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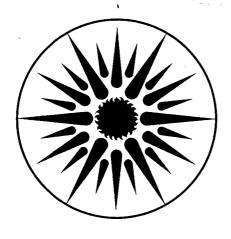
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VOLATILE ORGANIC EMISSIONS FROM ADHESIVES WITH INDOOR APPLICATIONS

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Abstract

Studies have shown that volatile organic compounds (VOC) emitted from building materials are a potentially important source of indoor air pollution. In this study, we investigated emissions of VOC from both solvent-and water-based adhesives. Adhesives were applied to an inert substrate and dried for at least a week. VOC were cryogenically trapped and identified by GC-MS or sorbent trapped, solvent extracted, and quantified by GC-FID. Among the compounds emitted by adhesives were toluene, styrene, and a variety of normal, branched, and cyclic alkanes. The measured emission rates ranged from below the limit of detection for some adhesives to a total alkane emission rate of over 700 $\,\mu g \, g^{-1} h^{-1}$ for a water-based adhesive. A simple, well-mixed tank model was used to assess the potential impacts of the adhesives studied and to demonstrate that adhesives can be significant sources of VOC.

Introduction

Because heating or cooling outdoor air as it enters an office building consumes a significant amount of energy, recent office construction practices have incorporated energy-conserving strategies to minimize the amount of outdoor air used for ventilation. These practices may contribute to the large number of non-specific health complaints thought to be related to poor indoor air quality. The underlying etiological agents as determined by the criteria of current health standards are unknown. While few studies of organic contaminants in indoor air have been made in non-industrial buildings, fragmentary evidence suggests that these contaminants can be

major contributors to poor indoor air quality (1,4,8). In the industrial indoor environment, exposure to elevated levels of organic contaminants have been shown to induce a variety of toxic, carcinogenic and teratogenic effects. The possibility that chronic exposure to these same compounds may occur in non-industrial indoor environments caused by the increased use of synthetic building materials and furnishings in office spaces makes it imperative that identities, levels and sources be characterized.

Previous studies have indicated the potential of building materials to contribute to poor indoor air quality by the emission of volatile organic compounds (VOC) (5,6). In addition, one of these studies identified adhesives as an important source of VOC in indoor air (6). Because of this and because adhesives are suspected of causing widespread health complaints in an office building (7), we investigated the emissions of VOC from a broad range of adhesives with indoor applications.

Experimental

Two procedures were used for the study of adhesive emissions (3). In the first phase of this study, the adhesives were vacuum extracted and VOC were cryogenically trapped. Preweighed plates were coated with approximately 1 g of wet adhesive and dried for 9-14 days under ambient conditions prior to extraction. Cryogenically trapped VOC were taken up in carbon disulfide and injected into a gas chromatograph-mass spectrometer for qualitative and semi-quantitative analysis. Compounds were identified by comparing unknown spectra with spectra contained in the EPA/NIH Mass Spec-

tral Data Base (2). This procedure allowed quick screening of samples to determine which adhesives emitted significant quantities of VOC of interest and, thereby, provided a basis for selection of materials for further study. Organic compounds emitted by 15 adhesives used for floor, wall and ceiling treatments were identified by the above procedure.

In the second phase of the study we measured the VOC emission rates of the eight adhesives which had significant emissions. Larger samples of adhesives were prepared as above using manufacturers' recommended coverages and were dried for 9-14 days at ambient conditions. Humidified (40% RH at 25°C), high purity air at room temperature and atmospheric pressure was passed through small chambers containing the adhesive samples and the VOC entrained were trapped with two-stage charcoal adsorption tubes for periods of one to two hours. The samples were desorbed with carbon disulfide and injected into a gas chromatograph equipped with a flame ionization detector for quantification.

Results and Discussion

Based upon screening by vacuum extraction, eight of the fifteen adhesives studied continued to emit significant amounts of VOC after one week of drying. The major compounds identified in the vacuum extracts of the solvent-based adhesives were toluene, styrene, and a variety of cyclic, branched and normal alkanes. Of these, toluene was the most abundant. Surprisingly, a large variety of alkanes were obtained from three of the water-based adhesives. A more complete listing of VOC emitted by the adhesives is found in Table 1. Two of the water-based adhesives, both from the

same manufacturer, emitted an almost identical, complex mixture of normal and branched alkanes and cyclohexanes, indicating a common solvent base. Two other adhesives, S-6 and W-3, emitted a different mixture of higher molecular weight, normal and branched alkanes. The similarity between the emissions from S-6 and W-3 is unexpected since they are supplied by different manufacturers, and one is a solvent-based asphalt adhesive and the other is a water-based natural rubber adhesive. After drying periods of 9-14 days, only five of eight adhesives had solvent emission rates detectable by the air exposure procedure. (The minimum emission rate measurable for a single VOC by this procedure is estimated to be 0.1 $\mu g g^{-1}h^{-1}$.) Three solvent-based adhesives had toluene emission rates between 0.6 and 62 $\mu g q^{-1}h^{-1}$. The two water-based adhesives, W-1 and W-2, emitted the same complex mixture of normal and branched alkanes and cyclohexanes in the emission rate tests as they had in the vacuum-extraction screening tests (see Figure 1). Because of the complexity of the mixture, emission rates were not determined for individual compounds. Instead, emission rates for total alkanes were calculated and are reported in Table 1. These emission rates ranged from 610 to 780 μg $g^{-1}h^{-1}$. When the recommended coverage of an adhesive (shown in Table 2) is multiplied by the appropriate emission rate listed in Table 1 for a specific adhesive, the emission rate based upon surface area is obtained. These rates range from 0.14 to 7.4 mg $m^{-2}h^{-1}$ for toluene and 140 to 180 mg $m^{-2}h^{-1}$ for total alkanes, in reasonable agreement with the VOC emission rates, 2.1 and 271 mg $m^{-2}h^{-1}$, reported by Mølhave for two adhesives (6).

Table 1. Volatile organic compounds emitted by adhesives with indoor applications for sales are given for tolugies or total alkanes for salested adhesives.

Adhesive Chemical Base	Application	Volatile Organic Compounds	Weight of Adhesive (10 ⁻² g cm ⁻²)	Drying Time (days)	Emission Rate (µg g ⁻¹ h ⁻¹)
Solvent-based					Toluene
Synthetic rubber S-1	Rigid plastic foams to walls, ceilings	toluene; styrene	6.70	9	0.59
Unapecified polymer S-2	Styrene, polyurethane, cork, celotex, carpet	low-molecular-weight alcohol?;toluene			
Synthetic polymer S-3	Plasticized rubber, vinyl carpet	toluene	0.93	13 13	6 2 4 8
Asphalt S-6	Roof shingles, mesonry cracks	n-decase,n-undecase, C ₁₀ -C ₁₁ branched alkanes(9+ compounds), C ₁₀ cyclohexanes (4 compounds)			
Unspecified polymer S-7	Subfloor bonding	methyl cyclopentane; cyclohexane; toluene	4.32 6.50	11 11	2.4
Weter-based					Total Alkanes
Synthetic rubber W-1	Foam backed indoor and outdoor carpet	n-octane, n-nonane, C ₈ -C ₉ branched alkanes (7+ compounds), methyl cyclohexane, C ₈ -C ₉ cyclohexanes (10+ compounds)	2.54 2.85	1 4 1 4	740 760
Synthetic rubber ¥-2	Foam backed indoor and outdoor carpet	same as compounds W-1	3.36 3.82	14 14	610 780
Natural rubber W-3	Poam, aponge backed indoor and outdoor carpet	toluene; n-nonane, n-decane, n-undecane, Clo-Cli branched alkanes (9+ compounde), Clo cyclohemane			

To assess the potential impacts of adhesives on indoor air quality, the emission rates determined in this study were used in a ventilation model, assuming a well-mixed tank. When applied to a hypothetical office space, the model results, as shown in Table 2, demonstrated that the two water-based adhesives could produce significant concentrations of YOC. However, model results must be interpreted with caution, since the adhesives as applied in an office space would be overlaid with carpet or other coverings which would undoubtably moderate the YOC emissions though, perhaps, extending the duration of emissions. Thus the model results may represent an upper bound on YOC concentration. A more complete description of these studies can be found in a recent report (3).

Table 2. Calculated steady-state concentrations of toluene and total alkanes produced by emissions from adhesives applied to an area equal to the floor area in a hypothetical office space at two ventilation rates assuming an occupant density of 7 workers per 100 m^2 .

Adhesive	Recommended_2 Coverage (g m ⁻²)	Yentilation Rate 233 m ³ h ⁻¹ **		
		Toluene Concentration (mg m^{-3})		
S-1 S-3 S-7	245 120 235	0.06 2.8 0.25	0.25 11 1.0	
		Total Alkane Concentration (mg m^{-3})		
W-1 W-2	240 225	76 67	310 270	

^{*} Assumes smoking permitted. Space ventilated at minimum suggested ASHRAE rate of 34 m h^{-1} (20 ft min) per occupant.

Conclusion

This study of volatile organic emissions demonstrates the potential of adhesives to contribute to poor indoor air quality. Emissions from water-based adhesives were unexpectedly high and may be significant in certain applications. Further studies of adhesives should be conducted with emphasis on determining emissions under a range of temperatures, humidities, and air flows while the adhesives are under simulated use, gluing appropriate materials such as carpets or tiles. These studies should also assess the effects of adhesive aging on emissions. There is also a need to compare concentrations derived from emission rates determined in small chambers and

^{**} Assumes no smoking permitted. 3 Space ventilated at minimum suggested ASHRAE rate of 8.4 m^{h-1} (5 ft³ min⁻¹) per occupant.

models with concentrations measured in large chambers under simulated use, and with concentrations at actual indoor sites where relevant parameters such as the age and amount of the adhesive, and environmental conditions are known.

Studies of organic emissions from building materials coupled with measurements of organic concentrations in commercial office buildings will provide the compound identification necessary for assessing the health effects of chronic exposure on occupants. These studies will indicate the range of organic concentrations to be addressed in health effect studies and may also suggest which compounds are commonly associated with each other, indicating the need for examining possible synergistic health effects. Finally, field studies of organic contaminants in office buildings under different conditions of ventilation, temperature, and humidity combined with laboratory studies of organic emissions from building materials under a similar range of conditions will provide a data base for architects and builders so that materials with low potential for organic emissions can be selected for use in new, energy-conserving office buildings and in remodeled office buildings.

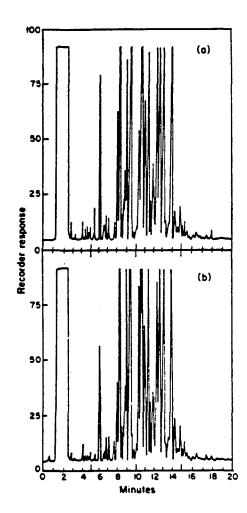
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Figure 1. Comparison of the gas chromatograph-flame ionization detector chromatograms of (a) the vacuum extract and (b) the charcoal tube $\,$ air sample of adhesive W-1.

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