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Keywords indoor air, material emission, VOCs, ammonia, formaldehyde, new buildings

Abstract

Indoor air concentrations and emissions from structures and interior materials were investigated in eight residential buildings during the time of construction and the first year of occupancy. Volatile organic compounds (VOCs), formaldehyde and ammonia concentrations and emissions as well as temperature, humidity, and ventilation were measured.

The total VOC (TVOC) concentration was generally above the S3-class limit of 600 µg/m^3 (FiSIAQ 2001) in the newly finished buildings but the concentration usually decreased below the S3-level and in some apartments below the S1-level of 200 µg/m^3 in six months. The concentrations of the major VOCs decreased most strongly during the first six months of occupancy, reaching mean concentration levels of 5–15 µg/m^3. Variables affecting the concentrations of indoor air gaseous pollutants in the buildings were the ventilation system, the floor covering material, the ceiling surface product, the wall surface product, the season, the relative humidity and temperature of the indoor air, and occupancy. The relative humidity (RH) affected ammonia and formaldehyde concentrations most strongly. Higher concentrations were measured when the RH was above 50% during the follow-up. The formaldehyde concentration did not significantly exceed the S2-class level of 50 µg/m^3 during the first year in any of the apartments. In some newly finished buildings and during the follow-up, the indoor air concentrations of ammonia were above the S3-level of 40 µg/m^3. The emission measurements performed from the complete floor construction showed that the emission was affected by all of its components, i.e., the structure, levelling agent, adhesive, and floor covering material. Significantly higher emissions were often measured on-site from the complete floor structure than from the single materials measured in the laboratory. The impact of adhesives on VOC emissions from the complete PVC- coated structures was clearly seen as higher emissions from those with a more permeable types of PVCs. The contribution of the average on-site measured emissions to indoor air concentration was ~550 µg/m^3 (~57% of the measured concentration) for TVOC and ~45/40 µg/m^3 (~100% of the measured concentration) for ammonia and formaldehyde in the newly finished building. The TVOC contribution from surfaces decreased to ~200 µg/m^3 in six months whereas the contribution of ammonia and formaldehyde remained about the same. The ceiling structure contributed by most to the concentration levels whereas the contribution from walls was lower than expected on the basis of large surface area. The study confirmed that the Finnish material classification system provides a basis to achieve good indoor air quality when comparing to the target values for pollutant concentrations given by the classification (FiSIAQ 2001) in real buildings; however, suggestions for its further development are given. Based on the indoor air and emission results, reference values, i.e. “normal” and “abnormal values”, were defined for the six- and twelve month-old buildings.

Järnström, Helena. Reference values for building material emissions and indoor air quality in residential buildings [Referenssiarvot rakennusmateriaalien emissioille sekä sisäilman laadulle asuinrakennuksissa]. Espoo 2007. VTT Publications 672. 73 s. + liitt. 63 s.

Avainsanat indoor air, material emission, VOCs, ammonia, formaldehyde, new buildings

Tiivistelmä

Sisäilman pitoisuuksia sekä rakenteiden emissioita tutkittiin kahdeksassa asuinrakennuksessa rakentamisen aikana sekä ensimmäisen vuoden aikana käyttöönoton jälkeen. Tutkimuksen aikana mitattiin orgaanisten haihtuvien yhdisteiden (VOC-yhdisteiden), formaldehydin ja ammoniakin sisäilman pitoisuudet ja emissiot rakenteista sekä lämpötilat, kosteus ja ilmanvaihto.

VOC-yhdisteiden kokonaismäärä (TVOC) oli tavallisesti yli S3-luokan eli 600 $\mu g/m^3$ (FiSIAQ 2001) juuri ennen luovutusta, mutta pitoisuus saavutti tavallisesti S3-luokkaa vastaavan tason, ja joissain asunnoissa jopa S1-luokan eli 200 $\mu g/m^3$ ensimmäisen kuuden kuukauden aikana. Yksittäisten VOC-yhdisteiden pitoisuudet laskivat eniten ensimmäisen kuuden kuukauden aikana asunnon käyttöönotosta, jolloin keskimääräiset pitoisuudet olivat tasolla 5–15 $\mu g/m^3$. Sisäilman pitoisuuksiin eniten vaikuttavat tekijät olivat ilmanvaihtojärjestelmä, lattiapinnoite, kattopinnoite, seinäpinnoite, vuodenaika, sisäilman lämpötila ja kosteus sekä asunnon toiminta.

Sisäilman suhteellinen kosteus (SK) vaikutti eniten formaldehydi- ja ammoniakkipitoisuuksiin. Korkeampia pitoisuuksia seurantajakson aikana mitattiin kun SK oli yli 50 %. Formaldehydipitoisuus ei ylittänyt S2-luokan tasoa eli 50 $\mu g/m^3$ missään tutkitussa asunnossa. Joissakin uusissa sekä asutuissa asunnoissa sisäilman ammoniakkipitoisuudet olivat yli S3-tason eli 40 $\mu g/m^3$. Emissiomittaukset valmiista rakenteesta osoittivat, että rakenteen emissioihin vaikuttavat kaikki sen eri komponentit eli runko, tasoite, liima ja lattiapinnoite. Oikeasta rakenteesta mitattiin usein merkittävästi korkeampia emissioita kuin yksittäisistä materiaaleista laboratorio-olosuhteissa. Asennuksessa käytetyn liiman vaikutus oli selvästi nähtävissä niillä muovimattopinnoitteilla, jotka olivat VOC-yhdisteitä läpäiseviä.

Paikan päällä mitattujen emissioiden vaikutus sisäilman pitoisuuksiin oli ~550 µg/m^3 (~57 % mitatusta pitoisuudesta) TVOC:lle ja ~45/40 µg/m^3 (~100 % mitatusta pitoisuudesta) ammoniakille ja formaldehydille juuri valmistuneessa rakennuksessa. TVOC:n vaikutus pieneni noin ~200 µg/m^3:aan kuuden kuukauden aikana, kun taas ammoniakin ja formaldehydin osuudet pysyttelivät samalla tasolla. Kattorakenteen vaikutus pitoisuuksiin oli suurin, kun taas seinien osuus oli odotettua pienempi suuresta pinta-alasta huolimatta.

Tutkimus osoitti, että suomalainen materiaaliluokitus luo perustan hyvän sisäilman saavuttamiselle, kun sisäilman pitoisuuksia vertaillaan luokituksen asettamiin tavoitearvoihin (FiSIAQ 2001) oikeissa rakennuksissa. Kuitenkin, tämän tutkimuksen perusteella esitetään ehdotuksia luokituksen edelleen kehittämiseksi. Sisäilma- ja emissiotulosten perusteella määritettiin referenssiarvot eli arvot “normaali” ja “poikkeava” kuuden kuukauden ja yhden vuoden ikäiselle rakennukselle.

Preface

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I	Järnström, H., Saarela, K., Kalliokoski, P., and Pasanen, A.-L. 2006. Reference values for indoor air pollutant concentrations in new, residential buildings in Finland. Atmospheric Environment 40, 7178–7191.
II	Järnström, H., Saarela, K., Kalliokoski, P., and Pasanen, A.-L. 2007. Reference values for structure emissions measured on site in new residential buildings in Finland. Atmospheric Environment 41, 2290–2302.
III	Järnström, H., Saarela, K., Kalliokoski, P., and Pasanen, A.-L. 2007. The impact of emissions from structures on indoor air concentrations in newly finished buildings: predicted and on-site measured levels. Sent for publication to Indoor and Built Environment in September 2007.
IV	Järnström, H., Saarela, K., Kalliokoski, P., and Pasanen, A.-L. 2008. Comparison of VOC and ammonia emissions from individual PVC materials, adhesives and from complete structures. Environment International 34/3, 420–427.

Abbreviations

AgBB	Ausschuss zur gesundheitlichen Bewertung von Bauprodukten
ASTM	American Society for Testing Materials
C	Carbon
c	concentration (µg/m^3)
HCHO	formaldehyde (chemical structure)
DBP	dibutylphthalate
DEHP	di- (2-ethylhexyl)phthalate
DIBP	di-isobutylphthalate
DPP	dipentylphthalate
ECA	European Collaborative Action
FISIAQ	Finnish Society of Indoor Air Quality
FID	flame ionization detector
FLEC	field and laboratory emission cell
GC	gas chromatograph
GEV	Gemeinschaft emissionskontrollierte Verlegewerkstoffe e.V.
GM	geometric mean
GUT	Gemeinschaft umweltfreundlicher Teppichboden
IAQ	indoor air quality; here, IAQ is often restricted to the quantity and quality of chemical pollutants in the indoor air
ICL	The Indoor Climate Labelling
ISO	International organization for standardization
L	loading (m^2/m^3)
LCI	lowest concentration of interest

LOD	limit of detection
M1	material class 1
MS	mass spectrometer
MSD	mass selective detector
n	number
PCA	principal component analysis
PVC	polyvinylchloride
q	air flow (l/s)
R^2	square of the correlation coefficient
RAL	Deutsches Institut für Gütesicherung und Kennzeichnung e.V.
RH	relative humidity
S1	indoor air class 1
S2	indoor air class 2
S3	indoor air class 3
SER	specific emission rate (µg/m^{2}h)
SVOC	semi-volatile organic compounds
TIC	total ion chromatogram
TXIB	2,2,4-trimethyl-1,3-pentanediol di-isobutyrate
TVOC	total volatile organic compounds
V	Volume (l)
VOC	volatile organic compounds
VTT	Technical Research Centre of Finland
VVOC	very volatile organic compound
WHO	World Health Organization

1. Introduction

It has been estimated that people spend more than 90% of their time indoors; consequently, indoor air quality (IAQ) is of great importance (ECA 2000). Besides causing adverse health effects, poor IAQ may reduce productivity and lead to large economic losses (Seppänen 1999).

IAQ has been under attention in non-industrial environments since the early 1970s, when health problems occurred because of high levels of formaldehyde in indoor air. Formaldehyde emissions originated from building materials containing urea-formaldehyde resins, such as particle board and urea-formaldehyde insulation foam. Because the sources of formaldehyde emissions were obvious, they were also relatively easy to control, and serious IAQ problems rapidly disappeared.

Since the 80s, volatile organic compounds (VOCs) have often been associated with poor IAQ and building-related symptoms. However, no clear health effects have been established for these compounds, which are diverse in their chemical and physical properties. The most common VOCs include acids, alkanes, aromatic hydrocarbons, alcohols, aldehydes, ketones, esters, glycols and glycol ethers, terpenes, and chlorinated hydrocarbons. By definition, these compounds have boiling points ranging from ~50 to 260 ºC. Compounds with a boiling point lower than this are called VVOCs (very volatile organic compounds) and compounds with a higher boiling point (260–380 ºC) are semi-volatile organic compounds (SVOCs, WHO 1989.)

Building materials are important sources of VOCs. Source control – i.e. lowering emissions from building materials in newly established or renovated buildings – is one way to improve IAQ. Consequently, several labelling systems have been introduced to restrain emissions from building materials (Wolkoff 2003). The harmonization work in Europe is still under development (ECA 2005).

The Finnish Indoor Climate Classification was introduced in 1995 and its revised version in 2000 (FiSIAQ 2001). The classification defines the design and target values for thermal comfort, ventilation, odour intensity, noise levels, concentration of indoor air pollutants, material emissions and components of ventilation systems. The classification also provides procedures for constructions work in new buildings (S, M and P classes). Though the classification is voluntary, the use of M1-classified, low-emitting materials has markedly increased and probably improved the IAQ in new Finnish buildings. To date, there are over 1,000 M1-classified building products on the market (The Finnish Building Information Foundation 2007).

Indoor air investigations as well as emission measurements from building materials in test chambers have been performed extensively in recent decades. Investigations on exposure to indoor (and outdoor air) pollutants have focused on source identification by using different source apportionment models (Sexton and Hayward 1987, Edwards et al. 2001a, Liu et al. 2006). However, there are very few investigations on emissions measured on site from real finished structures. The objective of this study was to investigate indoor air concentration levels and emissions in new residential buildings that represent the current building practice in Finland, and in which low-emitting building materials were used. Emissions of structures were determined *on site* from the real finished structure and the impact of them on the concentration levels was clarified. The evolution of material emissions and concentration levels was investigated throughout the first year of occupancy. The results can be utilized as quality control reference data to ensure proper construction practices and in solving problems related to indoor air. The results can also be used for the further development of the Finnish classification.

2. Review of the literature

2.1 Gaseous indoor pollutants

Inorganic gaseous pollutants in indoor environments include oxides of nitrogen (NO_x), sulphur (SO_x) and carbon (CO, CO_2). These compounds originate from combustion processes (heating and traffic, WHO 1989). In addition, ozone can occur in office environments where copying and printing machines are abundant. Outdoor air is also a source of ozone (Weschler 2000). Ammonia can accumulate in indoor air from degradation processes in structures (Karlsson et al. 1989, Gustafsson 1990, Bornehag 1991) and consumer products as well as from the activities of occupants (e.g. environmental tobacco smoke, Tidy and Cape 1993).

Volatile organic compounds (VOCs) and formaldehyde have been regarded as the most important organic gaseous pollutants in indoor air. Semi-volatile organic compounds (SVOCs) adsorb onto particles and surfaces indoors, and their indoor air concentration is usually very low, 10 $\mu g/m^3$ or less (Wensing et al. 2005). Amines (nitrogen containing organic compounds) have been detected at ng/m^3 levels indoors (Palmiotto et al. 2001, Zhu et al. 2004). Amines are also produced in the degradation of casein-containing building materials (Karlsson et al. 1989).

Building materials are important sources of organic compounds as will be discussed in Chapter 2.3. The materials are affected by their surroundings, i.e. by light, humidity and temperature. Other sources of VOCs are outdoor air (e.g. traffic emissions), activities of inhabitants (environmental tobacco smoke, cleaning agents, etc.), contaminated ventilation systems, microbes, and chemical reactions occurring indoors (Wolkoff 1995, Weschler 2000). Indoor air concentrations of VOCs and/or formaldehyde have been measured in several studies in recent decades, the most comprehensive of which are summarized in the following chapters.

2.2 Indoor air investigations

2.2.1 Established and new dwellings

In 1990, Seifert published the indoor air target values for TVOC and VOC groups on the basis of what can be achieved in real buildings as follows: TVOC 300 µg/m^3, alkanes 100 µg/m^3, aromatic compounds 50 µg/m^3, terpenes 30 µg/m^3, halogenated compounds 30 µg/m^3, esters 20 µg/m^3, aldehydes 20 µg/m^3, and other compounds 50 µg/m^3. The values are intended to be applicable in non-industrial environments outside the private sphere, such as offices, schools, kindergartens, etc. (Seifert 1990).

Brown et al. (1994) reviewed a total of fifty indoor air studies performed in established dwellings between 1978 and 1990 in several countries. Concentrations of VOCs were, in general, 5–20 times higher indoors than outdoors. The average indoor/ outdoor ratio for TVOC was 7. The weighted average geometric mean TVOC concentration in established buildings was 1,130 µg/m^3. Single VOC concentrations were below 50 µg/m^3, with most below 5 µg/m^3. The mean VOC and TVOC concentrations were higher in established dwellings compared to public buildings (Brown et al. 1994).

Table 1 summarizes indoor air measurements conducted after 1994. There are several different methods to determine the TVOC/ VOC concentration in air samples as shown in Table 1. Both carbon based adsorbents (i.e. charcoal sorbents) and polymeric (i.e. Tenax) have been used. In addition, both passive (diffusive air sampling) and active (air sampling with pumps) sampling methods are used. The TVOC has been determined by summing the individual VOCs or VOC groups, or by calculating the total area from the flame ionization detector (FID) between hexane/heptane to hexa/heptadecane. Consequently, the values in Table 1 are not fully comparable due to the differences in the methodology used.

Concentrations of individual VOC compounds typically ranged from 1–20 µg/m^3 in established dwellings. The mean TVOC concentration, or sum of target VOCs, was between 120–350 µg/m^3. Home indoor concentration levels of VOCs ranged from ~100 µg/m^3 to ~1000 µg/m^3 in five European cities in the EXPOLIS project. Aromatics were the most abundant compounds in all cities and their concentrations were from ~40 µg/m^3 (in Basel, Helsinki and Oxford) to 106–185 µg/m^3 (in Athens, Milan, and Prague; Saarela et al. 2003).

An analysis of all eluted compounds in Canada revealed that a large number of VOCs are present but remain unidentified. The identified compounds were associated with tobacco smoking, presence of consumer products and plastic materials indoors (Otson et al. 1994). Principal component analysis revealed traffic, indoor equipment and cleaning agents as possible sources of pollution in French dwellings (Kirchner et al. 2003). Multiple regression analysis with concentration and questionnaires showed that outdoor air, smoking indoors, house age, carpet, pets and mothballs were associated with pollutant concentrations in Japanese houses (Ohura et al. 2006).

The TVOC concentration was significantly higher in single-family houses than in multi-family houses in a Swedish study (Bornehag and Stridh 2000). The concentration of TVOC and aldehydes were highest in naturally ventilated buildings. The concentration of alkanes/ alkenes was highest in dwellings built before 1970. The concentrations of 18 VOCs were highest in dwellings with reported condensation on windowpanes. The relative humidity (RH) in indoor air was significantly positively correlated with the concentration of seven VOCs (formaldehyde, n-decane, limonene, C10- and C12-alkane, hexanal, and tridecane. TVOC levels were higher in buildings built after the 1990s than in older ones in England (Raw et al. 2004).

The average or GM indoor air formaldehyde concentrations varied between 12–22 µg/m^3 in the established dwellings (Table 1). The indoor air concentrations of formaldehyde were compared between buildings in Uppsala, Sweden, and Nagoya, Japan, using the same sampling and analytical method (Sakai et al. 2004). The maximum concentrations were higher in Nagoya than in Uppsala. The highest concentration in Uppsala was measured in a 24-year-old wooden detached house. The highest concentration in Nagoya was measured in a two-year-old concrete detached house with an un-vented kerosene space heater. The formaldehyde concentration was significantly higher in non-wooden houses than in wooden houses and higher in dwellings less than ten years old compared to the older ones. Higher formaldehyde levels were also measured in English homes built in the 90s than in older ones (Raw et al. 2004). High formaldehyde concentrations were detected in homes with particle board flooring and the concentration was significantly higher in autumn than in winter.

*Table 1. Summary of indoor air investigations in established dwellings (older than two years, *measurements from pre-selected dwellings, kindergartens, offices and schools).*

Country	**Number of dwellings**	**Sampling and quantification**	**Indoor air concentration (µg/m³)**			**Reference**
			TVOC	**Target VOCs**	**Formaldehyde**	
Canada	757	passive sampling on charcoal pads, 52 target VOCs	not measured	40 VOCs < 1–40 µg/m³, 12 VOCs < 1 µg/m³	not measured	Otson et al. 1994
Finland	50	active sampling on Tenax TA, 48 target VOCs	sum of 48 target VOCs: 121 µg/m³ (median)	toluene 20 µg/m³ limonene 9 µg/m³ α-pinene 8 µg/m³ xylene 6 µg/m³ hexanal 5 µg/m³ other VOCs < 5 µg/m³ benzene 3 µg/m³ (median)	not measured	Kostiainen 1995
Germany*	> 580	active sampling on Anasorb 747 TVOC: FID area between hexane and hexadecane	230 µg/m³ (median)	toluene 21 µg/m³ butanol 15 µg/m³ hexanal 12 µg/m³ xylene 9 µg/m³ (median)	not measured	Scholz and Santl 1999

Table 1. Continues...

Australia	27	Tenax TA/ Ambersorb XE 340/ activated charcoal (VOCs) chromotropic acid (formaldehyde) TVOC: FID area after 5 min (inc. acetone, ethanol, C_5- alkanes)- heptadecane	170 $\mu g/m^3$ (GM) < 300 $\mu g/m^3$ (75th percentile)	toluene 10 $\mu g/m^3$ ethanol 7 $\mu g/m^3$ nonanal 7 $\mu g/m^3$ acetone 6 $\mu g/m^3$ others < 1–5 $\mu g/m^3$ (GM)	not measured	Brown 2002
USA	150	active sampling on multisorbent tubes	not measured	1–36 $\mu g/m^3$ (mean, occurrence ratios larger than 0.06)	not measured	Zhang et al. 2003
Greece	50	active sampling on Tenax TA and active charcoal (Basel) microenvironment monitors were used	sum of VOC groups: 426 $\mu g/m^3$ (mean)	toluene 12–60 $\mu g/m^3$ benzene 2–11 $\mu g/m^3$ naphthalene 1–22 $\mu g/m^3$ (GM)	not measured	Saarela et al. 2003 (EXPOLIS-study)
Switzerland	50		sum of VOC groups: 109 $\mu g/m^3$ (mean)		not measured	
England	50		sum of VOC groups: 184 $\mu g/m^3$ (mean)		not measured	
Italy	50		sum of VOC groups: 283 $\mu g/m^3$ (mean)		not measured	

Table 1. Continues...

The Czech Republic	50		sum of VOC groups: 283 µg/m^3 (mean)		not measured	
Finland	201		sum of VOC groups: 169 µg/m^3 (mean) sum of 30 target VOCs: 150 µg/m^3	limonene 32 µg/m^3 toluene 20 µg/m^3 α-pinene 16 µg/m^3 hexanal 12 µg/m^3 butanol 12 µg/m^3 benzene 2 µg/m^3 other VOCs < 10 µg/m^3	not measured	Saarela et al. 2003 (EXPOLIS-study) Edwards et al. 2001
France	62/ 88 (VOCs/ aldehydes)	passive sampling on Carbograph adsorbents (VOCs) DNPH (aldehydes)	30 target VOCs	highest VOCs: hexanal 17 µg/m^3 toluene 16 µg/m^3 limonene 13 µg/m^3	21 µg/m^3 (GM) range 2–75 µg/m^3	Kirchner et al. 2003
Sweden	178	passive sampling on Tenax TA	350 µg/m^3 (mean), range 85–1050 µg/m^3	80% of the identified 120 VOCs: mean concentration < 10 µg/m^3	12 µg/m^3 (mean), range 0–62 µg/m^3	Bornehag and Stridh 2003
Sweden	27	triethanolamine passive gas tubes (formaldehyde), activated charcoal diffusion sampler (chlorinated VOCs)	not measured	chlorinated VOCs: 0.03–1.03 µg/m^3 (GM)	8 µg/m^3 (GM)	Sakai et al. 2004
Japan	37		not measured	chlorinated VOCs: 0.4–40 µg/m^3 (GM)	18 µg/m^3 (GM)	Sakai et al. 2004

Table 1. Continues...

England	876	passive sampling on Tenax TA (VOCs) TVOC: FID area between hexane and hexadecane	210 μg/m^3 (GM) 1010 μg/m^3 (95th percentile)	benzene 3 μg/m^3 toluene 15 μg/m^3 m/p-xylene 4 μg/m^3 limonene 6 μg/m^3 undecane 3 μg/m^3 Texanol 5 μg/m^3 TXIB 2 μg/m^3 (GM)	22 μg/m^3 (GM) 61 μg/m^3 (95th percentile)	Raw et al. 2004
Germany	2013	passive sampling on charcoal pads	sum of 30 target VOCs: 188 μg/m^3 (mean) 506 μg/m^3 (95th percentile)	toluene 30 μg/m^3 a-pinene23 μg/m^3 limonene 33 μg/m^3 (mean) other VOCs 0.5–10 μg/m^3	not measured	Schlink et al. 2004
Japan	25	passive sampling on activated charcoal (VOCs) DNPH (carbonyl compounds)	not measured	single VOCs < 1–3 μg/m^3 in general (GM) except: toluene 12 μg/m^3, formaldehyde 19 μg/m^3, acetaldehyde 9 μg/m^3 and p-dichlorobenzene 41 μg/m^3	19 μg/m^3 (GM)	Ohura et al. 2006

The VOC concentration levels in newly established or renovated buildings are generally several mg/m^3 (Table 2). Formaldehyde levels up to 400 µg/m^3 have been measured after completion. Levels of formaldehyde and alpha-pinene were higher in the newly built wooden framed houses than in the other houses in Japan. The reduction in VOCs is fastest during the first months but full stabilisation of levels may take two years (Park and Ikeda 2006).

The most effective strategy for controlling IAQ is pollution source control and the design of ventilation rates to handle uncontrollable sources (Guo et al. 2003). The study by Tuomainen et al., (2001, 2003) showed that the TVOC concentration in the study building, where the Finnish Classification was applied throughout the building process, was one tenth of the concentration in the reference building, which was built with conventional building materials.

Table 2. Summary of indoor air investigations in new dwellings (less than two years).

Country	**Number of dwellings**	**Sampling and analysis**	**Indoor air concentration (µg/m^3)**			**Reference**
			TVOC	**VOCs**	**formaldehyde**	
Denmark	2	diffusive samplers (21 VOCs) active sampling on water (formaldehyde)	sum of 21 VOCs 0–50 days: 4–5 mg/m^3 to 1 mg/m^3	0–15 months (mean): 6–487 µg/m^3 (occupied) 2–754 µg/m^3 (vacant)	up to 100 µg/m^3 (occupied) up to 400 µg/m^3 (vacant)	Wolkoff et al. 1991
Review	10 studies		4 500 µg/m^3 (weighted average GM)	3– 900 µg/m^3 (weighted average GM)	not reviewed	Brown et al. 1994
USA	11	active sampling on Tenax TA (52 VOCs), DNPH (aldehydes) TVOC: MS TIC area between heptane-heptadecane	1.5–2.7 mg/m^3	48 VOCs: 0.5 ppb – 15 ppb a-pinene, ethylene glycol, hexanal: > 15–50 ppb acetic acid: 80 ppb	49 µg/m^3 (GM)	Hodgson et al. 2000
Australia	1	Tenax TA/ Ambersorb XE 340/ activated charcoal chromotropic acid (formaldehyde) TVOC: FID area after 5 min (inc. acetone, ethanol, C_5- alkanes)-heptadecane	2 days – 35 weeks: 3000–200 µg/m^3	After 35 weeks: 23 VOCs: < 0.5–9.3 µg/m^3 methanol: 130 µg/m^3 ethanol: 120 µg/m^3 acetone: 29 µg/m^3	2 days – 35 weeks: 120–46 µg/m^3	Brown 2002

Table 2. Continues...

Australia	1	active sampling on Tenax GR TVOC: total FID area	3 months/ 5 months/ one year: 43/28/ < 5 µg/m^3	not measured	not measured	Guo et al. 2003
Japan		passive sampling (24 hours) on DNPH (7 aldehydes) and charcoal (18 VOCs)	sum of 18 VOCs 1–3 years: 328–166 µg/m^3	After 1 year: VOCs: 3–269 µg/m^3 aldehydes: 1–8 µg/m^3	1–3 years: 134 µg/m^3 to 86 µg/m^3	Park and Ikeda 2006
Switzerland	19	no information	not measured	not measured	44 µg/m^3 (mean) 103 µg/m^3 (95th perc.)	Thalmann and Coutalides 2006
Finland	12 + 12 (case + control building)	active sampling on Tenax TA (VOCs), DNPH (aldehydes) TVOC: FID area between hexane-hexadecane	0–5 months: 911–153 µg/m^3 (case) 4082–734 µg/m^3 (control) 1–3 years: 70–28 µg/m^3 (case) 931–265 µg/m^3 (control)	Most frequent VOCs after 1 year (case build.): limonene: < 1–37 µg/m^3 3-carene: < 1–16 µg/m^3 α-pinene: 4–11 µg/m^3 toluene: < 1–13 µg/m^3 nonanal: 2–8 µg/m^3 xylene: < 1–4 µg/m^3	0–5 months: 19–13 µg/m^3 (case) 13–18 µg/m^3 (control) 1–3 years: 12–16 µg/m^3 (case) 17–23 µg/m^3 (control)	Tuomainen et al. 2001, 2003

2.2.2 Seasonal trends

Seasonal trends of VOC concentrations were investigated by passive sampling during one year in twelve German homes. The total amounts of VOCs were about two to three times higher in the cold season than in the warm season. Elevated concentrations (above 20 μg/m^3) of ethyl acetate, 1,1,1- trichloroethane, and limonene could be assigned to indoor sources like adhesives, felt markers, correction fluids and cleaning agents. About one-half of the VOC concentrations could be assumed to be generated by the occupants or their activities. Benzene levels were similar in all homes and lower in summer time. Thus they were concluded to originate mainly from outdoor air (Seifert et al. 1989).

Similar results were gained in later German studies. Higher concentrations of target VOCs were measured in winter than in summer in Leipzig (Rehwagen et al. 2003). The sum of 30 VOCs varied between ~100–500 μg/m^3 in 1,499 indoor measurements in randomly selected apartments between 1994 and 2001. Similar seasonal patterns were also observed in Munich and Cologne (Schlink et al. 2004).

Ohura et al. (2006) also found higher concentrations of aromatic hydrocarbons and volatile organic halogenated compounds (except for *p*-dichlorobenzene) in winter than in summer in Shimizu, an industrial city in Japan. This was due to keeping the windows closed in the winter. On the other hand, concentrations of some aldehydes, such as formaldehyde and valeraldehyde, tended to be higher in summer in both indoor and outdoor samples. The increases were attributed to volatilization from emission sources as temperatures were higher. Higher formaldehyde concentrations were measured in autumn in English homes (Raw et al. 2004).

Formaldehyde concentrations were also higher from spring to autumn in Danish apartments during the first year after completion. The concentrations in vacant apartments were up to 400 μg/m^3 during the warm season. The concentration of some alkanes, terpenes, butanol, and butyl acetate were higher in the warmer period. A minor increase of Texanol (2, 4 ,4- trimethyl-1, 3-pentanediol monoisobutyrate) was also measured in a vacant apartment during the warm season (Wolkoff et al. 1991a).

Reponen et al. (1991) also measured considerable seasonal variations for formaldehyde concentrations in three newly built houses in Finland. The highest levels of up to 140 $\mu g/m^3$ were measured in the autumn. The seasonal variation almost disappeared when the formaldehyde levels were adjusted for temperature and humidity.

An analysis of the data on ambient conditions and concentrations of 26 VOCs collected in 754 residences in Canada during one year showed that only 13% of the variation of individual VOC concentrations can be directly explained by environmental factors such as outdoor temperature, differential temperature or relative humidity (Fellin and Otson 1993, 1994).

2.3 Building materials as sources of indoor air pollutants

2.3.1 Indoor air equation and emissions from structures

The indoor air concentration is determined by the (1) outdoor pollutant concentration, (2) ventilation rate, (3) indoor source strength, (4) inside mixing conditions, and (5) pollutant decay rate.

In general, the emission from building materials is composed of several rate-dependent steps like (1) organic vapour production, (2) transport through the bulk phase, (3) transport through a physical barrier, and (4) diffusional transport from the building material into the room air. Factors that affect various steps include temperature, humidity, barrier thickness and permeability, face velocities, and room concentration of the pollutant. In addition, ageing or depletion of the source affects emissions over a period of time (Hawthorne and Matthews 1987). The rate of release of a compound is mainly determined by the rate of diffusion of the compound in the material and the air circulation in the boundary layer just above the surface of the material. Evaporation controls the emissions of liquids, e.g. paints, in the drying phase (Wolkoff 1995).

The specific emission rate (SER*)* is a function of concentration C (mg/m^3) and air exchange rate n (h^{-1}) divided by the ratio of a surface to the room volume, i.e. loading L (m^2/m^3), which can also be signed as the area specific air flow rate q (m^3/ h m^2) as follows (ISO 2006a):

$$SER = n/L * C = q * C \qquad (1)$$

In real buildings, the outdoor concentration is subtracted from the indoor concentration to form C. For the emission testing equipment (see chapter 2.3.3) the outdoor concentration is zero, since high purity air is used.

The impact of emissions from interior surfaces on the indoor air concentrations can be estimated by means of a standard room. The standard room is the smallest room allowed ($V = 17.4\ m^3$) and is considered as the "worst case" since indoor air concentrations increase with increasing surface/volume ratio.

The standard room dimensions are defined in Table 3 (Mølhave 1982, ISO 2006a). The area-specific flow rates, when the air exchange rate is $0.5h^{-1}$, are shown in the same table.

Table 3. Standard room dimensions, loadings, and area specific flow rates (with $n = 0.5h^{-1}$; Mølhave 1982, Danish Standards Association 1994, ISO 2006a).*

Surface	**Area (m^2)**	**Loading (m^2/m^3)**	**Area specific air flow rate* (m^3/m^2h)**
Floor	7	0.41	1.3
Walls	24	1.41	0.4
Ceiling	7	0.41	1.3
Casing, frames, sills	4.2	0.24	2.1
Sealings, putty, etc.	0.2	0.012	44
Doors	2	0.1	4.5

2.3.2 The influence of temperature and RH on the emissions from building materials

No general model for describing the influence of temperature and humidity on emissions from building materials is feasible due to the chemical diversity of pollutants and building materials (Wolkoff 1998). The influence of the temperature and RH on emission rates has been studied most extensively for formaldehyde. The emission rate of formaldehyde measured from chipboard doubled on a 7 ºC increase or an increase from 30 to 70% RH at 22 ºC (Andersen et al. 1975). The formaldehyde emissions from pressed wood increased 12-fold when the temperature rose from 20 ºC to 36 ºC and the RH from 25% to 75% (Hawthorne and Matthews 1987). The increase of formaldehyde emission rate from new wooden materials (plywood, particle board) and ceiling tiles with increasing humidity was confirmed by van Netten et al. (1989). An opposite trend was observed for new gypsum board.

The emission of styrene and alkyl benzenes from a textile floor covering with styrene-butadiene backing increased very little when the temperature rose from 23 ºC to 71 ºC. On the contrary, benzothiazole, a less volatile and more polar compound, increased more than four-fold for the same temperature increase. The equilibrium concentrations of 15 different compounds were not affected by a change in the humidity from 0% to 45% except for aniline, for which the emission increased slightly with increasing humidity (Sollinger et al. 1994).

The initial emission rate and the source decay constant were found to increase by temperature for higher alkanes, 2-ethylhexanol and 4-phenylhexene measured from PVC, carpets or paint. The concentrations increased two- to three-fold over several weeks by increasing the temperature from 23 ºC to 30 ºC (van der Wal et al. 1997).

The emission of 1,2-propandiol, 2-ethylhexanol and dimethyloctanol increased when the RH increased to 50%. Plasticizers appeared to decompose at the high RH. Texanol emissions, however, were not affected by the RH.

Emissions of VOCs from varnish and paint were studied for different temperatures ranging from 15 ºC to 35 ºC at the RH of 3% and RHs ranging from 3% to 62% at 25 ºC. The polyurethane plastic finish varnish was tested at 1.2 h^{-1} with a loading of 0.33 m^2/m^3. The emission rate peaks appeared within 0–2 hours and were near depletion by the sixth hour. The TVOC emissions were higher at higher temperature and RH. However, the single VOCs sometimes showed deviating behaviour. Water-based gloss acrylic paint was tested at the same conditions with a loading of 0.8 m^2/m^3. The TVOC and VOC emissions increased at higher temperatures. However, due to humidity originating from the drying paint fluctuations in emission rates were observed at the beginning of the test (< 12 hours). The highest emission rates were measured for the RH 32% and fluctuations were seen up to 100 days. The impact of the RH on emissions of individual VOCs varied (Haghighat and De Bellis 1998).

The emission rate of di- (2-ethylhexyl) phthalate (DEHP) was not found to be influenced by the RH from 10% to 70% (Clausen et al. 2004). Recently, Clausen et al. (2007) published a physically-based emission model for SVOCs that was consistent with the experimental data and independent of the RH.

2.3.3 Emission testing methods and standards

The environmental chamber technique for the determination of VOC emissions was introduced in the 1980s and has been standardized in Europe (European collaborative action 1997, ISO 2006a & b). A corresponding method exists in the US (ASTM 1997). The emission test consists of sample preparation, sampling and chemical analysis. The environmental chamber parameters to be controlled include temperature, RH and air exchange rate. In addition, the air tightness of the chamber, air mixing (i.e. air velocity above the sample) and recovery of target compounds are defined in the standards. The most important factors that influence the emission rate are, however, the prehistory and the preconditioning of the material specimen prior to the sampling (Wolkoff 1995). When the impacts of air velocity, temperature, humidity and air (vs. nitrogen) on the emission from five different building products were studied, primary emissions appeared not to be affected by the air velocity after 2–4 days (Wolkoff 1998).

Environmental chamber sizes vary from a few litres to “walk-in” type room-size large-scale chambers. The use of small-scale environmental chambers has spread extensively for the determination of VOC emissions from building products (Wensing 1999).

The portable micro chamber field and laboratory emission cell (FLEC) was developed for identifying emission sources on-site (Figure 1, Wolkoff et al. 1991b). The FLEC method has been included in the emission standard (ISO 2006b). The FLEC is supplied with clean and humified air from an air supply control unit. Realistic room air velocities and specific ventilation rates (l/m^2s) are achieved for FLEC (Wolkoff 1995). Table 4 summarizes the specifications for the 1 m^3 environmental chamber and FLEC.

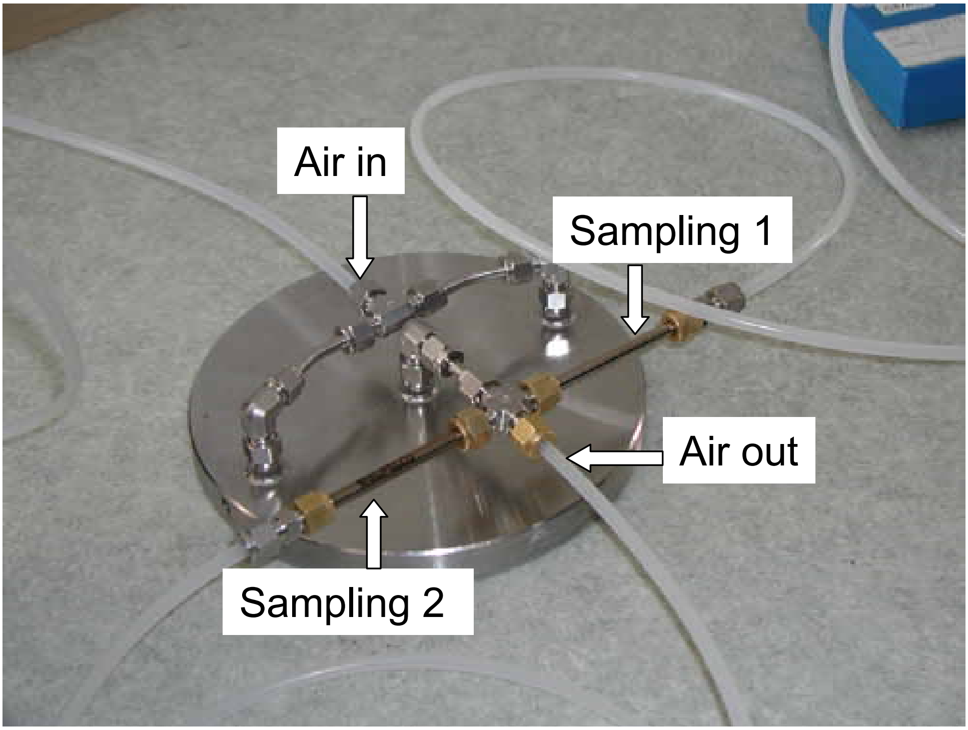

Figure 1. The field and laboratory emission cell (FLEC).

The performance (i.e. recovery, repeatability, influence of air velocity, temperature and humidity, comparison with the environmental chamber) of the FLEC has been evaluated in several studies. Roache et al. (1996) found excellent repeatability for parallel sampling with FLEC for liquid floor wax and latex paint. The emission results for floor wax applied to glass were also comparable with those measured in a small chamber. Inconsistency in the impact of air velocity was found for glycols. Afshari et al. (2003) also concluded from the emission tests with paints that the environmental chamber of $1m^3$ and FLEC gave similar results for the VOCs (aldehydes, alcohols) under the same temperature, RH, and airflow/ area conditions. A review of comparison studies between FLEC and chamber measurements concluded that the observed differences were mostly $\leq 15\%$. Higher differences were assigned to edge effects, sorption on walls, inhomogenity of the building products and analytical errors (Wolkoff et al. 2005).

Both the qualitative flow theory model and visualization showed that the air flow is not evenly distributed over the test surface in the FLEC. The variation in air velocity has an effect on measuring point sources, which appear as special cases (such as encapsulated solvent residues). However, for test specimens with homogenous surfaces the effect will average out (Uhde et al. 1998). The flow field of the FLEC was found laminar by three-dimensional computational fluid dynamics. In the case of internal diffusion material, the local VOC emission rate becomes uniform and the FLEC works well. However, in the case of evaporation type material or evaporation and diffusion controlled material, the FLEC is not suitable for testing because of the thin FLEC cavity. In these cases the concentration gradient to the surface is lower than normal, which lowers the emission rates (Zhu et al. 2007).

Table 4. Specifications for materials emission test chambers (ISO 2006a, b, Wolkoff 1995a).

Specification	**Environmental chamber**	**FLEC**
Volume, m^3	1	3.5×10^{-5}
Floor area, m^2	0.41	0.0177
Loading, m^2/m^3	0.41 (floor, ceiling) 1.41 (walls)	507
Air supply, l/min	8.3	0.3
Air exchange rate, h^{-1}	0.5	514
Air velocity, m/s	0.005	0.01
Area specific air flow rate, l/s $m^2/m^3/m^2$ h	0.1–0.3/0.4–1.1	0.3/1.1

2.3.4 Emissions measured from building materials

Emissions from building materials have been measured extensively since the 1970s. Table 5 summarizes the pollutants detected in common building materials (Wolkoff 1995). Emissions from PVC materials and adhesives are reviewed more thoroughly in this chapter.

Table 5. VOCs measured from common building materials (Wolkoff 1995).

Building material	**VOC compound/ compound group**
PVC floor covering	Alkanes, aromatic compounds, 2-ethylhexanol, TXIB
Parquet (wooden)	C_5–C_6-aldehydes, terpenes
Linoleum	C_5–C_{11}-aldehydes, aliphatic acids, benzaldehyde
Rubber carpet	Acetophenone, alkylated aromatic compounds, styrene
Adhesive	C_9–C_{11}-alkanes, toluene, styrene
Lacquer	Alkanes, aldehydes
Paint	Alkanes, glycols, glycolesters, Texanol
Sealant	Ketones, esters, glycols, polychlorinated biphenyls, siloxane
Particle board	Alkanes, aldehydes, ketones, butanol, formaldehyde

Girman et al. (1986) studied emissions from 15 different solvent and water-based adhesives after seven days of drying. Eight of the adhesives emitted VOCs at 0.1–700 µg/g h. Toluene was the most abundant VOC at 0.6–62 µg/g h. Other common VOCs were styrene and alkanes. Seifert et al. (1989) identified toluene, xylenes and ethylbenzene as the main VOCs determined by static head-space analysis of two water-based, acrylate copolymer adhesives.

A wide range of VOCs are emitted from vinyl/PVC floorings. These include plasticizers, solvent residues and their secondary reaction products. In the study by Yu and Crump (1998), TVOC emission rates varied from ca. 100 $\mu g/m^2h$ to more than 20,000 $\mu g/m^2h$ for less-than-two-year-old materials. Alkanes, aromatics, alcohols, ketones and esters were detected. Wall and floor adhesives emitted VOCs up to ~300 mg/m^2h tested after 24 hours

A set of PVC flooring materials produced in 1991–1994 (50 products) were investigated by Lundgren et al. (1999). The VOC emissions were determined for the 4-week-old and 26-week-old products. The median TVOC emissions were ~200–300 $\mu g/m^2h$ for the 4-week-old products and ~100 $\mu g/m^2h$ for the 26-week-old products. The main VOCs were 2-butoxyethanol, 2-(2-butoxyethoxy) ethanol, phenol, hepta- and octadecanes and trimethylbenzene. Reduction factors for VOCs were calculated as the difference between the 4-week and 26-week emissions divided by the 4-week emission. Reduction factors of less than 0.5 were found for 10 VOCs, including 2,2,4-trimethyl-1,3-pentanediol di-isobutyrate (TXIB, 0.17). These compounds may play a significant role in the indoor environment over a longer period (Lundgren et al. 1999).

Phthalates (SVOCs) have been measured abundantly from PVC materials. The emissions of phthalate esters were studied from six PVC-coated wall coverings in 1 m^3 glass chambers for a period of 14 days (Uhde et al. 2001). A fogging apparatus, i.e. a 15 x 15 cm stainless steel plate kept at 15 ºC, was mounted in each chamber. The chamber was heated for 48 hours at 70 ºC. Di-butylphthalate (DBP), di-ethylhexylphthalate (DEHP), di-iso- butylphthalate (DIBP) and di-pentylphthalate (DPP) were detected in the chamber air. The maximum concentrations were 0.5–5.1 $\mu g/m^3$ and they remained on an almost constant level during the measurement period. Maximum fogging values of 17.7 µg and 60.4 µg were measured for DPP and DEHP respectively.

The emission of DEHP from a PVC was studied for 472 days. A steady state emission was reached in 150 days. The chamber concentration was ~1 $\mu g/m^3$. The concentrations were also found proportional to the loadings. The emission of DEHP was, therefore, concluded to be limited by diffusion in the boundary layer. A dust layer was found to increase the emission rate because of uptake of DEHP: about four times as much DEHP was measured from the dust particles over a 68-day period than was emitted in the gas phase experiments without dust (Clausen et al. 2004).

The emission of phthalates from PVC flooring, polyolefine flooring, a refrigerator list, two electric cables, PVC skirting and floor wax was studied with a Climpaq and FLEC for a period of 6 to 250 days (Afshari et al. 2004). The emission of DEHP resulted in a maximum concentration of 1 $\mu g/m^3$ for all materials except for polyolefine covered with wax, which did not emit this compound. Instead, DBP was detected at 22 $\mu g/m^3$ from this material. A quasistatic equilibrium for DBP was reached after 60 days. The concentration was found to be almost independent of chamber and ventilation rate.

Wilke et al. (2004) studied VOC and SVOC emissions from low-emitting adhesives and PVCs as well as from complete structures. The TVOC emission rates from nine adhesives ranged between 900–10 000 µg/m^{2}h after 24 hours. The emission rates decreased to below 200 µg/m^{2}h in 28 days for eight adhesives; 2-ethylhexanol, acetic acid and phenoxy propanol were the most abundant compounds. At this point the SVOC emission rates were higher than the TVOC emission rates for three adhesives. The TVOC emission rate from the single PVCs (five products) measured at 28 days old varied between < 100 µg/m^{2}h and 1000 µg/m^{2}h. The main compounds were TXIB and alkylbenzenes (SVOCs).

2.3.5 On-site measured emissions and source identification

The emission of VOCs from a material is affected by the substrate and its surroundings as well as the material combinations in a real structure (Wolkoff 1998, Guo et al. 2003, Wilke et al. 2004, Wirtanen 2005). Secondary emissions can develop under the influence of humidity, ozone, UV light, etc. Weschler and Schields 1997, Weschler 2000). Hydrolysis reactions in the floor structure (PVC/adhesive/casein containing levelling agents) may produce 2-ethylhexanol, butanol and ammonia (Karlsson et al. 1989, Gustafsson 1990, Bornehag 1991). Low-alkaline levelling agents have been shown to protect against these reactions (Alexanderson 2004). Thus the on-site measured emission can differ from the emission measured from single materials under laboratory conditions.

There are few investigations of on-site measured emissions. The published results are usually based on measurements with the FLEC. Must (1996) measured VOCs from vinyl flooring with adhesive and parquet flooring without adhesive. The emission from concrete under the flooring was measured with the adhesive, after the adhesive was rubbed off and after airing for 3, 7 and 28 days. The TVOC level measured from the adhesive was up to 40, 000 µg/m^3. The TVOC decreased to below 6,000 µg/m^3 three days after the adhesive had been scrubbed off and the structure had been aired, and finally to ~400 µg/m^3 in 28 days. The TVOC measured under the parquet was clearly lower, below 200 µg/m^3 (Must 1996).

The emission rates from floor structures were determined in a newly built office building (Saarela et al. 2000). The TVOC emission rates measured from two concrete structures with low-emitting vinyl flooring increased from ~150–

400 μg/m^{2}h (4-week-old structure) to ~450–650 μg/m^{2}h during the first 13 weeks after installation. The emissions decreased to a level of ~100 μg/m^{2}h in 21 weeks.

Evaluation of emission sources in real buildings has also been done by comparing emissions from building products measured in test chambers with real room concentrations in a study by Zellweger et al. (1995). The expected room concentrations were calculated with the measured air exchange rate. The predicted levels of cyclohexane and hexanal were more than one order of magnitude higher than those measured in the indoor air. Application (i.e. substrate) and sink effects were possible explanations for the difference. A good agreement between the predicted and measured concentrations was found for 1-methyl-2-pyrrolidinone from parquet lacquer applied on wood and room concentration. On the contrary, the predicted concentration from parquet lacquer applied on glass was much higher than measured.

Cabinetry materials, passage doors, and plywood subfloor were the predominated sources of formaldehyde and other aldehydes in new manufactured houses in the US. This was concluded from chamber emission measurements performed with single materials. Terpenes emitted from the plywood subfloor (Hodgson et al. 2002).

Wilke et al. (2004) studied complete structures under laboratory conditions. Three materials, a PVC, linoleum and carpet, were studied in combination with the same adhesive, screed primer and concrete. The total emissions were lower for the tested compositions than the sum of total emissions from the single components.

2.3.6 Emission labels

Today, there are several labels for tested, low-emitting materials, including the German AgBB, RAL UZ (Blue Angel), GUT, Emicode, the Danish ICL, and the Finnish Classification of Indoor Air Climate (Wolkoff 2003, European collaborative action 2005). The harmonization work on the European level is still ongoing (European collaborative action 2005).

The German AgBB scheme evaluates emissions of VOCs and SVOCs from building products at three and 28 days. In addition, the lowest concentrations of interest values (LCI) are listed for selected VOCs and the sum of the concentrations divided with this value should not exceed one (AgBB 2006).

The Blue Angel product labelling was created in 1977 in Germany to promote products with better environmental and health characteristics than conventional products. Today, it includes 11 criteria documents (RAL-UZ) for construction products, furniture and electronic devices which limits emissions of VOCs and SVOCs. Emissions of Blue Angel products remain well or very well below the AgBB requirements (www.blauer-angel.com, European collaborative action 2005).

The AgBB scheme for the evaluation of VOC emissions is also adopted by the European GUT label for carpets since 2004. However, lower concentration criteria are used for TVOC and SVOC compared to the AgBB. The GUT members cover more than 80% of the European carpet production volume (www.gut-ev.de, European collaborative action 2005).

The Emicode system by German Adhesives Manufacturers (GEV) for adhesives and related material monitor substances with proven or suspected carcinogenic affects and TVOC after 1–10 days (www.emicode.com, European collaborative action 2005).

The Danish Indoor Climate labelling has a different approach. VOC concentrations need to decay below indoor relevant levels, which are defined using available toxicological information and odour thresholds. VOCs, including aldehydes, amines, isocyanates, acids and compounds containing sulphur, are at least determined. The results are given as a single indoor-relevant time-value, which is determined by converting the measured chamber concentrations to concentrations in a standard room and comparing them with odour thresholds and mucous membrane irritation thresholds; the odour and threshold values are given in a databank (“VOCBASE”) (Danish Standards Association 1994).

The Finnish Climate Classification, developed by the Finnish Society of Indoor Air Quality and Climate (FiSIAQ), defines the design and target values for thermal comfort, ventilation rates, odour intensity, noise levels, concentration of indoor air pollutants and material emissions, as well as procedures for construction work in new buildings (S, M and P classes). The classification defines three classes for IAQ: S1, S2, and S3. The S1 class corresponds to the best IAQ. The target values are defined for concentration of TVOC, formaldehyde and ammonia, as well as for temperature, humidity and air velocity. The target values for the emission of VOCs, formaldehyde, ammonia and carcinogens, and the odour intensity level are established for building materials at 28 days. The products should also be casein-free (FiSIAQ 2001). Although the classification is voluntary, the use of M1-classified, low-emitting materials has markedly increased and probably improved the IAQ in new Finnish buildings. To date, there are over 1,000 M1-classified building products on the market (The Finnish Building Information Foundation 2007).

3. Aims of the present study

The general aim of the current study was to create a database of the indoor air concentrations and structure emissions for VOCs, formaldehyde and ammonia in newly finished residential buildings.

The specific aims were:

- to determine reference values for structure emissions and indoor air concentrations for VOCs, formaldehyde and ammonia measured *on-site* (I, II)
- to investigate which parameters affect emission rates and indoor concentration levels. The investigated parameters were building materials, structure types and ventilation systems, as well as time of construction, season, climatic factors (temperature, RH, air exchange rate), ventilation system operation time (in the newly finished building) and time point from last construction work prior to use, (I, II)
- to investigate the emissions and their impact on indoor air quality during the first year after the building has been taken into use (III)
- to investigate the difference between material emissions measured *on-site* and in the laboratory (IV)
- to investigate how the Finnish material classification system (FiSIAQ 2001) works in practice in achieving good IAQ in real buildings, and to give suggestions for the further development of the material classification.

4. Methods

4.1 Study buildings

Fourteen apartments in eight residential buildings (seven apartment buildings and one two-family house), built according to the current Finnish building regulation code, were investigated. Seven of the buildings were located in Helsinki and one in Turku, both cities being located on the coast of Gulf of Finland. The buildings were built by three different construction companies. The buildings were chosen in order to represent their normal building practice. Also, the time point of construction was chosen so that the buildings were finished both during the summer and winter periods. The construction times varied between 12–15 months. In three buildings, 2–4 different apartments were measured because they had different floor covering materials. The description of building structures, finishing materials, the ventilation system and its operation time before occupation, surface areas and the time of building are summarized in publications I (Table 1), II (Table 1) and III (Table 1). Low-emitting, M1-classified materials were used in all the buildings – that is, the laboratory tests performed for 4-week-old samples have given TVOC, ammonia and formaldehyde emissions lower than 200, 30, and 50 $\mu g/m^2h$ respectively (FiSIAQ 2001). The RH of the structure was measured to be < 85% before the floor covering was installed. In seven buildings the walls were finished with screed and painted. Wallpaper was laid on the screed in building 4. Ceilings were finished with screed (2 different products). The floors were finished with fine screed (dispersal 2–5 mm) in the site-built buildings and with gross screed (dispersal 10–30 mm) in the manufactured buildings. Different types of PVC materials and parquets were used as floor covering materials. The installation of the floor covering material was the last construction work occurring 2–28 weeks prior to occupation, except in building 4 where painting was performed 12 weeks prior to occupation. Supply air filters of grade EU7 were used in the buildings with a mechanical supply and exhaust air system, i.e. 80% of particles of size 1 µm are filtered (Ministry of the Environment 2003). The average surface-to-volume ratios for the structures were as follows: floor covering 0.39 m^2/m^3, ceiling 0.44 m^2/m^3 and walls 0.81 m^2/m^3 (doors, windows and closets excluded).

Figure 2 summarizes the measurement scheme carried out in buildings 1–7. In building 8, only indoor air concentrations were measured. In building 3, only the emissions from the floor structure were measured along with indoor air concentrations.

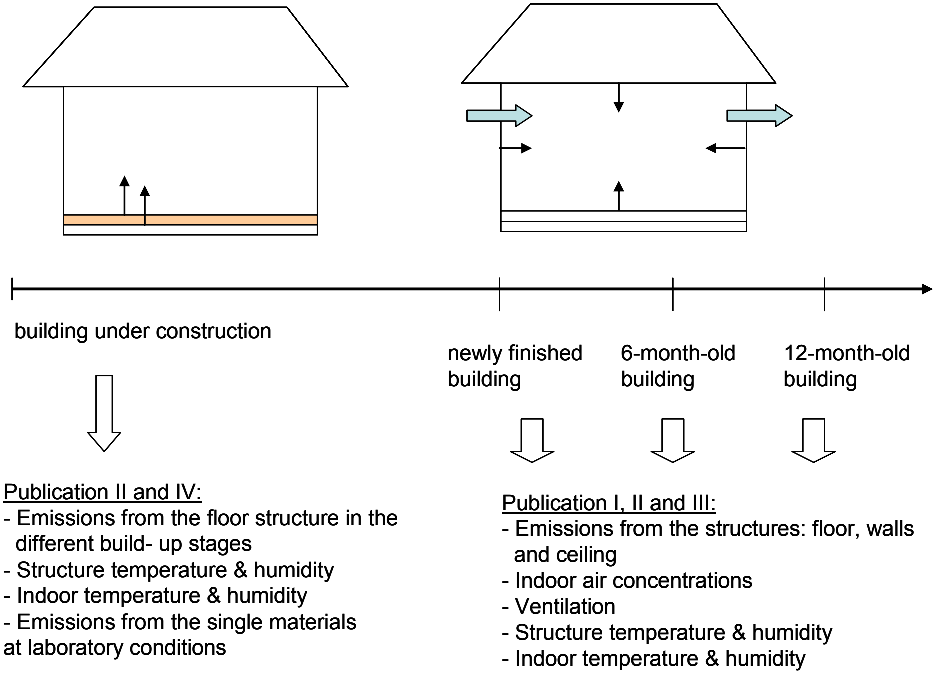

Figure 2. Set-up for the research.

4.2 Sampling and analysis

Indoor air samples were usually collected in the bedroom and before noon. VOC emission samples from the floor, walls and ceiling were collected 0–2 hours after sampling indoor air VOCs. Emission measurements of ammonia and formaldehyde lasted for up to six hours. Emissions were determined using the Field and laboratory emission cell (FLEC, ISO 2006b). VOCs were collected on Tenax TA adsorbent and analyzed with a gas chromatograph equipped with a mass selective detector (MSD, identification of VOCs) and a flame ionization detector (FID, quantification of VOCs). Sample volumes were 2–5 litres. TVOC was calculated from the FID chromatogram as the total integrated area between hexane and hexadecane and given as toluene equivalents (ISO 2004). The concentrations of SVOCs were calculated by summarizing the total integrated FID area after hexadecane and given as toluene equivalents. Background levels of diethyl phthalate (average amount of 5 ng/ tube) originating from the thermal desorption unit were subtracted from the results. Ammonia and formaldehyde samples were collected on an impinger (0.005 M sulphuric acid solution). Sample volumes were 100–300 litres (100 litres / 10 ml impinger). The ammonia concentration was determined with an ion selective electrode and the formaldehyde concentration with the spectrophotometrical acetyl acetone method. Indoor air humidity, temperature and exhaust air flows were measured simultaneously with indoor/ emission sampling. The details of the sampling and analysis procedures as well as the equipment used are described in Publications I and II. The experimental set up for the emission measurements of single PVCs (six different products) and adhesives (four different products) under laboratory conditions are described in Publication IV. Emission measurements were performed according to the emission test chamber standard (ISO 2006a). Analysis was performed as described above.

The limits of detection (LOD) for VOCs were determined by analyzing ~5 ng amounts of corresponding VOC standards. The LOD was determined by multiplying the noise level of the FID chromatogram with three (Christian 1994). The LOD for VOCs varied between 2–23 ng/ tube (0.5–5 $\mu g/m^3$ for a sampling volume of 5 litres, the higher LODs were for more polar compounds, e.g. organic acids). Non-detectable values were marked as LOD/2.

The ventilation rates were determined by measuring the total volumetric exhaust air flow rate. In the apartments provided with two-way mechanical ventilation, the supply air volumetric flow rates were also measured. Since all new buildings are required to be depressurised (Ministry of the Environment 2002), this ventilation rate was also assumed to correspond to the air exchange rate. This was verified to be true in all the apartments with combined mechanical supply and exhaust air ventilation in this study.

4.3 Quality assurance and validation of field sampling

Blank samples were treated in the same way as the real samples except no air was drawn through them. No breakthrough samples were collected; however, earlier laboratory work for accreditation purposes has shown that sample volume ranges were valid. No ozone scrubbers were used. The repeatability for on-site emission measurements (TVOC, formaldehyde, ammonia) was investigated by parallel sampling with two different FLECs. The parallel sampling was performed from the floor structure one and three days after the floor covering (parquet/ PVC) was removed in five different apartments. The relative standard deviation of parallel FLEC sampling was, on average, 9% (range 0.4–24%) for the TVOC emission and 10% (range 1–50%) for ammonia emission (Publication IV).

The conditioning time prior to VOC sampling was investigated by taking samples at four time points within 18–310 minutes after introducing clean air to the FLEC cell. The sampling was done from the uncovered floor structure (1 and 3 days after the removal of the floor covering material) on-site. The difference between the parallel samples decreased as the sampling time increased. A pre-conditioning time of 30 minutes was found appropriate for TVOC sampling (Publication IV).

The outdoor TVOC concentration near the air intake was 20 $\mu g/m^3$ in building 1, 4th floor. This site was considered to have the heaviest surrounding traffic and, additionally, it was located near a coal-fired power plant. The supply air TVOC/ ammonia/ formaldehyde concentrations at two measurement points in building 8 were 14–34 $\mu g/m^3$ / < 5 $\mu g/ m^3$ / < 10 $\mu g/ m^3$ respectively.

4.4 Statistical analysis

4.4.1 Principal component analysis (I)

Principal component analysis (PCA) was performed to reveal the main variables affecting IAQ. PCA is an orthogonal linear transformation that transforms the data to a new coordinate system. Thus the greatest variance by any projection of the data comes on the first coordinate (called the first principal component), the second greatest variance on the second coordinate, etc. The parameters investigated were floor covering (PVC/parquet), ventilation system (mechanical exhaust/ mechanical intake and exhaust) and the time of its operation (in the newly finished building), floor structure / levelling agent (on-site built+ fine screed/ manufactured+ gross screed), walls (paint/wallpaper), ceiling (two different products), time of construction and season (winter/summer, indoor air humidity), and time point from floor installation. The variables of TVOC, formaldehyde, ammonia and single VOC concentration, as well as the temperature, humidity, and air exchange rate, were included in the PCA model. Due to water damage in building 7, apartment 2, these results were excluded from the analysis. Models were calculated for the 0-, 6- and 12-month-old buildings. The automatic unit variance scaling was performed by the Simca software. The model details are presented in Table 6.

Table 6. PCA model details for the 0-, 6- and 12-month-old buildings (R^2 = explained variation, the range for R^2 is 0–1).

Model	**0-month-old building**	**6-month-old building**	**12-month-old building**
TVOC, ammonia, formaldehyde	13 measurements (apartments), R^2: 0.695 (2 components)	14 measurements (apartments) R^2: 0.812 (2 components)	14 measurements (apartments) R^2: 0.799 (2 components)
VOCs	VOCs occurring at $n \geq 3$ sites (76/238 compounds) R^2: 0.631 (3 components)	VOCs occurring at $n \geq 4$ sites (56/167 compounds) R^2: 0.576 (3 components)	VOCs occurring at $n \geq 4$ sites (61/189 compounds) R^2: 0.576 (3 components)

Factor rotations in the factor analysis are done in order to simplify the factor structure and make the interpretation easier and more reliable. Varimax rotation searches for a linear combination of the original factors so that variance of the loadings is maximized. High correlation (loadings) between compounds in the different sampling locations means that they have similar sources (Abdi 2003). Varimax rotation was performed for 30 target VOCs in the 12-month-old buildings. PCA was performed with SPSS 12.0.1 (Varimax rotation) and SIMCA-P 7.01 for Windows software.

4.4.2 Descriptives, correlations and tests

Statistical analysis and tests were performed with SPSS 12.0.1 for Windows software. Descriptives – i.e., mean, maximum, minimum, standard deviation, 95th percentiles – were summarized and the normal distribution of the data was analyzed with the Shapiro-Wilks W test. The null hypothesis of a normal distribution was rejected if the calculated W statistic was less than $W_{0.05}$. The data were also tested for lognormality.

Perason's correlations were use to study relationships between airborne concentrations/ emissions and temperature, humidity and volumetric exhaust air flow rates.

The non-parametric Mann-Whitney U test (at the significance level of $p < 0.05$) was used to clarify the statistical significance of the variables for the concentrations of TVOC, VOC groups, VOCs, formaldehyde and ammonia. Variables tested were ventilation system, floor covering material and structure. The apartments with a mechanical supply and exhaust air system were excluded in the tests with floor covering material and structure.

5. Results

5.1 Reference values for on-site structure emissions and indoor air concentrations (I, II)

The Shaphiro-Wilks W test suggested that the data fit quite well normal distributions. The W values were 0.77–0.93 for indoor air TVOC, VOC group, ammonia, and formaldehyde concentrations in both the 6- and 12-month-old buildings. The W value for SVOC concentrations was 0.83 in the 12-month-old building. The test statistic $W_{0.05}$ was not significant for w-values above ~0.86 (I). Because any of the test statistics were not clearly significant at $p = 0.05$ which could reject the null hypothesis of normality, the data was assumed to be distributed normally. Tables 7–9 summarize the TVOC, ammonia, and formaldehyde concentrations/emissions in the measured apartments.

Thus, the reference values were defined as follows:

1) The normal value is the mean concentration or emission, and
2) The limit for abnormal concentration or emission is the mean plus two standard deviations = 95th percentile.

The reference values for TVOC, VOC groups, formaldehyde and ammonia for 6- and 12-month-old buildings (I, II) are summarized in Tables 10 and 11. The mean concentrations of SVOCs remained rather stable (8–9 $\mu g/m^3$) during the monitoring period. The 12-month-values for emissions represent the targets that can be achieved in new buildings after the initial drying phase.

*Table 7. Indoor air TVOC, ammonia (NH_3) and formaldehyde (HCHO) concentrations (µg/m^3) in the studied apartments (*not measured or no result, ☼ not included in statistical analysis).*

	0 month-old building (µg/m^3)			**6-month-old building (µg/m^3)**			**12-month-old building (µg/m^3)**		
Site	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO
1	1 068	56	*	449	58	18	523	84	50
2	865	56	*	174	43	22	204	57	52
3	1 007	55	*	396	22	16	475	92	47
4	730	30	37	1 013	63	42	384	64	*
5	690	22	16	703	46	45	422	31	51
6	1 300	52	17	173	46	22	472	57	36
7	335	45	20	13	41	17	105	46	20
8	311	64	13	306	77	17	144	36	8
9	755	38	19	550	49	32	333	30	15
10	2 106☼	26	20	212	47	28	170	22	14
11	617	26	17	148	30	11	255	31	14
12	352	43	19	95	23	7	99	37	17
13	372	62	15	112	20	10	104	27	14
14	*	*	*	141	22	11	129	27	14

*Table 8. TVOC, ammonia (NH_3) and formaldehyde (HCHO) specific emission rates (SER, µg/m^2h) from the floor and ceiling surfaces (*not measured or no result).*

Site	**Floor**			**Ceiling**		
Newly finished building SER µg/m^2h						
	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO
1 (parquet)	129	6	9	530	*	*
2 (PVC)	*	*	*	616	*	*
3 (PVC)	*	*	*	655	*	*
4 (PVC)	926		8	919	40	56
5 (parquet)	93	12	< 5	*	*	*
6 (PVC)	447	14	< 5	544	57	0
7 (parquet)	74	8	*	376	24	20
8 (parquet)	16	14	10	200	29	14
9 (PVC)	104	18	18	684	59	63
10 (PVC)	245	11	*	1338	63	96
6-month-old building SER µg/m^2h						
Site	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO
1 (parquet)	80	10	6	238	51	*
2 (PVC)	174	1	*	130	25	*
3 (PVC)	71	2	*	516	24	*
4 (PVC)	336	2	10	539	21	*
5 (parquet)	18	2	4	*	*	*
6 (PVC)	84	13	6	296	68	37
7 (parquet)	38	18	*	123	37	14
8 (parquet)	184	6	< 5	210	86	17
9 (PVC)	45	11	< 5	346	67	109
10 (PVC)	106	16	< 5	172	76	34
12 -month-old building SER µg/m^2h						
Site	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO
1 (parquet)	33	10	5	238	59	30
2 (PVC)	119	1	10	83	48	34
3 (PVC)	60	5	*	260	36	28
4 (PVC)	191	20	*	239	21	16
5 (parquet)	47	37	8	199	43	46
6 (PVC)	125	6	*	205	114	40
7 (parquet)	21	7	6	26	20	13
8 (parquet)	12	6	< 5	136	61	15
9 (PVC)	30	3	5	239	42	21
10 (PVC)	179	2	< 5	192	42	34

*Table 9. TVOC, ammonia (NH_3) and formaldehyde (HCHO) specific emission rates (SER, $\mu g/m^2h$) from the wall surfaces (*not measured or no result).*

Site	**Painted walls**								
	Newly finished building SER $\mu g/m^2h$			*6-month-old building SER $\mu g/m^2h$*			*12-month-old building SER $\mu g/m^2h$*		
	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO	TVOC	NH_3	HCHO
1 (gypsum)	230	*	*	50	8	*	50	9	8
2 (gypsum)	140	*	*	90	< 5	*	25	< 5	< 5
3 (concrete)	977	*	*	49	< 5	*	42	18	< 5
3 (gypsum)	200	*	*	*	*	*	62	6	< 5
4 (concrete)	248	9	7	123	15	5	32	11	< 5
4 (gypsum)	*	*	*	*	*	*	76	11	< 5
7 (concrete)	82	13	8	11	5	< 5	9	8	8
7 (gypsum)	100	8	7	10	7	5	39	7	8
8 (concrete)	14	6	< 5	66	20	< 5	19	19	< 5
8 (gypsum)	20	6	5	42	< 5	15	37	3	< 5
9 (concrete)	104	25	11	37	18	*	19	19	20
9 (gypsum)	90	26	9	31	6	37	37	3	< 5
10 (concrete	*	*	*	*	10	*	21	9	< 5

Table 10. Reference values for indoor air concentration in 6- and 12-month-old buildings (I).

Compound or compound group	**Normal value ($\mu g/m^3$)**		**Limit for abnormal concentration ($\mu g/m^3$)**	
	6 months	**One year**	**6 months**	**One year**
Aromatic hydrocarbons	50	30	80	40
Aliphatic hydrocarbons	25	25	35	35
Cycloalkane	5	10	10	15
Alcohol	25	35	35	50
Aldehyde	35	35	50	50
Ketone	5	10	10	15
Ester	15	15	20	20
Glycol/ glycolether	25	25	45	35
Terpene	70	70	110	110
Acid	5	10	10	20
TVOC	270	270	400	400
Formaldehyde	25	30	30	40
Ammonia	45	45	60	60

Table 11. Reference values for structure emissions in 12-month-old buildings (II).

	Specific emission rate µg/m^{2}h							
	PVC		**Parquet**		**Levelled ceiling**		**Painted wall**	
	(n = 6)		**(n = 4)**		**(n = 10)**		**(n = 9)**	
	normal value	abnormal value	normal value	abnormal value	normal value	abnormal value	normal value	abnormal value
Acid	10	15	5	10	10	15	5	10
Alcohol	15	25	5	10	25	35	5	10
Aldehyde	15	25	5	10	25	35	5	10
Aliphatic hydrocarbon	20	40	< 5	5	20	35	10	15
Aromatic hydrocarbon	25	65	< 5	5	10	15	< 5	5
Cycloalkane	< 5	5	< 5	5	< 5	5	< 5	5
Ester	15	30	5	10	10	15	5	10
Glycol/ glycolether	25	50	5	< 10	20	30	10	15
Ketone	10	20	5	10	5	10	< 5	5
Terpene	< 5	5	< 5	5	30	45	< 5	5
TVOC	120	170	30	45	180	230	40	50
Ammonia	15	25	< 5	5	50	70	10	15
Formaldehyde	5	10	5	10	30	40	10	15

5.2 Factors affecting indoor air concentrations and emissions (I, II, III)

The PCA analysis revealed the most important factors that affected the IAQ during the first year after the completion of the buildings. The results are summarized in Table 12. The lowest TVOC concentrations were observed in the newly finished buildings with a mechanical supply and exhaust air system. The same levelling agent (product 2) was used on the ceiling structure in these buildings and the RH was low, less than 40%. The floor covering was parquet and the walls were painted. The lowest TVOC, ammonia and formaldehyde concentrations in the 6-month-old buildings were measured in the building where product 2 was used on the ceiling surfaces and the RH was less than 30%. The clustering of VOCs also occurred in these apartments. The same was observed in the 12-month-old buildings, where the clustering of VOCs was clear according to the ventilation system (mechanical supply), the ceiling surface (product 2), and the indoor air humidity (< 40%) (I, Figures 1 and 2).

Table 12. Summary of PCA results in the 0–12-month-old buildings (I).

0-month-old building	**6-month-old building**	**12-month-old building**
Lowest TVOC concentration: Ventilation system: mechanical supply and exhaust air system, ceiling surface: product 2, walls: painted, indoor air RH: < 40% VOC classification (clustering): Ventilation system: mechanical supply, ceiling surface: product 2	Lowest TVOC, ammonia and formaldehyde concentration: ceiling surface: product 2, indoor air RH: < 30% VOC classification (clustering): ceiling surface: product 2, indoor air RH: < 40%	Lowest TVOC, ammonia and formalde-hyde concentration: ventilation system: mechanical supply air system, ceiling surface: product 2, walls: painted, indoor air RH < 40% VOC classification (clustering): ventilation system: mechanical supply air system, ceiling surface: product 2, walls: painted, indoor air RH: < 40%

The Varimax rotation for target VOCs revealed six components in the 12-month-old buildings (Table 13). These accounted for 94% of the variability in the variance. The first component accounted for 44% of the variability and factor loadings above 0.4 for the first component were detected for nearly all VOCs (alcohols, aldehydes, aliphatic compounds, terpenes, 2-butoxy ethanol and styrene), except for benzene, ethyl benzene, toluene, xylene, trimethylbenzene and naphthalene; the latter were divided to the second and third components. The first component was characterized by material emissions and inhabitancy, the second by material emissions, and the third by traffic emissions (III).

Statistically significant differences for TVOC, VOC group, single VOC, formaldehyde and ammonia concentrations in the 0–12-month-old buildings were observed between the different ventilation systems and floor covering materials (Table 14). A statistically significant difference was found between tridecane concentrations in the newly finished buildings when the different building structures (on-site built/ manufactured) were compared. Only apartments with mere mechanical exhaust air were included in the statistical analysis of floor covering and structure since the ventilation system was found to influence the IAQ in the PCA analysis (I). A statistically significant ($p < 0.05$) higher SVOC concentration was observed in the rooms with PVC flooring than in the rooms with parquet flooring in the newly finished and 6-month-old buildings (III).

Statistically significant correlations between indoor air concentrations and temperature, RH, and air exchange rates were found (I, Table 3). TVOC, ammonia and formaldehyde concentration correlated positively ($p < 0.01–0.05$) with the RH in the 6- and 12-month-old buildings. Of single VOCs, aldehydes and butanol especially correlated with the RH. The air exchange rates correlated most with the formaldehyde and dodecane concentration in the 12-month-old buildings (I, Table 3).

Significant correlations between emissions and surface or structure humidity and temperature were detected (II, Table 3). There was, however, no clear trend in correlation patterns throughout the first year. The structure humidity correlated with TVOC and ketone emissions from the parquet and with aromatic compounds from the ceiling in the newly finished buildings. The structure humidity correlated with the formaldehyde emission from the ceiling in the 12-month-old buildings (II).

Table 13. Factor loadings for six components after Varimax rotation for indoor air concentrations of selected VOCs in the 12-month-old buildings (III).

Component matrix(a)	**Component**					
	1	**2**	**3**	**4**	**5**	**6**
Butanol	0.775	0.029	-0.037	-0.363	0.293	0.209
Hexanol, 2-ethyl	0.512	0.587	-0.362	0.457	-0.194	0.054
Propanol, 2-methyl	0.76	-0.118	0.184	0.185	0.396	-0.404
Benzaldehyde	0.875	0.08	-0.178	0.198	-0.002	0.372
Hexanal	0.94	-0.09	-0.08	0.136	0.233	0.152
Octanal	0.616	-0.393	-0.405	0.407	0.173	0.212
Decane	0.772	0.27	0.173	0.137	-0.502	-0.043
Nonane	0.738	-0.266	0.115	-0.557	-0.188	-0.01
Undecane	0.741	0.353	0.455	-0.26	-0.151	-0.019
Benzene	-0.49	0.262	0.65	0.127	0.181	0.047
Benzene, ethyl	0.69	0.534	0.256	0.289	-0.057	-0.177
Benzene, dimethyl	0.024	0.473	-0.861	-0.137	-0.03	-0.01
Naphthalene	0.223	-0.705	0.308	-0.093	-0.588	0.001
Benzene, propyl	0.841	0.199	0.001	-0.398	0.065	-0.017
Styrene	0.855	0.4	0.067	0.143	-0.138	-0.167
Benzene, methyl	-0.103	0.413	0.587	0.254	0.015	0.338
Benzene, trimethyl	-0.165	0.164	0.786	-0.045	0.211	0.295
Ethanol, 2-butoxy	0.79	0.321	-0.015	-0.48	0.126	0.017
delta-3-Carene	0.618	-0.441	0.309	0.347	0.034	-0.427
alpha-Pinene	0.595	-0.593	0.013	0.194	-0.138	0.452
dl-Limonene	0.777	-0.531	0.048	0.027	0.27	-0.078
Explanation%	**44%**	**15%**	**14%**	**8%**	**7%**	**6%**

*Table 14. Statistically significant (*p < 0.05, **p < 0.01) differences in indoor air concentrations, temperature and relative humidity in the 0–12-month-old buildings based on the Mann Whitney non-parametric test (I).*

<table>
<tr><th>Newly finished building</th><th>6-month-old building</th><th>12-month-old building</th></tr>
<tr><td colspan="3"><i>Ventilation system: mechanical supply and exhaust air system vs. mechanical exhaust air system</i></td></tr>
<tr><td>Mechanical supply and exhaust air system:
Lower TVOC**, glycol/ glycolether, ketone*, aliphatic hydrocarbon*, terpene**, alpha pinene**, toluene**, dodecane**, 2,2- ethoxy-ethoxy ethanol**, heptanal**, benzoic acid** concentration and temperature**</td><td>Mechanical supply and exhaust air system:
Lower TVOC**, formaldehyde**, acid* and terpene* concentration</td><td>Mechanical supply and exhaust air system:
Lower TVOC** and formaldehyde**, aliphatic hydrocarbon**, alcohol*, aldehyde*, aromatic hydrocarbon*, ester*, ketone* and terpene* concentration</td></tr>
<tr><td colspan="3"><i>Floor covering (8 apartments with mechanical exhaust air system included): PVC vs. parquet</i></td></tr>
<tr><td>Parquet flooring:
Higher aldehyde**, tetradecane*, benzaldehyde*, tridecane*, 2,2- ethoxy-ethoxy ethanol*, diphenylmethanone*, methylpentanone*, hexadecane* and acetic acid* concentration
PVC flooring:
Higher 2,2,- butoxyetoxyethyl acetate* concentration</td><td>PVC flooring:
Higher 2,2,- butoxyetoxyethyl acetate* concentration</td><td>Parquet flooring:
Higher formaldehyde**, dodecane*, butanol*, butylacetate*, 2-phenoxyethanol*, pentanol*, 2-methyl propanol*, 2,2- ethoxy-ethoxy ethanol*, trimethylbenzene*, methoxypropanol* and heptanone* concentration and relative humidity**</td></tr>
<tr><td colspan="3"><i>Structure: on-site built vs. manufactured</i></td></tr>
<tr><td>On-site built:
Higher tridecane* concentration</td><td></td><td></td></tr>
</table>

5.3 The difference between the on-site and laboratory measured emissions for materials (IV)

Up to thirty times higher specific emission rates were measured on-site from the complete floor structure than from the PVC single materials. For some product combinations, the emission rate was the same from the finished structure as for the single PVC. The impact of adhesives was clear on the VOC emissions from the complete floor structures. The results indicate that three factors increased the VOC emissions from the PVC flooring: 1) the permeability of the PVC product for VOCs, 2) the VOC emissions from the adhesive used, and 3) the VOC emission from the back of the PVC product.

Glycols/ glycol ethers and their esters were abundantly present in the emissions from the PVC coated floors and single adhesives. All the single PVC materials emitted 2-ethylhexanol. Adhesives contributed to some extent to the on-site measured emission of 2-ethylhexanol for the more permeable types of PVCs. TXIB was found from both single PVCs and from the complete floor structures, as well as from the underlying structures. High ammonia emissions were measured from single adhesives on a glass plate on the first day but the contribution of the adhesives to the emission from the complete floor structure was not as clear as for TVOC. The ammonia emissions were affected by both the adhesive and the PVC product.

5.4 The impact of emissions on IAQ during the first year after the building had been taken into use (III)

The expected indoor air concentrations for the pollutants originating from the interior surfaces were calculated by Eq. 1, in III.

The contribution of the average on-site measured TVOC emissions to indoor air concentration was about 550 $\mu g/\ m^3$ in the newly finished buildings. This was 57% of the measured average indoor air concentration. The contribution of TVOC emissions from surfaces decreased to ~200 $\mu g/m^3$ in six months and to 170 $\mu g/m^3$ in twelve months (59% and 54% of the measured mean TVOC concentrations). The TVOC emission from surfaces had higher impact on the indoor air concentrations in three buildings with mechanical supply and exhaust systems than in those with mere mechanical exhaust (75% vs. 45%). The concentrations were, however, lower in buildings with a mechanical supply and exhaust air system.

The concentrations of glycols/ glycol ethers, aliphatic hydrocarbons, esters and ketones were well explained by the emissions from surfaces. The emissions of alcohols, aldehydes and terpenes decreased steadily from surfaces, whereas their airborne concentrations did not, which indicates the presence of other sources for these compounds. Aromatic compounds also had other non-measured sources.

The impact of VOC emissions originating from the ceiling structure on the indoor air concentration levels was higher than predicted based on the M1-target values. The ceiling surface was finished by spraying, which results in a porous structure, i.e. a large surface area. This, in addition to the fact that the surface was measured from a complete concrete structure, most likely was the reason for the higher emission rates. The walls contributed less than expected since their VOC emission were well below the M1-target value of 200 $\mu g/m^2h$.

The contributions of ammonia and formaldehyde emissions from the main interior structures to indoor air concentrations were less than 45 $\mu g/m^3$ for ammonia and below 40 $\mu g/m^3$ for formaldehyde. These were much lower than the predicted values. Non-identified sources were responsible for about 0–35% of the ammonia and formaldehyde concentrations in indoor air.

6. Discussion

The TVOC concentration was generally above the S3-class limit of 600 µg/m^3 (FiSIAQ 2001) in the newly finished buildings. The TVOC concentration usually decreased below the S3 level in 6 months; in 43% of the apartments the TVOC concentration reached the S1-class level of 200 µg/m^3 during the first 6 months (I).

TVOC levels were higher in this study than in an apartment building where the Finnish climate classification was carefully applied throughout the building process (Tuomainen et al. 2001, 2003). There, the mean TVOC concentration was 153 and 70 µg/m^3 after occupancies of 5 and 12 months respectively. In this study the mean TVOC concentration was 329 µg/m^3 after six months and 247 µg/m^3 after twelve months of occupancy (I). It is also possible that the TVOC levels were not yet fully stabilized because the median TVOC (sum of 48 VOCs) concentration was 121 µg/m^3 in 50 apartments studied by Kostiainen (1995). These apartments (older than three years, no repairs in the previous 18 months) were located in Helsinki and their inhabitants had no symptoms. In measurements conducted in 201 randomly selected homes in Helsinki, the mean of the sum of 30 target VOCs was 150 µg/m^3 (Edwards et al. 2001b). The sum of the same VOCs in the one-year-old buildings in this study was 112 µg/m^3.

Energy efficiency demands will favour the use of combined mechanical ventilation with heat recovery during the winter period in new buildings (European community 2003). The results of this study showed that such systems are also beneficial for controlling the VOC concentration in newly finished buildings since clearly lower concentrations were measured in buildings with mechanical supply air (I). Higher ventilation rates, cleaner supply air or a longer operation time for the ventilation system did not explain the lower concentrations. Therefore, infiltration, i.e. the flow of outdoor air into the building through cracks and other unintentional openings and through the normal use of external doors for entrance and egress, affected probably less in buildings with mechanical supply air. The operation of the ventilation system should be started as soon as possible after the completion of the building. The same was concluded by Tuomainen et al. (2001, 2003). On the other hand, very early operation leads to contamination of the ventilation system with dust originating from the construction work. The lowest TVOC concentration prior to occupancy was measured in the building where the combined mechanical supply and exhaust ventilation had been operating for 2 weeks (I).

The concentrations of the major VOCs generally decreased most strongly during the first six months of occupancy, reaching mean concentration levels of 5–15 µg/m^3. No significant change was seen between the 6- and 12-month results. The number of different VOCs increased along with the occupancy time. Xylenes were the dominating VOCs in the newly finished buildings. The concentration of xylenes decreased strongly with occupancy time, whereas such a trend was not detected in the concentration of toluene, which reached a mean concentration of 11 µg/m^3 in the one-year-old buildings (I). Toluene was the major VOC in two studies conducted in Helsinki with the same mean concentration of 20 µg/m^3 (Kostiainen 1995, Edwards et al. 2001b). In the present study the concentration ratio of toluene and xylene was 1.4 in the 12-month-old apartments; it was 2–3 in the previous studies. The mean concentration of benzene was similar at 2–3 µg/m^3 in all of these studies. This was also detected in this study. The main source of indoor air benzene has earlier found to be outdoor air in Helsinki (Edwards et al. 2001a). The outdoor air benzene concentration was 2 µg/m^3 outside building 1, which is similar to the results of the earlier studies in Helsinki (Edwards et al. 2001b, Helsinki mean was 1.7 µg/m^3). The mean concentrations of terpenes showed a decreasing trend in spite of terpene emissions from furniture. No halogenated compounds were detected in this study (I).

The formaldehyde concentration did not significantly exceed the S2-class level of 50 µg/m^3 (FiSIAQ 2001) during the first year in any of the apartments. The concentration was below the S1-class level of 30 µg/m^3 in eight apartments. In some buildings the indoor air concentrations of ammonia were above the S3 level of 40 µg/m^3 (FiSIAQ 2001) in the newly finished buildings and during the follow-up (I). On the other hand, Tuomainen et al. (2001, 2003) measured lower S1- level formaldehyde and ammonia concentrations (30/30 µg/m^3 respectively, FiSIAQ 2001) during the first three years of occupancy in the building where the Finnish climate classification was applied. The sprayed roof structure was painted in this building, which was not the case in the study presented here.

Seasonal changes were observed. In general, higher formaldehyde and ammonia concentrations were measured in the summer when the RH was at least 50% (I). Similar seasonal variation of formaldehyde in buildings has also been reported earlier (Reponen et al. 1991, Raw et al. 2004, Schlink et al. 2004). The reason for the increased concentrations remain speculative, however, the influence of hydrolysis reactions and competitive displacement may increase the surface emissions of these compounds at higher RH. New formaldehyde sources, such as furniture, were common in the inhabited buildings. This caused a slightly increasing trend in the mean concentration and, especially, led to elevated maximum formaldehyde concentrations in the occupied apartments (I). Reactions of VOCs originating from new materials with ozone may also contribute to the elevation of formaldehyde levels (Weschler and Schields1997, Weschler 2000).

The reference values for indoor air TVOC concentrations presented in this study are somewhat higher than the levels measured in older, non-problem apartments in Finland (Kostiainen 1995, Edwards et al. 2001b, Chapter 2.2.1, Table 1). This difference is partly due to differences in the TVOC determination. The relatively small number of apartments investigated in this study causes that the reference values can only be considered as tentative ones. The case selection assumed independent observations since different building materials were used in all apartments. However, clustering effects were seen for apartments in the same buildings because of same structure type, ventilation system, environment etc. These were considered for when possible in the statistical analysis. Despite small sample size, some clear trends (e.g. the effect of the ventilation system) were observed. More measurement data, however, is necessary for validated statistical calculations.

Even though the reference values presented here cannot be considered to be fully representative of the Finnish housing stock in which low-emitting materials are used, the values based on the results in one-year-old buildings facilitate interpretation of the results of measurements conducted in established, residential buildings. The 12-month reference values also coincide quite well with earlier results from German and English studies (Seifert 1990, Raw et al. 2004, Schlink et al. 2004, Chapter 2.2.2, Table 2) and the target values defined in the German national building regulations (Bundesministerium für Verkehr, Bau-und Wohnungswesen 2001). The normal values for 6-month-old buildings are the most important practical result of the study. It could be concluded that

IAQ should be assessed as early as possible, and preferably before the occupants move in. However, the study showed that the concentrations of VOCs are high in the newly finished apartments despite selection of low-emitting materials and control of the building moisture. On the other hand, the concentrations decrease rapidly in the beginning. Even though the occupancy affects IAQ, six months is proposed as a compromise point of time for evaluating concentration levels.

As the summary of PCA, the variables associated to the concentrations of studied indoor air pollutants in the buildings were the ventilation system, the floor covering material, the ceiling surface product, the wall surface product, the season, the relative humidity and temperature of the indoor air, and occupancy (I). Edwards et al. (2001a) also found that the inhabitancy, traffic emissions and material emissions were associated to the level of the same VOCs. In this study the factor associated with materials was clearly dominant, which was not observed in the established buildings studied by Edwards et al. (2001a).

The emission measurements performed from the finished floor surface showed that the emission was affected by all of its components, i.e. the structure, levelling agent, adhesive and floor covering material (II, IV). The target value for TVOC (200 µg/m^{2}h, FiSIAQ 2001) was reached for the painted wall, parquet floor and for three PVCs, but not as well for the three other PVCs or ceiling surfaces in the newly finished buildings (II). The surface of the different PVC materials clearly differed with regard to the permeability to VOCs from the underlying structure (IV). A less permeable type of PVC may have been measured by Wilke et al. (2004) since the emissions from the complete, PVC-coated structure was lower than the sum of emissions from the single components. Secondary emissions can appear from PVC coverings and adhesives under humid conditions (Gustafsson 1990). Wilke et al. (2004) identified secondary emissions – i.e. aldehydes and organic acids – from adhesives 46–56 days after the experiment started. In this study the emission rate of a known degradation product, 2-ethylhexanol, was on an average level of ~15 µg/m^{2}h (toluene equivalents) from the 4-week-old PVC floorings. In the 12-month-old buildings the average emission from the same PVCs was 8 µg/m^{2}h (toluene equivalents). However, some of the single PVC materials and some adhesives emitted 2-ethylhexanol. Thus this compound is not merely a degradation product and its presence in indoor air at low concentrations does not necessarily indicate moisture damage.

The occurrence of the plasticizer TXIB in indoor air has been coupled with complaints on IAQ (Metiäinen et al. 2002). In this study (II, IV) TXIB was found both from single PVCs and from the complete structures, as well as from the underlying structures. One of the less permeable (for VOCs) products had clearly higher TXIB emissions than the others. Very high emissions were measured from its underlying structure compared to the other products.

High ammonia emissions were measured from single adhesives on a glass plate on the first day, but the contribution of the adhesives to the emission from the complete structure was not as clear as for TVOC (IV). The indoor air concentration of ammonia increased in the Finnish building stock in the 90s, probably due to the shorter construction times and, consequently, the shorter drying periods for the structure (Hiltunen 2000). The results of this study, however, showed that the increased use of adhesives and/or PVC flooring may be an additional factor for this trend.

The formaldehyde M1 target value (50 $\mu g/m^2h$, FiSIAQ 2001) was reached for both the floors and the walls, and quite close for the ceilings in the newly finished buildings (II). The target value for the ammonia emission (30 $\mu g/m^2h$, FiSIAQ 2001) was clearly exceeded from some ceilings. The M1 target values were reached for all other emissions, than ammonia emission from the ceiling during the first year.

The calculations showed that target values for the indoor air pollutant concentrations as defined in the Finnish indoor climate classification (FiSIAQ 2001) can be reached for TVOC and ammonia if the ventilation fulfils its S1 criterion (III). For formaldehyde, only S2 level could be achieved. If emissions from the surfaces correspond to the upper limits of the M1 values, no S levels can reached for ammonia or formaldehyde and only the S3 levels for the TVOC concentration with the lowest air exchange rate (0.5 h^{-1}) allowed.

The contribution of the average on-site measured emissions to indoor air concentration was lower for TVOC and much lower for ammonia and formaldehyde than predicted based on M1 target values (III). This was due to the fact that the ammonia and formaldehyde emissions from the walls and floors were well below the M1 target values. The average TVOC contribution from surfaces to the airborne concentration was as high as ~550 $\mu g/m^3$ in the newly finished buildings. Thus an air exchange rate of 2.9 h^{-1} would have been needed to reach the S1-level concentration for TVOC (200µg/m^3, FiSIAQ 2001) at this point. However, the S1 level was reached within 6 months on average if only emissions from surfaces contributed. For ammonia, the contribution from surfaces remained ~40 µg/m^3 throughout the first year and for formaldehyde, it decreased to less than 20 µg/m^3 in the same time.

7. Conclusions and recommendations

A database for the indoor air concentration and interior surface emissions of VOCs, formaldehyde and ammonia for new, residential buildings was created during this study. The reference values of on-site surface emissions and indoor air concentrations for VOCs, formaldehyde and ammonia were presented as the mean and 95 percentile values (Table 10 and 11). The former represent "normal" concentration/ emission levels, whereas the latter ones describe "abnormal" limit values. The normal values indicate levels that are anticipated in residential buildings with the current building practice by using low-emitting materials. In other words, the reference values are defined for buildings, in which M1-classified (FiSIAQ 2001) materials are used and the air exchange rate fulfils the building requirement of 0.5 h^{-1} (normally apartment buildings). The use of these values in the case of other types of buildings should be considered with care. If concentrations or emissions exceed the limit for abnormal concentration/ emission, further investigations are recommended. The reference values represent purely technical guidance values. Thus they cannot be used for health effect evaluation. In addition, the results remain tentative due to the small sample size (14 apartments).

The effect of inhabitancy also affected the airborne concentrations in the occupied buildings- this was most evident for the terpenes and this has been taken into consideration when applying the reference values; the apartments in which new furniture was installed were not included.

The lowest concentration levels were achieved in the 0–12-month-old buildings with a mechanical supply and exhaust air system, screed product nr 2 (a limestone-based product, maximum grain size 0.3 mm) as the ceiling surface, painted walls and parquet as the floor covering material. The indoor air concentrations of formaldehyde and ammonia increased as the RH increased, especially in the 6- and 12-month-old building. This was also the case for other aldehydes and butanol. In addition, occupancy clearly increased some pollutant (formaldehyde, terpenes) concentrations.

The contribution of TVOC emissions from structures to indoor air concentration decreased from 57% to 54% in twelve months. Surface emissions were responsible for about 65–75% of the ammonia and formaldehyde concentrations. The emissions from interior surfaces were found to change rapidly during the first year, especially during the first six months. Generally, the emission rates decreased, but new compounds also appeared due to occupancy.

Significantly higher emissions were often measured on-site from the complete floor structure than from the single materials (PVC and adhesive) measured in the laboratory. The impact of adhesives on VOC emissions was clearly seen as higher emissions for some complete structures. This means that the different product combinations (PVC+ adhesive) should be considered while selecting materials. In addition, special care should be taken when drawing conclusions concerning a suspected problem structure on the basis of the emissions since large variations was detected in normal, undamaged buildings.

The use of classified building materials did not guarantee good IAQ alone; effective ventilation was also required. To ensure this, the air flow rate recommendations given per floor area presented in the first classification version (FiSIAQ 1995) should be reintroduced. The contribution of sources other than surfaces was found to be significant. The importance of these sources increased with time. The planned extension of the Finnish classification system to include furniture and cleaning agents is thus welcome.

The testing for labelling purposes is done for 28-day-old single products in the Finnish M1 scheme (FiSIAQ). However, this is not appropriate for testing adhesives since floor covering is usually laid within a few hours of spreading the adhesive on the structure. A better time point to test adhesives would be 24–72 hours after spreading on a plate. The 72-hour time point for measuring VOC emissions was also proposed as a testing scheme in the evaluation of VOC emission from building products, reported within the EU (ECA 1997). Also, the testing of complete structures would provide a better approach to evaluate the actual emissions. It appears to be important to consider the compatibility of different products in the composite.

The study confirmed that the Finnish material classification system provides a basis on which to achieve good IAQ in real buildings. Nevertheless, it could be developed further. For example, the incorporation of target values for VOC groups and critical VOCs (e.g. 2-ethylhexanol and TXIB) in the material classification would be advantageous from the consumer's point of view. In addition, supportive testing of material combinations as real structures would be beneficial. The reference values suggested in this study should also be updated when enough new data exist. Then, reference values based on the low percentile (such as 25th) values could also be set to indicate goals for exquisite construction practice.

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Reference values for indoor air pollutant concentrations in new, residential buildings in Finland

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Abstract

Eight buildings, representing the present construction practice in Finland, were investigated to create numeric reference data for indoor air quality (IAQ) in new residential buildings. Low-emitting materials according to the "Finnish Classification of Building Materials" were used in all the buildings. The airborne volatile organic compounds (VOCs), formaldehyde, and ammonia concentrations as well as temperature, relative humidity, and the air exchange rate were determined in the newly finished buildings and after 6 and 12 months. Target values for the indoor air concentrations were not generally reached in newly finished buildings. The lowest concentration levels were measured in buildings with mechanical supply and exhaust air systems. Formaldehyde concentrations fulfilled best the target values. The TVOC concentration usually reached the S2/S3-class values within 6 months. However, the ammonia concentration remained above the S3 limit during the whole first year. The concentrations of ammonia and formaldehyde showed seasonal variations, i.e., higher concentrations were measured in summer. The concentrations of individual VOCs generally decreased most strongly during the first 6 months and the final mean concentration levels were generally less than $15\,\mu g\,m^{-3}$. As the occupancy period got longer, the VOCs originating from the construction phase were increasingly replaced by new ones. Reference values based on means and on 95 percentiles are presented to facilitate interpretation of the results of measurements done to ensure that proper construction practices have been applied or to investigate IAQ problems.

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Keywords: Indoor air; VOC; Formaldehyde; Ammonia; Reference values; New buildings

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1. Introduction

Discussion concerning the health risks of the indoor air quality (IAQ) has risen because complaints are common among occupants. The impact of chemical pollutants on the perceived IAQ has been investigated in several studies. The volatile organic compounds (VOCs) have been suspected to cause "sick-building" symptoms like headache, eye and mucous membrane irritation, fatigue, and asthmatic symptoms (Molhave, 2003; Wolkoff and Nielsen, 2001; Wieslander et al., 1997; Norrbäck et al., 1995, 2000). The European exposure study (EXPOLIS) showed that VOC exposure was higher

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at homes than outdoors or at work places (Edwards et al., 2001; Saarela, et al., 2003).

WHO air quality guidelines exist for major ambient air pollutants such as nitrogen dioxide and ozone as well as for a few organic pollutants including mainly chlorinated and aromatic hydrocarbons (World Health Organization, 2000). The scientific proof of adverse health effects is considered adequate for these compounds and exposure limits or unit risks (for carcinogens) have been determined. The International Agency of Cancer Research recently upgraded formaldehyde to Group 1, known human carcinogen (IARC, 2004). However, there is still inadequate information about health effects of other VOCs. The total amount of VOCs, TVOC, has not been proven to correlate with symptoms (Andersson et al., 1997; Molhave, 2003).

Many national building codes state that IAQ should be safe and healthy in newly established buildings (Boverket, 2002; Bundesministerium für Verkehr, Bau-und Wohnungswesen, 2001; Erhvervs-og byggestyrelsen, 1995; Ministry of the Environment, 2002). In addition, the Finnish and Swedish building codes state that there should not be any odours that decrease comfort. The guideline values defined by the Finnish building code are in some cases stricter than the ones given by the health authorities. For example, the building code includes a 1 $\mu g\,m^{-3}$ guidance value for styrene in new buildings whereas the limit value given by the Finnish health authority in established buildings is 40 $\mu g\,m^{-3}$ (Ministry of Social Affairs and Health, 2003).

Building materials are important emission sources of VOCs, especially in new buildings (Wolkoff, 1999). Consequently, the development of low-emitting materials has been of interest as the consumers' awareness and demands have increased. Today, there are several labels for tested, low-emitting materials including the German RAL UZ 38 (Blue Angel), RAL-GZ 479, Emicode and GuT, the Danish ICL, and the Finnish Classification of Indoor Air Climate (Wolkoff, 2003). The harmonisation work on the European level is still under work (Kephalopoulus, et al., 2003). The Finnish Climate Classification, developed by the Finnish Society of Indoor Air Quality and Climate (FiSIAQ), defines design and target values for thermal comfort, ventilation rates, odour intensity, noise levels, the concentration of indoor air pollutants and material emissions as well as procedures for constructions work in new buildings (S-, M- and P- classes). The classification defines three classes for IAQ: S1, S2, and S3. The S1 class corresponds to the best IAQ. Target values are defined for concentration of TVOC, formaldehyde, ammonia as well as for temperature, humidity, and air velocity. The concentration target values for the classes (S1/S2/S3) are as follows: TVOC 200/300/600 $\mu g\,m^{-3}$, ammonia 30/30/40 $\mu g\,m^{-3}$, and formaldehyde 30/50/100 $\mu g\,m^{-3}$. The target values for the emission of VOCs, formaldehyde, ammonia, carcinogens, and the odour intensity level are defined for building materials at the age of 28 days (FiSIAQ, 2001). Though the classification is voluntary, especially the use of M1-classified, low-emitting materials has markedly increased and improved the IAQ in new Finnish buildings. Up to this date, there are over 900 M1-classified building products (The Finnish Building Information Foundation, 2005).

VOC concentration levels in indoor air have been published in a number of studies (Seifert, 1990; Wolkoff et al., 1991; Scholz and Santl, 1999; Bornehag and Stridh, 2000; Rehwagen et al., 2003; Raw et al., 2004). Seifert published target values for TVOC and VOC groups on the basis of what can be achieved in real buildings as follows: TVOC 300 $\mu g\,m^{-3}$, alkanes 100 $\mu g\,m^{-3}$, aromatic compounds 50 $\mu g\,m^{-3}$, terpenes 30 $\mu g\,m^{-3}$, halogenated compounds 30 $\mu g\,m^{-3}$, esters 20 $\mu g\,m^{-3}$, aldehydes 20 $\mu g\,m^{-3}$, and other compounds 50 $\mu g\,m^{-3}$ (Seifert, 1990). These results coincide quite well with results gained in the Swedish house stocking (Bornehag and Stridh, 2000). The average TVOC level exceeded 300 $\mu g\,m^{-3}$ in homes under 2 years old in England (Raw et al., 2004).

In the United States, the concentrations of 54 VOCs in indoor air were determined in four 2.5–9-month-old manufactured houses and in seven on site built houses 1–2 months after completion (Hodgson et al., 2000). Both low-VOC and conventional building products were used on interior surfaces and the air exchange rates varied between 0.14 and 0.78 h^{-1}. The TVOC varied between 1.5 and 2.7 $mg\,m^{-3}$. The predominant compounds were α-pinene, ethylene glycol, formaldehyde, acetaldehyde, hexanal, and acetic acid with concentrations between 10 and 18 ppb and 117 ppb for acetic acid. At the on site built buildings, toluene, β-pinene, and 1-butanol were also detected.

An Australian study (Brown, 2002) showed that the airborne VOC levels were one to two orders of magnitude higher in new or renovated buildings than in established dwellings. The decay of VOCs with time correlated with molecular volumes indicating that emissions were limited by diffusion within the materials. In the old, established dwellings ($n = 27$), the indoor geometric mean concentration of TVOC was $170\,\mu g\,m^{-3}$. Single VOCs identified were toluene, ethanol, nonanal, acetone, limonene, *m*,*p*-xylene, *p*-dichlorobenzene, benzene, and ethylacetate. The TVOC-concentration reached $200\,\mu g\,m^{-3}$ in 9 months in a new dwelling where VOC sources had been limited and in which the air exchange rate was $0.35\,h^{-1}$ (Brown, 2002).

The Finnish climate classification has been applied in a few studies. In the Finnish "Allergy-house" office building, special attention was given to low-emitting materials and to the proper functioning of the air exchange system. The TVOC concentration reached $200\,\mu g\,m^{-3}$ in 2 months after completion. The air exchange rates were $1.8–2.8\,h^{-1}$ (Saarela et al., 2000, 2001). A 3-year follow-up study was performed in an index apartment building, where the climate classification was applied throughout the building process, and in a reference building, which was constructed by conventional building practice. The TVOC concentration was 10-fold lower in the index building compared to the reference building. The designed air exchange rates were $1.7\,h^{-1}$ (index building) and $0.8\,h^{-1}$ (reference building). The formaldehyde concentration was below $20\,\mu g\,m^{-3}$ and the ammonia concentration below $30\,\mu g\,m^{-3}$ in the newly finished index building. The TVOC concentration level decreased to the level of $200\,\mu g\,m^{-3}$ in the index building within 5 months (Tuomainen et al., 2001, 2003).

In this paper, the results of indoor air concentrations of TVOC, VOCs, formaldehyde, and ammonia collected in new, residential buildings in Finland in years 1999–2003 are summarised. The objective was to investigate the development of IAQ in buildings, which represent the current building practice in Finland and in which low-emitting building materials were used. The results can be utilised as quality control reference data both to ensure that proper construction practices have been applied and in problem cases. The mean concentrations (normal values) are presented to serve the former aim and the means added with two standard deviations (95 percentiles) to indicate abnormal conditions with possible need for remedial measures.

2. Materials and methods

2.1. Study buildings

Fourteen apartments in eight residential buildings (seven apartment buildings and one two-family house), built according to the current Finnish Building Regulation Code, were investigated. Seven of the buildings were located in Helsinki and one in Turku, both cities locating on the coast of Gulf of Finland. The construction times varied between 12 and 15 months. The building structure types, floor-covering materials, the types of air exchange system and the time of construction (counted from the point of time when the heating was on) are summarised in Table 1. Low emitting, M1-classified materials were used in all the buildings. That is, the laboratory tests performed for 4-week-old samples have given TVOC-, ammonia- and formaldehyde emissions lower than 200, 30, and $50\,\mu g\,m^{-2}\,h$, respectively (FiSIAQ, 2001). The relative humidity (RH) of the structure was determined to be $<85\%$ before the floor covering was installed. In seven buildings, the walls were finished with screed and painted. Wallpaper was laid on the screed in the building 4. Ceilings were finished with screed (2 different products). The floors were finished with fine screed (dispersal 2–5 mm) in the site built buildings and with gross screed (dispersal 10–30 mm) in the manufactured buildings. Different types of PVC materials and parquets were used as floor covering materials. The installation of the floor covering material was the last construction work occurring 2–28 weeks prior to occupation, except in building 4, where painting was performed 12 weeks prior to occupation.

Indoor air measurements were performed first in the newly finished building, when the ventilation was operating, but before the occupants had moved in. The measurements were repeated after 6 and 12 months. In addition, an investigation was done 6 weeks after the inhabitants had moved in building 1 (apartments 2 and 3). Outdoor air samples were collected in building 1 locating near a coal-fired power plant. At one apartment (building 7, apartment 2), a water leak took place a few weeks before the apartment was finished. The apartment was heated and ventilated for about 2 weeks after which the indoor air was measured. These VOC results were not included in the statistical analysis.

Table 1
Buildings studied

Building, apartment number, apartment, floor area	Structure	Floor covering material (in bedroom)	Air exchange system and operation time before occupation	Time of construction (heating on)
Building 1, apartment 1, 59.5 m^2	OSCC	Parquet 1	ME, 8 weeks	December 1999–August 2000
Building 1, apartment 2, 40.5 m^2	OSCC	PVC 1, adhesive 1*		
Building 1, apartment 3, 40.5 m^2	OSCC	PVC 2, adhesive 2		
Building 2, 78.5 m^2	MCCS	PVC 3, adhesive 3*	ME, 2 weeks	June 2000–February 2001
Building 3, 40 m^2	OSCC	Parquet 1	ME, 12 weeks	June 2000–March 2001
Building 4, 78 m^2	OSCC	PVC 4, adhesive 4	ME, 2 weeks	November 2000–June 2001
Building 5, 79 m^2	MCCS	Parquet 2	MES, 4 weeks	January 2001–July 2001
Building 6, 76.5 m^2	MCCS	Parquet 2	MES, 2 weeks	May 2001–December 2001
Building 7, apartment 1, 31.5 m^2	MCCS	PVC 5, adhesive 1*	ME, 4 days	June 2001–December 2001
Building 7, apartment 2, 31.5 m^2	MCCS	PVC 6, no adhesive		
Building 8, apartment 1, 126 m^2	OSCC	Parquet 3	MES, 1 week	December 2001–July 2002
Building 8, apartment 2, 98.5 m^2	OSCC	Parquet 4		
Building 8, apartment 3, 106.5 m^2	OSCC	Parquet 5		
Building 8, apartment 4, 232 m^2	OSCC	Parquet 6		

OSCC = on-site built concrete cast, MCCS = manufactured, cored concrete slab, ME = mechanical exhaust, MES = mechanical exhaust and supply system, * = not M1-classified.

2.2. Sampling and analysis

Indoor air samples were taken in a closed room (usually the bedroom) at the height of approximately 1.40 m before noon (10:00–12:00 a.m.). The follow-up measurements were done every time in the same room and in the same place. The air exchange rates were determined simultaneously. The inhabitants were asked to avoid cleaning, smoking, and the use of fragrances in the morning prior to the measurements. Additional ventilation through doors or windows 24 h before the measurement was discouraged. No smoking or pets were observed during the measurements.

Air samples of 2–5 l for VOCs were collected on Tenax TA adsorbent at the air flow rate of ~100 ml min^{-1} (ISO, 2004). The sampling of ammonia and formaldehyde was performed into a 0.005 M sulphuric acid-solution at 2–4 l min^{-1}. Field blanks were collected and analysed parallel to the real samples. The temperature and RH were registered using a Vaisala HMP41 moisture detector. The air exchange rate was determined simultaneously based on ventilation duct measurements with an Alnor AXD-530 thermo anemometer.

Tenax tubes were thermally desorbed at 260 °C for 6 min (cold trap temperature −30 °C) by using a PE ATD 400 apparatus and analysed with a HP 5890 series 2 gas chromatograph connected to a HP 5972 mass spectrometer and FID detector. The gas chromatograph was equipped with a double- capillary column HP PONA (50 m × 0.2 mm × 0.25 μm) and the sample was split 1:1 in the two columns, which were connected to MSD and FID detectors. The temperature programme of the GC oven was as follows: 40 °C (5 min) ⇒ (6 °C min^{-1}) ⇒ 280 °C (5 min), total run time 50 min. MSD in SCAN mode was used to identify single VOCs and the FID response was used for quantification. TVOC was calculated as toluene equivalents from the total integrated FID signal between hexane and hexadecane. An external toluene standard was used for calibration. Selected VOC compound standards were analysed to compare retention times and mass spectra. The ammonia concentration was determined with an ion-selective electrode and the analysis of formaldehyde was done with the spectrometric acetyl-acetone method (Salthammer, 1992).

2.3. Data handling

Statistical tests were performed with Simca-P 7.0 (principal component analysis (PCA)) and SPSS 12.0.1 for Windows software. PCA was first performed to reveal which variables affected IAQ most. The variables included floor covering (PVC/parquet), air exchange system (mechanical exhaust/mechanical intake- and exhaust) and the time of its operation (in the newly finished building), floor structure/levelling agent (on site built + fine screed/manufactured + gross screed), walls (paint/wall paper), time of construction and season (winter/summer, indoor air humidity) and time point from floor installation. The variables TVOC, formaldehyde, ammonia and single VOC concentration as well as the temperature, humidity, and air exchange rate were included in the model. Due to water damage in building 7, apartment 2, these results were excluded from the PCA and SPSS analysis of the newly finished buildings. The normal distribution of the data was analysed with the Shapiro–Wilks *W* test. The *W*-value is 1 when the distribution is perfectly normal. The non-parametric Mann–Whitney *U* test was then used to clarify the statistical significance of the variables for the concentrations of TVOC, VOCs, formaldehyde, and ammonia.

3. Results

3.1. TVOC and single VOCs

The TVOC concentration generally varied between ~300 and 1300 $\mu g\,m^{-3}$ in the newly finished buildings. An exceptionally high concentration of TVOC (2100 $\mu g\,m^{-3}$) was measured in one of the apartments in the building 7 and this was explained by a preceding "bake out "period (the apartment was heated to 30–35 °C for 2 weeks) done due to the water leakage. In general, the TVOC concentration was above the S3-class limit of 600 $\mu g\,m^{-3}$ (FiSIAQ, 2001). Type of ventilation was the parameter that by most affected the TVOC concentration: the lowest TVOC concentrations of 310–620 $\mu g\,m^{-3}$ were measured in the buildings 5, 6 and 8, where a mechanical supply and exhaust ventilation system was used. Totally about 240 different VOCs were identified. The most frequently occurring VOCs in the buildings are summarised in Table 2. Totally, 72 different VOCs occurring at least in four apartments were identified at a concentration above 2 $\mu g\,m^{-3}$. The outdoor TVOC concentration was 20 $\mu g\,m^{-3}$. The xylenes, with a mean concentration of 160 $\mu g\,m^{-3}$, were the dominating VOCs in the newly finished buildings. α-Pinene, with a mean concentration level of 64 $\mu g\,m^{-3}$, was the next. There were some deviant VOC-profiles in the new buildings. The total concentration of acids was 113 $\mu g\,m^{-3}$ (apartment 2) and 2-butanone oxime concentration 87–93 $\mu g\,m^{-3}$ in the buildings 9 and 10. Styrene occurred in six apartments with a mean concentration of 3 $\mu g\,m^{-3}$ and a maximum concentration of 6 $\mu g\,m^{-3}$.

The indoor air TVOC concentration in the occupied apartments in the building 1 had decreased 60–75% to a concentration level of 300–500 $\mu g\,m^{-3}$ during 6 weeks after the occupants had moved in. VOCs, which were not identified earlier, contributed to 70–160 $\mu g\,m^{-3}$ of the TVOC-value. β-Pinene, limonene, 5-methyl-2-hexanol, and 3-methyl-2-pentanone were most important of these new VOCs.

The TVOC concentration decreased below the level of 600 $\mu g\,m^{-3}$ in 6 months in all the apartments except in the apartment investigated in the building 2, where the TVOC concentration reached the level of 1000 $\mu g\,m^{-3}$. The explanation was the emissions of α-pinene and δ-3-carene from a newly installed wooden furniture (bookcase). In six apartments (buildings 1, 4, 5, 7 and 8), the TVOC concentration reached the S1-class level of 200 $\mu g\,m^{-3}$ during the first 6 months. The effect of the "bake out" period in one of the apartments in the building 7 could be seen as low TVOC concentration during the follow-up measurements. The concentrations of individual major VOCs generally decreased most strongly during the first 6 months of occupancy and the mean concentration levels were generally 5–15 $\mu g\,m^{-3}$. Totally 167 different VOCs were identified in the 6-month-old buildings. Thirty-nine different VOCs occurring at least in four apartments were identified at a concentration above 2 $\mu g\,m^{-3}$ (Table 2).

No significant change was seen between the 6- and 12-month results except in the building 4, where the TVOC concentration level rose. The fresh air supply was restricted prior to the measurement and this most likely affected the results. Totally 189 different VOCs and 54 different VOCs occurring at least in four apartments were identified at a concentration above 2 $\mu g\,m^{-3}$ in the 12-month-old buildings (Table 2).

The number of different VOCs increased along with the occupancy time; 111 VOCs occurred more often in the 12-month-old buildings than in the newly finished buildings. The occurrence of 2-ethylhexanol increased from one to nine apartments and the occurrence of 6-methyl-5-hepten-2-one from two to ten apartments in 12 months. Other VOCs, that were found most frequently in the 12-month-old building, included hexadecane, heptadecane, pentadecane, hexanal, decanal, limonene,

Table 2
The mean concentration, mean + 2x standard deviation and the Shapiro–Wilks W test value of TVOC, VOC groups, most frequently ($n \geqslant 4$) occurring VOCs (and TXIB), ammonia and formaldehyde, air exchange rate, temperature and humidity in the 0-, 6- and 12-month-old buildings

Compound/measurement	Indoor air concentration (μg m^{-3})								
	0 months ($n \geqslant 4$)			6 months ($n \geqslant 4$)			12 months ($n \geqslant 4$)		
	Mean	Mean +2 std. dev.	W-value	Mean	Mean +2 std. dev.	W-value	Mean	Mean +2 std. dev.	W-value
Aromatic hydrocarbons	*163*	*220*	0.877	***46***	***77***	0.670	***24***	***35***	0.843
Benzene	n.d			n.d			3	4	0.896
Benzene, methyl	20	28	0.896	5	6	0.666	11	16	0.863
Xylene, *o,p*	38	64	0.914/0.926	4	7	0.675/0.756	4	6	0.812/0.807
Xylene, *m*	72	122	0.794	24	114	0.589	4	6	0.941
Benzene, ethyl-	29	42	0.847	2	3	0.597	3	6	0.732
Benzene, C3–C4 substituted	24	43		2	3		2	3	
Styrene	3	4	0.921	2	3	0.665	3	5	0.604
Aliphatic hydrocarbons	*95*	*161*	0.657	***24***	***30***	0.947	***24***	***34***	0.883
Pentane	30	79	0.446	14	30	0.816	n.d.		
Heptane	3	4	0.935	2	3	0.872	2	3	0.947
Octane	4	6	0.973	n.d.			1	2	0.968
Nonane	12	24	0.633	1	1	0.922	1	1	0.931
Decane	32	60	0.609	n.d.		0.771	n.d.		
Aliphatic C10–C11, sum of isomeres	57	136							
Tridecane	4	8	0.766	3	6	0.758	2	2	0.808
Tetradecane	5	8	0.777	2	3	0.921	1	1	0.937
Pentadecane	2	3	0.793	2	2	0.874	1	2	0.893
Hexadecane	1	2	0.711	2	3	0.669	1	2	0.906
Heptadecane	n.d.			1	1	0.701	1	5	0.811
Dodecane	4	6	0.922	3	4	0.826	2	3	0.876
Undecane	18	32	0.595	4	6	0.776	3	4	0.868
Cycloalkane	*27*	*50*	0.683	***4***	***6***	0.767	***7***	***12***	0.609
Cyclohexane, butyl	7	16	0.582	n.d.			n.d.		
Alcohol	*43*	*66*	0.755	***21***	***31***	0.840	***34***	***48***	0.822
2-Propanol	38	85	0.643	14	35	0.635	6	10	0.787
1-Butanol	18	22	0.699	15	45	0.880	8	12	0.955
1-Propanol, 2-methyl	9	13	0.861	6	11	0.723	6	10	0.871
1-Pentanol	3	4	0.787	n.d.			5	9	0.781
2-Ethyl-hexanol	n.d.			1	1	0.881	2	3	0.766
Phenol	n.d.			3	5	0.876	2	2	0.944
Benzylalcohol	n.d.			1	1	0.849	n.d.		

Table 2 (*continued*)

Compound/measurement	Indoor air concentration (µg m^{-3})								
	0 months ($n \geqslant 4$)			6 months ($n \geqslant 4$)			12 months ($n \geqslant 4$)		
	Mean	Mean +2 std. dev.	*W*-value	Mean	Mean +2 std. dev.	*W*-value	Mean	Mean +2 std. dev.	*W*-value
Aldehyde	*45*	*70*	0.680	***31***	***44***	0.687	***34***	***46***	0.874
Butanal	5	9	0.868	n.d.			2	3	0.987
Pentanal	10	17	0.830	3	4	0.827	6	8	0.971
Hexanal	21	33	0.838	10	15	0.691	11	16	0.800
Heptanal	3	6	0.591	2	3	0.641	2	2	0.917
Octanal	5	8	0.644	3	6	0.897	3	5	0.891
Nonanal	8	11	0.867	6	8	0.844	7	9	0.904
Decanal	3	4	0.966	5	6	0.953	5	6	0.876
Benzaldehyde	5	7	0.772	4	7	0.837	3	4	0.760
Ketone	*15*	*25*	0.741	***4***	***7***	0.768	***6***	***10***	0.728
2-Butanone	6	10	0.831	2	3	0.881	2	3	0.849
2-Heptanone	3	7	0.710	n.d			n.d.		
Ethanone, phenyl	n.d.			1	1	0.863	2	2	0.777
6-Methyl-5-hepten-one	n.d.			2	3	0.839	2	3	0.907
Cyclohexanone	2	4	0.898	2	2	0.985	2	3	0.840
Geranyl acetone	n.d			1	2	0.787	2	3	0.900
Methanone, diphenyl	1	2	0.763	n.d			n.d.		
Pentanone, 4-methyl	12	20	0.952	n.d.			n.d.		
Ester	*22*	*42*	0.580	***11***	***16***	0.777	***9***	***12***	0.909
Butylacetate	5	8	0.808	6	10	0.857	3	5	0.829
Ethylacetate	n.d.			3	5	0.758	4	6	0.848
Ethanol, 2,2-butoxyethoxyacetate	40	137	0.709	n.d.			4	7	0.821
TXIB	1 ($n = 1$)			3 ($n = 1$)			3 ($n = 3$)		
Glycol/glycolether	*46*	*65*	0.863	***22***	***41***	0.570	***22***	***31***	0.922
1,2-Propanediol	24	34	0.875	30	58	0.703	14	21	0.928
2,2-Ethoxyethoxyethanol	13	21	0.840	n.d			5	11	0.693
Butoxyethanol	3	5	0.900	n.d.			2	4	0.820
Butoxyethoxyethanol	20	31	0.932	n.d.			n.d.		
Phenoxyethanol	n.d.			n.d.			3	5	0.725
1,3-Butanediol	n.d.			n.d			11	15	0.791
Terpene	*96*	*140*	0.877	***65***	***98***	0.838	***69***	***104***	0.790
α-Pinene	61	88	0.871	37	59	0.831	35	61	0.767
δ-3-Carene	18	26	0.858	18	30	0.854	19	35	0.770
β-Pinene	11	16	0.916	3	5	0.836	3	5	0.823
Limonene	12	26	0.759	10	16	0.820	12	25	0.903
Acid	*22*	*53*	0.418	***5***	***6***	0.862	***9***	***16***	0.682
Benzoic acid	3	4	0.965	3	4	0.903	3	4	0.843
2-Ethylhexanoic acid	n.d.			2	4	0.941	n.d.		
Acetic acid	3 ($n = 3$)	5	0.824	n.d.			8	13	0.873

Other									
Decalin, 2-methyl	5	9	0.954	1	1	0.988	n.d		
TVOC all sites	780	1103	0.857	**329**	**473**	0.814	**247**	**336**	0.880
TVOC mechanical exhaust only	1098	1543	0.790	**459**	**666**	0.903	**356**	**473**	0.910
TVOC mechanical supply and exhaust	397	509	0.724	**155**	**218**	0.739	**139**	**188**	0.735
Formaldehyde	19	26	0.835	**21**	**28**	0.900	**26**	**37**	0.790
Ammonia	42	51	0.918	**42**	**51**	0.931	**43**	**57**	0.867
	Average	Max	Min	Average	Max	Min	Average	Max	Min
Temperature (°C)	24	30	18	23	28	22	24	28	21
Indoor air humidity (%)	45	63	26	29	66	15	35	74	0.73
Air exchange rate (h^{-1})	0.95	1.61	0.82	0.98	1.45	0.79	0.91	1.39	23

(n.d. = not detected).The reference values for TVOC, VOC groups, formaldehyde and ammonia recommended in this paper are marked as bolds (VOC groups as bold italics). The mean concentration can be regarded as a "normal" value and the mean added with two standard deviations can be regarded as an limit for an "abnormal" value for newly finished and for 6- and 12-month-old apartments.

menthol, phenoxyethanol, phenylethanone, geranylacetone, and octamethylcyclotrisiloxane. Among aromatic hydrocarbons, the mean concentration of xylenes decreased from 110 to 8 μg m^{-3} during the first year whereas there was no clear trends in the concentrations of toluene and styrene. Benzene was detected only in the 12-month-old buildings with a mean value of 3 μg m^{-3}.

3.2. Formaldehyde

The indoor air formaldehyde concentration varied between 13 and 37 μg m^{-3} in the newly finished buildings. The formaldehyde concentration did not significantly exceed the S2- class level of 50 μg m^{-3} during the first year in any of the apartments (Table 2). However, seasonal changes were observed during the follow-up measurements in the buildings where a mere mechanical exhaust ventilation system was used. In general, higher formaldehyde concentrations were measured in summer time when the RH was at least 50%. There was also a slightly increasing trend in the formaldehyde concentration. Its mean value increased from 19 to 26 μg m^{-3} during the first year.

3.3. Ammonia

The indoor air concentrations of ammonia were 20–60 μg m^{-3} in the newly finished building (Table 2). The lowest ammonia concentrations were measured in apartments 2, 3 and 7, which were finished in wintertime. The ammonia concentration levels correlated with the season, i.e., with the RH. Ammonia concentrations of 60–90 μg m^{-3}, clearly above the S3- level of 40 μg m^{-3}, were measured in the 6–12-month-old buildings during the summer period, when the RH exceeded 50%. In the winter, the RH was below 40% and the ammonia concentrations were on the level of 20–50 μg m^{-3}.

3.4. Reference values

The results in Table 2 can be considered as reference values for TVOC, VOC groups, formaldehyde and ammonia for The mean concentration can be regarded as a "normal" value and the mean added with two standard deviations can be regarded as an limit for an "abnormal" concentration for 6- and 12-month-old apartments. Especially, the concentrations of VOCs changed rapidly during the first months after completion of the construction. Thus, the concentrations were unstable in the newly finished buildings and no reference values are presented for them.

3.5. Variables affecting the concentrations of indoor air gaseous pollutants

Two examples of PCA results are presented in Figs. 1 and 2. Lower TVOC concentration was observed in the newly finished buildings with a mechanical supply and exhaust air system (Fig. 1a and b). The same levelling agent (product 2) was also used on the ceiling structure in these buildings and the RH was low, less than 40%. The floor covering was parquet. The lowest TVOC, ammonia, and formaldehyde concentrations in the 6-month-old buildings were measured in the building where

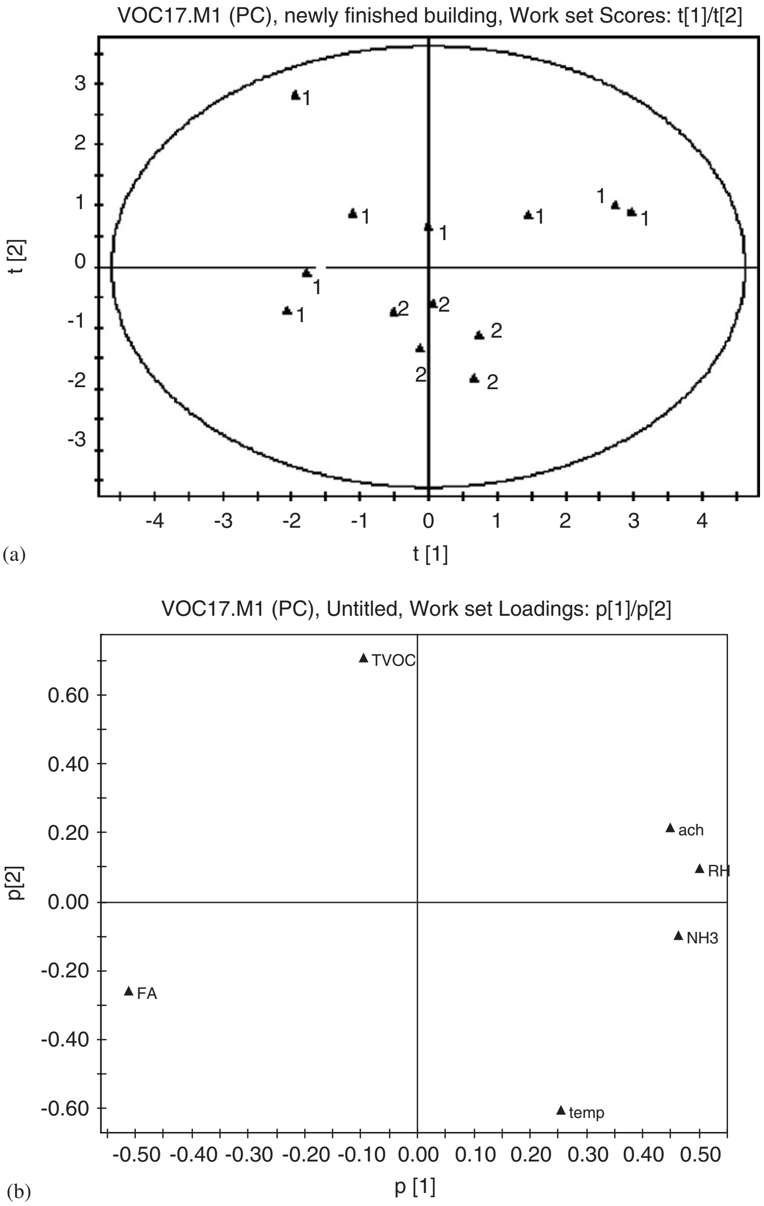

Fig. 1. Example of PCA results: IAQ in the newly finished building. The score plot (a) shows how the 13 measurement sites (1 = mechanical exhaust air system, 2 = mechanical supply and exhaust air system) are scattered in the model and the model contribution of the measured parameters is shown in the corresponding loading plot (b). The contribution of the measured parameter decreases as the distance of the parameter to the observation increases (TVOC = TVOC concentration, FA = formaldehyde concentration, NH_3 = ammonia concentration, RH = relative humidity, temp = temperature, ach = air exchange rate).

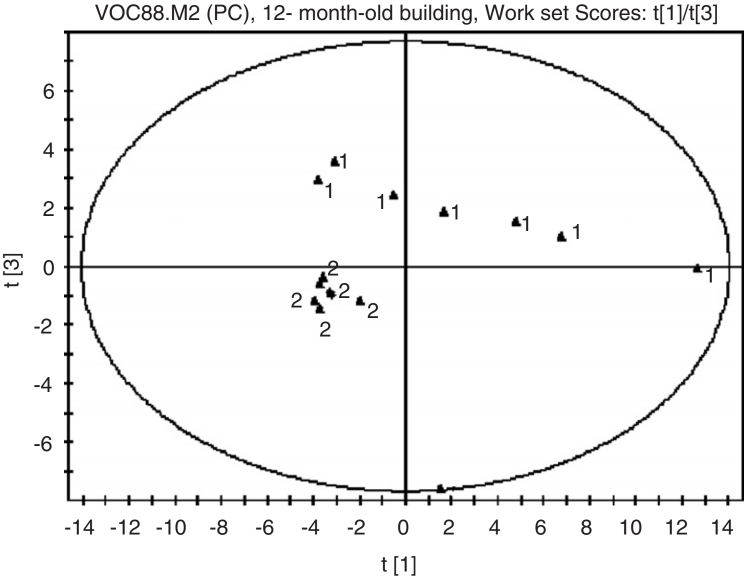

Fig. 2. Example of PCA results: classification of VOCs in the 12-month-old building (1 = mechanical exhaust air system, 2 = mechanical supply and exhaust air).

product 2 was used as ceiling surface product and when the RH was less than 30%. The clustering of VOCs also occurred at these apartments. The same was observed in the 12-month-old buildings, where the clustering of VOCs according to air exchange system (mechanical supply), ceiling surface (product 2), and indoor air humidity ($<40\%$) was quite clear (Fig. 2). As the summary of (10) PCA, variables affecting the concentrations of indoor air gaseous pollutants in the buildings were season, RH and temperature of indoor air, air exchange system, floor covering material, ceiling surface product, wall surface product, and occupancy.

3.6. The effect of season, relative humidity and temperature of indoor air

The mean temperature was 23–24 °C and the range was from 18 to 30 °C during the follow-up period. Seasonal changes were observed for the indoor air RH: in winter time the humidity could be as low as 15% whereas in summer the humidity could raise up to 70%. The RH was lowest ($p<0.05$) in the winter–spring period (November–April) in the newly finished buildings. Similar effect was also detected in the 6-month-old buildings in the concentrations of TVOC, ammonia, and formaldehyde. However, the season did not explain any of the concentration levels or temperature in the 12-month-old buildings. The correlations of TVOC, ammonia, formaldehyde, and selected VOCs with the outer conditions in the 6- and 12-month-old buildings are shown in Table 3.

3.7. The effect of the air exchange system

The air exchange rates varied between 0.7 and 1.5 h^{-1} during the first year. These were above the minimum required value of 0.5 h^{-1} in Finnish residential buildings (Ministry of the Environment, 2002). The correlations between air exchange rates and the airborne concentrations of the pollutants measured were unstable (Table 3). In the newly finished buildings, the TVOC concentration was significantly lower ($p<0.01$) in the buildings with mechanically supply and exhaust air than in the buildings with mechanical exhaust air. The temperature was also lower in these buildings ($p<0.05$). The TVOC and formaldehyde concentrations were also lower ($p<0.01$) in the 6- and 12-month-old buildings with combined mechanical ventilation.

In the newly finished building, the glycol/glycolether, ketone, and aliphatic hydrocarbon ($p<0.05$) as well as terpene ($p<0.01$) concentrations were lower in the buildings with combined mechanical ventilation than in the buildings with mechanical exhaust system only. Similar statistically significant differences were found for acid and terpene

Table 3
The correlation ($^{*} = p < 0.05$, $^{**} = p < 0.01$) of TVOC, ammonia, formaldehyde and VOCs in indoor air with the outer conditions in the 6- and 12-month-old buildings

Compound	6 months			12 months		
	Temp	RH	ACH	Temp	RH	ACH
TVOC	0.551^{*}	0.839^{**}	-0.033	0.459	0.623^{*}	0.370
Formaldehyde	0.467	0.814^{**}	-0.137	0.217	0.833^{**}	0.722^{**}
Ammonia	0.626^{*}	0.747^{**}	0.046	0.610^{*}	0.853^{**}	0.683^{*}
Undecane	0.292	0.211	0.248	-0.023	0.672^{**}	0.481
Tetradecane	0.325	0.573^{*}	0.131	0.152	0.489	0.335
Dodecane	0.379	0.192	0.225	0.164	0.758^{**}	0.756^{**}
Pentanal	0.497	0.958^{**}	-0.304	0.427	0.878^{**}	0.288
Hexanal	0.423	0.876^{**}	-0.279	0.486	0.824^{**}	0.135
Heptanal	0.764^{*}	0.721	0.148	-0.078	0.122	0.541^{*}
Octanal	0.331	0.534^{*}	-0.303	0.630^{*}	0.399	-0.344
Nonanal	0.543^{*}	0.854^{**}	0.003	0.456	0.444	-0.064
Decanal	0.504	0.846^{**}	0.113	0.454	0.401	0.097
Benzaldehyde	0.497	0.633^{*}	-0.095	0.617^{*}	0.808^{**}	-0.040
n-Butanol	0.177	0.747^{**}	-0.447	0.339	0.820^{**}	0.572^{*}
2-Ethylhexanol	0.325	0.441	0.141	0.338	0.544^{*}	-0.311
α-Pinene	0.639^{*}	0.756^{**}	-0.158	0.459	0.328	-0.146
δ-3-Carene	0.622^{*}	0.691^{**}	-0.003	-0.098	0.328	0.055
1,2-Propanediol	0.240	0.798^{**}	-0.395	0.529	0.624^{*}	0.131

temp = temperature, RH = relative humidity, ACH = air exchange rate.

concentrations ($p < 0.05$) in the 6-month-old buildings and for aliphatic hydrocarbon ($p < 0.01$) as well as alcohol, aldehyde, aromatic hydrocarbon, ester, ketone, and terpene ($p < 0.05$) concentrations in the 12-month-old buildings.

3.8. *The effect of the floor covering material*

To avoid the influence of the ventilation system on the results, only the buildings with mechanical exhaust air system ($n = 8$) were included in the analysis of the effect of the floor covering material. No significant differences in the TVOC, ammonia or formaldehyde concentrations were observed between the apartments with parquet and PVC flooring in the newly finished or the 6-month-old buildings. Higher formaldehyde concentration and humidity was measured in the apartments with parquet ($p < 0.05$) in the 12-month-age buildings. The mean concentration of aldehydes was higher in the newly finished buildings with parquet floors than those with PVC flooring ($p < 0.05$). The terpene concentration was higher in the apartments with parquet floorings during the whole follow-up period.

4. Discussion

This study showed that the S1 target values were not generally reached in the newly finished residential buildings despite of the use of classified, low-emitting building materials. The lowest concentration levels were measured in buildings with a mechanical supply and exhaust ventilation. The mean TVOC concentration was 780 $\mu g\,m^{-3}$ in the newly finished buildings. TVOC concentration reached, in general, the S2/S3-class values (300/600 $\mu g\,m^{-3}$) within 6 months. The decline of the TVOC concentration was rapid in the beginning but became rather stable (about 300 $\mu g\,m^{-3}$) after 6 months. On the other hand, the composition of the complex TVOC mixture went through a profound change during the follow-up period of 1 year. As the occupancy period got longer, the VOCs originating from the construction phase were increasingly replaced by new ones. The moving of occupants caused a considerable effect in the apartment where measurements were performed 6 weeks later. The TVOC concentration had already decreased 60–75% and the new VOCs constituted almost one third of the VOCs detected. As many as 111 VOCs occurred more often in the 12-month-old buildings than in the newly finished buildings. TVOC levels were higher in this study than in an apartment building, where the Finnish climate classification was very carefully applied throughout the building process (Tuomainen et al., 2001, 2003). There the mean TVOC concentration was 153 and 70 $\mu g\,m^{-3}$ after occupancies of 5 and 12 months, respectively. It is also possible that TVOC levels were not yet fully stabilised because the median TVOC concentration was 121 $\mu g\,m^{-3}$ in 50 apartments studied by Kostiainen (1995). These apartments (older than 3 years, no repairs in previous 18 months) were located in Helsinki and their inhabitants had no symptoms. In measurements conducted in 201 randomly selected homes in Helsinki, the mean of the sum of 30 target VOCs was 150 $\mu g\,m^{-3}$ (Edwards et al., 2001).

Among the aromatic compounds, the concentration of xylenes decreased strongly with occupancy time whereas such a trend was not detected in the concentration of toluene. The concentration of styrene was rather high, exceeding often the 1 $\mu g\,m^{-3}$ guidance value given by the Finnish building code. The level of styrene remained stable. Its possible sources are polystyrene based insulating material and/or the polyethene material filled with polystyrene pads, which were used under the parquet floor covering. Somewhat surprisingly, the concentration of benzene increased with time being $3\,\mu g\,m^{-3}$ after 1 year. The contribution was mainly from building 7 (two apartments) but its sources remain unknown. Among the other major VOCs, the mean concentrations of terpenes showed a decreasing trend inspite of terpene emissions from furniture. Toluene was a major VOC in two studies conducted in Helsinki with the same mean concentration of $20\,\mu g\,m^{-3}$ (Kostiainen 1995, Edwards et al., 2001). The concentration ratio of toluene to xylene, which was now 1.4 in the 12-month-old apartments, was 2–3 in the previous studies. The mean concentration of benzene was similar, $2–3\,\mu g\,m^{-3}$ in these studies. No halogenated compounds were detected in this and in the previous Finnish studies.

While the concentration of formaldehyde generally was below the S1-class limit of $30\,\mu g\,m^{-3}$, even the S3-class limit for the ammonia concentration ($40\,\mu g\,m^{-3}$) was usually exceeded. Both concentrations showed seasonal variations, i.e., higher concentrations were measured in summer, when the RH was above 50%. Seasonal variation of formaldehyde in buildings has also been reported earlier (Reponen et al., 1991; Raw et al. 2004). New formaldehyde sources, such as furniture, appeared in the inhabited buildings. This caused a slightly increasing trend in the mean concentration and, especially, led to elevated maximum concentrations in the occupied apartments. Reactions of VOCs originating from new materials with ozone may also contribute to the elevation of formaldehyde (Weschler et al., 1997, 2000a,b). However, the formaldehyde concentration did not significantly exceed the S2-class level of $50\,\mu g\,m^{-3}$ during the first year in any of the apartments.

Energy efficiency demands will favour the use of the combined mechanical ventilation with heat recovery during the winter period in new buildings (European community, 2003). The results of this study showed that such systems are also beneficial as regards to the VOC concentration in newly finished buildings. Therefore, the operation of the ventilation system should be started as soon as possible after the completion of the building. On the other hand, very early operation leads to the contamination of the ventilation system with dust originating from construction work. The lowest TVOC concentration prior to occupancy was measured in the building where the mechanical supply and exhaust ventilation system had been operating for 2 weeks.

The mean and 95 percentile values (in Table 2) rounded to reference values for TVOC, VOC groups, formaldehyde, and ammonia for 6- and 12-month-old apartments are summarised below. The reference values are purely technical guidance values. Thus, they cannot be used for health effect evaluation. The normal values indicate levels, which can be reached with the current building practice and by using low-emitting materials. If concentrations exceed the limit for abnormal concentration, the presence of indoor sources is possible and further investigations are recommended.

Compound or compound group	Normal value ($\mu g\,m^{-3}$)		Limit for abnormal concentration ($\mu g\,m^{-3}$)	
	Six months	One year	Six months	One year
Aromatic hydrocarbons	50	30	80	40
Aliphatic hydrocarbons	25	25	35	35
Cycloalkane	5	10	10	15
Alcohol	25	35	35	50
Aldehyde	35	35	50	50
Ketone	5	10	10	15
Ester	15	15	20	20
Glycol/glycolether	25	25	45	35
Terpene	70	70	110	110
Acid	5	10	10	20
TVOC	270	270	400	400
Formaldehyde	25	30	30	40
Ammonia	45	45	60	60

The reference values are somewhat higher than the levels measured in older, non-problem apartments in Finland. Even though the reference values presented here cannot be considered to be fully representative for the Finnish housing stock in which low-emitting materials are used, the values based on the results in 1-year-old buildings facilitate interpretation of the results of measurements conducted in established buildings. The 12-month reference values also coincide quite well with earlier results from German and English studies (Seifert, 1990; Raw et al., 2004; Schlink et al., 2004) and the target values defined in the German national building regulations (Bundesministerium für Verkehr, Bau-und Wohnungswesen, 2001).

The normal values for the 6-month-old buildings are the most important result of the study. It would be practical if IAQ could be assessed as early as possible, and preferably before the occupants move in. However, the study showed that especially the concentrations of VOCs are high in the newly finished apartments despite of selection of low-emitting materials and considering the building moisture. In addition, the concentrations decrease rapidly in the beginning. Even though the occupancy already affects IAQ, 6 months is proposed as a compromise point of time.

The study indicated the need to further develop the Finnish Climate Classification guidelines. The present system suits rather well for the control of formaldehyde, reasonably well for TVOC, but poorly for ammonia.

Acknowledgments

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PUBLICATION II

Reference values for structure emissions measured on site in new residential buildings in Finland

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ATMOSPHERIC ENVIRONMENT

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Reference values for structure emissions measured on site in new residential buildings in Finland

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Abstract

A 3-year research project was established in 1999 to create numerical reference data for material emissions during the time of construction and during the first year. Seven buildings, representing the present construction practice in Finland, were investigated. Material emissions were measured by using the field and laboratory cell (FLEC) during the time of construction, in the newly finished, and in the 6- and 12-month-old buildings. The emission rates for volatile organic compounds (VOCs), formaldehyde, and ammonia were determined.

The highest total VOCs (TVOC) emissions were measured in the newly finished buildings from the ceiling structure and from some of the PVC floor coverings. These emissions were up to 1300–2000 $\mu g\, m^{-2} h^{-1}$. Individual VOCs with emission rates above 50 $\mu g\, m^{-2} h^{-1}$ included 2-(2-butoxyethoxy) ethanol and its acetate, C4–C16-substituted alkylbenzenes, and xylenes. The mean TVOC emission decreased at least to the Finnish M1-class level (200 $\mu g\, m^{-2} h^{-1}$) from all surfaces and in all the buildings in 6–12 months. The ammonia and formaldehyde emissions from the ceiling structure were 20–60 $\mu g\, m^{-2} h^{-1}$ in the newly finished buildings and the M1-levels (30/50 $\mu g\, m^{-2} h^{-1}$) were exceeded in some cases. These emissions even increased in some buildings during the follow-up period indicating the difference between emissions measured in the laboratory and on site from real structures. Reference values based on the means and 95th percentile are presented to be utilised in both quality control and while investigating indoor air quality problems which are suspected to be caused by a defect structure.

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Keywords: Material emission; VOC; Ammonia; Formaldehyde; Reference value

1. Introduction

High pollutant concentrations have often been measured in newly built or renovated buildings (Hodgson et al., 2000; Tuomainen et al., 2001, 2003; Brown, 2002). Building materials are considered to be one of the major sources of pollution in the indoor environment. Therefore, emissions from different building materials have been investigated extensively. Since the 1980s, the emissions of formaldehyde have been characterised (Hawthorne and Matthews, 1987). The environmental chamber technique for the determination of volatile organic compounds (VOC) was introduced in the 1980s and

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it has been standardised in Europe (European collaborative action, 1991, 1993; ISO, 2006a, b). A corresponding method exists in the US (American Society for Testing Materials, 1997).

The use of small-scale environmental chambers with volumes ranging from a few litres to a few cubic metres has been increasing (Wensing, 1999). The weakness of these traditional chamber techniques is that they cannot be used to investigate emissions from existing real building structures. The emission of VOCs from a material in a real building structure is affected not only by this material but also by the environmental conditions and other surrounding materials (Wolkoff, 1998; Wilke et al., 2004; Wirtanen, 2005). Secondary emissions can develop under the influence of humidity, ozone, UV-light, etc. (Weschler and Schields, 1997; Wolkoff et al., 2000). Hydrolysis reactions in the floor structure (PVC/adhesive/casein containing levelling agents) can produce 2-ethylhexanol, butanol, and ammonia (Karlsson et al., 1989; Gustafsson, 1990; Bornehag, 1991). Thus, the emission measured on site can differ considerably from the emission measured from a single material under laboratory conditions.

The portable microchamber, field and laboratory emission cell (FLEC), which was developed for identifying emission sources on site (Wolkoff et al., 1991), does not suffer from this defect. The performance of the FLEC has been evaluated in many studies (Gunnarsen et al., 1993; Wolkoff et al., 1995; Roache et al., 1996; Wolkoff, 1995, 1998; Uhde et al., 1998), and the method has been utilised in several field investigations (Zellweger et al., 1995; Must, 1996; Saarela et al., 2000). The FLEC method is also included in the emission standard (ISO, 2006b).

Several labels for tested, low-emitting materials (RAL UZ 38, RAL-GZ 479, Emicode and GuT, ICL, the Finnish Climate Classification) exist and the harmonisation work on the European level is under work (Wolkoff, 2003; Kephalopoulus et al., 2003). The Finnish Climate Classification, developed by the Finnish Society of Indoor Air Quality and Climate (FiSIAQ), defines design and target values for thermal comfort, ventilation rates, odour intensity, noise levels, the concentration of indoor air pollutants, material emissions, and procedures for construction work in new buildings (S-, M- and P-classes). The target values of low-emitting M1-classified materials for the emission of total VOCs (TVOCs) ($200\,\mu g\,m^{-2}\,h^{-1}$), formaldehyde ($50\,\mu g\,m^{-2}\,h^{-1}$), ammonia ($30\,\mu g\,m^{-2}\,h^{-1}$), carcinogens ($<0.001\,\mu g\,m^{-2}\,h^{-1}$), and odour intensity (the percentage of dissatisfied to the odour less than 15%) are defined for building materials tested at the age of 28 days. The products should also bee casein-free (FiSIAQ, 2001). Though the classification is voluntary, the use of M1-classified, low-emitting materials has markedly increased in new Finnish buildings.

In this study, the emissions from different building structures were determined during the time of construction and during the first year of occupancy in residential buildings. The buildings were built following good modern construction practice, including structure humidity control. The corresponding indoor air concentration results have been published recently (Järnström et al., 2006). The emission results collected from the same buildings are presented here and are aimed to be used as reference values to support the quality control during the time of construction and later when searching for possible defect structures in indoor air problem cases.

2. Methods

2.1. Measurement sites

The measurement sites were chosen in collaboration with three construction partners. Emission measurements were performed in seven apartment buildings (four built on site and three manufactured). Five buildings have mechanical exhaust ventilation (buildings 1, 2, 3, 4 and 7) and two a mechanical supply and exhaust ventilation (buildings 5 and 6). The construction of the buildings lasted for 12–15 months during the years 1999–2002. The buildings are otherwise the same as studied by the authors in another paper (Järnström et al., 2006), except for building 8 where only the indoor air concentrations were measured and, therefore, it is not included in this paper. The emissions of TVOC, ammonia, and formaldehyde were determined for the floor structure during the following construction stages: concrete structure, levelled structure, and covered floor. The levelled floor structure was measured just before the floor covering was installed. The covered floor structure was measured 4 weeks after the installation of the covering to correspond to the material age when the M1-test is performed. The emissions from all surfaces (floor, walls, ceiling) were measured in the newly finished building (usually the bedroom), just before the occupants moved in. The measurements were repeated in the 6- and 12-month-old, inhabited building.

2.2. Materials

All buildings have slab-on-grade concrete construction. Three buildings were built on site and four were constructed from pre-manufactured cored concrete slabs. Low-emitting, M1-classified materials (RTS, 2006) were used in all the buildings. The floor structure was finished with calcium-silicate-based screed products (pH 10.5–11). Fine screed (grain size <0.3 mm, thickness 2–5 mm) was levelled on the on-site built structure. Gross screed (grain size <0.6 mm, thickness 5–50 mm) was used on the pre-manufactured slabs. PVCs (six cushion vinyl sheet products) and parquets (two pre-lacquered products, both oak) were used as flooring materials. Four different adhesives were used in the installation of the PVC materials. In building 7, apartment 2, no adhesive was used in the installation of the floor covering material. The walls were finished with calcium-silicate-based screed with polymeric binding agent (two different base/surface screed combinations, grain sizes 0.6/0.3 mm) and painted (five different products) with the exception of building 4 where wall paper was laid on the screed. Ceilings were finished with the same screed products as used on the walls (two different products). The screed was sprayed on the ceiling structure, resulting in a slightly porous appearance of the surface. The building materials and construction periods are summarised in Table 1. When the construction work was completed, the floor coverings were 8–22 weeks, the ceiling structures 17–27 weeks, and the wall structures 11–26 weeks old.

2.3. Sampling and analysis

Field measurements for the material emissions were performed using the Field and Laboratory Cell (FLEC) technique (ISO, 2006b). The FLEC was first tightened to the surface after which humidified (relative humidity, RH 50%) synthetic air was introduced to the cell at 300–400 ml min^{-1}. The air tightness was checked, i.e., the flow of the air coming out had to be $>95\%$ of the in-coming airflow. Sampling of VOCs was performed onto

Table 1
Measurement sites

Building, apartment, measured room area, total apartment area	Time of construction	Floor structure	Floor covering	Wall	Ceiling
1, apartment 1, 10 m^2, 59.5 m^2	December 1999–August 2000	OSCC	Parquet 1	Levelled and painted concrete/gypsum	Screed, product 1
1, apartment 2, 17 m^2, 40.5 m^2			PVC 1: +adhesive 1[a]		
1, apartment 3, 17 m^2, 40.5 m^2			PVC 2+ adhesive 2		
2, 12 m^2, 78.5 m^2	June 2000–February 2001	MCCS	PVC 3+ adhesive 3[a]		
3, 9.5 m^2, 40 m^2	June 2000–March 2000	OSCC	Parquet 1		
4, 11 m^2, 78 m^2	November 2000–June 2001	OSCC	PVC 4+adhesive 4	Levelled concrete/gypsum and wall paper	
5, 10 m^2, 79 m^2	January 2001–July 2001	MCCS	Parquet 2	Levelled and painted concrete/gypsum	Screed, product 2
6, 11 m^2, 76.5 m^2	May 2001–December 2001	MCCS	Parquet 2		Screed, product 1
7, apartment 1, 25 m^2, 31.5 m^2	June 2001–December 2001	MCCS	PVC 5 +adhesive 1[a]		
7, apartment 2, 25 m^2, 31.5 m^2			PVC 6 (no adhesive)		

OSCC, on-site built, concrete cast; MCCS, manufactured, cored concrete slab.

[a] Not M1-classified.

Tenax TA adsorbent from the out coming air 30–60 min after the air stream was introduced. VOC sampling rate was $100\,\mathrm{ml\,min^{-1}}$ and sample volume was 2–5 l. Parallel sampling was performed. After this, ammonia and formaldehyde were sampled simultaneously into a 0.005 M sulphuric acid solution with impinger. Sampling volume was 100 l/10 ml solution. Field blanks were collected. The FLEC blank was measured on a glass plate at the laboratory before the field measurements, after which the FLEC was sealed in a clean polyethene bag for transport. Field blank results above the limit of quantification were subtracted from the results. The limits of quantification are as follows: VOCs $1–10\,\mu\mathrm{g\,m^{-3}}$, ammonia $5\,\mu\mathrm{g\,m^{-3}}$, and formaldehyde $5\,\mu\mathrm{g\,m^{-3}}$.

The RH of the floor structure was determined with a Vaisala HMP44 moisture detector at 0.4 × structure thickness according to principles outlined by the Finnish Building Information Foundation (RTS, 1998). The surface temperature and RH were registered using a Vaisala HMP42 moisture detector. Tenax tubes were thermally desorbed at 260 °C for 6 min (cold trap temperature −30 °C) by using a PE ATD 400 apparatus and analysed with a HP 5890 series 2 gas chromatograph connected to a HP 5972 mass spectrometer and flame ionisation detector (ISO, 2004). The gas chromatograph was equipped with a double-capillary column HP PONA ($50\,\mathrm{m} \times 0.2\,\mathrm{mm} \times 0.5\,\mu\mathrm{m}$) and the sample was split 1:1 in the two columns, which were connected to MSD and FID detectors. The temperature programme of the GC oven was as follows: 40 °C (5 min) $\Rightarrow$ ($6\,^{\circ}\mathrm{C\,min^{-1}}$) $\Rightarrow$ 280 °C (5 min), total run time 50 min. MSD in SCAN mode was used to identify single VOCs and the FID response was used for quantification. TVOC was calculated as toluene equivalents from the total integrated FID signal between hexane and hexadecane. An external toluene standard was used for calibration. Standards of selected VOCs were analysed to compare retention times and mass spectra. The ammonia concentration was determined with an ion-selective electrode and the analysis of formaldehyde was done with the spectrometric acetyl-acetone method (Salthammer, 1992).

The specific emission rate $\mathrm{SER_a}$ ($\mu\mathrm{g\,m^{-2}\,h^{-1}}$) for the pollutant a was calculated as follows:

$$\mathrm{SER_a} = \frac{\mathrm{Q^*(C_a - C_{a0})}}{\mathrm{A}} \tag{1}$$

Q is the volumetric airflow into the FLEC ($\mathrm{m^3\,h^{-1}}$), $\mathrm{C_a}$ the concentration of pollutant a in the chamber air ($\mu\mathrm{g\,m^{-3}}$), $\mathrm{C_{a0}}$ the concentration of pollutant in the empty chamber ($\mu\mathrm{g\,m^{-3}}$), and A is the surface area ($0.0177\,\mathrm{m^2}$ for the FLEC).

2.4. Statistical analysis

Statistical analyses were performed by using SPSS 12.0 software package for Windows. Descriptive statistics were summarised and the parametric Mann–Whitney U-test was used to evaluate differences in emission levels of TVOC, VOC groups, ammonia, and formaldehyde from the different structures (concrete structure, levelling agent, floor covering). The correlations of emissions with temperature and humidity were investigated for all emission results. In addition, correlations between emissions and building structure humidity were examined. The RH measured in the floor structure was applied for all surfaces. Only statistically significant correlations are presented because of limited space. The normal distribution of the data was analysed with the Shapiro–Wilks W-test ($n < 50$). The W-value is 1 when the distribution is perfectly normal. The W-values for the TVOC/ammonia/formaldehyde emissions in the 12-month-old buildings were as follows: 0.982/0.982/0.727 (parquet), 0.935/0.777/0.976 (PVC), 0.944/0.931/0.623 (painted wall) and 0.862/0.828/0.946 (ceiling). The results were also tested for lognormality. The measured values that were below the limit of detection were marked as the limit of quantification (LOQ)/2 in the statistical analyses (Hornung and Reed, 1990).

3. Results

3.1. Floor structure during the construction stages

3.1.1. Concrete structure before levelling

Higher emissions of TVOC, aldehydes, ketones, esters ($p < 0.05$), and ammonia ($p < 0.01$) were measured from the on-site built structure than from the manufactured concrete slab, whereas the formaldehyde emissions ($15\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$ or less) were similar. The mean TVOC emission was $47\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$ (range $14–118\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$) from the manufactured structure and $146\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$ (range $101–259\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$) from the on-site built structure. Fig. 1 shows that aliphatic hydrocarbons were the most dominating VOCs. The highest emission rates ($35–48\,\mu\mathrm{g\,m^{-2}\,h^{-1}}$) were

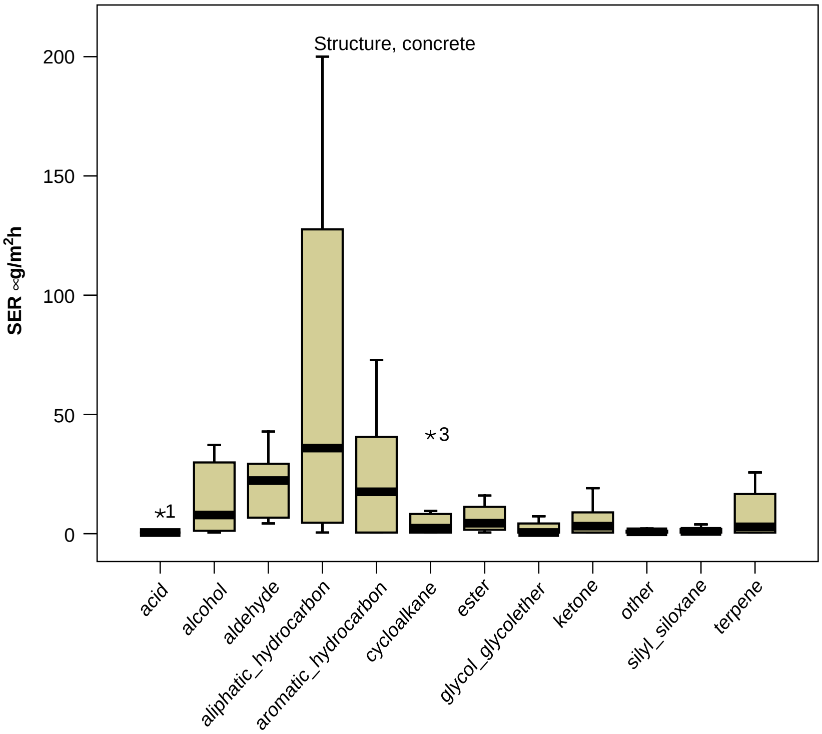

Fig. 1. Specific emission rate (SER, $\mu g\,m^{-2}\,h^{-1}$) of VOC groups measured from the concrete structure (sum of on-site built and manufactured concrete structures). The thick line shows the median value. The 25th and 75th percentile values are signed by the lower and upper end of the boxes. The lines show the minimum and maximum values and the stars stand for extreme values (= cases with values more than three times the interquartile range = 75th percentile−25th percentile).

measured for dodecane and undecane. The mean ammonia emissions were $14\,\mu g\,m^{-2}\,h^{-1}$ (range 4–$24\,\mu g\,m^{-2}\,h^{-1}$) from the manufactured and $70\,\mu g\,m^{-2}\,h^{-1}$ (range 31–$113\,\mu g\,m^{-2}\,h^{-1}$) from the on-site built structures. The TVOC emission correlated positively with the structure temperature ($r = 0.745$, $p < 0.01$). The ammonia emission correlated with the indoor air temperature ($r = 0.703$, $p < 0.05$). The humidity of the floor structure decreased from a level of >90% to 61–72% in 18–20 months after the structure was finished and the building was heated at >15 °C. In the building 1, a RH of 78% was measured after 20 months. This was unusually high since the humidity was 84–87% in the same building (three measurement points) 16 months earlier. A measurement error is a probable explanation for this result.

3.1.2. Levelled structure

Higher TVOC emissions ($p < 0.05$) were measured from the levelled manufactured structures (gross screed) than from the levelled on-site built structures (fine screed). The mean TVOC emissions were $974\,\mu g\,m^{-2}\,h^{-1}$ (range 308–$1990\,\mu g\,m^{-2}\,h^{-1}$) and $287\,\mu g\,m^{-2}\,h^{-1}$ (range 100–$650\,\mu g\,m^{-2}\,h^{-1}$), respectively. The same was true for ammonia and formaldehyde emissions although the differences were not statistically significant. The ammonia emissions were $31\,\mu g\,m^{-2}\,h^{-1}$ (range 14–$71\,\mu g\,m^{-2}\,h^{-1}$) for the levelled manufactured structures and $23\,\mu g\,m^{-2}\,h^{-1}$ (range 12–$39\,\mu g\,m^{-2}\,h^{-1}$) for the levelled on-site built structures. The corresponding mean formaldehyde emissions were $168\,\mu g\,m^{-2}\,h^{-1}$ (range 87–$274\,\mu g\,m^{-2}\,h^{-1}$) and $113\,\mu g\,m^{-2}\,h^{-1}$ (range 50–$175\,\mu g\,m^{-2}\,h^{-1}$). The concentrations of different VOC groups are summarised in Fig. 2. The dominating VOC groups were aliphatic hydrocarbons and glycols/glycolethers for both levelling agents. Although the concentrations of most VOC groups were higher from the structures with gross screed the differences were not statistically significant. The TVOC emission correlated positively with the surface temperature ($r = 0.751$, $p < 0.05$) and the formaldehyde emission with the indoor air humidity ($r = 0.842$, $p < 0.01$).

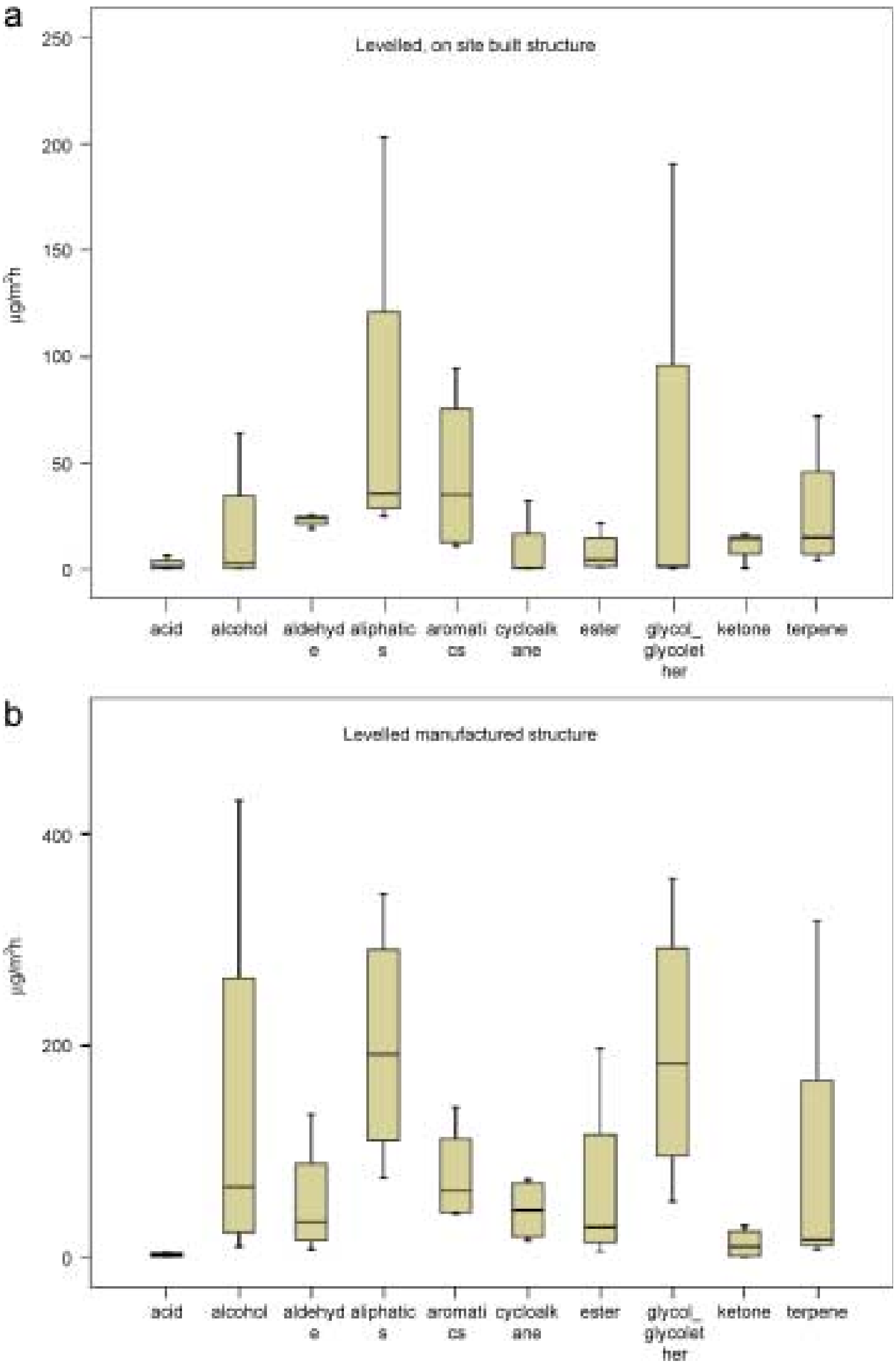

Fig. 2. (a, b) Specific emission rate (SER, $\mu g\,m^{-2}\,h^{-1}$) of VOC groups measured from the levelled concrete structure built on site and manufactured concrete structure before the floor covering was installed. The explanation for the boarders is shown in Fig. 1.

3.1.3. Four-week-old floor covering

The mean TVOC emission was 686 $\mu g\,m^{-2}\,h^{-1}$ (range 40–2070 $\mu g\,m^{-2}\,h^{-1}$) from the PVC-coated structures and 120 $\mu g\,m^{-2}\,h^{-1}$ (range 104–157 $\mu g\,m^{-2}\,h^{-1}$) from the parquet. The mean ammonia emission measured from the PVC was

$28\,\mu g\,m^{-2}\,h^{-1}$. It showed a large variation ranging from 5 to $69\,\mu g\,m^{-2}\,h^{-1}$. No ammonia emissions above $5\,\mu g\,m^{-2}\,h^{-1}$ were measured from the parquet. The formaldehyde emission was below $10\,\mu g\,m^{-2}\,h^{-1}$ for both structures. The highest emissions of individual VOCs measured from the PVC included 2-(2-butoxyethoxy) ethanol acetate (mean emission $250\,\mu g\,m^{-2}\,h^{-1}$, range 2–750 $\mu g\,m^{-2}\,h^{-1}$) and 2-phenoxy-ethanol (mean emission $471\,\mu g\,m^{-2}\,h^{-1}$, range 3–1405 $\mu g\,m^{-2}\,h^{-1}$). Aldehydes were typical for the parquet floor covering. The TVOC emission from the parquet correlated positively with the surface temperature ($r = 0.998$, $p < 0.01$).

3.2. Structures in the newly finished and 6- and 12-month-old buildings

3.2.1. Floor covering

The mean TVOC emission measured from the floor structure was ~100–930 (PVC) $\mu g\,m^{-2}\,h^{-1}$ and ~15–130 $\mu g\,m^{-2}\,h^{-1}$ (parquet) in the newly finished buildings (Table 2). The emissions from PVC-covered floors decreased rapidly being less than $350\,\mu g\,m^{-2}\,h^{-1}$ after 6 months and less than $200\,\mu g\,m^{-2}\,h^{-1}$ after 1 year in all the apartments. The mean ammonia emission from the PVC was about $15\,\mu g\,m^{-2}\,h^{-1}$ in the newly finished building and it remained at 10–15 $\mu g\,m^{-2}\,h^{-1}$ during the first year. The mean ammonia emission from the parquet flooring was on the level of $10\,\mu g\,m^{-2}\,h^{-1}$ in the newly finished building and decreased to $5\,\mu g\,m^{-2}\,h^{-1}$ during the first year. The formaldehyde emissions from both the PVC and parquet structure remained below $10\,\mu g\,m^{-2}\,h^{-1}$ during the whole measurement period.

Glycolethers, especially 2-(2-butoxyethoxy) ethanol and its acetate, were the main compounds measured from the PVC flooring. Higher emissions of aliphatic hydrocarbons, alcohols, and esters were measured from the PVC ($p < 0.05$) than from the parquet in the newly finished buildings. The difference in the emission levels decreased within the first year after completion and the final mean VOC levels were about 2–10 $\mu g\,m^{-2}\,h^{-1}$ for both PVC and parquet floor coverings. However, there were a few VOCs that were characteristic for PVC flooring in some of the apartments. These include TXIB (2,2,4-trimethyl-1,3-pentanediol diisobutyrate, mean emission rate $25\,\mu g\,m^{-2}\,h^{-1}$ in two apartments) and 2-phenoxy ethanol (emission rate $37\,\mu g\,m^{-2}\,h^{-1}$ in one apartment). Alkyl benzenes (C4–C10 substituted) were measured from three PVCs (average emission $10\,\mu g\,m^{-2}\,h^{-1}$). The emission of alkylbenzenes outside the TVOC range from PVC 6 was remarkabale, $150\,\mu g\,m^{-2}\,h^{-1}$. The esters ($p < 0.05$) were higher in the PVC emissions and the ketones ($p < 0.05$) in the parquet emissions in the 6-month-old buildings. The alcohol emissions were higher from the PVC ($p < 0.05$) in the 12-month-old buildings. The emission of various VOC groups from PVCs 3,4,5 and 6 decreased 25–87% during the first 6 months. The emissions measured from PVC types 5 and 6 showed no significant differences in the building 7, although the latter was installed without adhesive. The correlations of the emissions with the surface and structure temperature and the RH are summarised in Table 3.

3.2.2. Ceiling

The TVOC emission ranged from 200 to $1350\,\mu g\,m^{-2}\,h^{-1}$ in the newly finished buildings. The lowest emissions, 200 and $376\,\mu g\,m^{-2}\,h^{-1}$, were measured in buildings 5 and 6 although they had the newest structures (17-week-old). These buildings had mechanical supply and exhaust air systems. Dominating VOC groups included aliphatic and aromatic hydrocarbons, aldehydes, and glycols/glycolethers. The main VOCs included 2-(2-butoxyethoxy) ethanol and its acetate and xylenes. The TVOC emissions decreased to less than $600\,\mu g\,m^{-2}\,h^{-1}$ in 6 months and to less than $300\,\mu g\,m^{-2}\,h^{-1}$ in 12 months (Table 2). The mean ammonia emission varied from 45 to $51\,\mu g\,m^{-2}\,h^{-1}$ during the first year. The mean formaldehyde emission decreased from 42 to $28\,\mu g\,m^{-2}\,h^{-1}$ in the same time. The highest emission of $109\,\mu g\,m^{-2}\,h^{-1}$ was measured in the 6-month-old building 7, apartment 1. The correlations between the emissions and temperature and RH are summarised in Table 3.

3.2.3. Walls

The TVOC emission measured from the painted wall structure varied widely, from 15 to $1000\,\mu g\,m^{-2}\,h^{-1}$, in the newly finished buildings. The lowest emissions of 14–100 $\mu g\,m^{-2}\,h^{-1}$ were measured from the 12- and 16-week-old walls painted with two different products in the same buildings 5 and 6 where the lowest ceiling emissions were also detected. The highest emission was measured from a 23-week-old structure where re-painting had probably occurred although this was not reported. Aliphatic and aromatic hydrocarbons as well as glycols and

Table 2
The mean value and range for TVOC, VOC groups, formaldehyde and ammonia emissions measured from the structures in the 0–12-month-old buildings

Specific emission rate SER ($\mu g\,m^{-2}\,h^{-1}$)

Compound/compound group	Floor covering: PVC			Floor covering: parquet			Ceiling			Walls		
	0 month	6 months	12 months	0 month	6 months	12 months	0 month	6 months	12 months	0 month	6 months	12 months
Acid	16, 7–22	10, 2–18	9, 3–17	11, 4–20	5, 3–7	4, 3–5	8, <1–15	8, 3–14	7, <1–14	8, 3–23	5, 4–7	4, 1–7
Alcohol	27, 12–50	19, 4–38	13, 3–28	6, 3–8	8, 1–22	3, <1–7	59, 24–139	27, 7–45	21, 3–48	8, 1–36	6, 1–26	3, 1–5
Aldehyde	15, 4–28	11, 1–31	14, 2–29	16, 2–25	7, 2–10	5, 1–10	58, 32–86	48, 24–82	25, 3–45	13, 4–71	5, 1–10	4, 1–6
Aliphatic hydrocarbon	19, 5–28	10, 1–23	20, 3–48	2, 2–2	33, 1–11	2, 1–3	88, 26–165	29, 9–60	21, 3–41	24, 1–185	5, 1–15	8, 1–17
Aromatic hydrocarbon	51, 12–137	12, 4–25	12, 3–36	20, 2–65	2, 1–2	1, 1–1	112, 46–165	34, 7–69	7, <1–22	28, 1–204	4, 3–5	1, 1–1
Cycloalkane	2, 1–3	2, 1–5	n.d.	6 (n = 1)	n.d.	n.d.	9, 2–17	5, 5–5	1, <1–1	13, 1–32	1, n = 1	n.d.
Ester	178, 19–497	23, 3–79	15, 3–48	7, 4–13	5, 3–8	6, 4–10	37, 6–167	17, 5–46	8, 2–24	25, 2–158	7, 2–20	5, 2–12
Glycol/glycolether	90, 16–235	48, 4–110	20, 1–43	19, 1–51	4, 2–9	5, 2–8	98, 5–399	28, 6–100	20, 5–36	68, 5–183	11, 1–65	9, 1–24
Ketone	9, 1–16	3, 1–4	6, 1–19	20, 7–29	22, 13–29	6, 3–9	28, 8–67	13, <1–29	4, <1–12	4, 1–14	9, 1–45	2, 1–4
Terpene	5, 3–8	6, 3–8	2, n = 1	4, 1–7	2, 1–4	2, 2–2	42, 8–109	116, 12–297	28, <1–62	40, 1–224	2, 1–6	1, 1–2
TVOC	431, 104–926	136, 45–336	117, 30–191	78, 16–129	80, 18–184	28, 12–47	651, 200–1338	286, 123–539	182, 26–260	200, 14–997	51, 10–123	36, 9–76
Ammonia	13, 6–18	7, <5–16	14, <5–37	10, 8–12	10, <5–16	5, <5–6	45, 24–63	51, 21–86	49, 20–114	13, 6–26	9, 2–20	10, <5–19
Formaldehyde	9, <5–18	5, <5–10	7, 6–10	7, <5–10	5, <5–6	5, <5–8	42, 5–96	42, 14–109	28, 13–46	7, <5–11	13, <5–37	9, <5–20

n.d. = not detected.

Table 3
Statistically significant (*$p<0.05$, **$p<0.01$) correlations between TVOC, VOC groups, ammonia, formaldehyde and RH (surface and structure) and temperature (surface and structure) in the 0–12-month-old buildings

	0 month	6 months	12 months
PVC	RH surface and Aldehydess: 0.979* Terpenes: 0.974*	RH surface and Aldehyde: 0.832* Aromatic comp.: 0.888* Formaldehyde: 0.984** T surface and Ester: 0.821*	No correlations $p<0.05/0.01$
Parquet	T surface and Aromatic comp.: 0.965* RH structure and TVOC: 0.979* Ketones: 0.986*	No correlations $p<0.05/0.01$	RH surface and Aliphatic comp.: 0.970*
Painted gypsum walls	No correlations $p<0.05/0.01$	No correlations $p<0.05/0.01$	T surface and Acids: −0.762* Glycol/glycolether: 0.808* RH surface and Glycol/glycolether: 0.865*
Painted concrete walls	T structure Acids: 0.998*	RH surface and Ammonia: 0.784* Glycol/glycolether: 0.782*	T structure and Aldehydes: −0.773*
Ceiling	T surface and Aldehyde: 0.821* RH surface and Cycloalkanes: 0.823* RH structure and Aromatic comp.: 0.717*	No correlations $p<0.05/0.01$	RH structure and Formaldehyde: 0.642*

glycolethers were the major VOC groups. Among the individual compounds, 2-(2-butoxyethoxy) ethanol and its acetate were again the most important ones. The mean TVOC emission level decreased to the level of 50 $\mu g\,m^{-2}\,h^{-1}$ within the first 6 months (Table 2). The ammonia emission remained below 30 $\mu g\,m^{-2}\,h^{-1}$ in all the buildings during the first year. The mean formaldehyde emission was about 10 $\mu g\,m^{-2}\,h^{-1}$ and varied from less than 5–40 $\mu g\,m^{-2}\,h^{-1}$. The TVOC measured from the wall structure on which wall paper was laid (building 4) was 230 $\mu g\,m^{-2}\,h^{-1}$. Its ammonia emission was on the level of 70 $\mu g\,m^{-2}\,h^{-1}$ and formaldehyde emission 40 $\mu g\,m^{-2}\,h^{-1}$. The correlations between the emissions and the temperature and RH are presented in Table 3.

4. Discussion

4.1. Floor structure

In general, higher emissions were measured from the on-site built structures than from the manufactured structures. The TVOC and formaldehyde emissions remained within M1-class (200/50 $\mu g\,m^{-2}\,h^{-1}$) for both structures whereas the mean ammonia emission from the on-site built structure exceeded the M1-target value ($<30\,\mu g\,m^{-2}\,h^{-1}$) by two-fold. The ammonia emission from the manufactured structure was within the M1-class requirements. Much higher VOC and formaldehyde emissions were measured from the floor structure levelled with gross levelling agent than from the structure treated with fine levelling agent. The reason for this difference was probably the thicker layer of gross levelling.

The emission measurements performed after the completion of floor construction showed that the emission was affected by all its components, i.e., the structure, levelling agent, adhesive, and floor covering material. The emission also depended on the time elapsed before the floor covering was installed on the levelled structure. This time varied from <1 week to 4 weeks. The TVOC emissions were quite high (~250–650/~2000 $\mu g\,m^{-2}\,h^{-1}$) from the structures levelled with the fine/gross screed, when the structure was measured 1 day after the levelling. The M1-target value for TVOC was reached in two buildings in which the fine screed had been levelled 1 and 3 weeks earlier. The highest TVOC emissions were measured from 4-week-old floors coated with three PVC products (1, 3 and 4). These products were concluded to be permeable to VOCs originating from the underlying structure based on emission tests done with the adhesive (not presented here) on a glass plate in laboratory. The laboratory tests showed that 2-(2-butoxyethoxy) ethanol acetate and 2-phenoxy ethanol were the most important compounds. Those compounds also dominated in the emission of 4-week-old complete floor structure but were absent in the emissions from single PVCs. These emissions clearly exceeded the M1-target level of 200 $\mu g\,m^{-2}\,h^{-1}$ for the TVOC emission. On the other hand, the emission of TVOC from an other PVC product (2) was very low, about 100 $\mu g\,m^{-2}\,h^{-1}$ during the whole 1-year follow-up period. This result agrees with the finding done by Wilke et al. (2004) who studied emissions from complete floor structures in the laboratory. The TVOC emission from the permeable PVC structures also reached the M1 target level during the first year of occupancy. The decrease of the TVOC emission was fast especially during the first 6 months (68%). This emission decreased further during the next 6 months but slower (19%). The TVOC emission from the parquet remained within M1-class all the time. The formaldehyde and mean ammonia emissions measured from the different floor covering materials were within the M1-class requirements during the whole follow-up period.

Secondary emissions can appear from PVC coverings and adhesives under humid conditions (Gustafsson, 1990). In this study, the emission of a known degradation product, 2-ethyl-hexanol, was on the average level of 14 $\mu g\,m^{-2}\,h^{-1}$ from the 4 week-old PVC floorings (six products, max: 28 $\mu g\,m^{-2}\,h^{-1}$). In the 12-month-old buildings, the average emission from the same PVCs was 9 $\mu g\,m^{-2}\,h^{-1}$ (six products, max: 19 $\mu g\,m^{-2}\,h^{-1}$). In addition, 2-ethylhexanol was measured from the parquet but these emissions were less than 5 $\mu g\,m^{-2}\,h^{-1}$. No correlations of these compounds with structure RH was found in this study. Emissions of 2-ethylhexanol are also measured from single PVC materials and adhesives at laboratory conditions (Lundgren et al., 1999; Wilke et al., 2004).

4.2. Ceiling

The mean TVOC emission from the ceiling structure was clearly above the M1-class limit in the newly finished building. Lowest emissions were measured in two buildings provided with mechanical supply and exhaust air system. This emission decreased fast (56%) during the first 6 months and the M1-class limit was, in general, reached within the first year. Terpene emissions increased in nine apartments out of ten during the measurement period. The increase is probably due to wooden furniture installed in the apartments. The levelled ceiling structure has a slightly porous character and consequently, sorption processes are there more important than on the other surfaces (Neretnieks et al., 1993). As many as 35 new VOCs (occurring at least in three buildings) were identified in the 6- and 12-month buildings. The highest ammonia emissions were released from ceilings in the newly finished buildings. These emissions were generally above the M1-target value. In addition, the ammonia emission increased during the 6–12-month follow-up measurements.

Formaldehyde emissions showed large variations and emissions up to 100 $\mu g\,m^{-2}\,h^{-1}$ were measured in the newly finished and 6-month-old buildings. However, the M1-target value was reached in all buildings during the first year. The seasonal variation of the RH may have affected the formaldehyde emission as been found in earlier studies (Reponen et al., 1991). No correlation could be found between indoor air RH and formaldehyde emission in this study but this may be due to the fact the emission measurements were always done at the RH of 50%. On the other hand, the RH of indoor air did affect some other emissions.

4.3. Walls

The TVOC emissions from the painted walls (concrete/gypsum board) were within the M1-target value in the newly finished buildings. The TVOC emission from the wall structure, which was finished with wall paper (building 4), was high in the new building but reached the M1-target value during the first year. Similar difference appeared in the ammonia and formaldehyde emissions. The ammonia emissions from the painted wall fulfilled M1-class requirements but those from the wall paper structure were on the level of 40–60 $\mu g\,m^{-2}$. The formaldehyde emissions from the painted wall structure were generally within the M1-class during the entire measurement period. The formaldehyde emission from the wall, which was finished with wall paper, remained on the level of 40 μg m^{-2} h^{-1}. The higher emissions measured from the wall paper structure were probably due to the adhesive.

5. Conclusions and implications, reference values

The material classification systems are useful to facilitate the selection of low-emitting building materials. However, this does not guarantee good indoor air quality because the classifications are based solely on laboratory tests of individual materials. The emissions are known to change after installation due to interactions with other materials and the environment in the building. In addition, ageing, occupancy, and moisture damage affect the emissions.

The emissions from structures were found to change rapidly during the first year, especially during the first 6 months. Generally, the emission rates decreased but emissions of new compounds also appeared due to occupancy.

Reference values for concentrations of microbes were presented in Finland already in 1992. These values have been found to be very useful while interpreting measurement data. Earlier, we suggested similar limits for airborne concentrations of TVOC, formaldehyde, ammonia, and ten VOC groups. The corresponding reference values are now presented in Table 4 for emissions from structures in the 1-year-old building. The mean emission rate measured from the structure is again used as the "normal value" and the 95th percentile is the "abnormal limit" value. The 12-month-values represent the targets which can be achieved in new buildings after the initial drying phase. The results can be used both in quality control and to verify the role of chemical emissions in suspected problem cases.

The emission results presented here confirm that the Finnish material classification system works in real buildings. Nevertheless, it could be developed further. For example, the incorporation of target values for VOC groups and critical VOCs (e.g., 2-ethylhexanol and TXIB) in the material classification would be advantageous from the consumers point of view. In addition, supportive testing of material combinations as real structures would be beneficial.

The target value for TVOC was reached well for the painted wall structure and parquet floor but not as well for the PVC floor or ceiling structure. Formaldehyde target values were reached well for both floor structures and walls and quite well for the ceiling structure. The target value for ammonia emission from the ceiling structure was clearly exceeded in some cases.

Table 4
Reference values for the emission of organic pollutants measured from structures in the 12-month-old building

	Specific emission rate (μg m^{-2} h^{-1})							
	PVC (n = 6)		Parquet (n = 4)		Levelled ceiling (n = 10)		Painted wall (n = 9)	
	Normal value	Abnormal value	Normal value	Abnormal value	Normal value	Abnormal value	Normal value	Abnormal value
Acid	10	15	5	10	10	15	5	10
Alcohol	15	25	5	10	25	35	5	10
Aldehyde	15	25	5	10	25	35	5	10
Aliphatic hydrocarbon	20	40	<5	5	20	30	10	15
Aromatic hydrocarbon	15	25	<5	5	10	15	<5	5
Cycloalkane	<5	5	<5	5	<5	5	<5	5
Ester	15	30	5	10	10	15	5	10
Glycol/glycolether	20	45	5	10	20	30	10	15
Ketone	10	20	5	10	5	10	<5	5
Terpene	<5	5	<5	5	30	45	<5	5
TVOC	120	170	30	45	180	230	40	50
Ammonia	15	25	<5	5	50	70	10	15
Formaldehyde	5	10	5	10	30	40	10	15

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PUBLICATION III

The impact of emissions from structures on indoor air concentrations in newly finished buildings: predicted and on-site measured levels

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The impact of emissions from structures on indoor air concentrations in newly finished buildings – predicted and on-site measured levels

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Short title: The impact of emissions from structures

Key words: new buildings, emissions, indoor air, VOCs, formaldehyde, ammonia

Abstract

The impact of surface emissions (VOCs, ammonia and formaldehyde) on the indoor air quality (IAQ) in newly established buildings was examined. Six apartment buildings, in which low-emitting, classified building materials were used, were investigated 0, 6, and 12 months after completion of construction work. The predicted indoor air concentrations based on the on site measured emissions (floor, walls, ceiling) and air exchange rates were in general lower than the measured indoor air concentrations. The contribution of other sources was on the average about 50% for VOCs and 25–35% for ammonia and formaldehyde. The ceiling structure was the main source of pollutants throughout the first year. PVC floor covering affected IAQ significantly in the newly finished buildings. The contribution of walls was relatively low in spite of large surface area. The impact of VOC emissions from surfaces on indoor air clearly decreased during the follow-up period whereas the impact of ammonia and formaldehyde emissions remained about the same. Higher SVOC

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concentration was measured in the buildings with PVC flooring compared to the rooms with parquet flooring in the 0–6-month-old buildings.

1. Introduction

The impact of pollutant emissions from new materials on indoor air quality (IAQ) is considerable and consequently, evaluation procedures, and measurement techniques for material emissions have been developed [1, 2, 3, 4]. Classification and labelling systems have been established for building materials in many European countries [5, 6]. Different approaches have been adopted, for example, in Germany and Denmark. In the German AgBB system, individual VOC compounds are considered on the basis of lowest concentration of interest (LCI)-values in addition to total volatile organic compounds (TVOC), semi-volatile organic compounds (SVOC), and carcinogens [7]. Available toxicological information and odour thresholds are utilized in the Danish Indoor Climate labelling system [8].

The aim of the Finnish Indoor Classification is to encourage the use of low-emitting building materials and air handling components by providing target values for emissions and airborne concentrations of pollutants. These target values are based on practical experience of levels that are possible to achieve by careful material selection. Even though the health effects have not been considered as such, the aim to maintain airborne pollutant concentrations as low as possible is naturally beneficial for health as well. The target values for TVOC, ammonia, and formaldehyde emissions for M1-classified material are 200, 30, and 50 $\mu g/m^2h$, respectively, tested at the age of 28 days [9]. The use of M1-classified materials shall imply that the S1 target values for IAQ are also reached. The corresponding S1-values are 200, 30, and 30 $\mu g/m^3$. The S1-requirement for the air flow is 12 l/s/ person [9]. The first version of the Finnish classification from the year 1995 also included air flow rates per floor area to ensure control of building material emissions. This was removed from the present version because the use of low-emitting materials was considered to be sufficient and ventilation would be needed only to control human effluents. The testing of furniture and cleaning agents is under development [10].

The IAQ in new or renovated buildings has been investigated in many studies. The decay of pollutants is fastest within the first months and the concentration levels have shown to approach those of established buildings within 2 years [11, 12, 13, 14].

SVOCs have been under interest in recent years considering IAQ and health effects. Because of their low vapour pressure, SVOCs are adsorbed on surfaces and particles and, consequently, their indoor concentration is usually very low, 10 µg/m^3 or less [15]. On the other hand, SVOC emissions tend to last for a long time [16].

Attempts to identify VOC sources have been done in earlier studies [17, 18]. A mass balance model was used to study indoor and outdoor contributions to carbonyl concentrations in the United States. Of ten carbonyls investigated, formaldehyde and acetaldehyde had the strongest indoor sources [17]. Principal component analysis was applied for VOC source identification for personal and residential microenvironment samples in Helsinki within the European exposure (EXPOLIS) study. Variability in residential indoor VOC concentrations was associated with indoor cleaning products, traffic emissions, long-range transport of pollutants, and product emissions [18].

The results presented in this paper are based on indoor air concentrations and on-site measured material emission for TVOC, VOC groups, ammonia, and formaldehyde in newly established residential buildings with low-emitting materials, which have been reported recently [19, 20]. The contribution of pollutants from different structures (floor, walls, and ceiling) to indoor air concentration levels are summarised during the first year after the building has been taken into use. Aspects on the further development of the material emission classification are also presented.

2. Materials and methods

The studied buildings with low- emitting, M1-classified building materials and measurement schemes as well as sampling and analysis techniques have been previously presented in detail [19, 20]. The numbering is kept consistent with the earlier publications. Building 3 is not included since only the floor structure was investigated in this building. The field measurements were usually done in the bedroom. The room sizes and the air exchange rates are shown in Table 1. The ventilation rate measurements were done with Alnor AXD-530/ Swema Air 300/ Alnor GGA 26 pressure difference meters. The pressure differences were measured for the exhaust air vents.

Table 1. Study buildings: room sizes and air exchange rates in the 0-, 6- and 12-month-old buildings

Building	**Floor area**	**Wall area**	**Ceiling area**	**Room volume**	**Air exchange rate 0/6/12 month-old**		
	m^2	m^2	m^2	m^3	h^{-1}		
1, apartment 1	10	21	11.5	26.5	1.06	1.26	0.87
1, apartment 2	12.5	24	14	32.5	1.61	1.45	1.31
1, apartment 3	12.5	24	14	32.5	1.47	1.31	1.39
2	12	25	13	30	1.01	0.93	0.97
4	11	22	13	30	0.82	0.82	0.82
5	10	24	10.5	24	1.0	1.02	0.98
6	11	30	12	27.5	0.91	0.94	0.73
7, apartment 1	23	39.5	28	67.5	1.03	0.95	0.80
7, apartment 2	23	39.5	28	67.5	0.99	0.93	0.83
average	14	28	16	38	1.08	1.03	0.97

The airborne concentrations and emissions of volatile organic compounds, ammonia, and formaldehyde were determined as described earlier [19, 20]. In addition, the concentrations of SVOCs were calculated by summarising the total integrated FID area after hexadecane and given as toluene equivalents. Background levels of diethylphtalate (average amount of 5 ng/tube), originating from the thermal desorption unit, were subtracted from the results.

The expected concentration C for the pollutant *a* (µg/m^3) originating from the building structures was calculated as follows:

$$C_a = \frac{L * SER_a}{n} \qquad (1)$$

n is the air exchange rate in the room (h^{-1}), SER_a is the specific emission rate (µg/m^2h), and *L* is the surface-to-volume ratio (loading factor, m^2/m^3) for the measured surface. The average surface-to-volume ratios for the structures were as follows: floor covering 0.39 m^2/m^3, ceiling 0.44 m^2/m^3, and walls 0.81 m^2/m^3 (doors, windows and closets excluded).

Statistical analyses and principal component analysis (PCA) were performed by using SPSS 12.0 software package for Windows. The parametric Mann Whitney U-test was used to evaluate differences in SVOC concentration levels.

3. Impact of emissions from surfaces on indoor air concentration

3. 1 TVOC, ammonia and formaldehyde

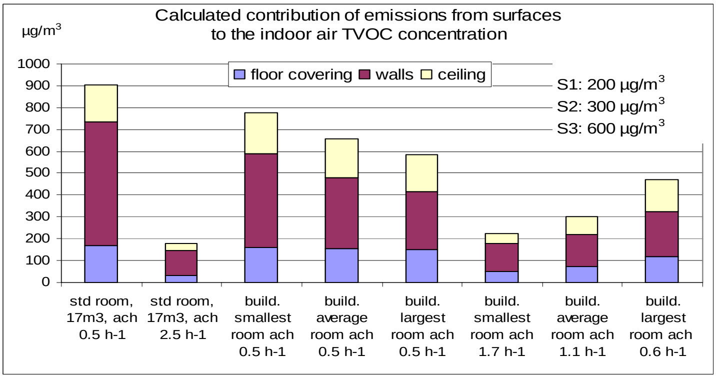
The theoretical impact of various structure emissions on the airborne TVOC, ammonia, and formaldehyde concentrations if the emissions were on the M1-limit levels are shown in figures 1a–c. The calculations have been performed for four different room sizes with the air exchange rate of 0.5h^{-1} (minimum required ventilation rate for residential buildings as given in the national building code [21]) and 0.6–2.5 h^{-1} (S1-requirement with one person in the room [9]): 1) the standard room [2, 3] , i.e., the smallest allowed room (area 7m^2, height 2.4 m, volume 17 m^3), 2) the minimum, 3) average, and 4) maximum room sizes in the buildings studied.

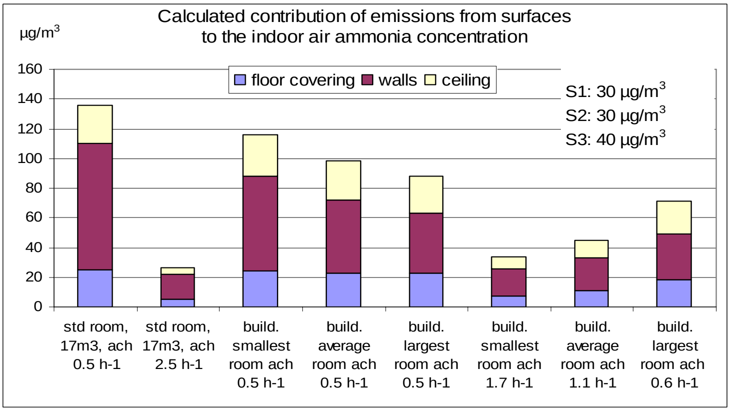
The predicted concentration levels of TVOC, ammonia, and formaldehyde for the smallest (standard) room with an air exchange rate corresponding to the S1-requirement (2.5 h^{-1}) would be 178, 27, and 44 µg/m^3, respectively. The concentrations of TVOC and ammonia are within the S1-target values (200/30 µg/m^3). The concentration of formaldehyde fulfills the S2-criterion (50 µg/m^3). The corresponding predicted values were 302 µg/m^3 (TVOC), 45 µg/m^3 (ammonia), and 75 µg/m^3 (formaldehyde) for the average room size in the buildings studied. However, it would be natural to use two persons/room as the design basis for these larger rooms. In this way, S1-criteria will be again fulfilled for TVOC and ammonia and S2-criterion for formaldehyde. However, the concentrations calculated with the minimum required ventilation rate (0.5h^{-1}) would be on the average even above the S3-levels (600/40/100 µg/m^3).

The contributions of TVOC, ammonia, and formaldehyde based on the measured emissions and air exchange rates in the 0, 6, and 12-month old buildings are presented in Table 2. The measured indoor air concentrations are shown in the same table. The average predicted and measured concentrations are summarized in Figures 2a–c. The TVOC results are also divided according to the ventilation system (mechanical exhaust air system and mechanical supply and exhaust air system).

The average contribution of TVOC emission from the interior surfaces (floor, walls walls, and ceiling) was 538 µg/m^3 in the newly finished buildings (the average air exchange rate of 1h^{-1} was used in the calculations). This was 57% of the measured indoor air average concentration (~900 µg/m^3), which indicates the presence of other sources. The largest difference between predicted and measured concentrations was observed in the building 7 (two one-room apartments with open-to-the -room kitchens). Possible sources include structures which were not measured (e.g., wooden window frames, skirting etc.) and kitchen furniture. The contribution of TVOC emissions from surfaces decreased to 225 µg/m^3 in six months and to 170 µg/m^3 in twelve months (Fig. 2a). These values were 59% and 54% of the measured mean TVOC concentrations. The TVOC emission from surfaces had higher impact on the indoor air concentrations in two buildings with mechanical supply and exhaust systems being 73%, 91%, and 76% of the TVOC concentration in the 0, 6, and 12-month-old buildings, respectively.

The contributions of ammonia and formaldehyde emissions from main interior structures were much lower than the theoretical values in Fig. 1b and 1c, less than 45 µg/m^3 for ammonia and below 40 µg/m^3 for formaldehyde (Fig. 2b and 2c). Also, ammonia and formaldehyde had other, non- identified sources that were responsible for about 25–35% of the indoor air concentration. No significant differences in the ammonia or formaldehyde concentrations were observed between the two air exchange systems. The average correlation between calculated and measured concentrations in the 0–12 month-old separate apartments were 0.77 for TVOC (range 0.16–0.99), 0.50 for ammonia (range 0.24–0.89), and 0.73 for formaldehyde (range 0.02–0.99).





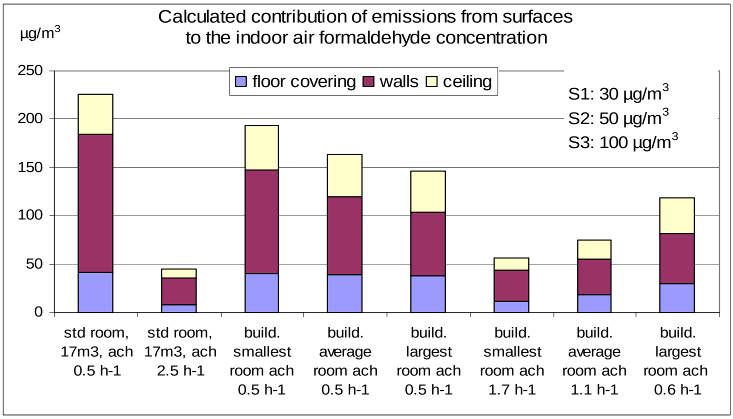


Figure 1a–c. Contribution of the emissions from surfaces to the indoor air TVOC, ammonia, and formaldehyde concentration as calculated for the standard room (V = 17 m^3) and the study buildings (min, max, and average) with SER = 200/30/50 µg/m^{2}h (M1- class target values [9]), ACH = 0.5–2.5 h^{-1}. The S1 target values are also shown [9].

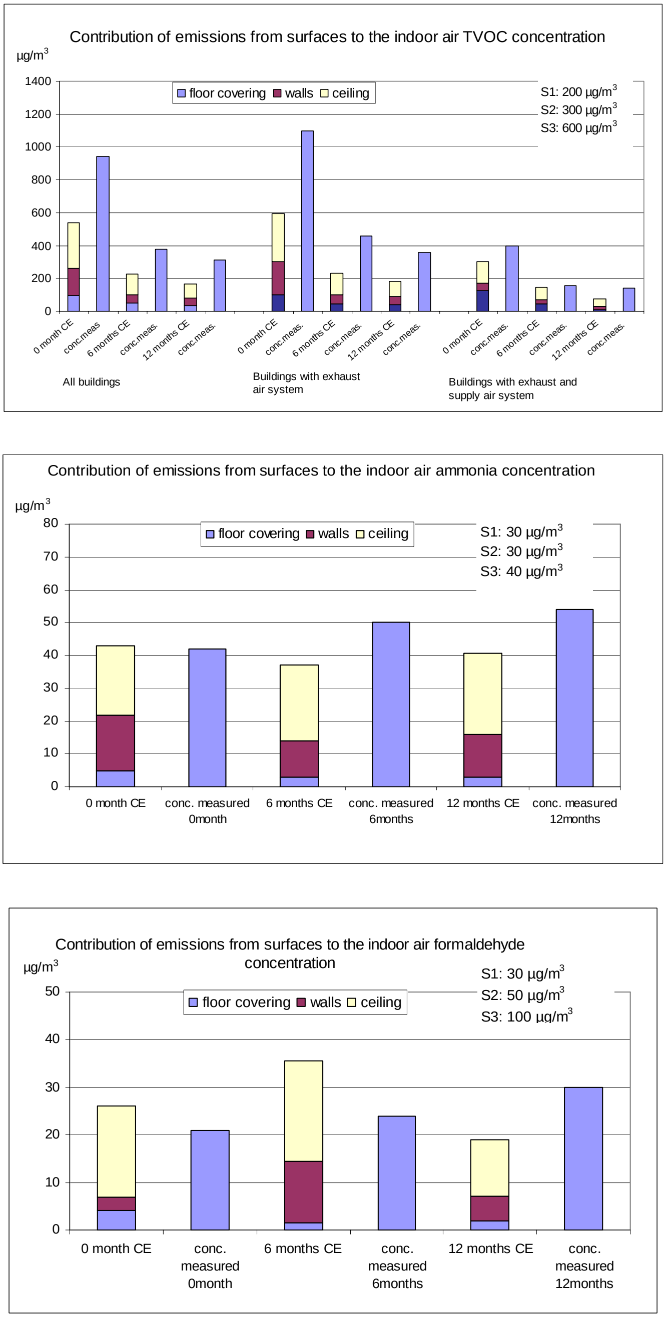


Figure 2a–c. Contributions of the emissions from surfaces to the indoor air TVOC, ammonia, and formaldehyde concentrations based on the on-site measured emissions, air exchange rates, and the airborne concentrations in the 0-, 6- and 12-month-old buildings. The S1 target values are also shown [9].

Table 2. Contribution of TVOC/ammonia/formaldehyde emissions from surfaces in the 0–12 -month-old buildings- measured and calculated values ([a] only VOCs measured, [b] only VOCs and ammonia measured from surfaces).

TVOC	**Measured indoor air concentration (μg/m^3)**	**Calculated indoor air concentration (μg/m^3)**	**Emission contribution (μg/m^3)**			
			floor	**concrete wall**	**gypsum wall**	**ceiling**
	0-month-old building (no inhabitants)					
Build.1,ap.1[a]	1068	448	47	118	59	223
Build.1,ap.2[a]	865	275	42	43	22	167
Build.1,ap.3[a]	1007	514	19	332	34	195
Build. 2	730/37/37	950/29/32	361/5/3	81/3/2	121/4/ 3	388/17/24
Build. 4	1300/53/17	1066/85/5	210/7/1	287/27/1	265/19/1	303/32/1
Build. 5	335/53/20	272/22/19	29/3/4	26/4/2	63/5/4	156/10/8
Build. 6	311/35/13	121/27/16	7/6/4	8/4/3	12/4/3	94/14/7
Build. 7, ap.1	755/38/19	411/50/38	39/7/4	20/5/2	40/12/4	310/27/29
Build. 7, ap.2	2106/37/20	790/51/58	95/4/7	21/5/2	42/12/4	631/30/45
avergae	**942/42/21**	**538/44/28**	**94/5/4**	**104/8/2**	**73/9/3**	**274/21/19**
	6-month-old building (inhabited)					
Build.1,ap.1[b]	449/58	159/27	25/2	39/4	11/2	84/18
Build.1,ap.2[b]	174/43	106/9	47/<1	13/1	6/<1	39/8
Build.1,ap.3[b]	396/22	223/14	21/1	19/2	9/1	174/11
Build. 2[b]	1013/63	498/24	142/1	43/5	65/8	247/10
Build. 4	173/46/22	332/85/57	40/6/3	70/23/17	58/18/17	165/38/21
Build. 5	130/41/17	75/28/9	15/7/1	3/2/1	6/4/2	50/15/6
Build. 6	306/77/17	234/54/18	77/3/1	38/11/1	24/1/9	96/39/8
Build. 7, ap.1	550/49/32	211/44/73	18/4/1	8/4/1	15/3/18	170/33/54
Build. 7, ap.2	212/47/28	188/53/20	44/7/1	18/2/1	40/5/1	86/38/17
average	**378/50/23**	**225/38/35**	**48/3/1**	**28/6/4**	**26/5/9**	**123/23/21**
	12-month-old building (inhabited)					
Build.1, ap.1	523/84/50	235/51/22	15/4/2	31/14/2	67/3/2	122/30/15
Build.1, ap.2	204/57/52	72/17/14	35/<1/3	4/4/1	5/1/1	28/12/9
Build.1, ap.3	475/92/47	125/25/13	17/1/1	15/6/1	11/2/1	82/15/11
Build. 2[b]	384/64	232/12	77/8	11/4	39/6	105/9/7
Build. 4[b]	472/57/36	299/104/43	59/3/1	109/33/18	18/5/1	114/63/22
Build. 5	105/46/20	48/18/16	8/3/3	3/3/3	25/5/6	11/8/5
Build. 6	144/36/8	127/55/14	6/3/1	14/14/2	27/2/2	80/36/9
Build. 7, ap.1	333/30/15	180/32/21	14/1/2	5/5/5	21/2/1	139/25/12
Build. 7, ap.2	170/22/14	207/32/23	83/1/2	5/2/1	12/5/1	108/24/19
average	**312/54/30**	**170/40/20**	**35/3/2**	**22/9/4**	**25/3/2**	**88/25/12**

The average relative contributions from the structures are summarised in Table 3. The contribution of TVOC emission from the ceiling structure dominates in the 0- and 6-month-old buildings but no more in the 12-month-old buildings. The impact of ammonia and formaldehyde emissions from surfaces remained approximately at the same level during the follow-up. The ceiling structure contributed most strongly to the ammonia and formaldehyde concentrations throughout the first year. However, it has to be pointed out that the FLEC measurements were always done at the RH of 50%. In real buildings, the indoor air the RH varied between 15–74%, which has not been considered in the emission calculations.

Table 3. The average contributions (%) of TVOC, ammonia, and formaldehyde from structures.

	Floor covering		**Ceiling (PVC/parquet)**	**Walls (PVC/parquet)**
TVOC	**PVC**	**parquet**		
0 month	28%	7%	44/57%	26/34%
6 month	28%	19%	50/57%	20/23%
12 month	34%	11%	37/50%	41/36%
Ammonia				
0 month	17%	12%	50/54%	31/33%
6 month	9%	13%	70/75%	21/13%
12 month	9%	20%	60/53%	28/24%
Formaldehyde				
0 month	15%	9%	61/66%	21/23%
6 month	6%	13%	68/63%	26/25%
12 month	10%	12%	72/68%	17/20%

3.2 VOC groups and single VOCs

Principal component analysis (Varimax rotation) for 30 target VOCs revealed six components in the 12-month-old buildings. These accounted for 94% of the variability in the variance. The first component accounted for 44% of the variability and correlation coefficients above 0.4 to the first component were detected for nearly all VOCs (alcohols, aldehydes, aliphatic compounds, terpenes, 2-butoxy ethanol and styrene), except for benzene, ethyl benzene, toluene, xylene, trimethylbenzene, and naphthalene. The latter ones were divided to the second and third components. The first component was characterised by material emissions and inhabitancy, the second by material emissions, and the third by traffic emissions. Edwards et al. [18] also found factors associated to the inhabitancy, traffic emissions and material emissions for the same VOCs. In this study, the factor associated with materials was clearly dominating, which was not the case in the established buildings studied by Edwards et al. [18].

The development of the contribution of surface emissions was different for different VOC groups. There were three main types of behaviour. Glycol/glycolethers, terpenes, and aromatic compounds are used to illustrate these in Figures 3a–c.

Glycols/glycol ethers were one of the groups for which the surface emissions represented almost 100% of the measured concentration levels in the 6 and 12-month-old buildings. Acids, aliphatic hydrocarbons, ketones, and esters behaved similarly. On the contrary, the emissions of terpenes, alcohols, and aldehydes from surfaces decreased steadily whereas their air concentration remained stable. The emission contributions were 88/106/45 % (alcohols), 91/93/55% and 53/83/21% (terpenes) in the 0/6/12 month-old buildings. The concentration and emission of aromatics decreased steadily but the emissions from surfaces explained only 30–50% of the indoor air concentration. The difference was probably due to traffic emissions and emissions from painted surfaces which were not investigated.

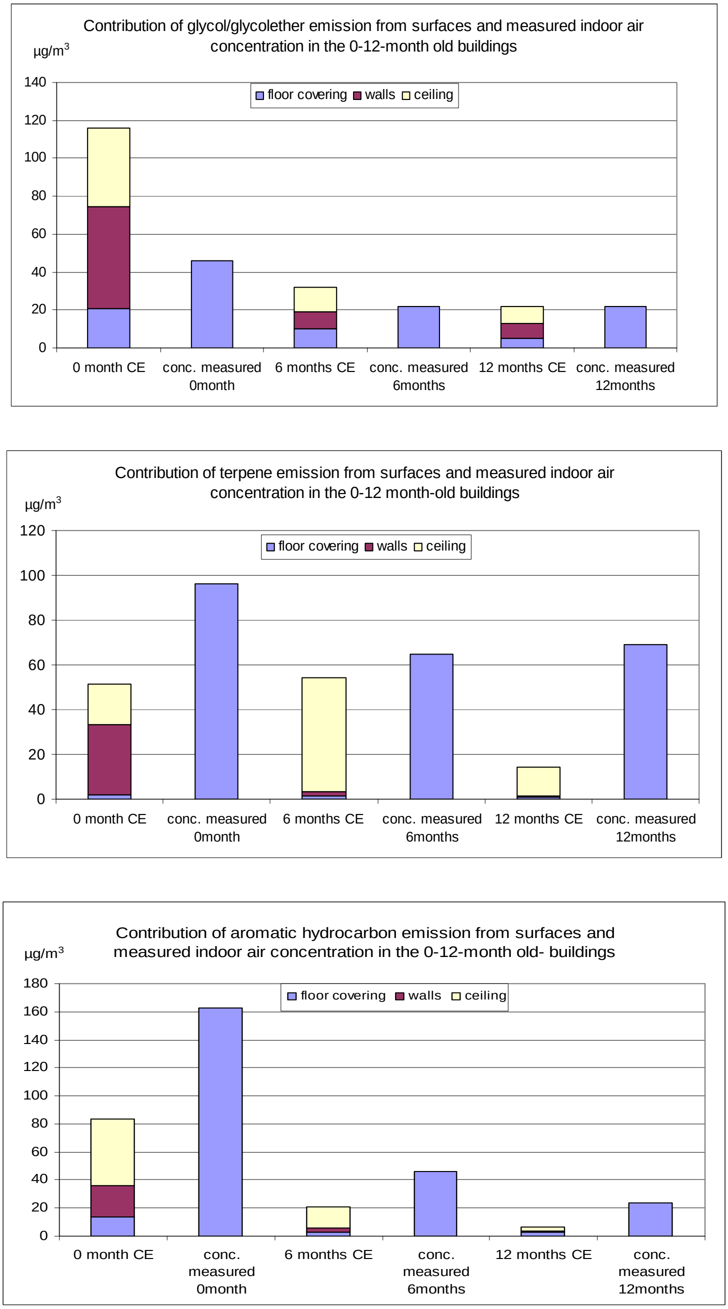


Figure 3a–c. Contributions of the glycol/glycolether, terpene, and aromatic hydrocarbon emissions from surfaces to the indoor air concentration in the 0-, 6- and 12-month-old buildings. The on-site measured emissions are corrected for the air exchange rates measured on site.

3. 3 SVOC compounds

The indoor air concentrations of SVOCs (C16–C22) in the study buildings are shown in Figure 4. The mean concentrations were 8, 9, and 8 µg/m^3 in the 0, 6, and 12 month-old buildings, respectively. An exceptionally high concentration of 61 µg/m^3 was measured in one of the newly finished buildings (Building 1, apartment 2). The concentrations became quite well normally distributed during the follow-up (W-value: 0.833). Statistically significantly ($p<0.05$) higher SVOC concentration was observed in the rooms with PVC flooring compared to the rooms with parquet flooring in the newly finished and 6-month-old buildings. The differences between the mean SVOC concentrations in PVC and parquet rooms were 14, 4, and 3 µg/m^3 in the 0, 6, and 12 month-old buildings, respectively.

The SVOC/TVOC ratio was on the average 0.01 in the newly finished buildings. The ratio increased to 0.03 in the six month and one year old buildings. Identification of individual compounds was difficult because the chromatograms contained a large number of small peaks in the SVOC time window. However, phthalate compounds (m/z 149) were not abundant.

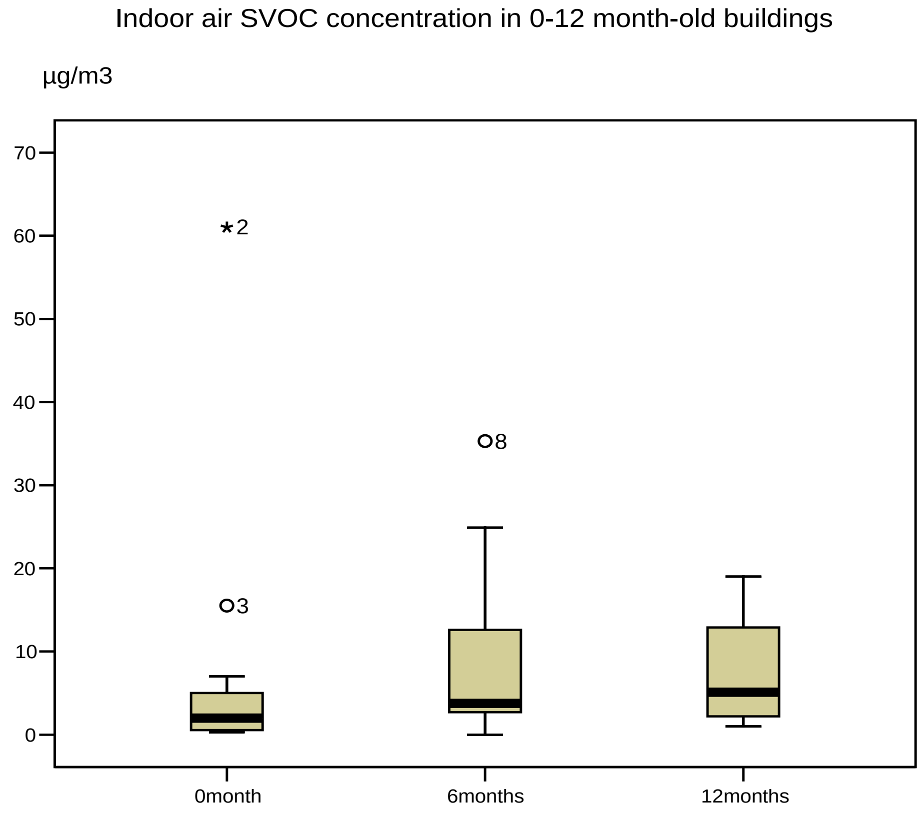


Figure 4. Indoor air SVOC concentration in 0–12- month-old buildings. The thick line shows the median value. The 25 percentile and 75 percentile values are signed by the lower and upper end of the boxes. The lines show the minimum and maximum values, the O stands for outliers (=cases with values more than 2 box lengths from the upper or lower edge of the box, the box length is the interquartile range) and the stars stand for extreme values (=cases with values more than 3 times the interquartile range).

4. Discussion and conclusions

The use of low-emitting, classified materials is an important means to achieve good IAQ. The indoor air concentrations should also be predictable in buildings where such materials are used if the ventilation is operating continuously. The calculations presented in this paper showed that target values (S1–S3- levels) for the indoor air pollutant concentrations as defined in the Finnish indoor climate classification can be reached for TVOC and ammonia if ventilation fulfills its S1-criterion. There was a slight inconsistency for formaldehyde, only the S2-level could be achieved. On the other hand, if emissions from surfaces correspond to the upper limits of M1- values no S-levels can reached for ammonia or formaldehyde and only the S3-levels (600 µg/m^3) for the TVOC concentration with the lowest air exchange rate (0.5 h^{-1}) allowed. Thus, the use of classified building materials does not guarantee good IAQ alone but effective ventilation is also required. To ensure this, the air flow rate recommendations given per floor area of the first classification version should be reintroduced.

The average on-site measured contribution of emissions to indoor air concentration was lower (~20%) for TVOC and much lower (~55–80%) for ammonia and formaldehyde than predicted. The mean reason for this difference is that the ammonia and formaldehyde emissions from the walls and floor structure were well below the M1- target values. The contribution of non-measured sources increased for ammonia and formaldehyde during the follow-up period of one year. However, the average concentration of formaldehyde remained within the S2- limit whereas the airborne average concentration of ammonia did not fulfill even the S3- requirement. Airborne concentrations and emissions of ammonia have received relatively little attention in IAQ studies. This study showed that, in addition to the emissions of from surfaces, ammonia has other important sources. Ammonia sources like wastewater drains and inhabitancy have also earlier found to affect the IAQ [13].

The average TVOC contribution from surfaces to the airborne concentration was as high as 540 µg/m^3 in the newly finished buildings. Thus, an air exchange rate of 2.9 h^{-1} would have been needed to reach the S1-level concentration for TVOC (200µg/m^3) at this point. However, the S1-level was on the average reached within 6 months if only emissions from surfaces contributed.

The impact of VOC emissions originating from the ceiling structure to the indoor air concentration levels was higher than predicted based on the M1-target values. The ceiling was also found to act as a sink for VOCs originating from other sources (e.g. furniture) during the first year of inhabitancy [20]. On the contrary, the walls contributed less than expected. The contribution of VOCs from the PVC floor covering in the newly finished buildings was important in comparison to the parquet floor covering. The difference in the contribution between the flooring materials decreased in six months. The effect of other, non-measured TVOC sources was remarkable (45%) for the buildings with a mechanical exhaust air system whereas the non-measured sources contributed to only 25% of the indoor air TVOC in the buildings provided also with mechanical supply air. Brown et al. [12] concluded that ~70% of the TVOC concentration indoors arose from unidentified sources in 27 established buildings.

The concentrations of glycols/glycol ethers, aliphatic hydrocarbons, esters, and ketones were well explained by the emissions from surfaces. On the other hand, the emissions of alcohols, aldehydes, and terpenes decreased steadily from surfaces whereas their concentration did not, which indicates the presence of other sources for these compounds. For example, the average emissions of α-pinene from structures were 46% and 58% of the average concentrations in the 0- and 12-month-old buildings. In this time, the concentration decreased from 61 µg/m^3 to 35 µg/m^3, respectively. Brown et al. [12] measured a steeper concentration decrease from 52 µg/m^3 to 9 µg/m^3 in ~8 months. Aromatic compounds also had other non-measured sources.

The concentration of SVOCs was on the average 10 µg/m^3 throughout the first year. This is in good agreement with earlier studies [15]. The maximum values decreased from 60 µg/m^3 to 20 µg/m^3 within this period of time.

The study confirmed that the slowly evaporating but toxic glycol ethers and their acetates are extensively used in paints and adhesives in Finland. In Sweden, common use of 2-(2-butoxyethoxy) ethanol in building materials has also been reported [22]. Glycol ethers are used in water based paints because of their good solvent properties [23]. Uptake of glycol ethers occurs efficiently via all routes but due to their rapid dermal penetration and slow evaporation the skin often constitutes a major route of entrance to the body. Glycol ether esters are rapidly hydrolysed to the corresponding glycol ethers in blood and body tissues [24]. Studies have shown that VOC exposure during painting work or from newly painted (with water based paint) surfaces can cause eye and asthmatic symptoms [25, 26, 27]. Even though the glycol ether concentrations were about 100 µg/m^3 in the newly finished buildings in this study, which is quite low compared to occupational exposure levels, it should be noted that exposure to glycol ethers continues for several months in residences. The EU risk assessment made on 2-

butoxyethoxy ethanol concluded that there is a need to limit its risks exposure risks for consumers and workers [28].

In conclusion, the Finnish material classification seems to work fairly well in achieving low pollutant levels in newly finished buildings where the emissions from the main interior surfaces dominated. However, there appeared to be reasons for slight modifications. Especially, the recommended air flow rates per floor area should be reintroduced to ensure good IAQ. In addition, the contribution of other sources than surfaces was found to be significant. The importance of these sources increased with time. The planned extension of the Finnish classification system to furniture and cleaning agents is thus welcome.

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PUBLICATION IV

Comparison of VOC and ammonia emissions from individual PVC materials, adhesives and from complete structures

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Comparison of VOC and ammonia emissions from individual PVC materials, adhesives and from complete structures

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Abstract

Emission rates of volatile organic compounds (VOCs) and ammonia measured from six PVC materials and four adhesives in the laboratory were compared to the emission rates measured on site from complete structures. Significantly higher specific emission rates (SERs) were generally measured from the complete structures than from individual materials. There were large differences between different PVC materials in their permeability for VOCs originating from the underlying structure. Glycol ethers and esters from adhesives used in the installation contributed to the emissions from the PVC covered structure. Emissions of 2-ethylhexanol and TXIB (2,2,4-trimethyl-1,3-pentanediol diisobutyrate) were common. High ammonia SERs were measured from single adhesives but their contribution to the emissions from the complete structure did not appear as obvious as for VOCs. The results indicate that three factors affected the VOC emissions from the PVC flooring on a structure: 1) the permeability of the PVC product for VOCs, 2) the VOC emission from the adhesive used, and 3) the VOC emission from the backside of the PVC product.
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Keywords: PVC; Adhesive; VOC; Material emission

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1. Introduction

Emissions of pollutants from interior structures are important contributors to the indoor air quality (IAQ) as elevated concentration levels of chemical pollutants can cause adverse health effects and discomfort. To improve IAQ, labeling schemes for tested, low-emitting building materials have been introduced (ECA, 2005). Standardized methods for environmental emission test chambers have been established (ISO, 2006a,b). Emission profiles of volatile and semi volatile organic compounds (VOCs and SVOCs) have been reported for several building materials including floor coverings and adhesives (Wolkoff, 1995; Yu and Crump, 1998; Lundgren et al., 1999; Wilke et al., 2004). Recently, Clausen et al. (2007) published a physically based emission model for SVOCs that was consistent with the experimental data and independent of relative humidity. However, emissions from real building structures are affected by the surroundings due to sorption, hydrolysis, and oxidative processes, which can lead to the production of secondary emissions (Gustafsson, 1990, 1996; Wolkoff, 1998; Wilke et al., 2004). These emissions, which are developing after a delay and are often long lasting, are not usually considered in the labeling schemes, which focus on primary emissions.

A wide range of VOCs and SVOCs are emitted from PVCs. These include plasticizers, solvent residues, unreacted monomers, and secondary degradation products. Plasticizers are often SVOCs (Saarela, 1999). Color changes in PVC floor structures and/or discomfort to IAQ have often been detected in humid conditions, especially in the presence of alkaline concrete. As an example, elevated structure humidity in alkaline conditions has been shown to cause degradation of the 2-diethylhexylphthalate (DEHP) in the PVC polymer (Gustafsson, 1990, 1996). Special attention has been paid to 2-ethylhexanol, which is generally considered to be a degradation product of DEHP and has been

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measured in buildings with indoor air complaints, e.g. bad odor and/or irritation symptoms (Brown et al., 1993; Norrbäck et al., 2000; Metiäinen et al., 2003; Tuomainen et al., 2004).

The plasticizer TXIB™ (2,2,4-trimethyl-1,3-pentanediol diisobutyrate) has also been detected in the indoor air in case studies where irritancy in the throat and eyes have occurred (Brown et al., 1993, Metiäinen et al., 2002). Odor and chemosthesis investigations, however, indicated that TXIB could contribute to odor but hardly to sensory irritation at these concentration levels (Cain et al., 2005).

The TVOC specific emission rates (SER) from vinyl/PVC floorings have been shown to range from 10^2 to 10^4 μg/m^2 h (Yu and Crump, 1998). A set of PVC flooring materials (n=50) produced in the 90's were investigated by Lundgren et al. (1999). The main VOCs were 2-butoxyethanol, 2-(2-butoxyethoxy) ethanol, phenol, hepta and octa decanes, and trimethylbenzene. Reduction factors (=the difference between the 4 week and 26 week emissions) of less than 0.5 were found ten compounds including TXIB indicating that these compounds may affect the IAQ over a long period of time (Lundgren et al., 1999). In another study, the TVOC SER from the single 28-day-old PVCs (five products) varied widely including SERs up to 1000 μg/m^2 h. The main compounds were TXIB and alkylbenzenes (SVOCs). Besides the single materials (floor covering materials and adhesives) the emissions from complete structures were also measured. The total SERs were lower for the tested compositions than the sum of total SERs from the single components (Wilke et al., 2004).

Only a few studies have published where the emissions have been tested both from separate materials in laboratory and from the complete structures on site. Zellweger et al. (1995) compared emissions measured in test chambers with concentration levels measured on site. The expected room concentrations were calculated with the air exchange rate measured. The predicted levels of cyclohexane and hexanal were more than one order of magnitude higher than those measured in the indoor air. Application (i.e., substrate) and sink effects were possible explanations for the difference. A good agreement between the predicted and measured concentrations was found for N-methyl-2-pyrrolidone from parquet lacquer applied on wood and room concentration. On the contrary, the predicted concentration from parquet lacquer applied on glass were much higher than the measured ones.

In this paper, emissions of VOCs and ammonia measured from single PVC materials and adhesives were compared to the results from on-site measured complete structures. The study is a part of a larger investigation of material emissions and indoor air concentrations in newly established residential buildings with low-emitting materials, the results of which have been published partially recently (Järnström et al., 2006, 2007).

2. Methods

The study buildings and materials as well as the sampling and analysis techniques have been presented in the other previous publications (Järnström, 2006, 2007). The analysis of ammonia is non-specific and is a total measure of ammonia including "aliphatic" amines. For consistency, the same numbering is used for the buildings as in the previous publications. The buildings where PVC was used as the floor covering material included buildings 1 (two apartments), 2, 4, and 7 (two apartments). No adhesive was used in the installation of the floor covering in one of the apartments in building 7. The PVCs (products 5 and 6) used in this building were otherwise similar products with a polyurethane reinforced surface. The PVC 2 had also a similar surface finish. The PVC products (1–6) and adhesives 2 and 4 were M1-classified, i.e. the SERs from the 28-day-old, single product measured at 23 °C, 50% RH and 0.5 h^{-1} do not exceed the following target values: 200 μg/m^2 h (TVOC), 30 μg/m^2 h (ammonia) and 50 μg/m^2 h (formaldehyde, FiSIAQ, 2001).

Emissions were first measured on site from the floor structure with the Field and laboratory emission cell (FLEC, ISO, 2006b) four weeks after the floor covering was installed and construction work was still going on. Then, the floor covering and adhesive were removed and the emission was measured from the underlying floor structure after one and three days (the residues of PVC and adhesive were removed with a spatula prior to the FLEC measurement).

In the laboratory, the adhesives were spread on a glass plate (spreading with a specific trowel in a 45° angle) and emissions were measured at 23 °C, 50%

Table 1
Specific emission rates (SER, μg/m^{2}h) of TVOC and ammonia from parallel sampling on site from the structure with FLEC (sd=standard deviation, rsd=relative standard deviation)

Building, age, measured structure	TVOC SER (μg/m^2 h)			Ammonia SER (μg/m^2 h)		
	FLEC1	FLEC2	sd (rsd %)	FLEC1	FLEC2	sd (rsd%)
Building 2 (PVC 3)						
6 month-old, uncovered floor structure, day 1	315	265	25 (9)	42	29	7 (18)
6 month-old, uncovered floor structure, day 3	212	264	26 (11)	24	8	8 (50)
Building 7, apartment 1 (PVC 5)						
6 month-old, uncovered floor structure, day 1	2600	2212	194 (8)	231	221	5 (2)
6 month-old, uncovered floor structure, day 3	1228	1983	378 (24)	90	107	9 (9)
12 month-old, uncovered floor structure, day 1	1745	1862	59 (3)	93	97	2 (2)
12 month-old, uncovered floor structure, day 3	1097	806	146 (15)	37	42	3 (6)
Building 7, apartment 6 (PVC 6)						
6 month-old, uncovered floor structure, day 1	1863	1816	24 (1)	207	219	6 (3)
6 month-old, uncovered floor structure, day 3	1208	1236	14 (1)	112	116	2 (2)
12 month-old, uncovered floor structure, day 1	1572	1586	7 (0.4)	101	108	4 (3)
12 month-old, uncovered floor structure, day 3	1056	807	125 (13)	56	55	1 (1)

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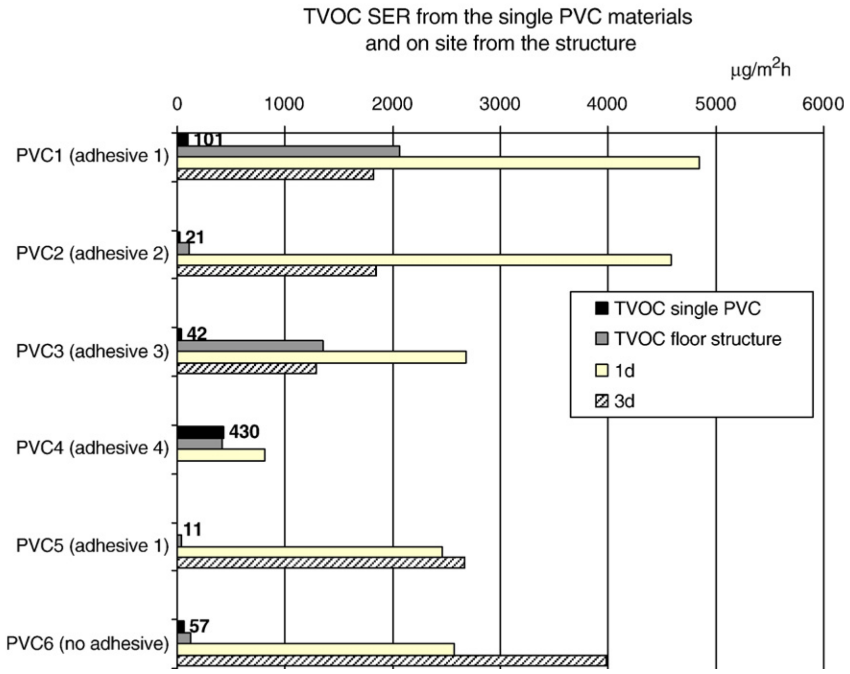

Fig. 1. Specific emission rate of TVOC (SER, µg/m^2 h) measured from the single PVCs 1–6 in the laboratory (column 1), on site from the complete structure (column 2), the underlying structure 1 day (column 3) and 3 days (column 4) after the removal of the floor covering (the values measured from the single materials are shown).

relative humidity (RH), and air exchange rate of 0.5 h^{-1} in emission test chambers (ISO, 2006a). The measurements were done 1, 3, 10, and 28 days after spreading. The PVC samples for the laboratory testing (with FLEC, ISO, 2006b) were taken from the same dispatch that was installed on the structure. The adhesives were supplied by the manufacturers or bought from a hardware store.

The repeatability for the on-site performed emission measurements (TVOC, ammonia) was investigated by parallel sampling with two different FLEC equipments. The parallel sampling was performed from the floor structure (within an area of 0.5 m^2) one and three days after the floor covering was removed (PVCs 3, 5, and 6). The pre-conditioning time prior to VOC sampling was investigated by taking samples at four time points ranging from 18–313 min after the clean air was introduced to the FLEC cell. Principal component analysis was performed with Simca-P 7.0 to reveal the impact of the PVCs and adhesives on indoor air concentrations.

3. Results

3.1. Validation of field sampling

As shown in Table 1, the relative standard deviation between parallel FLEC samplings with two FLECs was on the average 9% (range 0.4–24%) for the TVOC SER and 10% (range 1–50%) for ammonia SER. These are in good agreement with the repeatability of 11% for a TVOC SER from a PVC material measured with FLEC equipments at the laboratory, which has been measured earlier (results not published here).

A pre-conditioning time of 30 min was found appropriate for TVOC sampling. This time was also found sufficient for single PVC samples in earlier tests.

3.2. Laboratory versus field measurements

3.2.1. Single PVCs

The TVOC SER ranged from 11 to 101 µg/m^2 h from the single PVCs, except for PVC 4 the SER of which was 430 µg/m^2 h (Fig. 1). Even PVC 4 has a M1-label (the best class in the Finnish labeling system), and its TVOC SER was expected to be <200 µg/m^2 h (FiSIAQ, 2001). Four separate PVC samples emitted 2-ethylhexanol (Table 2). The SERs were below 30 µg/m^2 h. TXIB was emitted from the same four single PVCs with SERs up to ~15 µg/m^2 h (Table 2). The SERs of ammonia were 11–38 µg/m^2 h from the single PVCs (Fig. 2).

3.2.2. Adhesives on a glass plate

The TVOC SERs from the adhesives 1 and 3 were 9000–12 000 µg/m^2 h measured after one and three days (Fig. 3). These two products were not M1-classified. The TVOC SER from the M1-classified adhesive 4 also clearly exceeded the M1-limit. The adhesive 2 reached the M1-requirement already after the first day. The main VOCs from adhesives were glycol ethers (up to 9000 µg/m^2 h) and esters (up to 7000 µg/m^2 h). The main VOCs included 2-ethylhexanol, 2-(2-butoxyethoxy) ethyl acetate, phenoxy ethanol, and 2-ethylhexyl ethyl acetate. All adhesives emitted 2-ethylhexanol. The SERs of 2-ethylhexanol had large variations, from ~5 µg/m^2 h to up to ~2400 µg/m^2 h (Table 2).

The single adhesives emitted up to 100-250 µg/m^2 h ammonia after the first day (Fig. 4). The M1-limit (30 µg/m^2 h) was reached within ten days for all products. The SER from the adhesive 1 slightly increased between the 10- and 28-day measurements whereas the adhesives 2 and 3 had very low final SERs (<5 µg/m^2 h).

3.2.3. PVC on the structure

Only the TVOC SER of PVC 4 was on the same level from the 4-week-old structure and from the separate PVC sample whereas the SERs of the other PVCs were much higher from the complete structures than from the corresponding single PVCs. However, the SERs measured from the structures with PVCs 2, 5 and 6 were very low (Fig. 1).

Esters and glycol/glycol ethers contributed most to the on-site measured TVOC. These VOC groups were dominating for the adhesives used (Table 2).

No significant difference was observed in TVOC SERs between structures coated with PVCs 5 and 6, although the latter was installed without adhesive. Actually, SERs of C4–C16 alkylated benzenes were slightly higher from PVC 6.

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Table 2
The specific emission rates (SER, μg/m^2 h) of VOC groups and selected VOCs measured from the single PVC (PVC lab), from the PVC covered floor structure (PVC+adhes. field), and from the adhesive measured on a glass plate (adhes. lab 1d)

VOC group/ VOC	PVC1 lab	PVC1+ adhes1. field	adhes1. lab 1d	PVC2 lab	PVC2+ adhes2. field	adhes2. lab 1d	PVC3 lab	PVC3+ adhes3. field	adhes3. lab 1d	PVC4 lab	PVC4+ adhes4. field	adhes4. lab 1d	PVC5 lab	PVC5+ adhes1. field	adhes1. lab 1d	PVC6 lab	PVC6 field
Acid	10	<1	41	<1	<1	27	2	14	873	22	6	17	<1	<1	41	8	2
Acetic acid	<1	<1	19	<1	<1	26	<1	<1	868	<1	<1	13	<1	<1	19	8	<1
Alcohol	30	13	338	3	9	32	16	40	2463	42	12	184	<1	8	338	5	19
2-ethylhexanol	11	27	327	3	7	6	10	28	2406	29	9	122	<1	1	327	<1	14
Aldehyde	6	15	14	4	6	12	4	17	92	24	20	3	2	1	14	2	2
Benzaldehyde	1	4	8	<1	3	1	3	4	4	7	4	<1	2	1	8	1	1
Aliphatic hydrocarbon	7	54	<1	<1	21	7	1	29	4	53	19	5	<1	1	<1	4	8
Tetradecane	1	8	<1	<1	<1	2	1	11	<1	3	2	<1	<1	<1	<1	2	3
Aromatic hydrocarbon	38	435	12	<1	2	16	13	38	6	131	16	7	<1	9	12	61	97
Dodecylbenzene	28	<1	<1	<1	<1	<1	1	7	<1	65	4	<1	<1	<1	<1	30	48
Cycloalkane	<1	<1	<1	<1	<1	<1	<1	<1	<1	25	<1	17	<1	<1	<1	0	0
Ester	<1	20	295	<1	56	2	3	777	7206	17	249	1395	2	4	295	5	2
Ethanol, 2-(2-butoxyethoxy)-, acetate	<1	<1	6	<1	<1	<1	<1	750	6613	<1	246	1301	<1	2	6	<1	2
Acetic acid, 2-ethylhexyl ester	<1	15	168	<1	<1	<1	<1	6	422	<1	<1	20	<1	15	168	<1	<1
TXIB	13	6	<1	7	53	<1	1	<1	<1	4	<1	<1	<1	<1	<1	<1	<1
Glycol/glycolether	5	1453	9084	<1	5	129	<1	476	1001	13	55	285	<1	<1	9084	6	2
Ethanol, 2-phenoxy-	2	1405	9075	<1	<1	<1	<1	6	9	<1	3	<1	<1	<1	9075	<1	<1
Ketone	10	6	14	7	16	5	2	2	21	2	4	<1	<1	<1	14	9	7
2-Hexanone, 5-methyl-5-phenyl	7	4	<1	<1	<1	<1	1	2	<1	<1	<1	<1	<1	<1	<1	6	7
Silyl/siloxane	1	<1	14	<1	1	1	3	1	<1	1	1	3	2	2	14	2	1
Terpene	<1	3	<1	<1	<1	8	<1	19	<	6	9	48	<1	<1	<1	<1	<1
alpha-Pinene	<1	3	<1	<1	<1	<1	<1	9	<1	1	4	6	<1	<1	<1	<1	<1

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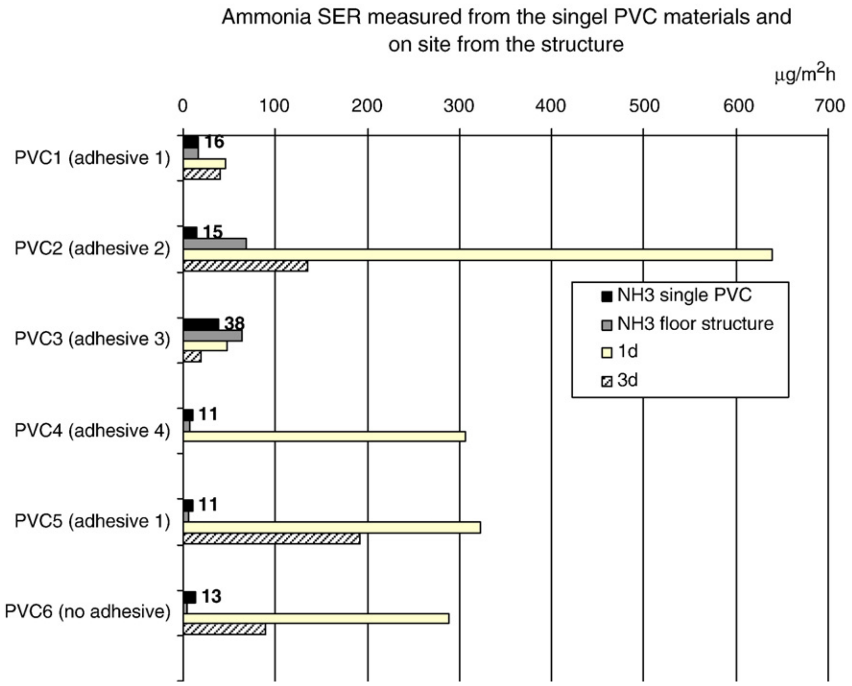

Fig. 2. Specific emission rate of ammonia (SER µg/m^2 h) measured from the single PVCs 1–6 in the laboratory (column 1), on site from the complete structure (column 2), the underlying structure 1 day (column 3) and 3 days (column 4) after the removal of the floor covering (the values measured from the single materials are shown).

For 2-ethylhexanol, the median SER was slightly higher from the 4-week old structure than from the single PVCs (Fig. 5). The highest increase, from a level of ~10 µg/m^2 h (single PVC) to ~30 µg/m^2 h (complete structure), was measured from PVC 1 and 3. The SERs measured from the adhesives (1 day old on a glass plate) used in the installation of these PVCs were ~300–2400 µg/m^2 h. Detectable TXIB emissions appeared from two 4-week-old structures (6 µg/m^2 h, PVC 1 and 53 µg/m^2 h, PVC 2).

The ammonia SERs measured from the complete structures coated with PVC 2 and 3 were 2–5 times higher than those from the single PVCs. These SERs were about two times higher than the M1-target value (30 µg/m^2 h, FiSIAQ, 2001). Ammonia SERs from PVCs 4, 5,

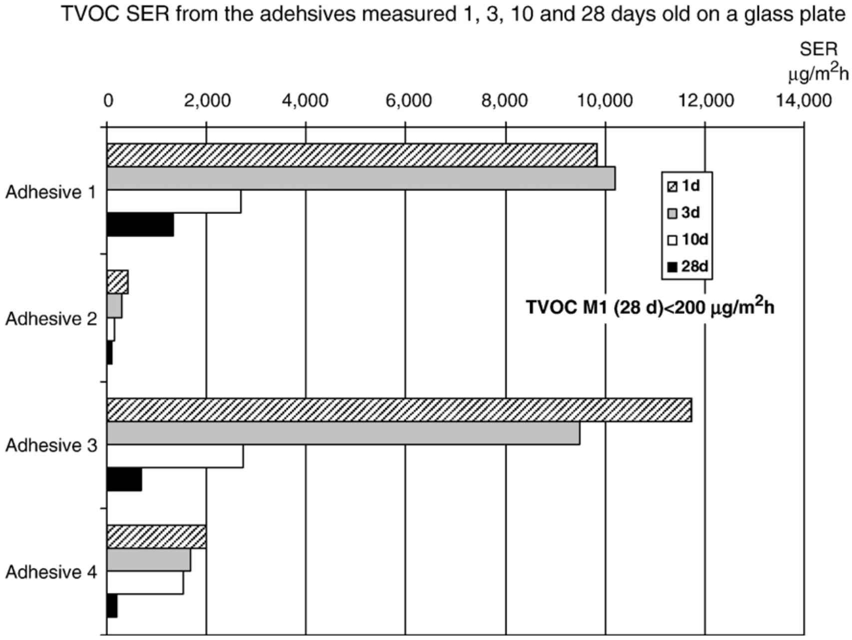

Fig. 3. Specific emission rate of TVOC (SER, µg/m^2 h) measured from the adhesives after 1, 3, 10, and 28 days on a glass plate. The M1 target value for the TVOC emission (28-day-old product) is shown (FiSIAQ, 2001).

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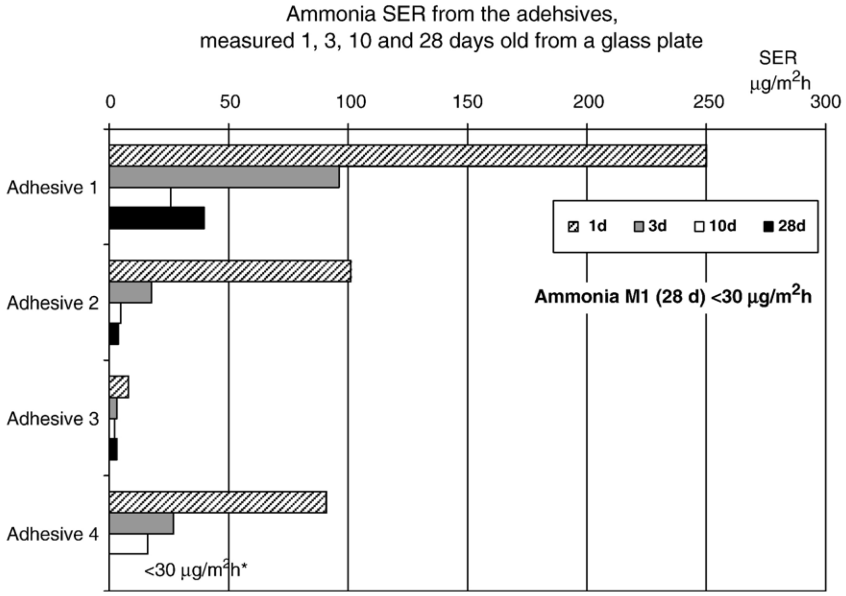

Fig. 4. Specific emission rate of ammonia (SER, µg/m^2 h) measured from the adhesives 1, 3, 10, and 28 days old from a glass plate. The M1-class target value for the ammonia emission (28-day-old product) is shown (FiSIAQ, 2001, * = no result, M1-class target value).

and 6 were 5–7 µg/m^2 h on site being somewhat lower than the SERs from the corresponding single PVCs. The SERs from the single PVC and the complete structure were similar for PVC 1 and within M1-class. The adhesives did not significantly contribute to the ammonia emissions from the structures coated with PVCs 1, 3, 4, and 5 (PVC 6 was installed without adhesive).

3.2.4. Structure under the PVC

The TVOC SERs measured from the underlying structure showed large variations, from 900 to 5000 µg/m^2 h on the first day after the removal of PVC (Fig. 2). The TVOC SERs measured from the underlying structures coated with PVCs 1 and 3 originated largely from the adhesives used, and 2-(2-butoxyethoxy) ethanol and its acetate and phenoxy ethanol were the most abundant VOCs. On the contrary, the contribution of the adhesives to the equally high TVOC SERs from the underlying structures of PVCs 2 and 5 was low. As high TVOC SER as 3000 µg/m^2 h was measured from the structure under PVC 6 which was installed without any adhesive. Emission measurements from the background side of the PVC (not presented here) confirmed that the PVC material itself was the main source of TVOCs in these cases. The main VOCs were TXIB (PVC 2) and alkylated benzenes (PVC 5 and 6).

SERs of 2-ethylhexanol, TXIB, and alkylated benzenes three days after the floor covering had been removed from the 4-week-old floor structure are summarized in Table 3. These compounds were detected from the floor structures under all PVCs. The SERs of 2-ethylhexanol and TXIB were exceptionally high under PVC 2. On the other hand, the SERs of C2–C10-alkylated benzenes were strikingly high under PVCs 5 and 6. About one third of these compounds were outside the TVOC time window (i.e., were SVOCs).

The ammonia SERs were ~20–650 µg/m^2 h from the underlying structure. The highest ammonia SERs were detected under PVCs 2, 4, 5, and 6. The effect of the adhesives was again smaller than that on VOCs even though, for example, PVC 1 was installed with an adhesive that emitted 250 µg/m^2 h of ammonia after the first day on the glass plate. The SERs from the complete and opened structures were,

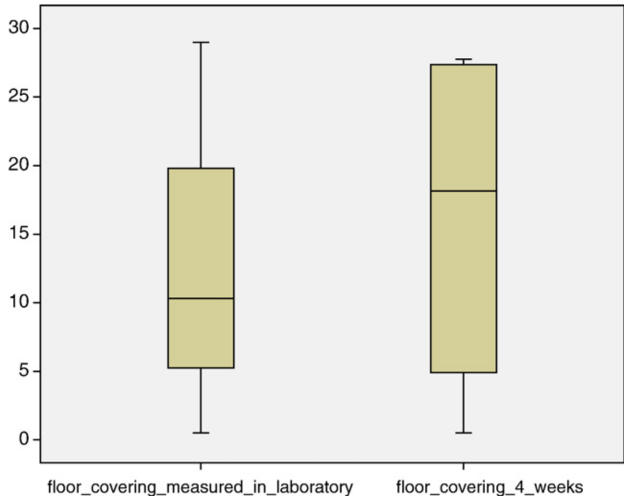

Fig. 5. Specific emission rate of (SER, µg/m^2 h) of 2-ethylhexanol from the single PVCs and from the PVC coated, 4-week-old floor structure. The thick line shows the median value. The 25 percentile and 75 percentile values are signed by the lower and upper end of the boxes. The lines show the minimum and maximum values.

Table 3
The specific emission rates (SER, µg/m^{2}h) of 2-ethylhexanol, TXIB, and alkylated benzene compounds measured from the 4-week-old floor structure three days after the floor covering was removed (*measured after one day)

PVC	SER (µg/m^2 h)		
	2-Ethylhexanol	TXIB	Alkylated benzenes
PVC 1	27	17	532
PVC 2	228	854	192
PVC 3	34	33	30
PVC 4*	30	2	148
PVC 5	7	5	1284
PVC 6	10	5	1473

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however, quite low. No significant differences were observed between the TVOC or ammonia SERs measured from the structures under PVC 5 and 6 (no adhesive). The impact of adhesive was therefore concluded to be insignificant as regards to these two structures.

3.3. The effect of PVCs on indoor air quality

The use of the impermeable PVC types (PVCs 2, 5, and 6) obviously lead to the retention of VOCs in the underlying floor structure. The later migration of these compounds into indoor air is possible. The principal component analysis actually showed that the concentrations of aromatic compounds were slightly higher in the apartments with the tighter PVCs but no statistically significant differences were observed. The airborne concentrations of aromatic compounds were 43 $\mu g/m^3$ in the apartments with the less permeable PVCs and 34 $\mu g/m^3$ in the apartments with the more permeable types in the 12-month-old buildings. However, the concentrations of aromatic compounds may also have been influenced by traffic pollution. No differences were observed in the concentrations of the other compounds.

4. Discussion

Significantly higher SERs were often measured on site from the complete structure than from the single PVC materials. Adhesives increased the VOC emissions from complete structures. The M1-label of PVC products is given for a 28-day-old product (FiSIAQ, 2001), which underestimates the contribution of emissions from adhesives in real structures. On the other hand, VOCs were found to migrate into the substrate, i.e. the levelled floor structure, which may lower emissions.

The results indicate that three factors affected the VOC emissions from the PVC flooring: 1) the permeability of the PVC product for VOCs, 2) the VOC emissions from the adhesive used, and 3) the VOC emission from the back of the PVC product. Glycols/glycol ethers and their esters were abundantly present in the emissions from the PVC structures and single adhesives. Wilke et al. (2004) also measured phenoxy ethanol SERs up to 200 $\mu g/m^2$ h from a less than five days old PVC structure. These compounds were measured in the indoor air in the studied buildings (Järnström et al., 2006).

All the single PVC materials emitted 2-ethylhexanol. Thus, this compound is not merely a degradation product and its presence in indoor air at low concentrations does not necessarily require moisture damage. Adhesives contributed to some extent to its on-site measured emission for the more permeable types of PVCs. The plasticizer TXIB was found both from single PVCs and from the complete structures as well as from the underlying structures. One of the less permeable (for VOCs) products had clearly higher TXIB SERs than the others. Very high SERs were measured from its underlying structure compared to the other products.

High ammonia SERs were measured from single adhesives on a glass plate on the first day but the contribution of the adhesives to the emission from the complete structure was not as clear as for TVOC. Single PVCs also emitted ammonia. The origin of ammonia in the PVCs was not further investigated during this study.

The ammonia SERs measured from the 4-week-old PVC coated structures exceeded the M1-value of 30 $\mu g/m^2$ h by two fold in two cases. As for the VOCs, the ammonia SERs measured from the structure after the floor covering was removed showed large variations. The emissions were again affected both by the adhesive and PVC product.

The indoor air concentration of ammonia has increased in the Finnish building stock in the 90's. The cause for the elevated ammonia has been addressed to the shorter construction times and consequently, the shorter drying periods for the structure (Hiltunen, 2000). The results gained during this project, however, showed that the impact of adhesives and/or PVC types may be an additional factor.

5. Conclusions

Higher emissions were often measured from complete PVC coated structures and the contribution from adhesives used in the installation was clear. The emission results implied that it is necessary to carefully specify the different product combinations (PVC+adhesive) when the emissions from a complete floor structure are aimed to be low initially. High emissions were emitted from the structure under unpermeable type PVCs, and they were concluded to originate from the PVC itself. For such structures, high SERs measured from the structure under the floor covering does not necessarily imply any degradation of the structure.

For labeling purposes, adhesives would be better tested in a complete composite. The testing of complete structures would also in general provide a better approach to evaluate the actual emissions from the combined materials.

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Series title, number and report code of publication

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Author(s)
Järnström, Helena

Title
Reference values for building material emissions and indoor air quality in residential buildings

Abstract
Indoor air concentrations and emissions from structures and interior materials were investigated in eight residential buildings during the time of construction and the first year of occupancy. Volatile organic compounds (VOCs), formaldehyde and ammonia concentrations and emissions as well as temperature, humidity, and ventilation were measured.

The total VOC (TVOC) concentration was generally above the S3-class limit of 600 µg/m^3 (FiSIAQ 2001) in the newly finished buildings but the concentration usually decreased below the S3-level and in some apartments below the S1-level of 200 µg/m^3 in six months. The concentrations of the major VOCs decreased most strongly during the first six months of occupancy, reaching mean concentration levels of 5–15 µg/m^3. Variables affecting the concentrations of indoor air gaseous pollutants in the buildings were the ventilation system, the floor covering material, the ceiling surface product, the wall surface product, the season, the relative humidity and temperature of the indoor air, and occupancy. The relative humidity (RH) affected ammonia and formaldehyde concentrations most strongly. Higher concentrations were measured when the RH was above 50% during the follow-up. The formaldehyde concentration did not significantly exceed the S2-class level of 50 µg/m^3 during the first year in any of the apartments. In some newly finished buildings and during the follow-up, the indoor air concentrations of ammonia were above the S3-level of 40 µg/m^3. The emission measurements performed from the complete floor construction showed that the emission was affected by all of its components, i.e., the structure, levelling agent, adhesive, and floor covering material. Significantly higher emissions were often measured on-site from the complete floor structure than from the single materials measured in the laboratory. The impact of adhesives on VOC emissions from the complete PVC- coated structures was clearly seen as higher emissions from those with a more permeable types of PVCs. The contribution of the average on-site measured emissions to indoor air concentration was ~550 µg/m^3 (~57% of the measured concentration) for TVOC and ~45/ 40 µg/m^3 (~100% of the measured concentration) for ammonia and formaldehyde in the newly finished building. The TVOC contribution from surfaces decreased to ~200 µg/m^3 in six months whereas the contribution of ammonia and formaldehyde remained about the same. The ceiling structure contributed by most to the concentration levels whereas the contribution from walls was lower than expected on the basis of large surface area. The study confirmed that the Finnish material classification system provides a basis to achieve good IAQ when comparing to the target values for pollutant concentrations given by the classification (FiSIAQ 2001) in real buildings; however, suggestions for its further development are given. Based on the indoor air and emission results, reference values, i.e. “normal” and “abnormal values”, were defined for the six- and twelve month-old buildings.

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Tekijä(t)
Järnström, Helena

Nimeke

Referenssiarvot rakennusmateriaalien emissioille sekä sisäilman laadulle asuinrakennuksissa

Tiivistelmä

Sisäilman pitoisuuksia sekä rakenteiden emissioita tutkittiin kahdeksassa asuinrakennuksessa rakentamisen aikana sekä ensimmäisen vuoden aikana käyttöönoton jälkeen. Tutkimuksen aikana mitattiin orgaanisten haihtuvien yhdisteiden (VOC-yhdisteiden), formaldehydin ja ammoniakin sisäilman pitoisuudet ja emissiot rakenteista sekä lämpötilat, kosteus ja ilmanvaihto.

VOC-yhdisteiden kokonaismäärä (TVOC) oli tavallisesti yli S3-luokan eli 600 µg/m^3 (FiSIAQ 2001) juuri ennen luovutusta, mutta pitoisuus saavutti tavallisesti S3-luokkaa vastaavan tason, ja joissain asunnoissa jopa S1-luokan eli 200 µg/m^3 ensimmäisen kuuden kuukauden aikana. Yksittäisten VOC-yhdisteiden pitoisuudet laskivat eniten ensimmäisen kuuden kuukauden aikana asunnon käyttöönotosta, jolloin keskimääräiset pitoisuudet olivat tasolla 5–15 µg/m^3. Sisäilman pitoisuuksiin eniten vaikuttavat tekijät olivat ilmanvaihtojärjestelmä, lattiapinnoite, kattopinnoite, seinäpinnoite, vuodenaika, sisäilman lämpötila ja kosteus sekä asunnon toiminta.

Sisäilman suhteellinen kosteus (SK) vaikutti eniten formaldehydi- ja ammoniakkipitoisuuksiin. Korkeampia pitoisuuksia seurantajakson aikana mitattiin kun SK oli yli 50 %. Formaldehydipitoisuus ei ylittänyt S2-luokan tasoa eli 50 µg/m^3 missään tutkitussa asunnossa. Joissakin uusissa sekä asutuissa asunnoissa sisäilman ammoniakkipitoisuudet olivat yli S3-tason eli 40 µg/m^3. Emissiomittaukset valmiista rakenteesta osoittivat, että rakenteen emissioihin vaikuttavat kaikki sen eri komponentit eli runko, tasoite, liima ja lattiapinnoite. Oikeasta rakenteesta mitattiin usein merkittävästi korkeampia emissioita kuin yksittäisistä materiaaleista laboratorio-olosuhteissa. Asennuksessa käytetyn liiman vaikutus oli selvästi nähtävissä niillä muovimattopinnoitteilla, jotka olivat VOC-yhdisteitä läpäiseviä.

Paikan päällä mitattujen emissioiden vaikutus sisäilman pitoisuuksiin oli ~550 µg/m^3 (~57 % mitatusta pitoisuudesta) TVOC:lle ja ~45/40 µg/m^3 (~100 % mitatusta pitoisuudesta) ammoniakille ja formaldehydille juuri valmistuneessa rakennuksessa. TVOC:n vaikutus pieneni noin ~200 µg/m^3:aan kuuden kuukauden aikana, kun taas ammoniakin ja formaldehydin osuudet pysyttelivät samalla tasolla. Kattorakenteen vaikutus pitoisuuksiin oli suurin, kun taas seinien osuus oli odotettua pienempi suuresta pinta-alasta huolimatta.

Tutkimus osoitti, että suomalainen materiaaliluokitus luo perustan hyvän sisäilman saavuttamiselle, kun sisäilman pitoisuuksia vertaillaan luokituksen asettamiin tavoitearvoihin (FiSIAQ 2001) oikeissa rakennuksissa. Kuitenkin, tämän tutkimuksen perusteella esitetään ehdotuksia luokituksen edelleen kehittämiseksi. Sisäilma- ja emissiotulosten perusteella määritettiin referenssiarvot eli arvot "normaali" ja "poikkeava" kuuden kuukauden ja yhden vuoden ikäiselle rakennukselle.

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Indoor air concentrations and emissions from structures and interior materials were investigated in eight residential buildings during the time of construction and the first year of occupancy. Volatile organic compounds (VOCs), formaldehyde and ammonia concentrations and emissions as well as temperature, humidity, and ventilation were measured. The study confirmed that the Finnish material classification system provides a basis to achieve good IAQ when comparing to the target values for pollutant concentrations given by the classification in real buildings. Based on the indoor air and emission results, reference values, i.e. "normal" and "abnormal values", were defined for the six- and twelve month-old buildings.

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