INDOOR AIR POLLUTION BY VOLATILE ORGANIC COMPOUNDS (VOC) EMITTED FROM FLOORING MATERIAL IN A TECHNICAL UNIVERSITY IN SWITZERLAND

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ABSTRACT
A case study was conducted at a technical university where employees and students complained about deteriorated indoor air after the building had been renovated. Some employees even suffered from sickness and headache. Indoor air concentrations of total volatile organic compounds (TVOC) were elevated in both laboratory halls and reference rooms. Concentrations of most chemicals decreased considerably after six months. The smell intensity decreased in parallel but still was rated as highly annoying. The source of the odorous compounds and most probably of the health problems was identified to be the vinyl (PVC) flooring. The vinyl flooring emitted, among others, phenol and 2-ethylhexanol. Emission factors correlated inversely with the frequency of room usage. The results provide an example of the use of unsuitable building products with respect to healthy indoor air. In future projects it is suggested to focus on the effect of co-exposure to multiple component mixtures of pollutants.

INDEX TERMS
VOC, Phenol, Health effects, Vinyl flooring, Combined exposure

INTRODUCTION
The concentration of volatile organic compounds (VOCs) measured in indoor air are often significantly higher than outdoors. There are various potential origins such as human activities, infiltration from outdoor air, heating ventilation and air conditioning (HVAC) systems (Wolkoff, 1995; Fanger, Lauridsen, Bluyssen, *et al.*1988), bacterial and fungal activities (Rivers, Pleil and Wiener, 1992; Batterman, Bartoletta and Burge, 1991) and product emissions (Wallace, Pellizzari, Leaderer, *et al.* 1987; Wolkoff 1995; Wolkoff, Jensen, Kjaer, *et al.* 1996; Hodgson, Rudd, Beal, *et al.* 2000). The concentrations are usually orders of magnitude below the occupational threshold limit values (TLVs). However, some VOCs may be present above their human odor thresholds (OTs). Beside odor annoyance, VOCs at sub-TLV level may cause non-specific health effects such as eye and upper respiratory airway irritation, headache and increased weariness (Molhave, 1986; Molhave, 1991; Seifert, 1999 and references there in; Pitten, Bremer and Kramer, 2000).

The extensive use of petrochemical based organics and bio-organics in building products paralleled by low natural air exchange rates in nearly air-sealed buildings often results in elevated indoor concentrations of VOCs. A large number of studies were done on flooring materials, e.g. rubber, carpets, linoleum and vinyl (Wolkoff, 1995 and references therein). Linoleum emits mainly aliphatic and olefinic aldehydes and aliphatic acids (Wokoff, 1995; Wolkkoff, Clausen and Nielson, 1995; Jensen, Wolkoff, Willkins, *et al.* 1995; Jensen, Wolkoff, and Willkins, 1995). PVC, on the other hand, emits various types of VOCs

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(Wolkoff, 1995; Wolkoff, Jensen, Kjaer, *et al*. 1996). Lundgren (Lundgren, Jansson and Ek-Olaussen, 1999) identified 30 compounds with an emission rate ranging from 5 to 350 µg $m^{-2} h^{-1}$ at 4 weeks after manufacturing. Among these 2-(2-butoxyethoxy) ethanol, butoxyethanol, phenol, hepta- and octadecanes were found to be the predominant compounds which were suggested for further investigation with respect to their role as indoor air toxicants.

This field study was initiated after employees and students of a technical university in Switzerland complained about deteriorated indoor air quality since renovation of the building. Some people suffered from sickness, headache and increased weariness after having worked in the odorous indoor air of the laboratory halls which were equipped with PVC flooring. The study aimed at identifying components emitted by the building products which could be responsible for the observed health effects. Field data based evidence for health effects of individual VOC components emitted from building products are to our knowledge rarely described in the literature.

METHODS

Study Site

The university was located in the suburban countryside of Switzerland. Classrooms and laboratories of the investigated department were placed on the 2nd and 3rd floor of a three-floor industrial building in an industrial zone. Several small businesses remained on the 1st floor and in the vicinity such as e.g. a tinsmith or a car painter. The building was renovated in spring 1998. The walls and ceilings were made of white painted concrete. The floors of the laboratory halls were covered with a chemically inert PVC flooring, while in all other rooms a linoleum flooring was built in. All rooms were naturally vented. Ordinary laboratory benches and metal cupboards were installed in the laboratories.

The indoor temperature and the relative humidity during the air sampling campaigns ranged between 22.8 and 23.4°C and between 23 and 30%, respectively, on January 3 and at about 26°C and 45% on June 23. During emission measurements the temperature ranged between 23.1 and 24.5°C. The humidity of the air used for emission sampling was held at 50%.

Sampling and analyses

VOCs in indoor air were measured using a thermodesorption / gaschromatography / mass spectrometry / FID (TD/GC/MS/FID) method similar to that published by Wolkoff (Wolkoff, Jensen, Kjaer, *et al*. 1996) and by Mogl (Mogl, Haas and Knutti, 1995). Emission factors of flooring materials were determined using the field and laboratory emission cell (FLEC) (Wolkoff, Clausen and Nielsen, 1995).

Three different rooms were selected for investigation. 1) The biology laboratory hall (PVC flooring) on the 2nd floor, 2) the chemistry laboratory hall (PVC flooring) on the 3rd floor and, 3) the staff room (linoleum) on the 3rd floor for comparison. Indoor-air samples were collected on January 3, 2000 and on June 23, 2000. On-site FLEC samples of the built-in flooring materials were collected on September 13 and September14, 2000. Back-up samples of both the PVC and the linoleum flooring material were sampled in the FLEC-chamber on September 22, 2000. Back-up samples were of the same batches as the built-in material. They were stored in the university under unknown conditions. On January 3, 2000, an outdoor-air sample was collected outside the window of the chemistry laboratory hall in order to check for potential infiltration.

RESULTS

TVOC/VOC concentrations in indoor air

The total volatile organic compound (TVOC) concentration decreased from the first to the second campaign, i.e. from ½ year to 1 year after renovation, by a factor of two to three (Table 1). The VOCs could be divided in two groups. The first group consisted of relatively highly volatile compounds of decreasing concentrations, e.g. acetone, isopropanol, 2-butanone, and some aromatic and aliphatic hydrocarbons, which showed rapidly decreasing concentrations in all sampling locations. The second group consisted of compounds which did not decrease significantly, or even increased during the study period. The concentration of phenol, for example, remained constant in the air of the two laboratory halls and increased in the staff room. The concentration of 2-ethylhexanol increased in all rooms. Aldehydes increased mainly in the staff room.

Table 1. VOC concentrations (🡣: decreasing; ➔: steady, ➚: increasing) in toluene equivalents [$\mu g/m^3$] in samples collected on January 3, 2000 and June 23, 2000.
Chemlab: Chemistry laboratory hall, *Biolab:* Biology laboratory hall.

Sampling site Flooring type	*Outdoor*	*Chemlab PVC*		*Biolab PVC*		*Staff room Linoleum*	
Date of sampling	*Jan. 3*	*Jan. 3*	*June 23*	*Jan. 3*	*June 23*	*Jan. 3*	*June 23*
🡣 Ethanol	4	190	37	130	5	11	5
🡣 Acetone/i-propanol	40	250	55	330	18	130	38
🡣 2-Butanone	6	140	19	85	6	59	12
🡣 Benzene	10	14	1	13	2	14	3
🡣 1-Butanol	6	27	15	61	12	33	16
🡣 n-Heptane	10	30	4	33	2	46	17
🡣 Toluene	29	210	61	261	61	212	47
🡣➚ Hexanal	-	15	6	28	6	3	16
🡣 Ethylbenzene	3	52	4	48	3	37	7
🡣 Xylenes	16	210	19	190	16	150	31
🡣 n-Nonane	2	10	1	9	3	7	2
🡣 α-Pinene	-	18	2	39	1	12	1
🡣 Benzaldehyde	-	17	2	23	1	-	1
➔➚ C_3-Benzenes	6	44	10	34	15	12	20
➔➚ Phenol	3	13	13	34	33	5	10
🡣 Decane	6	20	4	22	3	13	4
➔ Octanal	-	3	2	4	2	4	6
➚ 2-Ethylhexanol	-	5	10	5	17	4	12
🡣 Limonene	2	19	2	21	1	43	3
🡣 Undecane	-	9	3	11	3	7	3
➚ Nonanal	2	4	9	4	8	4	14
🡣 TVOC*	**172**	**1160**	**410**	**1400**	**320**	**1030**	**400**

*) total of components which are chromatographically eluted between hexane and hexadecane
-: < 1 $\mu g/m^3$

VOC emission from flooring material

The major components emitted by the PVC flooring were phenol, 2-ethylhexanol, and a number of low volatile hydrocarbons, while the linoleum flooring emitted a homologous series of linear aldehydes and carbon acids (not quantified) (Table 2). Both indoor air concentrations and emission factors of phenol and 2-ethylhexanol were significantly higher in the less frequently used and vented biology laboratory than in the chemistry laboratory.

Table 2. Emission factors [$\mu g \cdot m^{-2} \cdot h^{-1}$] of the flooring material.
On-site: On-site FLEC-sampling of VOCs emitted from built-in floorings in the room indicated on September 13 and 14.
FLEC-chamber: FLEC chamber sampling of VOCs emitted from unused back-up samples of the same batches as the built-in material on September 22.

	On-site			*FLEC chamber*	
Flooring type	*PVC*		*Linoleum*	*PVC*	*Linoleum*
	Chemlab	*Biolab*	*Staff room*	*Back-up samples*	
2-Butanone					5.8
1-Butanol		13	1.7	1.0	
Pentanal				1.7	21
Toluene		2.2			
Hexanal	0.3	0.1	1.5	1.0	33
Heptanal					11
2-Butoxyethane				3.0	
Phenol	3.4	13		85	
Decane				2.5	
Octanal				0.6	24
2-Ethylhexanol		22		16	
N-Methylpyrrolidone				7.7	
Octylformate				0.7	16
Undecane				2.1	
Nonanal	1.8		1.1	2.0	23
Decanal				1.0	
Decenal	1.6			0.6	7.7
2-Phenoxyethanol		6.0			
Hydrocarbons C>12	2.5	7.0	0.9	54	7.6

Empty fields: <0.1 $\mu g\ m^{-2}\ h^{-1}$

DISCUSSION
The decrease in TVOC concentrations with time was well in agreement with former results of VOC monitoring in new or renovated buildings (Wolkoff, 1995). Even though the TVOC concentrations decreased by more than a factor of two, and single VOCs were present below odor thresholds (OT), the perceived air quality in the two laboratory halls was rated extremely annoying. The odor quality was equal in all laboratory facilities and did not depend on any of the chemicals used for training and teaching. The group of VOCs which showed no clear decline or even an increase in concentration suggested the presence of large indoor reservoirs which emitted VOCs under diffusion or reaction controlled conditions (Hodgson, Rudd, Beal, *et al.* 2000). The FLEC measurements supported the presence of VOC reservoirs. Phenol and 2-ethylhexanol were clearly emitted by the PVC flooring (Lundgren, Jonsson and Ek-Olausson, 1999), whereas aldehydes were predominantly emitted by the linoleum (Jensen, Wolkoff, Willkins, *et al.* 1995; Jensen, Wolkoff and Willkins, 1995).

Malodorous air was present in all rooms which were equipped with the PVC flooring. The odor intensity, however, rated variable and seemed to correlate with three parameters: 1) the concentration level of phenol and 2-ethylhexanol, 2) the on-site measured emission factors of phenol and 2-ethylhexanol, and 3) negatively with the frequency of room usage. Reproduced odor quality of the laboratory halls using the unused PVC back-up sample in a sealed glass jar was very close to that of a simple mixture of phenol, 2-ethylhexanol and N-methylpyrrolidone. However, phenol and 2-ethylhexanol had different emission characteristics. The highest emission factor for phenol (85 $\mu g\ m^{-2}\ h^{-1}$) was obtained from the unused back-up sample. Readings decreased to 13 and 3.4 $\mu g\ m^{-2}\ h^{-1}$ in the biology laboratory and the chemistry laboratory, respectively. The emission of 2-ethylhexanol, on the other hand, increased from 16 $\mu g\ m^{-2}\ h^{-1}$ in the back-up sample to 22 $\mu g\ m^{-2}\ h^{-1}$ in the biology laboratory. It was not detectable in the flooring of the chemistry laboratory. This finding may indicate hydrolysis reaction of the di(ethylhexyl)phtalate (DEHP) used in the PVC flooring as plasticizer. The hydrolysis rate and, hence, emission rates presumably were dependent on environmental conditions such as humidity, alkalinity and temperature.

CONCLUSIONS AND IMPLICATIONS

The observed disease symptoms were strictly building related. The spatial coincidence of symptoms with the odor annoyance suggests that the compounds which were emitted by the PVC flooring caused the sickness. Even though direct causality cannot be proven, results indicate that phenol and 2-ethylhexanol, which both persisted in parallel with the symptoms, have played a predominant role in the complex VOC mixture. This conclusion contradicts the “Lowest Concentration of Interest” (LCI) concept (Bluyssen, Cochet, Fischer, *et al.* 1997). Phenol and 2-ethylhexanol are listed at an LCI of 400 $\mu g/m^3$ and 1000 $\mu g/m^3$, respectively. The LCI given for phenol is reasonably close to the OT (427 $\mu g/m^3$) used by Fang (Fang, Clausen and Fanger, 1999), but far above the OT of 0.016 ppm (about 70 $\mu g/m^3$) published by the National Toxicological Program (NIEHS 2001). In this study, phenol in the VOC mixture of the biology laboratory was already perceived at 30 $\mu g/m^3$ in combination with other organics suggesting a synergistic effect. Similarly, Wolkoff (Wolkoff, Jensen, Kjaer, *et al.* 1996) published perceived air quality (PAQ) results of low emitting building products. Despite low emissions, determined PAQs were rated considerable.

Regarding health, evaluating the effects of single compounds may not always be adequate (WHO 1989). Ziem (Ziem and Davidoff, 1992) reported that only 2% of at least 60’000 chemicals that are used widely have been comprehensively studied for toxic effects and that they have rarely been studied in combined exposure, which actually exists in the real world. Furthermore, he stated that the minimum exposure, levels necessary for a specific toxic effect are rarely known and the minimum toxic dose estimates for general population grossly overestimate the doses that could affect sensitive individuals. As a consequence more rigid use of product labeling, careful selection of building products and construction surveys are of major importance in reducing indoor air pollutants to the lowest level possible.

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