the emission tests show that all the materials were found to emit VOCs that could contribute to the total VOCs measured in the indoor environment except samples of PVC skirting board, polythene spacer and PVC extrusion (thermal stop for window frame attachment, filled with polyurethane foam). A total of 50 compounds were found in the 5 major compounds emitted by each of the 30 materials. A number of compounds including C₉-C₁₄ aliphatic hydrocarbons, C₅-C₁₀ aldehydes and aromatics such as xylenes, were found to be among the 5 major compounds emitted after 24 h from a number of materials tested in the environmental chamber. Some compounds such as Texanol, TXIB (2,2,4-trimethyl-1,3-pentanediol diisobutyrate), terpenes (α -pinene, limonene, carene), naphthalene and ethyl acrylate were specifically related to one or a few types of materials.

The composition and the amount of the emission from each material changed with time. For example, the emission from the acrylic adhesive and spray-on adhesive declined rapidly to near the detection limit in 1 day. Emission from emulsion paints receded slowly after an initial rapid decline, maintaining a steady rate of emission which was almost constant over the last 8.5 days of a 10-day test. The rate of emission from wood stain and gloss paints declined more rapidly and concentrations in the chamber were at the detection limit within 7 days.

Most of the walling, ceiling and curtaining materials produced a relatively low rate of VOC emission. The emission from the bituminised materials should not be important for the indoor atmosphere since these materials were used on the exterior structures of the house. The type of supporting substrate had a strong influence on the emission from the emulsion paint. When comparing the GC/FID chromatograms of the emitted VOC components of 3 water-borne paints applied on glass and on plas-

Table 2. Frequency compounds occur as the dominant peak in the air of the main bedroom (%) over the 2-year monitoring period

Compound	House T ₁	House T ₂	House M ₃	House M ₄
Undecane	31	68	68	31
Decane	0	0	16	52
Texanol 2	27	16	4	4
<i>o</i> -Xylene	19	4	0	0
<i>m/p</i> -Xylene	0	0	0	9
Pentadecane	12	8	0	0
Texanol 1	8	4	4	4
TXIB (butan-1-ol)	0	0	4	0
(RT 5.56)	0	0	4	0
Unknown (RT 3.4)	4	0	0	0

() = Possible identity; RT = gas chromatographic retention time in minutes.

terboard, a number of the compounds were reduced and some not detectable due to absorption into the plasterboard. This effect could be significant for long-term release in the indoor air. This is especially true for a compound such as Texanol which is the dominant component of the emission from emulsion paint. Emission rates from the flooring materials were low but were nearly constant over the whole weeks of test, thus demonstrating their importance as long-term sources of VOCs.

Timbers were found to be long-term sources of terpenes and aldehydes. A wide array of VOCs were emitted from treated timber, the soft wood that was used to build the partitioning frame of a house known as studd and packing support batten. These were shown to maintain a relatively high and steady emission rate for at least 2 weeks after their emission curves had reached a constant level.

The gap sealing foam could have a short-term impact on the indoor air concentration of VOCs. This was demonstrated by an abrupt rise in the toluene and xylene concentration in houses M₃ and M₄ during 1 month in winter when the houses were being made more air-tight by filling in all the gaps and ducts in the house. The toluene and xylene concentration rose sharply in the living rooms from 2 $\mu\text{g}\cdot\text{m}^{-3}$ for toluene and 8 $\mu\text{g}\cdot\text{m}^{-3}$ for xylenes to 445 $\mu\text{g}\cdot\text{m}^{-3}$ and 308 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The VOC concentration soon declined to its former constant level during the next sampling event, illustrating the short-term impact of the VOC emission from this material.

Table 3. Major VOCs emitted from materials used in the BRE test houses

Materials		Major VOCs emitted after 24 h of chamber test					Comments
		1	2	3	4	5	
Solid flooring materials	vinyl ¹	unknown (RT 35.04)	unknown (RT 30.83)	C ₁₄	C ₁₃	naphthalene	the TVOC emission rates were typically below 55 µg·m ⁻² ·h ⁻¹ which was maintained more or less constant during the 10-day chamber test; aldehydes were emitted from chipboard; a near steady state was reached after 6 days of test. A steady rate of emission was maintained from all flooring materials over 14 days
	carpet ¹	nonanal	C ₁₃	C ₁₀	diethyl benzene	TXIB	
	underlay	TXIB	Texanol	C ₁₀	C ₁₅	C ₁₆	
	chipboard ¹	hexanal	<i>m</i> - <i>p</i> -xylene	toluene	nonanal	octanal	
Adhesives and rubber sealant	black rubber + adhesive backing	phenol	TXIB	terpinene	C ₁₃	C ₁₀	VOCs such as dichloromethane and t-butyl-methyl-ether from sprayed adhesive and methyl vinyl ketone and methyl cyclohexane from acrylic adhesive disappeared rapidly during the first 6 h after application; the emission rate reached a low continuous level after 1 day
	spray adhesive for carpet underlay	2,6-di-t-butyl- <i>p</i> -cresol	t-butyl methyl ether	(phthalic anhydride)	unknown (RT 38.7)	unknown (RT 31.4)	
	acrylic adhesive for vinyl	C ₉	<i>o</i> - and <i>m</i> - <i>p</i> -xylene	toluene	1,2,4-trimethyl benzene	cyclohexane	
Wall and ceiling materials	plaster-board	hexanal	(methylcyclo-hexane)	C ₉	C ₁₀	C ₇	the TVOC emission rates for all the boards were relatively low and constant; the maximum rate of emission for plasterboard was 6 µg·m ⁻² ·h ⁻¹ ; and 15-mm and 6-mm plywood, 12 µg·m ⁻² ·h ⁻¹ and 10 µg·m ⁻² ·h ⁻¹ , respectively
	15-mm plywood	hexanal	α-pinene	limonene	<i>m</i> - <i>p</i> -xylene	unknown (RT 4.96)	
	6-mm plywood	C ₁₀	C ₉	unknown (RT 40.49)	<i>m</i> - <i>p</i> -xylene	(linalool)	
	bituminised fibreboard	hexanal	C ₁₁	C ₁₀	nonanal	octanal	complex mixtures of aliphatic and aromatic hydrocarbons, aldehydes, terpenes, phenol, alcohols and naphthalene were emitted; the peak TVOC emission rate was about 30 µg·m ⁻² ·h ⁻¹ which remained more or less constant during the 5 days of test
	asphalt	unknown (RT 26.49)	terpinene	nonanal	C ₁₃	octanol	
	PVC skirting board	ND	ND	ND	ND	ND	the TVOC emission rate was below the detection limit of the analysis. However, when heated to 40 °C, a relatively low TVOC emission of 4 µg·m ⁻² ·h ⁻¹ was measured
	polythene spacer	ND	ND	ND	ND	ND	
	rockwool (cavity wall)	C ₁₀	<i>m</i> - <i>p</i> -xylene	C ₉	<i>o</i> -xylene	hexanal	the TVOC emission rates from these materials were below 15 µg·m ⁻² ·h ⁻¹ , the emission rate declined slowly over the 5 days of test
	mineral wool (loft)	nonanal	decanal	undecanal	toluene	(diacetone alcohol)	
	water-borne emulsion paint ¹	Texanol 1	Texanol 2	1,2-propane-diol	2(2-butoxy-ethoxy) ethanol	decanal	adsorption and absorption by plasterboard substrate could reduce initial emission rates but later desorption would add to the total indoor concentration; the VOC peaked rapidly during the first 2 h but declined to a constant level after 1 day; a 2-4 mg·m ⁻² ·h ⁻¹ emission rate is probably sustained over a long period of time beyond the 10-day chamber test

The TVOC emission profile and emission rate can be used to classify the materials as short-term or long-term and high, moderate or low emitters. Table 4 classifies the test house materials that had been kept either in sealed containers or wrapped in foil prior to emission testing according to

these categories. One further category exists, which is very low or no detectable emission. Long-term emitters are the materials that still emit at the end of a 14-day chamber test at a rate that demonstrates a continuing slow decline. Short-term emitters are materials where the emission declines to

Table 3 (continued)

Materials	Major VOCs emitted after 24 hours of chamber test					Comments	
	1	2	3	4	5		
Windows and doors	curtain	Texanol 1	Texanol 2	<i>m</i> -/ <i>p</i> -xylene	C ₁₀	<i>t</i> -butylbenzene	curtain emitted a low level of TVOC (2 µg·m ⁻² ·h ⁻¹); possible that Texanol results from contamination during storage; putty emitted a higher level of TVOCs (8.6 mg·m ⁻² ·h ⁻¹) at a constant rate; although not included in the top five, aldehydes were present in the VOC emission from curtain
	putty	<i>m</i> -/ <i>p</i> -xylene	<i>o</i> -xylene	pentane	decene	undecane	
	thermal-stop (PVC extrusion containing PUR foam)	ND	ND	ND	ND	ND	
	wood stain ^{1,2}	C ₁₀	C ₁₁	C ₁₂	C ₁₃	trimethylbenzene	
	gloss ^{1,3}	C ₁₀	C ₁₁	1,2,3,5-tetramethylbenzene	hexanal	C ₁₃	components of white spirit containing mixtures of aliphatic and aromatic hydrocarbons, were emitted; the VOC emission rate reached a peak after 30 min for woodstain on plywood and 5 min for gloss on undercoats; the emission rate declined to a near constant level after 8–10 days; decane and undecane could still be detected after 10 days; the emission rate was less than 0.1 mg·m ⁻² ·h ⁻¹
Timber	pillar	α-pinene	terpinene	hexanal	limonene	carene	terpenes and aldehydes were the dominant compounds emitted from untreated timbers; the treated types (the batten and the studd) emitted a higher level of VOC; the initial TVOC emission rate was rapid during the first day of exposure but subsequent emission was slow and took 2 weeks to reach a near constant emission rate; 200 µg·m ⁻² ·h ⁻¹ for the batten and 30 µg·m ⁻² ·h ⁻¹ for the beam
	beam/joist	α-pinene	hexanal	limonene	terpinene	(4-methyl-2-pentanone)	
	batten (packing support)	hexanal	C ₁₁	C ₁₂	C ₁₃	C ₁₄	
	studd	hexanal	<i>m</i> -/ <i>p</i> -xylene	pentanal	C ₁₀	α-pinene	
Other polymerics	caulk	C ₁₀	C ₁₁	(nonene)	(camphene)	4-ethyl-1-methylbenzene	a multi-stage emission profile was evident, representing the initial fast emission of various ranges of volatile compounds; the emission rate took 5 days to reach a constant level of 0.3 mg·m ⁻² ·h ⁻¹
	foam sealant	dichloromethane	<i>m</i> -/ <i>p</i> -xylene	<i>o</i> -xylene	toluene	unknown (RT 47.1)	the peak emission occurred during the first 30 min of exposure but declined from 22.2 mg·m ⁻³ to 0.18 mg·m ⁻³ after 13 days; xylene was then dominant
	water-proof emulsion	naphthalene	C ₁₄	1-methylnaphthalene	acenaphthene	biphenyl	a relatively high emission rate was measured during the first day of analysis (307.5 mg·m ⁻² ·h ⁻¹ TVOCs and 166.6 mg·m ⁻² ·h ⁻¹ naphthalene); the emission rate declined quite slowly and took 5–7 days to reach a fairly constant level
	polystyrene floor insulant	HCFC 142	styrene	ethylbenzene	toluene	ND	a constant low level of emission was detected; the maximum TVOC emission rate was 0.2 mg·m ⁻² ·h ⁻¹ and 0.1 mg·m ⁻² ·h ⁻¹ for styrene

() = possible identity according to GC retention time.

¹ Testing in 1-m³ chamber.

² Plywood was used as substrate.

³ Gloss was applied on 2 layers of undercoats on primer on plywood.

the detection limit of the chamber test within 1–5 days of exposure. High, moderate and low emitters are the materials that have a maximum TVOC emission rate of at least 100 mg·m⁻²·h⁻¹, 10 mg·m⁻²·h⁻¹ and 0.01 mg·m⁻²·h⁻¹, respectively, during the chamber test.

Formaldehyde emission was measured separately from the VOCs because of the different method of analysis. The

following materials were found to emit formaldehyde in order of decreasing emission rate: chipboard, carpet, 6-mm plywood, plasterboard, bituminised fibreboard, 15-mm plywood, mineral wool (insulant) and curtain.

The linking of indoor air concentrations of VOCs to information about sources provided by emission testing of materials using chambers is a complex process. Factors

Table 4. Classification of materials based on their TVOC emission rate

Long-term			Short-term		
high	moderate	low	high	moderate	very low/not detectable
water-proof bituminous emulsion	water-borne emulsion paints, putty, treated timber	vinyl flooring, carpet, underlay, chipboard, bituminised fibreboard, asphalt, plywood, untreated timber	spray-on adhesive, acrylic adhesive, wood stain, gloss paints and undercoats, caulk	foam sealant	plaster board, PVC skirting board

that need to be taken into account include the amount of material in contact with the indoor air environment, the emission rate in situ, which would be influenced by the micro-environment of the material, outdoor concentrations, ventilation characteristics and adsorption and re-emission of VOCs to and from indoor surfaces. Møhlave et al. [15] in a study of two new houses simplified the problem by measuring the emissions from composite building elements (e.g. wall composed of particle board, glue, wallpaper and paint) and not including the first 32 days after construction when emission rates and concentrations change most rapidly. They used the results for TVOCs and formaldehyde to compute predictions of indoor concentrations based on the emission data and a modified 'EXPOSURE' model and report that reasonably accurate predictions were achieved. Zellweger et al. [16] demonstrated the difficulties of relating room concentrations and emission data for one source, a flooring adhesive, and Jenson and Nielson [17] discussed the importance of one aspect of the micro-environment for determining emission rates, that of surface air velocity.

The complexity of the composition of emissions from many of the materials used in the BRE test houses as well as the complexity of the mixture of VOCs in the indoor air, especially during the first 6 months after construction, produce difficulties for identifying the major sources of particular VOCs found in the indoor air. No single material can be identified as having a dominating influence upon the VOC composition and concentration in the indoor air, and many of the compounds are common to more than one material. Similar findings were reported by Wolkoff et al. [18] who found 120 different VOCs during headspace analyses of 8 building materials and 20 building products used in the construction of two test houses.

The highest emission rates from materials used in the BRE test houses are associated with wet products such as paints and adhesives. Their emission declines rapidly during the first 24 h after application and this appears to be much more rapid than the changes in the indoor concentration in the houses. The influence of these products on the indoor air may be prolonged by the much greater adsorption and re-emission processes in a house compared with in a chamber. Undecane was most frequently the dominant compound in both types of houses. The most important sources were gloss paint and wood stain, but other materials such as timber, caulk and bituminised fibreboard may have contributed. The dominant aromatic hydrocarbons in the indoor air were xylenes and these were amongst the top five compounds for several materials, but were also present in the complex mixtures from other products such as gloss paint and wood stain. Texanol-1 and Texanol-2 were major compounds in the indoor air and the strongest source of these compounds was the emulsion paint applied to the walls and ceilings. Pentadecane and TXIB were released by flooring materials. No clear link was found between the TVOCs in the test house and emissions from the materials. Further work is ongoing to apply indoor air pollution models to further examine the relationships between the material sources and the indoor air concentration in the test houses.

Conclusions

Building and furnishing materials make an important contribution to the concentration of a wide range of VOCs including formaldehyde in the indoor air. During the first year following construction of the test houses, the mean

annual concentrations of TVOCs were above 95-percentile values measured in occupied UK homes and formaldehyde concentrations were between 75- and 95-percentile values. TVOC concentrations declined rapidly during the first 4 months after construction whereas formaldehyde exhibited a slower rate of decline with strong seasonal changes in concentrations.

Outdoor air was the major source of toluene and benzene in the indoor air. All but 3 of 30 of the materials used in construction and furnishing of the buildings were found to be potential sources of VOCs. For many of the compounds such as aliphatic hydrocarbons and xylenes it is difficult to link the occurrence in the indoor air to release from a particular material as several materials are potential sources. Texanol in the indoor air was predominantly due to paints used on the walls and ceiling and TXIB and pentadecane were from flooring materials. Several compounds significant in occupied homes including toluene, limonene and some VVOCs were not major com-

pounds in the indoor air of the test houses. This may reflect the importance of consumer products as sources of these compounds.

There was no significant difference in the concentration of TVOC, benzene, toluene, xylene, undecane and formaldehyde between the two types of test houses and concentrations at different locations within each house were similar. The composition of the TVOC did show some differences between houses, with decane being dominant during 16 and 52% of sampling events in the masonry-walled houses and on no occasions in the timber-framed houses. By contrast Texanol was more frequently dominant in the timber-framed houses. Seasonal changes in concentrations were observed with highest concentrations in summer months for those compounds with major indoor sources. This is probably due to higher summer temperatures resulting in an increased rate of emission of VOCs from materials in the buildings.

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