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DISSERTATION

Indoor Emissions - A Study on Various Sources of Volatile Organic Compounds in a Close-to-reality Model Room

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Abstract

This thesis deals with different aspects of indoor air quality in wood-based constructions. In a first step, two model rooms built from wood-based construction products were designed and constructed. This model rooms were used for different tests concerning indoor in quality in real rooms but under laboratory conditions. In both model rooms long-term VOC emissions were examined. The VOC emissions declined over a period of six months. The VOC profiles emitted by the wood-based materials were comparable to those determined in other studies. Climate parameters inside the model rooms were monitored to prove the suitability of the model room as a large-scale test set-up. Next, the model rooms were stepwise equipped with interior fittings. VOC emissions were analyzed after each fitting step to gain further information about the influence of different fitting stages on indoor air quality. Mutual influences of materials installed together in a room and their effect on indoor air quality were investigated. Both VOC concentration and VOC profile vary with the installed materials which partly also act as VOC sinks or barriers. Finally, six user scenarios were simulated to investigate the influence of user activities on indoor air quality. Beside VOC also aldehydes, particulate matter and CO_2 were analyzed. The results show distinct changes of these parameters due to user activities. The results to some extent exceeded those measured from building products but decreased quickly after the end of the activity. Altogether, the results of the present study offer new insights into indoor air quality in different utilization phases of wood-based constructions.

Kurzfassung

Die vorliegende Arbeit beschäftigt sich mit verschiedenen Aspekten der Innenraumluftqualität in Holzgebäuden. In einem ersten Schritt wurden zwei Modellräume aus Holzwerkstoffen konzipiert und errichtet. Mit Hilfe dieser Modellräume wurden Versuche zur Innenraumluftqualität im Realmaßstab unter Laborbedingungen durchgeführt. Zunächst wurden beide Modellräume langfristig hinsichtlich ihrer VOC-Emissionen untersucht. Es wurde festgestellt, dass in beiden Räumen die VOC-Emissionen über einen Zeitraum von sechs Monaten deutlich absinken. Untersuchungen zur Stabilität der Klimaparameter in den Modellräumen belegen ihre Eignung als Testsystem im Großmaßstab. In der nächsten Phase wurden beide Modellräume schrittweise mit Innenausbauten versehen. Nach jedem Ausbauschritt wurden die VOC-Emissionen analysiert. Dies lieferte neue Informationen über den Einfluss verschiedener Bauphasen auf die Innenraumluftqualität und die Veränderung durch das gemeinsame Einbringen verschiedener Materialien. Es wurden materialabhängige Änderungen der VOC-Emissionen sowie Senken- und Barriereneffekte beobachtet. Zuletzt wurden sechs Nutzerszenarien simuliert, um den Einfluss von Tätigkeiten des Raumnutzers auf die Innenraumluftqualität zu untersuchen. Neben VOC-Emissionen wurden im Rahmen dieses Versuchs Aldehydemissionen, Feinstaub- und CO_2-Konzentration erhoben. Die Ergebnisse zeigen, dass Nutzertätigkeiten zu sehr stark erhöhten Werten aller angeführten Parameter führen können. Die Werte überschreiten die Emissionen von Baustoffen teilweise deutlich, sinken jedoch nach Beendigung der Tätigkeit rasch wieder ab. Die Ergebnisse der vorliegenden Arbeit liefern neue Erkenntnisse zur Innenraumluftqualität in verschiedenen Nutzungsphasen von realen Holzgebäuden unter Ausschluss unerwünschter Umgebungseinflüsse.

Eigenständigkeitserklärung

Ich bestätige mit meiner Unterschrift, dass ich die Dissertation selbstständig verfasst, andere als die angegebenen Quellen und Hilfsmittel nicht benutzt und mich auch sonst keiner unerlaubten Hilfe bedient habe. Weiters bestätige ich, dass ich diese Dissertation bisher weder im Inland noch im Ausland in irgendeiner Form als Prüfungsarbeit vorgelegt habe.

Wien, 8. März 2016 Dipl.-Ing. Eva Höllbacher

Danksagung

Auch wenn eine Dissertation eine eigenständige wissenschaftliche Arbeit ist, so ist man bei aller Selbstständigkeit meist doch nicht von allen verlassen. Es lohnt sich daher, dem einen oder anderen seinen Dank auszusprechen.

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Abbreviations

µ-CTE	Microchamber
ACS	Air conditioning system
AgBB	Ausschuss zur gesundheitlichen Bewertung von Bauprodukten (Committee for Health-related Evaluation of Building Products)
BRI	Building related illness
C	Concentration in the emission test chamber
CEN	Comité Européen de Normalisation (European Committee for Standardization)
C_i	Concentration of substance i in the emission test chamber
CLT	Cross laminated timber
CPR	Construction Products Regulation
DAD	Diode array detector
DICL	Danish Indoor Climate Label
DIN	Deutsches Institut für Normung (German Institut for Standardization)
DNPH	2,4-dinitrophenylhydrazine
ECA	European Collaborative Action
EN	European Standard
FID	Flame ionization detector
FLEC	Field and Laboratory Emission Cell
GC	Gas-phase chromatograph
HDF	High density fiber board
HPLC	High performance liquid chromatography
I.D.	Inner diameter
IAQ	Indoor air quality
ISO	International Standardization Organization
L	Loading factor
LCI	Lowest concentration of interest
MAK	Maximale Arbeitsplatzkonzentration (workplace exposure limit)
MDF	Medium density fiber board
MS	Mass spectrometer

n	Air exchange rate
OSB	Oriented strand board
PM	Particulate matter
prEN	European draft standard
PTV	Programmed temperature vaporizing injector
q	Area specific air flow rate
RH	Relative humidity
RW	Richtwert (guide value)
SBS	Sick building syndrome
SER	Specific emission rate
SER_A	Area specific emission rate
SVOC	Semivolatile organic compound
T	Temperature
TC	Technical Committee
TDS	Thermal desorption system
TE	Toluene equivalents
TRK	Technische Richtkonzentration (technical reference concentration)
TS	Technical Specification
TVOC	Total volatile organic compounds
UV	Ultraviolet
VDI	Verein Deutscher Ingenieure (Association of German Engineers)
VOC	Volatile organic compound
VVOC	Very volatile organic compound
WHO	World Health Organization
X-lam	Cross laminated timber

1 Introduction

The wood working industry is an important economic factor in Austria and Europe. In 2011 this branch of industry had a share of 1 % of the whole Austrian economic performance and was one of the biggest employers in industry (Wolf, 2012). Its main products are sawn timber and engineered wood products, which are further processed to construction products or furniture. Traditionally, the Austrian wood working industry is an export oriented sector with an export share of 68 % in 2014. The biggest part of all products is exported to the European Union (Holzindustrie, 2015). In Austria, timber constructions gain more and more importance both in residential and public buildings. In 2008 39 % of all building projects were timber constructions compared to 25 % in 1998. Most of these building projects were private residences followed by agricultural functional buildings (Stingl et al., 2011). This is a clear increase within ten years, which is not only restricted to Austria, but reflects a pan-European trend to sustainable building. Still, there is one important characteristic of wood products, which has to be kept in mind during planning and construction: Natural and processed wood emit volatile organic compounds (VOC). These VOC differ depending on the wood species and the manufacturing process (Risholm-Sundman et al., 1998, Jensen et al., 2001). However, wood shares this characteristic with many other products for indoor use (Salthammer and Bahadir, 2009). VOC emissions are of interest in the indoor environment, because they strongly influence indoor air quality. In times of rising health awareness, indoor air quality is an important factor associated with human well-being (Jones, 1999). The increasing attention, which is centered on indoor air quality, is based on health concerns associated with the indoor environment, which have increased distinctly since the 1970s (Horvath, 1997). At that time the oil crisis initiated efforts to reduce energy consumption. One approach to reach this goal was to reduce energy loss through building envelopes. More airtight and therefore more energy-efficient buildings were created, which resulted in an accumulation of indoor pollutants as a consequence of reduced air exchange within the buildings (Zhang and Smith, 2003). A lot of research work was done during the last decades, identifying numerous sources of indoor air contaminants (Weschler, 2009). But still new substances and reaction processes are identified in the indoor environment, which are partly not very well studied (Weschler, 2011).

Several approaches were developed to guarantee good indoor air quality. This also includes analytical methods to measure and quantify VOC emissions from construction and furnishing products. Based on these measurement procedures, certifications and lawful regulations have been established to provide consumers with low emission products for indoor use. The common method to evaluate VOC emissions is the testing of single materials under controlled climate conditions in a laboratory environment. However, it is known that different materials and human activities tend to influence their mutual VOC emissions (Salthammer and Bahadir, 2009). Very often sources of VOC also act as sinks, which alter the VOC concentration and profile in an indoor environment by retaining certain substances and reemitting them at a later point in time (Tichenor et al., 1991, Meininghaus et al., 2000, Won et al., 2000). Laboratory tests on single materials, therefore, do not necessarily reflect the reality situation. On the other hand, investigations in real buildings are difficult and often hindered by environmental effects such as climate, season or contaminated outdoor air (Järnström et al., 2006, ECA, 2003). Therefore, a special test design is needed to study the various influences and mutual interactions of different VOC emission sources in a realistic indoor environment but to the exclusion of unwanted environmental effects. This problem was successfully addressed in the present thesis by designing and constructing a special test set-up that fulfilled the testing requirements and allowed for investigations on VOC emissions in a conventionally equipped environment under conditions as close as possible to those in a laboratory. For this purpose, two model rooms were constructed and adapted for several research tasks dealing with possible influence factors on indoor air quality with a focus on the use of wood products. The results of this work contribute to a better understanding of interactions of wood and other materials as VOC emission sources and sinks in the indoor environment.

2 State of the art

This chapter comprises a short survey of research on indoor air quality, possible sources of VOC in the indoor environment and consequences of an affected indoor environment. VOC emissions from wood-based products are discussed in more detail. Furthermore, standardized test procedures and national as well as international regulation and classification schemes are presented.

2.1 VOC and indoor air quality

2.1.1 Definition of VOC

The abbreviation VOC means volatile organic compound and comprises substances with either a low boiling point or a high vapor pressure. Which substances are to be classified as VOC is defined in several different definitions. The World Health Organization (WHO, 1989) describes a classification according to the boiling point of a substance (see Table 1).

Table 1: Classification of volatile substances according to their boiling point (WHO, 1989)

description	Abbreviation	Boiling point area [°C]
very volatile organic compounds	VVOC	< 0 to 50-100
volatile organic compounds	VOC	50-100 to 240-260
semivolatile organic compounds	SVOC	240-260 to 380-400
organic compounds associated with particulate matter or particulate organic matter	POM	> 380

ECA report No. 19 (ECA, 1997b) recommends a modified VOC definition. It includes all substances which can be found in the retention area between n-hexane (C6) and n-hexadecane (C16) after gas chromatographic separation on a non-polar column. All substances detected in this range shall be included in the cumulative value of all VOC, the total volatile organic compounds (TVOC). The test standard ISO 16000-6 (DIN, 2004) and the German AgBB scheme (AgBB, 2015) also refer to this definition.

2.1.2 VOC in the indoor environment

In Europe, people spend approximately 90 % of their time indoors (Schweizer et al., 2006). About 60 % of the day is spent at home while the rest of the time is divided between the work place and other enclosed spaces (Schweizer et al., 2006, Brasche and Bischof, 2005). Indoor air quality in living or working spaces is influenced by indoor pollutants which are released from a high number of possible natural and anthropogenic sources (Kostiainen, 1995, Wolkoff, 2013). These indoor pollutants include mainly gases like CO and CO_2, as well as nitrogen oxides, particulate matter and VOC (Salthammer and Bahadir, 2009). Indoor pollutants have undergone severe changes within the last decades. On the one hand dangerous substances have been reduced or even banned from the indoor environment. This trend led for instance to a clear decrease of occupants' exposure to formaldehyde from composite wood products or lead and mercury from paints. On the other hand new pollutants, mainly new VOC, arise and raise new challenges to obtain good indoor air quality (Weschler, 2009). Many scientific studies deal with VOC in indoor air and the identification of possible sources (Hodgson et al., 2002, Missia et al., 2010, Reitzig et al., 1998, Parra et al., 2008). These sources comprise outdoor air, building and furnishing materials as well as various user activities (Salthammer and Bahadir, 2009, Wolkoff and Nielsen, 2001). Remarkable amounts of VOC are so-called secondary VOC which are generated as reaction products in indoor air (Uhde and Salthammer, 2007, Weschler and Shields, 1997).

Investigations on indoor air quality of buildings identify a high number of different VOC. Kostiainen (1995) detected more than 200 individual substances in the indoor air of occupied houses, mainly alkylbenzenes, alkanes, terpenes, aldehydes and chlorinated aliphatic hydrocarbons. Comparable results were obtained by the AIRMEX study which surveyed different buildings in eleven European cities over a time period of five years. The authors identified acetone, different alkanes, formaldehyde and hexanal as the most abundant VOC in indoor air (Geiss et al., 2011). The AIRMEX study also focused on seasonal influences on indoor air quality. Increased VOC concentrations were measured in winter due to lower ventilation rates (Geiss et al., 2011). This is confirmed by other studies who also observed seasonal variations in VOC concentration due to changing ventilation rates (Missia et al., 2010, Liang et al., 2014b). Schlink et al. (2010) even identified the season as the most important influence factor on the overall VOC concentration in indoor air. Generally, low ventilation rates are said to execute a strong influence on VOC concentration as it leads to an accumulation of VOC indoors (Jia et al., 2008, Zhang and Smith, 2003). Low air exchange rates are usually attributed to energy-efficient, airtight constructions (Jones, 1999, Weschler, 2009). However, good indoor air quality was observed in both passive and conventional houses with sufficiently high ventilation rates due to mechanical ventilation (Langer et al., 2015).

In unoccupied houses building products and furnishings are the main sources of VOC and are supposed to decrease over time (Brown et al., 1994). Hodgson et al. (2000) for instance identified elevated concentrations of α-pinene, ethylene glycol, formaldehyde, acetaldehyde, hexanal and acetic acid in newly finished but unoccupied buildings. Most of these substances could be attributed to the use of wood products in construction and furnishing and generally decreased over time (Hodgson et al., 2002). Comparable results were obtained by Park and Ikeda (2006) who investigated more than 300 new and older houses over a period of three years. Here, VOC concentrations were rather high after the completion of new buildings or renovation actions, but decreased dramatically during the first year and reached values comparable to those in older homes. Investigations in lately renovated buildings also show elevated concentrations of different VOC of which most could be attributed to certain materials introduced during renovation (Reitzig et al., 1998). Studies carried out in different construction stages of buildings identified typical VOC related to each construction stage and the used building materials (Järnström et al., 2007, Liang et al., 2014a). However, occupancy may change the VOC profile tremendously. Järnström et al. (2006) examined indoor air quality in flats in Finland directly after completion and six and twelve months after occupancy. Although, the VOC concentration decreased during the first six months, a distinct change in the VOC profile could be observed. New substances arose due to the influence of occupants and replaced the VOC emitted by the applied building products. This trend was also observed by other authors (Yamaguchi et al., 2006, Liang et al., 2014b). According to findings of Derbez et al. (2014) these newly detected VOC increase temporarily when certain human activities take place while emissions from building materials show a clear decrease over time. This conclusion is supported by Missia et al. (2010) who examined emissions of BTEX, terpenes and carbonyls in the indoor air of different building types in five European cities and found strong evidence that the hydrocarbon emissions from building materials may get insignificant over time compared to other indoor sources. It is additionally stated by several studies, that although, outdoor air also influences indoor air by infiltration, indoor sources are the prevailing origin of VOC in the indoor environment (Edwards et al., 2001, Missia et al., 2010, Geiss et al., 2011). Especially in permanently occupied buildings, like private apartments, human activities and consumer products associated with them are an important VOC source. Although, the high influence of user behavior on indoor air quality is known from various studies (Steinemann et al., 2011, Nazaroff and Weschler, 2004, Petry et al., 2014) research on VOC for a long time concentrated on continuous, steadily decreasing sources like building products, which have been studied thoroughly during the last decades (ECA, 1997a, Gehrig et al., 1994, Lin et al., 2009, Guo et al., 2004b, Cheng et al., 2015). The influences of consumer products are manifold and not limited to VOC emissions. One commonly known source of indoor pollutants is tobacco smoke, which contains various substances like hydrocarbons, formaldehyde and acetaldehyde (Izu et al., 2014, Loefroth et al., 1989), but also leads to elevated particle emissions (Sahu et al., 2013). Other indoor combustion processes like burning candles or open fireplaces also generate high amounts of indoor pollutants (Schripp et al., 2014, Guillaume et al., 2013, Derudi et al., 2012, Petry et al., 2014). High emissions of VOC have been identified from scented consumer products like cleaning agents, air fresheners or personal care products (Steinemann, 2015, Singer et al., 2006b). These products are of special interest, as their VOC emissions are likely to form secondary pollutants due to reactions with indoor oxidants (Nazaroff and Weschler, 2004, Norgaard et al., 2014, Singer et al., 2006a). In the beginning, research on indoor air quality focused on primary emissions, but until now deeper knowledge was gained concerning reaction products of VOC and their influence on indoor air (Weschler, 2011). Literature describes various reaction pathways which are involved in the formation of secondary pollutants. Especially oxidation reactions of VOC, emitted by building or consumer products, are of interest (Uhde and Salthammer, 2007). Oxidation of unsaturated hydrocarbons (e.g. terpenes) by ozone or hydroxyl radicals, for instance, leads to the formation of aldehydes and ketones which have a low odor threshold and a higher irritation potential than their precursors (Weschler and Shields, 1997). Additionally, reactions between certain terpenes and ozone are a source of indoor particles. Weschler and Shields (1999) identified a ten times higher particle concentration in offices with a slight limonene source than in the control office. Secondary pollutants are of special interest as they are suspected to have a strong negative influence on human well-being, exceeding those of primary VOC (Wolkoff et al., 1997). Wolkoff and Nielsen (2001) criticized that investigation on indoor air quality focus mainly on non-reactive primary VOC, while reactive substances and secondary indoor pollutants, which are an important cause of indoor health complaints, are partly ignored by the common sampling techniques.

2.1.3 Health effects and perceived indoor air quality

As already mentioned in the above chapter, indoor air quality affects human health in various ways. Generally, possibly harmful indoor pollutants can be divided in pollutants from biological sources like allergens or bacteria and viruses, and pollutants from non-biological sources which include VOC, particles, fibers and inorganic gases (Jones, 1999). The negative health effects of poor indoor air quality are manifold. Frequently described symptoms are irritations of mucous membranes of eyes, nose and throat as well as headaches (Wolkoff and Nielsen, 2001). These symptoms are often related to two clinical pictures: the building related illness (BRI) and the sick building syndrome (SBS). Although, the naming may hypothesize an analogy between the two diseases, BRI is well distinguished from SBS. BRI describes clear medical conditions for which a specific cause can be found. Horvath (1997) groups BRI in three categories. These categories comprise allergic or immunologic diseases, infections and diseases due to exposure to chemicals or other substances. The symptoms are always well-defined and clearly related to a certain cause. In contrast there is no universally accepted clinical definition of SBS (Redlich et al., 1997). SBS is much more common than BRI and usually comprises a collection of non-specific symptoms which only occur in particular buildings, but for which no cause is apparent. These symptoms include mucous membrane irritations, neurotoxic effects, respiratory symptoms, skin irritation and chemosensory changes. The symptoms and their severity vary greatly in cases of SBS in different buildings, but also variations within the same building are possible due to different microenvironments and host effects (Horvath, 1997, Redlich et al., 1997). Most cases of SBS are multifactorial in origin and are related to indoor air contaminants like VOC, dust or bioaerosols, physical factors like temperature or relative humidity, an inadequate ventilation rate or functioning of the ventilation system and to psychosocial factors like stress or job dissatisfaction. Additionally, increasing awareness of chemical hazards and therefore increasing anxiety support the occurrence of SBS (Redlich et al., 1997, Jones, 1999, Horvath, 1997, Skov et al., 1990). There is some evidence that persons suffering from SBS are more sensitive towards indoor air quality than other subjects (Norbäck et al., 1990, Kjærgaard et al., 1991, Lin-lin et al., 2011). All cases of SBS have in common, that the symptoms improve when the person concerned leaves the building or when measures are taken to improve indoor air quality (Redlich et al., 1997). Many scientific studies deal with the reasons and consequences of SBS. Frequently, the attribution to a certain defined cause is not possible. Salonen et al. (2009) identified no exceptional emission sources or elevated concentrations when investigating VOC, formaldehyde and ammonia in office buildings affected by SBS. Comparable results were obtained by Hutter et al. (2006) who found a strong correlation between SBS symptoms and floor dust in a new office building, but could not clarify whether the symptoms where due to irritation by contents of the dust or psychological factors. However, the removal of the flooring which caused the dust led to a distinct improvement of the complaints. Wargocki et al. (1999) exposed 30 female subjects to an indoor emission source while performing office work. The test persons performed less well and experienced increased SBS symptoms when the pollution source was present. As no changes in TVOC could be observed compared to the empty room, the negative influence of the emissions source must be correlated to an altered VOC profile.

Indoor health complaints but also a negative perception of indoor air quality are mainly attributed to VOC emissions (Wolkoff, 2005). Different diseases like asthma and rhinitis but also increasing lifetime cancer risks have been correlated to exposure to elevated VOC concentrations (Billionnet et al., 2011, Guo et al., 2004a, de Blas et al., 2012). Possible health effects of VOC are therefore well studied. Mølhave et al. (2000, 1986) carried out several investigations on human reactions to different VOC. Varying results were obtained depending on the examined substances and the study design. Mucous membrane irritation was observed due to exposure to high concentrations of a mixture of 22 common indoor VOC (Mølhave et al., 1986), while no effect could be stated at high concentrations of single terpenes (Mølhave et al., 2000). However, in both cases the presence of VOC at elevated concentrations was clearly correlated with odor perception. Comparable results were obtained by Gminski et al. (2011) who exposed human subjects to high VOC concentrations from pinewood. The short-term exposure caused no negative health effect while the recognized odor was rather classified as more pleasant than unpleasant. Many VOC are odorous. This odor may lead to a positive as well as to a negative perception of indoor air quality. However, obtaining sound information about the perception and consequences of odorous VOC in indoor environments is difficult. Wolkoff et al. (2006) list possible problems related to analysis of odor which include personal and cultural factors influencing odor perception, confusion between odor perception and sensory irritation as well as analytical problems due to very low odor thresholds which are partly found to be below the limit of detection. As most products used indoors do not emit only one single VOC but a mixture of various different substances, odor perception from indoor materials also depends on alterations in VOC concentration and profile. Studies carried out on different materials showed that alterations in the VOC profile clearly influence odor perception. Generally, odor intensities showed no or only a modest decrease over time while primary VOC emissions clearly declined. This is caused by the formation of secondary VOC (Knudsen et al., 1999, Knudsen et al., 2003).

Various concepts have been developed for the assessment of indoor air quality like the Air Quality Guidelines for Europe of the WHO (WHO, 2000) or the TVOC concept (ECA, 1997b, Mølhave, 1991) which is widely used in adapted versions (Salthammer, 2011), although, it is criticized because it has no toxicological relevance and no direct cause relation could be stated between TVOC and indoor health complaints (Andersson et al., 1997, Mølhave et al., 1997). Two national concepts to assess indoor air quality in industrial and non-industrial environments are in the following described as examples. In Austria and Germany work place exposure limits are applied to evaluate health hazardous pollutants in industrial environments (Arbeitsinspektion, 2014, ABrBI, 2006). In Austria the work place exposure limits comprise so-called MAK and TRK values which are defined in a special regulation, which is called “Grenzwerteverordnung” (BMASK, 2011). However, these limit values are usually not applied in non-industrial environments. In both Austria and Germany indoor guide values exist for the assessment of indoor air quality. These guide values are not legally binding but justified by health reasons. Indoor guide values for different substances and the TVOC have been developed by the German Committee on Indoor Guide Values. These guide values are divided in two groups, RW I and RW II. RW II is related to possible health effects and based on toxicological and epidemiological knowledge. RW I is the concentration of a substance in indoor air at which even at livelong exposure time no negative health effects are expected (Umweltbundesamt, 2012). A slightly different concept was developed in Austria. The indoor guide values for VOC comprise tetrachloroethylene, styrene, toluene and the TVOC. For the three single substances guide values with different reference periods from one hour to seven days are published (BMLFUW, 2010d, BMLFUW, 2010c, BMLFUW, 2010b). TVOC is classified in five groups from low to very high (BMLFUW, 2010a).

2.2 VOC from wood-based materials

Wood products, which are widely used in construction work, interior fittings and furnishing today, are known to emit specific VOC into the indoor environment (Salthammer and Bahadir, 2009, Yu and Kim, 2012). These VOC differ with the wood species and the relevant production process (Jensen et al., 2001). Softwood, which is widely used in the production of engineered wood products, emits mainly substances belonging to the group of aldehydes and terpenes (Roffael, 2006). In untreated softwood, up to 90 % of the VOC emissions are terpenes (Englund and Nussbaum, 2000). Steckel et al. (2010) measure 85 % terpenes in the VOC emissions from Norway spruce. The most abundant substance is α-pinene, followed by β-pinene. This is also confirmed by studies of Wajs et al. (2007), who identify a comparable VOC profile in European larch. For Scots pine, terpenes amount to an even higher share of up to 98 % of the VOC emissions. Here, 3-carene and α-pinene are the most abundant substances (Englund and Nussbaum, 2000, Wajs et al., 2007). Hardwood shows a different emission behavior, with organic acids and aldehydes being the most abundant substance groups, whereas normally no terpenes are emitted (Risholm-Sundman et al., 1998, Paczkowski et al., 2013). In the VOC profile of most hardwood species, hexanal is the most abundant aldehyde (Paczkowski et al., 2013). Especially oak and cherry show high emission rates of acetic acid (Risholm-Sundman et al., 1998). The emission of terpenes from natural wood is due to the fact that terpenes are to a great extent present within the wood extractives. Whereas softwood extractives contain high shares of monoterpenes, which are responsible for the typical odor, mainly higher terpenes are identified in the extractives of hardwood (Fengel and Wegener, 2003).

VOC emissions from engineered wood products are highly influenced by the production process and therefore often show a different emission behavior, when compared to unprocessed wood. Wood products, composed of multiple layers of timber bonded together like glue laminated timber, usually show VOC emissions comparable to those of the wooden raw material (Höllbacher et al., 2015). On the other hand, thermal treatments during production processes lead to an alteration of VOC emissions. OSB (oriented strand boards) for instance emit increased amounts of aldehydes due to the hot pressing production process (Salthammer et al., 2003). Makowski et al. (2005) measure a maximum share of 64 % aldehydes in the VOC emissions of OSB. However, this magnitude is reached eight weeks after production. Earlier measurements identify terpenes as the most abundant substance group. These results are supported by findings of Wilke et al. (2013). Here, six OSB samples of unknown production date are evaluated. Although, all samples show different VOC concentrations, the most concentrated substances are aldehydes (Wilke et al., 2013). Studies on particleboards show, that aldehydes are the predominant substance group, followed by terpenes. More than 50 % aldehydes are measured in particleboards made of pinewood. Hardwood particleboards even emit about 90 % aldehydes. In all cases, the most abundant aldehyde is hexanal (Baumann et al., 2000). The terpene profile from particleboards shows huge differences depending on the processed wood species (Baumann et al., 1999). In medium density fiberboards (MDF) the most abundant substances usually belong to the groups of aldehydes and ketones, whereas no or only very small amounts of terpenes are emitted (Baumann et al., 2000, Baumann et al., 1999, Koontz and Hoag, 1995). Baumann et al. (2000) generally stated lower VOC emissions from MDF compared to particleboard, except MDF made from Southern pine. Investigations on the VOC emissions of MDF shortly after production show no substance concentrations above 5 µg m^{-3} (Brown, 1999). VOC emissions from hardboards are even lower concentrated than those from MDF. Que et al. (2013) identify only very small amounts of terpenes and traces of hexanal in the VOC emissions of hardboards. The very low amounts of terpenes in both MDF and hardboards can be explained by the severe thermal treatments during the production process, including the refining and the hot-pressing process, where especially terpenes with low boiling points are removed due to elevated temperature, pressure and humidity (Que et al., 2013, Baumann et al., 1999). Thermal wood modification, which is applied to modify wood resistance and color, is also known to distinctly influence the VOC emissions. Mainly increasing emissions of furfural and acetic acid are measured in both softwood and hardwood (Graf et al., 2004, Manninen et al., 2002). Fumigation with ammoniac for color adaption leads to even stronger emissions of acetic acid. Additionally, high amounts of acetamide are identified in fumigated oak wood (Volkmer et al., 2014).

Further processing of engineered wood products again alters the VOC emissions. Results from Wiglusz et al. (2002) demonstrate, that particleboards covered with a decorative paper foil emits significantly less VOC than the uncovered particleboards. Comparable effects are shown for floorings by Kim (2010). Here, laminate flooring produced from high density fiberboards (HDF) and plywood covered with fancy veneer, respectively, is studied. The findings of this investigation show clearly lower VOC emissions from the finished laminate flooring than from both HDF and plywood. Additionally, a further reduction of VOC emissions is obtained for the plywood flooring after the fancy veneer is coated with a UV cured coating. However, Cecchi (2014) identifies increased VOC emissions in prefinished composite parquet due to the application of adhesives and finishes.

2.3 Regulation and certification of VOC emissions in Europe

Besides the assessment of indoor air quality, several European countries developed systems to limit the VOC emissions from building and furnishing materials and partly also from consumer products. Different approaches were chosen to evaluate and, if possible, restrict VOC emissions. Generally, these approaches can be divided in two groups: limitation and information. Systems based on the limitation approach normally define limit values for VOC emissions which must not be exceeded. Products which do not fulfill the requirements are either banned from the market or denied a certification. Informative systems rely on the sensibility and decision of the consumer by informing about the amount of VOC emitted. The relevant information is usually transferred by different emission classes. Another factor dividing the existing schemes in Europe is whether they are mandatory or voluntary. Some countries define national regulations for VOC emissions from products of daily live, while others offer voluntary certification systems. Additionally to mandatory and voluntary systems that focus exclusively on VOC emissions there are several European ecolabels which include limit values for indoor pollutants in their guidelines. Generally, ecolabels cover a broad range of criteria and products. The main aim of these labels is to guarantee sustainable and environmentally friendly products which represent no health hazard for consumers.

2.3.1 Mandatory systems

2.3.1.1 Germany

AgBB is the relevant scheme for the evaluation of VOC emissions from building products in Germany. The scheme was evaluated by the Committee for health-related evaluation of building products (Ausschuss zur gesundheitlichen Bewertung von Bauprodukten, AgBB) and first published in 2000 and is based on an evaluation scheme for VOC emissions from solid flooring materials presented in ECA Report No 18 (ECA, 1997a). AgBB gets updated regularly, the latest version was published in 2015 (AgBB, 2015). AgBB is part of the general technical approval in Germany for floorings, flooring adhesives and coatings as well as underlays for floorings and decorative wallcoverings (DIBt, 2015). In the area of wood and wood products at the moment laminate floorings, parquet and wood floorings except solid wood floorboards are included in AgBB (n.n., 2010). VOC measurements are conducted according to the ISO 16000 series. Tests are performed in an emission test chamber. The relevant standards for sampling and testing are 16000-9 (DIN, 2008) and 16000-11 (DIN, 2006a) or CEN/TS 16516 (CEN, 2013), respectively. VOC analysis is done as defined in ISO 16000-6 (DIN, 2004). Excepted hereof are aldehydes which are sampled and analyzed by the DNPH method as described in ISO 16000-3 (DIN, 2010a). VOC concentrations determined in the emission test chamber are extrapolated to a model room with dimensions of 3 x 4 x 2.5 m^3. Air exchange rate is 0.5 h^{-1} and the uniformly claimed detection limit is 1 µg m^{-3}. All VOC with a concentration equal to or above 5 µg m^{-3} have to be considered in the assessment of single substances as well as in the calculation of the TVOC. AgBB refers to the definition of VOC as described in ISO 16000-6 (DIN, 2004) (see Chapter 2.1.1). AgBB defines limit values for TVOC, TSVOC and carcinogenic substances. TVOC is the sum of the concentration of all individual substances with concentrations equal to or above 5 µg m^{-3} in the retention range from C_6 to C_{16}. TSVOC is the sum of the concentration of all individual substances with concentrations equal to or above 5 µg m^{-3} in the retention range from above C_{16} to C_{22} (Däumling, 2012). Additionally, single substances are assessed according to their toxicological relevance by a risk factor, which is below described in more detail. A limit value exits for the sum of all substances which cannot be subsumed by this risk factor. The limit values as defined by AgBB can be seen in Table 2. AgBB is a mandatory limitation system. Therefore, products may only gain access to the German market, if they fulfill the requirements of the limit values defined within AgBB. Products which exceed the limits are excluded from the market.

Table 2: Limit values according to AgBB (AgBB, 2015)

Substance class	day 3	day 28
TVOC [µg m^{-3}]	≤ 10000	≤ 1000
Carcinogenic substances EU-Category 1A and 1B [µg m^{-3}]	≤ 10	≤ 1
TSVOC [µg m^{-3}]	-	≤ 100
R value	-	≤ 1
Sum of VOC without LCI [µg m^{-3}]	-	≤ 100

VOC tests can be stopped seven days after the start, if the measured VOC concentrations are less than 50 % of the limit values for day 28. There must not be any significant concentration increase of single VOC compared to day 3.

For the toxicological assessment of single substances threshold values are defined. Below this so-called lowest concentration of interest (LCI) no negative effect of a specific VOC is expected. It is assumed, that the potential negative effects of single VOC in a mixture are additive. That means that their potential health risks have to be summarized. Therefore, a risk factor R, as described in ECA Report No. 18 (ECA, 1997a), is determined. For each VOC i with a concentration above 5 µg m^{-3}, the ratio R_i of the concentration C_i to the relevant LCI value is calculated by the following formula:

$$R_i = \frac{C_i}{LCI_i} \tag{1}$$

R_i = risk factor R of compound i

C_i = concentration of compound i

LCI_i = lowest concentration of interest of compound i

For $R_i < 1$ no negative consequences are assumed. If several compounds with an existing LCI value are identified their R_i-values are summed to the R-value which also may not exceed 1 (Däumling, 2012). The current list of LCI values is attached to the AgBB-scheme and contains LCI values for approximately 184 substances (Umweltbundesamt, 2015a). The list is regularly updated.

Additionally to the determination of VOC concentrations sensory evaluations of odorous emissions shall get mandatory in future. These sensory tests are performed after 28 days by a trained panel according to ISO 16000-28 (DIN, 2010b) and VDI 4302 (VDI, 2012). Further knowledge about the suitability of the testing method for different building products shall be gained from a pilot phase which started in 2012. The results will provide the basis to decide about the way in which sensory tests will be included in the AgBB scheme in future (AgBB, 2015).

2.3.1.2 France

Since January 2012 a VOC label is mandatory for building and decoration products in France. This label is designated for all floorings, wall coverings and ceilings, partition walls, inserted ceilings, insulation materials, doors and windows, paints and lacquers as well as all materials necessary for the preparation or installation of the above mentioned products (Ministère de l'écologie, 2014). Each product is assigned to an emission class (A+, A, B, C). The classes are defined by limit values for TVOC and selected VOC (see Table 3). All products that exceed the limit values of classes A+ to B are assigned to class C (Ministère de l'écologie; du développement durable; des transports et du logement, 2011).

Table 3: Limit values for the emission classes after 28 days [µg m^{-3}] (Ministère de l'écologie; du développement durable; des transports et du logement, 2011)

substance	C	B	A	A+
TVOC	> 2000	< 2000	< 1500	< 1000
formaldehyde	> 120	< 120	< 60	< 10
acetaldehyde	> 400	< 400	< 300	< 200
toluene	> 600	< 600	< 450	< 300
tetrachloroethylene	> 500	< 500	< 350	< 250
xylene	> 400	< 400	< 300	< 200
1,2,4-trimethylbenzene	> 2000	<2000	< 1500	< 1000
1,4-dichlorbenzen	> 120	< 120	< 90	< 60
ethylbenzene	> 1500	< 1500	< 1000	< 750
2-butoxyethanol	> 2000	< 2000	< 1500	< 1000
styrene	> 500	< 500	< 350	< 250

The emission tests are performed in an emission test chamber according to ISO 16000-9 (DIN, 2008) or in an emission test cell according to ISO 16000-10 (DIN, 2006c). Sampling, sample storage and preparation of test specimen are done according to ISO 16000-11 (DIN, 2006a). Sampling and analysis of formaldehyde and acetaldehyde is done according to ISO 16000-3 (DIN, 2010a). All other substances are analyzed according to ISO 16000-6 (DIN, 2004). The chamber results are extrapolated to the European reference room as defined in CEN/TS 16516 (CEN, 2013). The usual test duration is 28 days. However, sound data from shorter VOC tests may be accepted (eurofins, 2015d, Ministère de l'écologie; du développement durable; des transports et du logement, 2011).

The aim of the French VOC label is not the ban of certain products from the market, but the information of consumers. The label is mandatory only for products designated for commerce (Ministère de l'écologie; du développement durable; des transports et du logement, 2012).

2.3.1.3 Belgium

In August 2014 a regulation concerning VOC emissions from building products was published in Belgium. The regulation comprises all kinds of floorings and products needed for the installation of floors like coatings, adhesives and screeds. Floorings completely made from natural stone, ceramics, glass and steel are exempted of this regulation (eurofins, 2015a). Sampling and testing is done according to ISO 16000-11 (DIN, 2006a) and CEN/TS 16516 (CEN, 2013).

The Belgian VOC regulation defines limit values after a testing period of 28 days. Limit values are set for TVOC, TSVOC, carcinogenic substances and selected VOC (see Table 4). Additionally, the risk factor R is calculated in the same way as in the AgBB scheme and must not exceed 1. However, the Belgian regulation does not define its own LCI values as the AgBB scheme does, but refers to the harmonized list of the European Joint Research Center as described in ECA Report No. 29 (ECA, 2013, n.n., 2014).

Table 4: Limit values according to the Belgian VOC regulation (n.n., 2014)

Substance class	day 28
TVOC [µg m^{-3}]	≤ 1000
Carcinogenic substances EU-Category 1A and 1B [µg m^{-3}]	≤ 1
TSVOC [µg m^{-3}]	≤ 100
R value	≤ 1
acetaldehyde [µg m^{-3}]	≤ 200
toluene [µg m^{-3}]	≤ 300
formaldehyde [µg m^{-3}]	≤ 100

2.3.2 Voluntary systems

2.3.2.1 Finland – M1

M1 is a voluntary emission classification system for building materials. It defines two emission classes (M1, M2). Each class has specific limit values for TVOC, carcinogenic substances, formaldehyde, ammonia and odor (see Table 5). All materials that do not comply with the limit value for class M2 are classified as M3. Emissions from products which normally cover small areas (2 m²) like doors, windows and grout and very small areas (0.4 m²) like sealants may also be expressed as air concentrations in the European reference room (CEN, 2013). Their relevant limit values for M1 can be seen from Table 5, column 3. No restrictions are placed on the use of brick, natural stone, ceramic tile, glass, metal surfaces, board and log surfaces of unprocessed wood (excluding hardwood). The VOC emissions of fresh wood may nevertheless exceed the M1 limit. Only tested materials are granted are granted the M1 label (n.n., s.a.).

Measurements are either performed in an emission test chamber according to ISO 16000-9 (DIN, 2008) and CEN/TS 16516 (CEN, 2013) or with an emission test cell according to ISO 16000-10 (DIN, 2006c). The minimum test chamber volume has to be 0.02 m³. VOC sampling and analysis is done according to ISO 16000-6 (DIN, 2004). Formaldehyde is analyzed according to ISO 16000-3 (DIN, 2010a). Sensory evaluation is done according to ISO 16000-28 (DIN, 2010b) or using a CLIMPAQ as described by Gunnarsen et al. (1994). Sampling, sample storage and preparation of test specimen is done according to ISO 16000-11 (DIN, 2006a). Building products are tested after 28 ± 2 days (VTT, 2015).

Table 5: Limit values for the emission classes after 28 days (n.n., s.a.)

Examined qualities	**M1 [µg $m^{-2}h^{-1}$]**	**M2 [µg $m^{-2}h^{-1}$]**	**M1 small areas [µg m^{-3}]**
TVOC	< 200	< 400	< 20
Formaldehyde	< 50	< 125	< 10
Ammoniac	< 30	< 60	< 10
Carcinogenic substances EU-Category 1A and 1B	< 5	< 5	< 1
Odor (dissatisfaction with odor shall be below 15%)	not odorous	not odorous	not odorous

2.3.2.2 Denmark – DICL

The Danish Indoor Climate Label (DICL) mainly focuses on the duration it takes until emissions from a new, freshly installed building product in a standard room are below an acceptable level. A time factor is calculated from the VOC concentration and a sensory evaluation (Danish Society of Indoor Climate, 2000). A sample is placed in an emission test chamber. Air samples are taken at defined time points and analyzed on their VOC content. The relevant substances comprise aldehydes, amines, isocyanates, acids and sulfurous compounds. The measured chamber concentrations are extrapolated to a model room as defined in ISO 16000-9 (DIN, 2008). The determined VOC concentrations are compared to odor and irritation thresholds. The result of the slowest emitting substance with the lowest odor or irritation threshold is relevant for the calculation of the time factor. This time factor is the duration until the moment when the relevant substance reaches half its odor or irritation threshold in the model room. Subsequently, odor intensity and acceptability are evaluated by sensory tests. Additionally, particle emissions are determined for ceiling structures. The product may be certified if the emissions are classified as low or average (Danish Society of Indoor Climate, 2000).

2.3.2.3 France – AFSSET

The voluntary French guideline “Protocole AFSSET 2009” was published by the French environmental agency AFSSET (today called ANSES). It is the aim of this guideline to reduce VOC concentrations in indoor air (eurofins, 2015e).

Specific emission factors of a product are determined after three and 28 days in an emission test chamber according to ISO 16000-9 (DIN, 2008) or in an emission test cell according to ISO 16000-10 (DIN, 2006c). VOC sampling and analysis is done according to ISO 16000-6 (DIN, 2004). Formaldehyde is analyzed according to ISO 16000-3 (DIN, 2010a). Limit values are defined for TVOC, carcinogenic substances and the sum of unidentified VOC (see Table 6). The risk factor R is calculated in the same way as in the AgBB scheme and must not exceed 1. A list of 165 LCI values is available for AFSSET. Additionally, allergenic substances have to be determined. The existence of such substances in a product has to be indicated as an information for allergic and sensible persons (AFSSET, 2009).

Table 6: Limit values according to AFSSET (AFSSET, 2009)

Substance class	day 3	day 28
TVOC [µg m^{-3}]	≤ 10000	≤ 1000
Carcinogenic substances EU-Category 1A and 1B [µg m^{-3}]	≤ 10	≤ 1
R value	-	≤ 1
Sum of unidentified VOC or VOC without LCI [µg m^{-3}]	-	≤ 100

2.3.3 Ecolabels

2.3.3.1 Austrian Ecolabel

The Austrian Ecolabel was founded in 1990 with the vision to support sustainable and environmentally friendly products and services in Austria (BMLFUW, s.a.-b). Products from the areas of building and living, household and cleaning, gardening, office, paper and print, green energy and mobility are awarded with the Austrian Ecolabel. Limit values for VOC emissions exist mainly for products from the area of building and living like mattresses, furniture, floorings, wood and wood products and paints and lacquers (BMLFUW, s.a.-a).

2.3.3.2 The Blue Angel

The Blue Angel is the oldest ecolabel worldwide. It covers 80 product categories and more than 12000 environmentally friendly products and services (eurofins, 2015b, RAL-gGmbH, s.a.-a). As the Austrian Ecolabel, the Blue Angel also defines criteria for VOC emissions mainly for interior building and furnishing products like floorings and related products, insulation materials, wall paints, furniture, sealants, mattresses and wood products (eurofins, 2015b).

2.3.3.3 Natureplus

Natureplus is an international label for high quality, sustainable and environmentally friendly building and furnishing products. High demands on quality regarding hygiene, health and environmental protection are part of the basic criteria of Natureplus. Therefore, all certified products must guarantee low emissions of VOC, odors, radioactivity, dust and fibers to the indoor environment (natureplus, 2011). Specific limit values are defined for each product in a product specific award guideline (natureplus, s.a.).

2.3.3.4 European Ecolabel

The European Ecolabel, also called European Flower, is the ecolabel of the European Community and recognized throughout Europe. Its aim is to guarantee high quality, healthy and environmentally friendly products and services (RAL-gGmbH, s.a.-b). The EU ecolabel defines labeling criteria for different product groups for indoor use, like building and furnishing products, household and consumer products or electric devices (RAL-gGmbH, 2015). Requirements for content and emissions of VOC and formaldehyde exist for textile and wooden floorings, mattresses and paints and varnishes (eurofins, 2015c).

2.4 Standardized VOC test methods in Europe

2.4.1 ISO 16000 series

In 1994 standardization organizations all over the world established the technical committee ISO/TC 146/SC 6 at the International Standard Organization to face the increasing need for internationally accepted testing procedures for indoor air quality. Since then about 40 international standards have been published which include also the ISO 16000 series (Nehr, 2015). A selection of standards from this series dealing with VOC testing and sampling in indoor air and laboratory test facilities and the relevant procedures for VOC analysis are presented in this chapter.

The ISO 16000 series covers not only testing of VOC emissions from construction products, but also indoor air measurements. Therefore, general standards are necessary to describe the basic requirements for planning measurements of air pollutants and to define the framework for a good sampling strategy in the indoor environment. A general overview over indoor air measurements is given in ISO 16000-1, which deals with the aspects which have to be considered when planning evaluations of indoor air quality (DIN, 2006b). More detailed descriptions concerning VOC are given ISO 16000-5. This standard is the link between the very general descriptions from ISO 16000-1 and the detailed analytical techniques from ISO 16000-6. It gives an overview over sources and origin of VOC and the typical emission characteristics of different products. Furthermore, various reasons for indoor VOC measurements are described together with their relevant boundary conditions. Additionally, possible sampling strategies are suggested (DIN, 2007). The analytical procedure for determining VOC in the air of real rooms and test chambers is given in ISO 16000-6. It defines both the sampling and analysis method. Air samples are taken on sorption tubes filled with Tenax®TA. For this purpose a clearly defined air volume is drawn over the tubes. Additionally, to the loaded sampling tubes, some tubes have to be used as field blanks. These sorption tubes are treated in the same way as the sampling tubes but are not loaded with air. The sorption tubes are subsequently analyzed using a thermodesorption unit combined with a GC system with either an FID or MS detector. It is not determined which kind of capillary column has to be used.

However, the use of 100 % dimethyl polysiloxane columns is recommended. ISO 16000-6 furthermore contains the correct handling and preparation of sorption tubes before and after sampling and describes the identification and quantification of VOC as well as the calculation of TVOC, which is based on the TVOC concept as described in ECA Report No 19 (ECA, 1997b) (see Chapter 2.1.1). For the correct determination of the TVOC the whole area of the chromatogram between n-hexane and n-hexadecane is quantified in toluene equivalents and the obtained concentrations are summed. For tests in emission test chambers the background value from the chamber and for field measurements the results of the field blank have to be subtracted (DIN, 2004).

Sampling, sample storage and preparation of test specimens from building and furnishing products for VOC emission testing in laboratory test devices is described in ISO 16000-11. This standard defines three product categories, solid products, liquid products and combined products and describes correct sampling methods for different products from each group (DIN, 2006a). An emission test chamber as a universally applicable laboratory testing method for determining VOC emissions from building and furnishing products is presented in ISO 16000-9. Test chambers made from stainless steel or glass are operated under constant climate conditions. Test chamber volume is not defined. The chamber has to be airtight and supplied with purified air. Good mixture of chamber air must be guaranteed. Duplicate air samples are taken after 72 ± 2 hours and 28 ± 2 days. Climate parameters are set to 23 ± 2°C and 50 ± 5 % relative humidity. Air velocity above the sample surface must be in the range from 0.1 to 0.3 m s^{-1}. A constant air exchange is mandatory and has to be reported; however, no exact value is defined for the air exchange rate. Annex B of ISO 16000-9 gives information on typical loading factors of different products depending on their use indoors (e.g. as flooring or wall paneling). The loading factor [m^2 m^{-3}] is the ratio of the surface area of a product to the volume of the room where it is installed. The loading factor is used to calculate the area specific air exchange rate q [m^3 $m^{-2}h^{-1}$] which is done by dividing the air exchange through the loading factor. Annex B suggests an air exchange of 0.5 h^{-1} (DIN, 2008). Similar conditions apply to another testing procedure: the emission test cell. Descriptions of emission test cells usually refer to the Field and Laboratory

Emission Test Cell (FLEC) as described by Wolkoff et al. (1993). A cylindrical, mobile test cell is placed on the plain surface of a sample material. The volume above the surface is 35 cm³. The relevant standard for emission test cells is ISO 16000-10 which defines the testing procedure, materials and climate conditions. According to this standard an emission test cell has to be made from stainless steel or glass. The edge of the emission test cell is covered with a sealant to guarantee airtightness. Only sealants with low emission and adsorption potential may be used. Other requirements are comparable to those for emission test chambers as defined in ISO 16000-6 (DIN, 2006c). Generally, the results from both emission test chamber and emission test cell are given as chamber air concentrations [µg m^{-3}]. The results can also be expressed as area specific emission rates (SER_A), which depend on time and on the surface area of the test specimen. The SER_A is calculated as follows:

$$SER_A = \frac{C \times n}{L} = C \times q \qquad (2)$$

SER_A = area specific emission rate [µg $m^{-2}h^{-1}$]

C = air concentration [µg m^{-3}]

n = air exchange [h^{-1}]

L = loading factor [m^2 m^{-3}]

q = area specific air exchange rate [m^3 $m^{-2}h^{-1}$]

2.4.2 prEN 16516

In 2011 the European Parliament and the Council of the European Union published the Construction Products Regulation (CPR, Regulation (EU) No 305/2011). This regulation replaced the Construction Products Directive (89/206/EWG) from the year 1989 (EU, 2011). Main aim of the CPR is the reduction of trade obstructions by harmonizing the requirements for construction products in the European Union (Umweltbundesamt, 2015b). The regulation defines seven basic requirements for construction works (EU, 2011). Technical details shall be defined in harmonized European standards and technical specifications. Hereby, it shall be guaranteed that the CE sign which allows for unrestrained trade throughout Europe is only awarded to products which fulfill the requirements of the CPR (Umweltbundesamt, 2015b).

The third basic requirement of the CPR deals with hygiene, health and environment. This requirement prohibits the emission of VOC or dangerous particles into indoor air (EU, 2011). The Technical Committee CEN/TC 351 of the European Committee for Standardization is requested to provide suitable testing procedures for all building products until 2015. The first testing method published in 2013 was CEN/TS 16516 “Construction Products - Assessment of release of dangerous substances - Determination of emissions into indoor air” (Umweltbundesamt, 2015b, CEN, 2013). This Technical Specification was published as a draft standard in July 2015 (CEN, 2015) and is in the following described in more detail.

The newly published prEN 16516 specifies a horizontal test method for the determination of VOC emissions from construction products to indoor air. It includes specifications and requirements of the test method, sampling and sample preparation, analytical methods and calculation of VOC concentrations, TVOC and SER.

The relevant test method defined in prEN 16516 is the emission test chamber. The test set-up is oriented at ISO 16000-9 (DIN, 2008), but further specifications are made. Generally, ventilated test chambers with a minimum volume of 20 L are requested. No upper limit is set for the test chamber volume. The volume of the chamber must be suitable to accommodate a test specimen big enough to be representative for the relevant product. It is suggested rather not to use small chambers for inhomogeneous products as small test specimens may not be representative. The test chamber must be supplied with clean air at a ventilation rate of 0.25 h^{-1} to 2.0 h^{-1}. Within this range SER are assumed to remain constant. The climate conditions within the test chamber are set to 23 ± 1°C and 50 ± 5 % relative humidity. The air velocity above the surface of the test specimen has to be between 0.1 and 0.3 m s^{-1}. Within this standard a European reference room is described. This reference room defines a universally applicable emission scenario. The dimensions of the reference room are 3 x 4 x 2.5 m^3. Door and window cover areas of 1.6 m^2 and 2 m^2, respectively. For products used only in very small areas like sealants a surface area of 0.2 m^2 is assumed. Emission test chambers are generally smaller and therefore treated as scaled-down models of the reference room. Loading factors of products in the test chamber are calculated by using the dimensions of the product in the reference room according to its relevant location within the room. However, deviations of the loading factor of a product from the reference room are possible. These deviations may not fall below 50 % or exceed 200 % of the specified loading factor. Within this range constant SER are assumed. Additionally, product standards may define new loading factors if none of the given factors are suitable. Temperature and relative humidity are set to 23°C and 50 %, respectively. The climate conditions within the test chamber are oriented at the settings for the model room. Only the air exchange rate in the model room differs from the test chamber, as it is fixed to 0.5 air exchanges per hour. Therefore, all results of VOC measurements reported as air concentrations [µg m^{-3}] have to be recalculated to an air exchange of 0.5 h^{-1}. Sampling and handling of product samples is done according to ISO 16000-11 (DIN, 2006a), but further requirements are added mainly to avoid sample contamination during transport and storage. Additionally, a sampling strategy shall be defined for each product group within the relevant product standards. The requested objective of sampling is to obtain a sample representative for the whole construction product, which meets the testing requirements. VOC are sampled and analyzed according to ISO 16000-6 (DIN, 2004). A defined volume of chamber air is collected at a controlled air flow rate on sampling tubes filled with Tenax®TA. The tubes are analyzed using a thermodesorption unit connected to a GC-MS system equipped with a 5 % phenyl

/ 95 % methyl polysiloxane capillary column. SER_A and TVOC are calculated according to ISO 16000-6 (DIN, 2004) and ISO 16000-9 (DIN, 2008) as described in Chapters 2.4.1 and 2.1.1. Sampling and analysis of carbonyl compounds is done according to ISO 16000-3 (DIN, 2010a) on DNPH cartridges which are analyzed by an HPLC system equipped with UV detector or diode array detector (CEN, 2015).

Additionally to the standardized method of measuring VOC emissions in an emission test chamber, prEN 16516 also specifies a selection of so-called indirect methods. These methods are intended to be used for comparative measurements of known products and comprise emissions cells like the FLEC, microchambers, thermal extraction, headspace analysis and VOC content determination (CEN, 2015).

3 Objectives

The overall aim of this thesis was to design, construct and operate a new test set-up tailored for investigations on VOC emissions in a common and realistic indoor environment but to the exclusion of any unwanted environmental influences. Hereby, new insights into the different indoor processes influencing VOC and, thus, indoor air quality should be gained. The success of this research work is documented in a series of peer-reviewed scientific papers that are combined in this thesis and can basically be divided into four relevant sections.

The first section comprises the design and construction of the model rooms. Two model rooms were constructed exclusively of the wood-based construction products X-lam and OSB, respectively. An external air conditioning system provided both rooms with purified and conditioned air at a fixed air exchange rate. The rooms were subsequently evaluated regarding their suitability for the further research tasks and were as far as technically possible optimized. The major focus here was to develop a reliable large-scale test set-up for simulating indoor environments and activities (see Publication I).

In the second section the long-term development of the VOC emissions in the model rooms was investigated. VOC were measured regularly over a period of six months in the empty, unfurnished model rooms to gain further knowledge about the emission behavior of the applied wood-based construction materials (see Publications I and II).

In the third section the model rooms were successively equipped with interior fittings. VOC sampling occurred on day 7 and 14 after each completed interior construction step. In parallel, the materials' specific emissions were determined in microchambers. In a next step, the model rooms were equipped with furniture and textiles, followed by another measurement period of two weeks. The aim was to determine the quantitative and qualitative changes of the VOC content in a room caused by gradual interior constructions. Changes of total VOC concentration, changes of individual substances and the appearance of new substances in the course of common building steps were investigated (see Publication III).

The fourth section comprises the simulation of different user scenarios in the model rooms, which are assumed to influence indoor air quality. Besides VOC, also CO_2, particulate matter, and carbonyl compounds were investigated. Blank samples were taken before each user scenario to determine the VOC concentration prior to the individual procedure. First sampling was performed during or directly after the scenario itself. Further samples were taken for the determination of the decline pattern of the individual indoor pollutants. The aim was to determine the influence of these selected human activities on different parameters of air quality in an idealized indoor environment (see Publication IV).

4 Materials and methods

This chapter provides a short overview of all methods used to obtain the results discussed in this scientific work. Detailed descriptions of the relevant materials and analytical methods can be found in the scientific publications cited in this chapter and appended to the present thesis.

4.1 Model rooms

The scientific publications included in this thesis focus on the model rooms as newly designed and operated test set-ups simulating real rooms in terms of size and configuration but under controllable conditions close to those in a laboratory. The model rooms were adapted and operated for answering various research questions. Different indicators of indoor air quality were measured in the course of different research tasks. All analyses carried out in the model rooms are briefly described below. Their relevance for the different research tasks can be seen from the enclosed scientific publications (see Publications I, II, III, IV).

4.1.1 Construction

Both model rooms were completely constructed from wood-based building products. One model room was built as a light-frame construction covered with Oriented Strand Board (OSB). For the other model room prefabricated elements of Cross Laminated Timber (X-lam) were used. The model rooms were thermally insulated with 10 cm of foamed polystyrene (XPS). Each room was equipped with three sampling points at which air samples were drawn. An additional sampling point was located in the exhaust air duct. An external air conditioning system supplied both model rooms with purified and conditioned air. The rooms met the requirements of the European reference room as described in prEN 16516 (CEN, 2015). Diameters of and test conditions in the model room are shown in Table 7. Publication I gives further details concerning the basic construction and functioning of the model rooms and the air conditioning system. During further investigations the rooms were equipped with interior fittings and furnishings. Details concerning the interior constructions can be found in Publication III.

Table 7: Test conditions in the model room

Parameter	Value
Temperature [°C]	23 ± 2
Relative humidity [%]	50 ± 5
Length [m]	4
Width [m]	3
Height [m]	2.5
Room volume [m³]	30
Door [m²]	1.6
Window [m²]	2
Air inlets/outlets [m²]	0.13
Floor area [m²]	12
Ceiling area [m²]	12
Wall area [m²]	31.3
Total surface area [m²]	55.3
Loading factor [m²/m³]	1.8
Air exchange rate [h^{-1}]	0.5 ± 0.005

4.1.2 VOC sampling

Room air was sampled on desorption tubes filled with Tenax®TA, using air sampling pumps (Chematech, Markes). 1 µL of the internal standard consisting of toluene-d8 and cyclodecane was injected into desorption tubes prior to sampling. Additionally, one desorption tube was used as field blank. This tube was treated the same way as all others but not loaded with room air.

4.1.3 Aldehyde sampling

Aldehydes were sampled on DNPH cartridges. A defined air volume was pumped over cartridges filled with high-purity silica-gel coated with 2,4-dinitrophenylhydrazine (DNPH).

4.1.4 Particulate matter sampling

For determining the particulate matter concentration, room air was taken in by a circular suction head and analyzed by an aerosol spectrometer. The results for the classes PM10, PM2.5 and PM1.0 in µg m^{-3} were automatically calculated from the number of particles and the process-specific C-coefficient. The default value of the C-coefficient was 1.

4.1.5 CO_2 sampling

CO_2 concentration was determined continuously using a multifunction analyzer (data logger for CO_2, temperature and relative humidity). The applied measurement procedure was the non-dispersive infrared spectroscopy (NDIR).

4.2 Microchamber

4.2.1 Set-up and test conditions

The microchamber (µ-CTE, Markes International, UK) consisted of six individual cells made of stainless steel and arranged in a heating block. To ensure constant temperature conditions, the microchambers were placed in a conditioning cabinet. The heating block of the microchamber was not used. Each cell was equipped with a sensor to measure temperature and relative humidity. Inlet air was controlled by a flow meter outside of the microchamber. The construction of the microchamber ensured the supply of all cells with the same amount of inlet air. Inserted samples were cut to a diameter equal to the cell diameter. The insertion of samples into the cells resulted in a significant reduction of the free volume due to the small cell volume. Therefore, the loading factor within the cell largely depended on the sample thickness and could not be influenced by the operator. Only temperature and relative humidity were identical for all samples. Table 8 shows the overall test conditions in the microchamber (see Publication III).

Table 8: Test conditions in the microchamber

Parameter	Value
Temperature [°C]	23 ± 1
Relative humidity [%]	50 ± 5
Chamber diameter [m]	0.045
Chamber volume [m³]	0.000048

4.2.2 VOC sampling

Samples were drawn at the air discharge openings of the cells. Desorption tubes filled with Tenax®TA were fixed at the openings and outgoing air passed through the tubes. The sample volume was calculated from the air flow rate passing through the cell and the sampling duration. Each tube was spiked with 1 µL of the internal standard consisting of toluene-d8 and cyclodecane. One tube was placed over an empty cell and used as blank (see Publication III).

4.3 Gas chromatographic analysis – VOC

Tenax$^{®}$TA desorption tubes from model room (see Chapter 4.1) and microchamber (see Chapter 4.2) sampling were analyzed using a thermodesorption unit linked to a GC-MS system (see Publications I, II, III, IV). The detected substances were identified by comparison of the mass spectra to spectra libraries. Peak areas were used for quantification. Toluene-d8 and cyclodecane were used as internal standards. Substance specific quantification and quantifications as toluene equivalents (TE) were performed.

4.4 HPLC analysis – Aldehydes

DNPH cartridges were eluted against the loading direction using 5 mL acetonitrile. The aldehyde hydrazones in the eluate were subsequently identified using HPLC-DAD. Aldehydes were quantified using an external standard (see Publication IV).

5 Summary of scientific publications

The first publication entitled “A large-scale test set-up for measuring VOC emissions from wood products under laboratory conditions in simulated real rooms” comprises the design and construction of the model rooms as well as the development of the long-term emissions of the basic construction material. It was the aim of this publication to determine the suitability of the model room as a test set-up for long-term VOC measurements on a realistic scale. Two model rooms were completely constructed from OSB and X-lam, respectively. Both rooms had dimensions of 3x4x2.5 m^3. The rooms were supplied with purified and conditioned air at an air exchange rate of 0.5 h^{-1} through an external air conditioning system. Temperature and relative humidity were set to 23±2°C and 50±5%rH, respectively. 13 VOC measurements were performed during a test duration of 23 weeks. TVOC as well as the sum of terpenes and aldehydes were calculated in toluene equivalents. Temperature and relative humidity were supervised during the whole test. Mean values of 22°C and 50.8%rH were measured in the OSB model room and 22.5°C and 49.7%rH in the X-lam model room. The deviations from the target values were larger in the OSB model room due to a lower buffering capacity compared to the X-lam model room. TVOC clearly declined over the whole test duration in both model rooms. In the OSB model room TVOC decreased by 72% from 442 µg m^{-3} TE to 124 µg m^{-3} TE. The most abundant substance group were aldehydes, accounting, on average, for 52% of the TVOC. In the X-lam model room TVOC decreased by 64% from 115 µg m^{-3} TE to 41 µg m^{-3} TE. Terpenes were the most abundant substance group reaching, on average, 80% of the TVOC. In both rooms a steep decline of the TVOC was measured during the first three weeks of testing. Comparison to literature results showed that a faster decline in the TVOC concentration was measured in emission test chambers compared to the model room. Yet, the emission profile was comparable to other studies. The study demonstrates the suitability of the model rooms for VOC determination under conditions comparable to those in real buildings.

The second publication “Measuring VOC emissions from wood-based building products under real room conditions in idealized model rooms” goes into further detail concerning the long-term emissions from the construction materials of the model rooms. A set of six aldehydes and four terpenes, commonly identified in VOC emissions from wood products, was chosen and analyzed for both model rooms. This analysis comprised a substance specific quantification of each selected substance and the monitoring of their development over the whole test period. As already determined in the first publication, the most abundant substances in the X-lam model room belong to the group of terpenes. α-Pinene shows the highest concentration followed by β-pinene. All four selected terpenes could be identified in the X-lam model room and showed a clear decrease over time. For the sum of all identified terpenes a decline rate of 83% could be determined. Only two aldehydes, pentanal and hexanal, were identified at a rather low concentration. However, these substances still declined by 82% over the whole testing period. In the OSB model room aldehydes were higher concentrated than terpenes. Five aldehydes were identified in the OSB model room with hexanal being the most abundant substance. From the selected terpenes α-pinene and Δ-3-carene were detected within the OSB model room. The sum of all identified aldehydes declined by 79% over a period of 23 weeks, whereas, terpenes declined by 61%. Their concentration was clearly lower than that of the measured aldehydes. All substances showed a clear decline over time. The findings of this part of the study are similar to those of other authors and once more prove the suitability of the model room as a test set-up and the comparability of the results to former research work.

In the third publication entitled “Influences of interior fitting processes on the indoor air quality of two model rooms” the rooms described in the first publication were used to simulate an interior fitting process and to examine the development of the VOC emissions to the exclusion of unwanted environmental effects. The installation of the selected materials was performed stepwise. VOC samples were drawn at three sampling points seven and 14 days after each installation step. The OSB model room was equipped with a dry lining comprising vapor barrier sheet, gypsum plasterboard, and limewash. In the next step a solid parquet made of larch wood was laid. The room was subsequently furnished with office furniture and in the last step equipped with textiles. In the X-lam model room an acoustical ceiling made of larch wood was installed, followed by a solid parquet made of oak wood. The walls were subsequently painted with a water-based wood stain for indoor use. In the next step the room was furnished with bedroom furniture made of solid pinewood and finally equipped with textiles. All used materials were additionally qualitatively analyzed in a microchamber (µ-CTE™). In the OSB model room the TVOC decreased during the whole interior fitting processes from 119 µg m^{-3} to 26 µg m^{-3}. No distinct changes in the VOC profile could be observed after any fitting step. In the X-lam model room, a change of the VOC concentration and/or the VOC profile could be observed after each fitting step. The highest concentration of 225 µg m^{-3} was reached after the installation of the acoustical ceiling. In both rooms, barrier and sink effects could be observed for various materials. The installation of textiles for instance led to distinct reduction of VOC in both model rooms. In the OSB model room the concentration of all identified VOC dropped below the quantification limits and a strong decrease of the concentration of all identified substances were measured in the X-lam model room. Additionally, in the OSB model room a clear reduction of the VOC concentration was determined after the installation of the dry lining, most likely due to the barrier effect of the used materials. The results of the study show that indoor air quality in real buildings is strongly affected by interactions of different materials.

The fourth publication “Emission of indoor pollutants from six user scenarios in a model room” focusses on the influence of user activities on indoor air quality. Six precisely defined user scenarios were performed in the model room. This user activities comprise the use of cleaning agent, an electric air freshener, cosmetics, an ethanol fireplace, smoking cigarettes, and peeling oranges. The common indoor air pollutants VOC, PM, carbonyl compounds and CO_2 were investigated during this user scenarios. The highest VOC concentration of 3547 µg m^{-3} was measured during the peeling of oranges. Cigarette smoking led to a strong increase of carbonyl compounds, especially formaldehyde which reached a concentration of 76 µg m^{-3} and PM, with a maximum concentration of 378 µg m^{-3}. Most user scenarios only slightly affected CO_2 except the use of the ethanol fireplace which resulted in strongly elevated CO_2 concentrations reaching maximum values of 1612 ppm. Every user scenario led to an increase of at least one of the investigated parameters. Generally, changes in the concentration of indoor pollutants after a user scenario were distinct but only of short duration as emissions decreased rapidly after the end of the scenario. The study clearly shows the strong influence of user activities on indoor air quality, although, interactions between different parallel user activities were not investigated.

6 Conclusions

It was the main aim of this thesis to gain further knowledge about air quality in real indoor environments and the influence of different possible sources of VOC. To reach this aim the model rooms were designed and constructed as large-scale test set-ups, resembling as far as possible a realistic room but to the exclusion of unwanted environmental effects. This test set-up is preferable to a man-size emission test chamber made from stainless steel, as it resembles a realistic building not only in size but also in construction and configuration. Thus, the model rooms represent a new test method, which combines the advantages of laboratory testing with the indoor processes of a real building and, therefore, enables investigations close to reality but unaffected by outward influences (see Publications I and II). It could be demonstrated that the development of VOC emissions in the model room differs from that in common emission test chambers. Generally, the initial VOC concentration from the applied construction materials was lower and the reduction of VOC slower and less distinct than in the emission test chamber as described in Publication I. This behavior is attributed to the realistic construction and configuration of the model room. It is assumed that the results obtained in the model room have a higher similarity to those in a real building than results from an emission test chamber. The investigations performed in the model rooms, therefore, offer more reliable information on the influence of indoor environment and activities on uncontaminated air in a conventional wood construction.

To guarantee reliable results, the model rooms were investigated for their suitability to measure and determine VOC in the indoor environment. Additionally, their original construction was used to observe long-term development of VOC emissions within buildings constructed exclusively from wood-bases products. The results which can be seen from Publication I clearly show that the performance of the model rooms was suitable for the further research tasks because fluctuations of climate parameters could be kept in narrow ranges and VOC measurements experienced no obstructions. The VOC concentration within the empty model rooms showed a steady and distinct decline which lasted for the whole measurement period and therefore distinctly exceeded the standardized testing time of 28 days.

During interior construction processes further influence factors arise within a building (see Publication III). Each construction step performed in the model rooms provided new alterations in both VOC concentration and profile. These alterations were manifold and may be assigned to source, sink and barrier effects of different materials. Changing the interior fittings within a room, therefore, means to impair indoor air quality. However, the consequences of these interventions cannot be completely foreseen. Although, testing of the applied materials in advance to their installation provides some information about their potential as a VOC emitter and makes it easier to retrace VOC sources, no knowledge about their interactions with other materials may be gained by this measure. The same is true for human activities as sources of indoor pollutants. Within this research work only exemplary experiments were performed under idealized conditions as can be seen from Publication IV. In this case the selected user scenarios led to high peak values of different indoor pollutants, but these pollutants diminished quickly after ending a certain activity. As all tests were performed separately, no conclusions about possible interactions between pollutants from different anthropogenic sources of VOC may be drawn. Still, there is strong evidence that the effects of human activities on indoor air quality may rather be accumulative than not.

Indoor air quality is influenced by numerous factors, some of which have been investigated extensively throughout this research work. The focus was mainly on VOC, which emit from various sources. The findings show that construction products as well as interior fittings and furnishings contribute to indoor VOC emissions. Furthermore, human activities and thus additionally inserted materials are possible and relevant sources of indoor air contaminants. Previous studies partly rate the influence of people on air quality in the indoor environment higher than that of construction products and this conclusion can also be drawn from the results of this thesis (see Publication IV). Despite this knowledge from previous and actual research, standardized VOC measurements are still performed as single substance tests. The generated data enable a good comparison between different materials. However, a straightforward conclusion on indoor air quality cannot be drawn from such comparisons because emissions out of any product into the room are altered or superimposed by combination with other materials, as was demonstrated here (see Publications III and IV). The results show that the influence factors on indoor air quality are manifold. A clear assignment of a substance detected in indoor air to a defined source is therefore difficult or almost impossible. User activities for instance emit high amounts of substances, which can also be defined as wood specific VOC (e.g. terpenes). This relationship is also true for other materials. It can therefore be assumed that, when evaluating indoor air quality, both the plurality of possible emission sources as well as the actual influence of an individual product on the indoor air quality have to be considered.

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8 Annex: peer reviewed scientific publications

Publication I

Höllbacher, E., Rieder-Gradinger, C., Stratev, D. and Srebotnik, E. (2015) A large-scale test set-up for measuring VOC emissions from wood products under laboratory conditions in simulated real rooms. Holzforschung 69 (4), pp. 457-462

Contribution of Eva Höllbacher:

Conception of the Study, Selection of Test Methods, Sampling and Analysis, Data Interpretation, Literature Research, Writing and Editing of the Manuscript

Publication II

Höllbacher, E., Rieder-Gradinger, C., Stratev, D. and Srebotnik, E. (2014) Measuring VOC emissions from wood-based building products under real room conditions in idealised model rooms. International Wood Products Journal 5 (4), pp.207-211

Contribution of Eva Höllbacher:

Conception of the Study, Selection of Test Methods, Sampling and Analysis, Data Interpretation, Literature Research, Writing and Editing of the Manuscript

Publication III

Höllbacher, E., Rieder-Gradinger C., Ters, T. and Srebotnik, E. (2016) Influences of interior fitting processes on the indoor air quality of two model rooms. European Journal of Wood and Wood Products, accepted

Contribution of Eva Höllbacher:

Conception of the Study, Selection of Test Methods, Sampling and Analysis, Data Interpretation, Literature Research, Writing and Editing of the Manuscript

Publication IV

Höllbacher, E., Rieder-Gradinger C., Ters, T. and Srebotnik, E. (2016) Emissions of indoor pollutants from six user scenarios in a model rooms. Environment International, submitted

Contribution of Eva Höllbacher:

Conception of the Study, Selection of Test Methods, Sampling and Analysis, Data Interpretation, Literature Research, Writing and Editing of the Manuscript

Further Publications

Höllbacher, E., Rieder-Gradinger, C., Stratev, D., Srebotnik, E. (2014): Measuring VOC emissions from wood-based building products under real room conditions in idealised model rooms. *In: Proceedings of the International Panel Products Symposium 2013 (IPPS 2013)*, Llandudno, Wales, UK, 9-10 October 2013

Höllbacher, E., Rieder-Gradinger, C., Stratev, D., Srebotnik, E. (2014): Influence of VOC emissions from wood and wood-based materials on indoor air quality. *In: Proceedings of the 13th International Conference on Indoor Air Quality and Climate (Indoor Air 2014),* Hong Kong, China, 7-12 July 2014

Höllbacher, E., Rieder-Gradinger, C., Ters, T., Srebotnik, E. (2014): Using a small-scale testing tool (µCTE) for rapid prediction of VOC from building products. *In: Proceedings of the 3rd International Conference on Processing Technologies for the Forest and Bio-based Products Industries (PTF BPI 2014),* Kuchl, Salzburg, Austria, 24-26 September 2014

Sinic, J., Ters, T., Jocham, C., Jury, S., Müller, U., Kandelbauer, A., Höllbacher, E. (2016): VOC-Emissionen und Oberflächeneigenschaften von lackierten MDF für Küchenmöbel. *Holztechnologie*

A large-scale test set-up for measuring VOC emissions from wood products under laboratory conditions in simulated real rooms

Eva Höllbacher, Cornelia Rieder-Gradinger, Daniel Stratev, Ewald Srebotnik

Abstract

A large-scale test set-up was designed to evaluate the VOC emissions of building materials in a real room situation but under laboratory conditions. Two model rooms (ModR) with a volume of 30 m^3 each were constructed of the wood-based building materials X-lam and OSB, respectively. Temperature and relative humidity (RH) inside the ModR were kept in a range of 21 to 25°C and 45 to 55% RH. VOCs were collected at 13 different times over a period of 23 weeks, and the total volatile organic compound (TVOC) concentration was calculated from GC/MS data. Results were quantified as toluene equivalents (TE). In the X-lam-ModR, the TVOC concentration decreased by 62% over the whole measurement period from 115 to 43 µg m^{-3}TE. Terpenes were the most abundant substance group and accounted in average for 80% of the TVOC concentration. In the OSB-ModR, the TVOC concentration decreased by 72% from 443 to 124 µg m^{-3}TE. Aldehydes showed the highest concentrations accounting in average for 52% of the TVOC, while 38% were terpenes. The results show that this type of tests provides realistic data for the praxis.

Introduction

Building products are indoor sources of volatile organic compounds (VOC) (Wolkoff and Nielsen, 2001, Jones, 1999). Particularly in airtight, energy-efficient buildings with low ventilation rates, a considerable amount of VOCs may accumulate indoors (Missia et al., 2010, Lin-lin et al., 2011). The rising awareness of possible health effects of indoor air quality is the driving force for VOC research.

Wood and wood-based building products with their specific emissions are also sources of long-lasting indoor VOC emissions (Salthammer and Bahadir, 2009, Yu and Kim, 2012). Softwoods emit mainly terpenes and aldehydes (Risholm-Sundman et al., 1998), but other wood species may also set free other substances (Jensen et al., 2001). Nevertheless, terpenes and aldehydes belong to the most relevant indoor VOCs (Hodgson et al., 2000, Kostiainen, 1995).

VOCs from building products are generally measured by means of inert test chambers with a volume of 1 m^3 or less as defined in ISO 16000-9 (DIN, 2008) or similar test set-ups. Though these methods are well defined and the results are well comparable, there is continuing debate on whether or not such test set-ups are representative for real room situations. On the other hand, measuring VOCs in real buildings is complicated by the fact that the control of climate conditions in such a setting is usually limited or even impossible. Moreover, environmental effects as well as surrounding materials have a strong influence on the measured VOC concentration (Järnström et al., 2007). It would therefore be desirable to test the VOC emission of building products in large-scale but under laboratory conditions.

In the present work, model rooms (ModR) were constructed and tested as large-scale test-ups for determining VOC concentration. The ModR resembled real rooms in terms of size and configuration and were supplied with purified and humidified air by an external air conditioning system. Thus, the VOCs can be measured under controllable climate conditions, similar to those in a laboratory. The aim of the study was to determine whether a continuous operation of these ModR is possible under well-defined, constant climate conditions. The results of the measurements should be evaluated for their plausibility compared to realistic

VOC emissions in everyday life, and the expectation was that this type of VOC tests are more reliable than the hitherto applied laboratory tests.

Materials and methods

Two model rooms (ModR) were constructed and installed inside a technical hall (Figure 1). The dimensions of the model room and the set target values for temperature (T), relative humditiy (RH) and air exchange rate meet the requirements of the European reference room as defined in CEN TS 16516 (CEN, 2013). Each ModR had dimensions of 4 m in length, 3 m in width and 2.5 m in height, resulting in a room volume of 30 m³. Additionally, two inert areas resembling a window and a door were installed. The window was made of glass and covered an area of 2 x 1 m². The door was made from stainless steel with dimensions of 2 x 0.8 m². For connecting the ModR to the ventilation system, two air inlets and two air outlets with a diameter of 0.2 m were installed. The dimensions specified above resulted in a total indoor surface area of 55.3 m². Three VOC measuring sites were installed. An additional measuring site was located in the exhaust air duct.

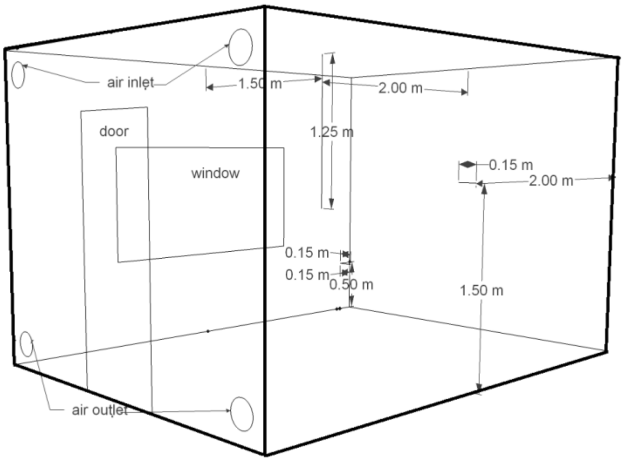

Figure 1: 3-D drawing of the model room, including inert areas, air inlets and outlets and the exact positions of the measuring sites located 1) in the center of the room, 2) in a corner in a height of 0.5 m and a distance of 0.15 m to the adjacent walls, and 3) at a wall in a height of 1.5 m and a distance of 0.15 m to the wall. Air inlets are installed at the upper part, air outlets at the lower part of the front wall.

The X-lam-ModR and OSB-ModR were completely constructed from the materials indicated. OSB with a thickness of 22 mm was manufactured of Scots pine (*Pinus sylvestris* L.), bonded with polymeric methylene diphenylene diisocyanate (pMDI). X-lam with a thickness of 95 mm was made of Norway spruce (*Picea abies*), bonded with polyurethane and melamine urea formaldehyde (MUF) resin. Both materials were delivered directly by the manufacturer. The OSB panels were manufactured 10 weeks before delivery, whereas the X-lam elements were freshly produced in the week prior to the start of the construction work which took another 2 weeks. The ModRs were constructed on-site. Light-frame construction was used to built the OSB-ModR. The panels were connected by tongue and groove joints. Additionally, the joints were sealed with aluminum tape. Prefabricated elements were taken for the X-lam-ModR. Sealing band was inserted in the overlap between the elements. To minimize surrounding temperature influences, the ModRs were insulated with 0.1 m thick panels of foamed polystyrene on the outside.

VOC measurements were performed under standardized climate conditions with consideration of the requirements of ISO 16000-9 and CEN TS 16516 (DIN, 2008, CEN, 2013). Climate conditions of 23 ± 2°C and 50 ± 5% RH were ensured inside the ModR by means of an air conditioning system (ACS) outside the ModR (Figure 2). Outside air was aspirated and led through a pre-cleaner into the ACS. The precleaner consisted of two bag filters for coarse and fine dust, respectively. The air was then led into a mixing chamber, where it was mixed with conditioned air that was recirculated in the ACS. Subsequently, the air was dehumidified in a cooling chamber which was operated with a heat exchanger. Then, the air was heated (electric heating) in a first heating unit, humidified by a vaporizer humidification unit and heated to the target temperature in a second heating unit. Air cooling was done in a vapor-compression refrigeration system. At the end of the air conditioning line, the amount of air which was not needed for the supply of the ModRs was recirculated in the ACS. The necessary airflow of 30 m^3 h^{-1} was purified with a carbon filter and then split to supply each ModR with 15 ±1 m^3 h^{-1} of air. This set-point value was manually controlled by iris diaphragms and monitored by measuring the air velocity in the exhaust air duct. Temperature and RH of the airflow were measured by sensors installed prior to the carbon filter and connected to a control unit that also allowed a manual control of the target values. Each ModR was equipped with a sensor for logging temperature and RH data every 15 min. Average temperature and RH were calculated from the data for each day over the whole measurement period of 23 weeks. The sensor readings also served for fine-tuning of the climate inside the ModR by manually adjusting of the set-points at the control unit.

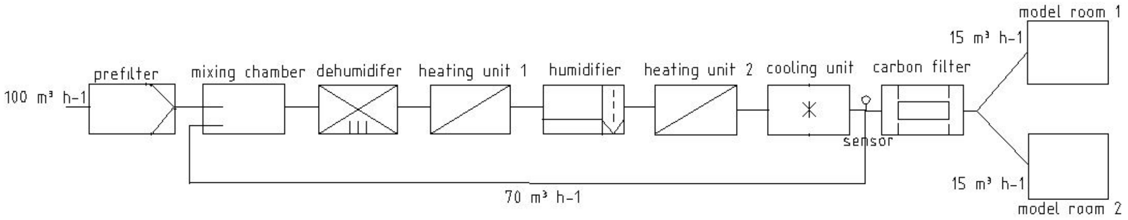

Figure 2: Scheme of the external air conditioning system which was designed for an airflow of 100 m³ h^{-1} to allow for a broad range of air exchange rates and up to six model rooms. For the present study, however, an airflow of only 30 m³ h^{-1} was needed and therefore the remaining 70 m³ h^{-1} were circulated in the air conditioning system.

Indoor air was sampled according to the requirements of ISO 16000-6 (DIN, 2004). Sorption tubes, made of glass and filled with Tenax TA®, were connected to PTFE tubes, which led to the measuring sites through stainless steel pipes. Over a time period of 23 weeks (161 days), VOCs were regularly measured. In total, 13 measurements were performed in both ModRs. Two 6 L samples of indoor air were drawn at each of the 4 measuring sites. Sampling was done with a FLEC® pump (Chematech, Denmark) at an airflow rate of 100 mL min^{-1}. Before sampling, each tube was spiked with 1 µL of internal standard which consisted of 0.37 µg toluene in methanol. Additionally, one sorption tube served as field blank, which was handled in the same way as the other sorption tubes, but was not loaded with indoor air to detect possible contaminations at the measuring sites.

VOCs were analyzed by a thermal desorption unit (Chromtech, Germany, TDAS 2000) coupled with the programmed temperature vaporizing (PTV) injector of a GC/MS system (Agilent Technologies, USA, 7890A/5975C). The analytes were thermally desorbed from the sorbent tube at 250°C for 5 min. The desorbed substances were then cryo-focused in the PTV injector at -15°C and abruptly heated to 280°C at a heating-up rate of 720°C min^{-1}. The PTV injector was operated in split mode with a split of 30:1. The analytes were separated on a methyl siloxane column (Agilent Technologies, USA, HP-PONA, 50 m x 0.2 mm x 0.5 mm) with He as carrier gas (12.85 cm s^{-1}). Temperature program: 3 min at 35°C, → 160°C (10°C min^{-1}), → 310°C (20°C min^{-1}). The MS data were scanned over a m/z range of 12–400. Detected compounds were identified by comparison with data of mass spectral libraries and quantified as toluene equivalents (TE).

The average toluene response for aldehydes and terpenes was 0.6 and 1.3, respectively. The target substance hexanal showed a toluene response of 0.7, while the response of α-pinene was 1.3. It may therefore be assumed that – compared to substance specific quantification – the aldehyde concentration was slightly underrated and the terpene concentration was slightly overrated. The total VOC (TVOC) concentration was calculated for each sample as the sum of all substances with a concentration ≥ 1 µg m^{-3}TE. The minimum concentration of 1 µg m^{-3}TE represents the detection limit of the GC/MS system. Additionally, the sum of terpenes and aldehydes was calculated. No significant difference was observed between the results of the individual measuring sites as determined by the Kruskal-Wallis test. Therefore, the arithmetic mean of all drawn samples (n = 8) was taken for further analysis. TVOC as well as the sum of terpenes and aldehydes included all substances in the retention area between C6 (hexane) and C16 (hexadecane), with the exception of acetic acid, which was included in the TVOC value regardless of its earlier retention time.

Results and discussion

Temperature and relative humidity

The temperature and relative humidity (RH) data for the X-lam- and OSB-ModR are shown in Figure 3. Average temperature in the X-lam-ModR over the whole testing period from January to June (23 weeks) was 22.5°C. TheT_{min} and T_{max} data were 20.2°C and 25.6°C, respectively. The average RH was 49.7%, while the RH_{min} and RH_{max} values were 39.8% and 58.3%, respectively. Temperature and humidity deviated from the allowed range at 14 days (8.7%) and 8 days (5%), respectively.

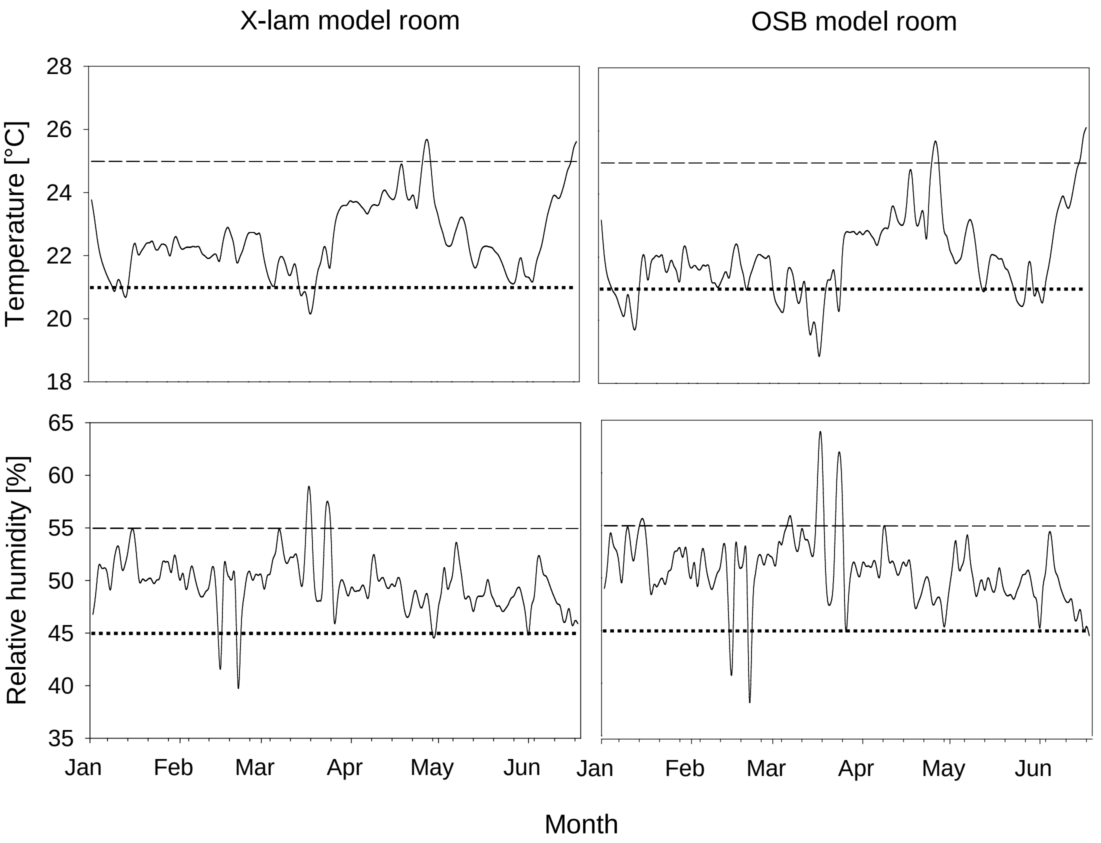

Figure 3: Temperature [°C] and relative humidity [%rH] in the X-lam and OSB model rooms over the whole measurement period from January to June 2013. Dashed and dotted lines indicate upper and lower limits for temperature and relative humidity ranging from 21 °C to 25 °C and 45 %rH to 55 %rH, respectively.

In the OSB- ModR, the average temperature was 22°C, while theT_{min} and T_{max} data were 18.9 and 26.1°C, respectively. Average RH was 50.8%, while RH_{min} and RH_{max} values were 38.2 and 63.2%, respectively. Temperatures and humidity deviated from the allowed ranges at 42 days (26%) and 14 days (8.7%), respectively.

When comparing the graphs, it is obvious that the time courses of indoor temperature and RH were similar in both ModRs. The main reason for the fluctuating temperature was the insufficient insulation, which did not prevent temperature changes from the outside hall. As cold air can take up less humidity, RH concomitantly rose with decreasing temperature and vice versa. This can be seen at two dates in March. A decline of the room temperature occurred due to decreasing outside temperature after a break-down of the heating system of the technical hall. In parallel with the temperature decline, RH increased. The fast decline of RH, which occurred on two days in February was caused by a breakdown of the humidification unit due to limescale. The RH measured on these dates were 41.4% and 38.2% for the OSB-ModR and 41.8 and 39.8% for the X-lam-ModR. This technical problem was solved by the installation of a new deionized water supply. The deviations from the limit values were more distinct in the OSB-ModR than in the X-lam-ModR, most likely caused by the higher buffering capacity of the X-lam construction material due to its greater thickness resulting in a balancing effect on indoor temperature and RH. Summing up, it is obvious that the air conditioning system (ACS) worked reasonably well and fulfilled the requirements for both parameters during 74% and 90% of the whole measurement period in the OSB- and in the X-lam-ModR, respectively.

Total amount of VOC (TOVC)

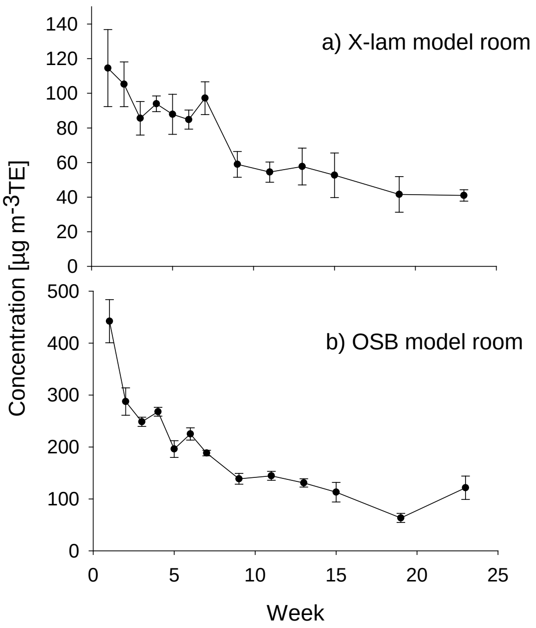

Figure 4: Time course of TVOC-concentration [µg m-3] in the a) X-lam and b) OSB model room. Data points represent the arithmetic mean and standard deviations of all 8 air samples drawn from the model room at weeks 1, 2, 3, 4, 5, 6, 7, 9, 11, 13, 15, 19, and 23.

VOCs were measured 13 times in both ModRs over a period of 23 weeks (Figure 4). Starting from an average value of 115 µg m^{-3}TE (100%), the concentration in the X-lam-ModR decreased by 25% during the first 3 weeks. Subsequently, fluctuations could be observed resulting in an increase to a maximum of 97 µg m^{-3}TE (85%) in week 7. However, based on the StdD, none of these fluctuations are statistically significant. Between week 7 and week 9, the TVOC concentration declined to 53%. During the following 5 measurements, the TVOC concentration further decreased to 41 µg m^{-3}TE (36%) at week 23. Overall, a distinct decrease could be observed for the TVOC concentration in the X-lam-ModR. The starting value is comparable to the findings of Steckel et al. (2010), who determined TVOC concentrations from Norway spruce being between 103 and 265 µg m^{-3} in a 23 L emission test chamber. However, in the test chamber the TVOC concentration declines more rapidly than in the ModR investigated here. Steckel et al. (2010) determined a reduction of about 90% after 28 days of emission testing. A comparable reduction of about 85% within 28 days was determined by Larsen et al. (2000). However, in the ModR the TVOC concentration is only reduced by 18% within the first 4 weeks. A reduction of 90% is reached after 13 weeks.

Similarly to the X-lam-ModR, a steep decline during the first weeks occurred in the OSB-ModR. The starting value amounted to 442 µg m^{-3}TE (100%) and was reduced to 56% within the first 3 weeks. Fluctuations occurred that are statistically insignificant and less pronounced than those observed in the X-lam-ModR. From week 7 to 19 the TVOC concentration decreased to its lowest value of 70 µg m^{-3}TE (16%). Overall, the TVOC concentration was reduced by 72% over the whole measurement period. Literature data show a very broad range of TVOC concentrations measured from OSB in emission test chambers. For example, Wilke et al. (2013) determined TVOC concentrations from 0.16 to 3.03 mg m^{-3} from 6 different OSB samples after 3 days in a 24 L test chamber. After 28 days, the values ranged from 0.14 to 0.99 mg m^{-3}. In average, the TVOC concentration declined by 51% within 28 days. In the ModR described here, reductions of 39 and 56% were reached in week 4 and 5, respectively. However, other studies report on different VOC emissions data: According to Makowski et al. (2005), long-term TVOC from OSB was reduced by 78% within 28 days and after 59 days the TVOC concentration was 8% of the starting value.

Wood-specific substance groups

Table 1: Wood-specific substance groups in model rooms based on the concentration of total volatile organic compounds (TVOC)

	X-lam-ModR (% TVOC)		OSB- ModR (% TVOC)	
Weeks	Aldehydes	Terpenes	Aldehydes	Terpenes
1	13	87	56	32
2	9	91	57	35
3	15	83	60	32
4	12	79	49	36
5	16	73	49	35
6	14	80	53	34
7	15	82	54	36
9	17	83	57	36
11	15	85	48	41
13	17	80	48	41
15	17	81	45	46
19	20	76	46	41
23	14	74	43	47
Average	15	80	52	38

Aldehydes and terpenes are the most important VOCs emitted from softwoods in the indoor environment (Risholm-Sundman et al., 1998, Uhde and Salthammer, 2007). Hence, the contribution of this substance group to the TVOC was determined (Table 1). Accordingly, the share of terpenes amounted to 80% of the TVOC value in X-lam-ModR. The percentage of terpenes varied between 68 and 91%, while the maximum was reached in week 2 with 91%. The most abundant terpene was α-pinene. On the other hand, the aldehyde concentration varied from 9 to 20%. The average percentage over 23 weeks amounted to 15%, while the highest values were reached in week 19 (20% of the TVOC concentration). Hexanal was the most abundant aldehyde. These data are consistent with the literature. For example, Steckel et al. (2010) measured 85% terpenes in the VOC emissions of spruce wood and Englund and Nussbaum (2000) detected similar VOC data from Norway spruce.

The TVOC emission profile in the OSB-ModR is different. Aldehydes constituted the most abundant substance group, followed by terpenes. Aldehydes accounted for 52% of the TVOC in average over 23 weeks and the data ranged from 43% to 60%. In week 3, 60% of the TVOC were aldehydes. In average, 38% of the TVOC from OSB were terpenes. The highest terpenes contribution was measured in week 23 (47%), whereas the lowest value (32%) occurred in week 1. Overall, hexanal was the most abundant aldehyde during the whole measurement period, while α-pinene and 3-carene showed the highest concentration of all detected terpenes. This composition of TVOC is not consistent with the results for solid Scots pine, which generally show higher amounts of terpenes, comparable to the findings for spruce wood (Englund and Nussbaum, 2000, Risholm-Sundman et al., 1998). However, other studies ascertained an elevated aldehyde concentration in OSB. This effect is said to be the result of the hot-pressing production process (Wilke et al., 2013, Ze-Li et al., 2013, Salthammer et al., 2003). Makowski et al. (2005) measured up to 64% aldehydes emitted by OSB panels and also concluded that the aldehyde concentration is highly influenced by the production process.

Conclusions

The results of the VOC measurements show a decrease of the TVOC value for both ModRs. While the observed fluctuations were statistically insignificant, an overall reduction of the TVOC concentration in the ModRs over a period of six month was evident. The composition of the TVOC measured in the ModRs was comparable to literature data. However, the decline of the TVOC concentration in the ModRs differs to those in an emission test chamber, where faster TVOC reduction rates were observed. The starting values measured from X-lam are comparable to the results of former studies, whereas the TVOC concentration from OSB seems to be quite low compared to literature. This may be explained by the 10 weeks storage time of the OSB panels prior to the construction of the ModR. It can be concluded that the model room experiments are suitable for VOC determination under conditions more comparable to those in a real building than small scale laboratory experiments.

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Measuring VOC Emissions from wood-based Building Products under Real Room Conditions in an Idealised Model Room

Eva Höllbacher, Cornelia Rieder-Gradinger, Daniel Stratev, Ewald Srebotnik

Abstract

Two model rooms were constructed out of OSB and CLT and tested as a large-scale test method for emissions of volatile organic compounds (VOC) from wood-based building products over a period of 23 weeks. The measured VOC emissions show terpene and aldehyde emissions in both model rooms. Emissions from all identified substances declined over time to a low value. In the CLT model room terpene emissions exceeded aldehyde emissions. Terpenes declined from 65 µg m^{-3} to 11 µg m^{-3} (83%), whereas aldehydes declined from 28 µg m^{-3} to 5 µg m^{-3} (82 %). In the OSB model room aldehyde emissions were higher and declined from 247 µg m^{-3} to 51 µg m^{-3} (79%). Terpene emissions decreased from 59 µg m^{-3} to 23 µg m^{-3} (61 %).

Introduction

Today, building products are considered to be an important parameter influencing indoor air quality (Wolkoff and Nielsen, 2001). Wood-based materials such as Cross Laminated Timber (CLT) and Oriented Strand Board (OSB) are widely used in the construction of buildings. For this reason, it is likely that they to a certain extent influence the quality of indoor air. Terpene and aldehyde emissions are classified as playing an important role in indoor air quality (Hodgson et al., 2000). Origins of these emissions are wood-based building products (Hodgson et al., 2002). In fact, wood and wood-based building products emit several specific VOC which differ depending on wood species and production process (Jensen et al., 2001).

To control and to limit the amount of VOCs emitted into indoor air, various regulation schemes have been launched in Europe, such as the AgBB evaluation scheme in Germany or the French classification scheme for construction and decoration products (AgBB, 2012, Ministère de l'écologie; du développement durable; des transports et du logement, 2011). Regarding the applied testing methods for determining VOC emissions from building products, both guidelines refer to the ISO 16000 series. The most widely used test method is the emission test chamber method as specified in ISO 16000-9. This standard defines several criteria for emission testing, such as limit values for temperature, relative humidity and air exchange rate (DIN, 2008). Test chambers therefore allow VOC measurements under ideally controlled laboratory conditions. However, it is still unclear to what extent test chamber data can be transferred to real room situations. On the other hand, analysing the influence of VOC emissions from building products in real buildings is usually hindered by environmental effects and a hardly controllable climate.

Therefore a large-scale testing method was developed, which allows for VOC measurements under controllable climate conditions in idealised model rooms that simulate real rooms in terms of size and configuration. The construction of the model rooms was carried out – as far as possible – in compliance with the construction of a real building in an attempt to obtain VOC data from two widely used wood-based building materials in a real room scenario.

Materials and Methods

The two model rooms were completely constructed of Oriented Strand Boards (OSB) manufactured of Scots pine and bonded with pMDI with a thickness of 22 mm, and Cross Laminated Timber (CLT) produced of Norway spruce and bonded with polyurethane and MUF resin with a thickness of 95 mm. To make the rooms less sensitive to outside temperature variations, they were built and operated inside a technical centre and thermally insulated with 0.1 m thick panels of foamed polystyrene.

Each model room had dimensions of 3 m in width, 4 m in length and 2.5 m in height and was equipped with two inert areas, one door and one window. The window was made of glass and covered an area of 2 m^2 (2 m x 1 m). A stainless steel-plate of 2 m in height and 0.8 m in width served as the door area. Each room was additionally equipped with 2 inlets and 2 outlets for the ventilation system, each with a diameter of 0.2 m. The dimensions specified above resulted in a room volume of 30 m^3 and a total wooden surface of about 55.3 m^2, leading to a loading factor of 1.8 m^2 m^{-3}. For performing VOC measurements, three measuring sites were installed at different positions within the model room. One site was located in the middle of the room, one in a corner at a height of 0.5 m and one next to the wall at height of 1.50 m. One additional measuring site was positioned in the exhaust air duct.

According to the requirements of ISO 16000-9 (DIN, 2008), a standardised climate of 23 ± 2 °C temperature and 50 ± 5 % relative humidity must be guaranteed inside the model rooms. To reach these requirements, an air conditioning system was installed outside of the model rooms. External air was drawn in over a pre-filter and led through a column equipped with a heating and cooling unit, a dehumidifier and a humidifier. The conditioned air was then cleaned by a carbon filter. The air stream was split and each model room was supplied with an air flow rate of 15 m^3 h^{-1} resulting in the required air exchange rate of 0.5 h^{-1}. To reach sufficient mixing of the indoor air, an infinitely variable ceiling fan was installed in each model room.

Before starting with the measurement the air conditioning system was run for two weeks to stabilise the climate within the model rooms. A sensor was positioned in the middle of each room to control temperature and relative humidity (rH). The sensor logged the values of both parameters in an interval of 15 min. For each day the mean value of temperature and relative humidity was calculated. The results show that temperature and relative humidity remained within the allowable range most of the time. The mean temperature in the CLT model room was 22.5 °C, the allowed range was left at 14 days. The relative humidity shows a similar behaviour, but left the allowable range on only 8 days. The mean relative humidity value was 49.7 % rH over the whole test period. In the OSB model room, the allowable temperature range was left on 42 days, reaching a mean value of 22 °C which met the requirements. This is also true for the relative humidity which reached a mean value of 50.77 % rH. Altogether, the relative humidity fell below or exceeded the limit values on 14 days.

VOC concentration in the model rooms was measured regularly over a period of 23 weeks with a total of 13 measurements for each model room.

Air sampling was conducted as follows: At each measuring site, two samples were drawn on glass tubes filled with Tenax TA®. The sorbent tubes were loaded with 6 L of indoor air at an air flow rate of 100 mL min^{-1} using a FLEC® pump (Chematech, Denmark). Before sampling each tube was spiked with an internal standard of 0.37 µg toluene-D8 dissolved in methanol. The sorbent tubes were connected to PTFE tubes which led to the measuring sites through stainless-steel pipes.

After sampling the sorbent tubes were thermally desorbed with a thermal desorption unit (Chromtech® TDAS 2000, Germany) coupled with a programmed temperature vaporising (PTV) injector of a GC/MS (Agilent® 7890A/5975C, USA). Desorption was carried out at a temperature of 250 °C for five minutes. The desorbed analytes were cryo-focused in the PTV injector at -15 °C and abruptly heated to 280 °C. The PTV injector was operated in split mode by a split of 30:1. The analytes were separated on a methyl siloxane column (Agilent® HP-PONA, 50 m x 0.2 mm x 0.5 mm) using helium as carrier gas at an average velocity of 12.85 cm s^{-1}.

The following temperature program of the oven was used: 3 min at 35 °C, then ramp to 160 °C at 10 °C min^{-1} and ramp up to 310 °C at 20 °C min^{-1}. The MS detector was set to scan over a m/z range of 12–400.

Compounds were identified by comparison to spectra libraries specialised on wood specific substances. All identified substances were quantified as toluene equivalents (TE). Additionally, substance specific quantification was done for a selection of six wood specific aldehydes and four terpenes. The quantified aldehydes were pentanal, hexanal, benzaldehyde, octanal, 2-octenal and nonanal, the quantified terpenes were α-pinene, β-pinene, Δ-3-carene and limonene. No clear differences were observed among the results of the single measuring sites. Therefore, mean values (n=8) were calculated for each measurement for further analysis.

Results and discussion

As aldehydes and terpenes are two very typical substance groups emitted by wooden materials, their emission rates were studied in detail. Figure 5 shows the development of the terpene and aldehyde emission rates in the CLT model room. The emissions of terpenes were higher throughout the 23 weeks. The graphs show a higher starting value for the terpene emissions that approximated aldehyde emission rates with advancing measuring period. Starting at a value of 65 µg m^{-3} the terpene emissions decreased by 83 % to 11 µg m^{-3}. The fastest reductions appeared at week three and week nine where the concentration was reduced by 38 % and 53 %, respectively. The considerable fluctuations between week three and week seven appear to smoothen after the second steep decline. On the other hand the aldehyde emissions decreased by 82 % from 28 µg m^{-3} to 5 µg m^{-3} showing the most distinct decline to 9 µg m^{-3} (68%) between week one and week two. Afterwards aldehyde emissions tend to fluctuate considerably until week seven followed by a slight decrease until week 23.

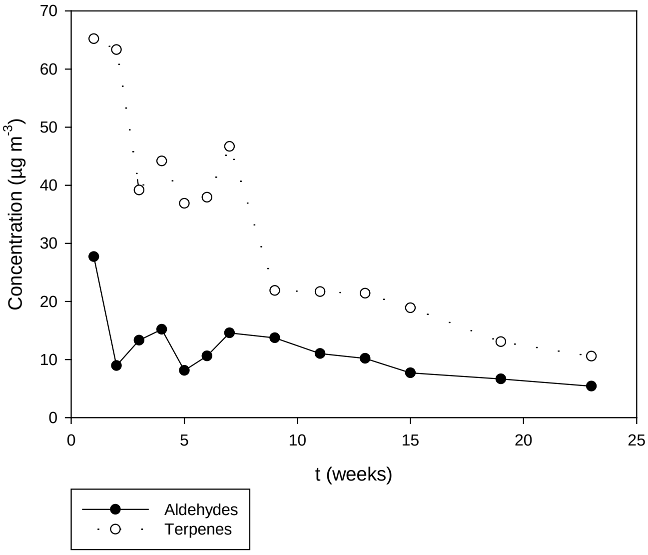

Figure 1: Aldehyde and terpene concentrations in the CLT model room

All four calibrated terpenes could be detected in the CLT model room. The most abundant substance was α-pinene followed by β-pinene. Δ-3-carene and limonene occured at similar concentrations (see Figure 6). All four determined terpene compounds showed a clear reduction over time. The highest overall reduction of 94 % was reached by β-pinene. The concentration declined from a value of 17 µg m^{-3} to 1 µg m^{-3}. However, α-pinene showed the highest emissions of all identified terpene compounds with a starting value of 35 µg m^{-3} in week one. The last measured value after 23 weeks was 7 µg m^{-3} corresponding to a total reduction of 28 µg m^{-3} or 80%. Similar relative reductions of 83 % and 71 % were measured for Δ-3-carene and limonene, respectively.

Fluctuations of the total terpene emissions (Figure 5) mainly reflect fluctuations of α-pinene and β-pinene. Δ-3-carene and limonene showed one significant increase in week seven. Starting from week nine, emissions of Δ-3-carene and limonene remained nearly constant at a very low level (1 – 2 µg m^{-3}).

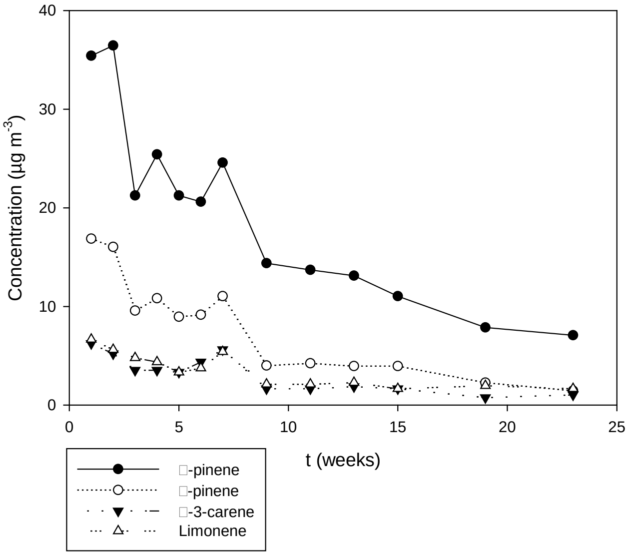

Figure 2: Terpene concentrations in the CLT model room

Three of the five selected aldehydes, namely pentanal, hexanal and nonanal were detected in the CLT model room. Hexanal emissions showed the highest concentrations for all time points but were lower than the emissions of α-pinene and β-pinene. Hexanal and pentanal emissions tended to fluctuate just as the terpene emissions (see Figure 7). The hexanal emissions were reduced by 64 % from 11 µg m^{-3} to 4 µg m^{-3} over the whole test period. The reduction was even higher for pentanal, reaching a value of 75 %. However pentanal emissions were quite low, starting at a value of 8 µg m^{-3} in week 1 and declining to 2 µg m^{-3} at week 23.

Nonanal emissions were too low for accurate quantification. However, the measured emissions showed no significant increase or decrease (data not shown).

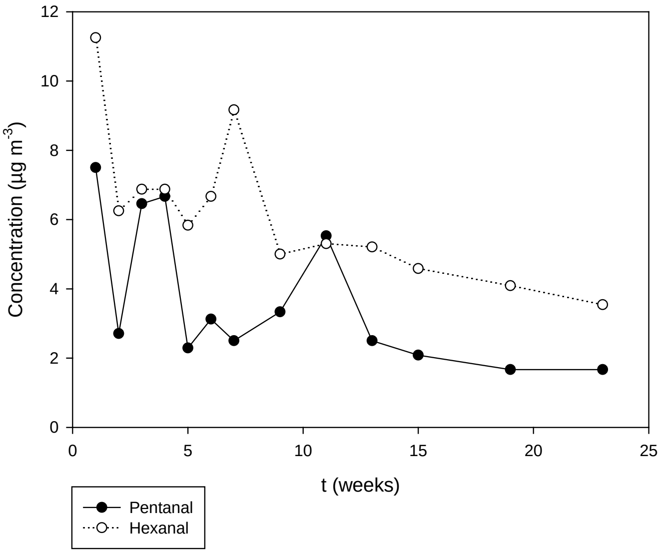

Figure 3: Aldehyde concentrations in the CLT model room

The emissions of aldehydes and terpenes in the OSB model room are depicted in Figure 8. The graphs show considerably higher concentrations of aldehydes over the whole measurement period of 23 weeks. An aldehyde concentration of 247 µg m^{-3} was measured after one week. Subsequently, the concentration declined rapidly to a value of 101 µg m^{-3} in week five and increased again over a period of two weeks to a value of 137 µg m^{-3} followed by another decrease (week seven to week 19). At the last measurement the aldehyde concentration was again increased, resulting in an emission level of 51 µg m^{-3}.

The terpene emissions were much lower than the aldehyde emissions. Starting with a concentration of 59 µg m^{-3} they declined to a value of 32 µg m^{-3} within three weeks. Just as for the aldehyde emissions, the terpene emissions also raised from week 19 to the last measurement in week 23 by 130 %.

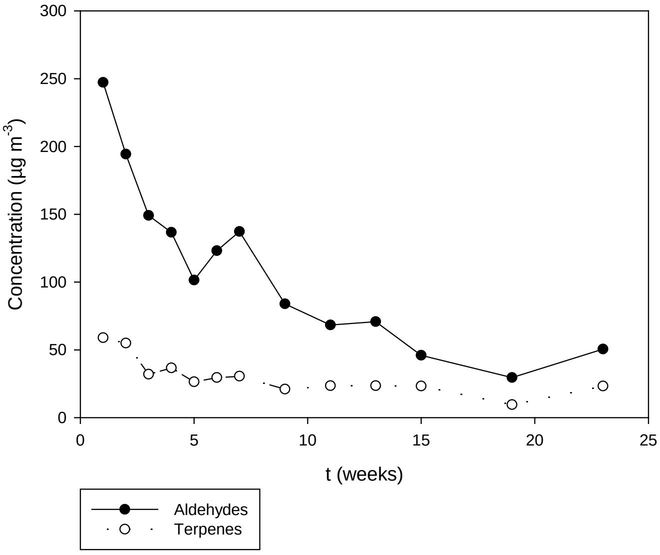

Figure 4: Aldehyde and terpene concentrations in the OSB model room

From all quantifiable terpene compounds, only α-pinene and Δ-3-carene have been detected in considerable concentrations in the OSB model room. α-pinene showed a higher concentration at the beginning but then decreased more rapidly than Δ-3-carene. α-pinene reduction over the whole test period was 69 % from a starting value of 35 µg m^{-3} to 11 µg m^{-3} after 23 weeks. Δ-3-carene reduction was only 35 % from 20 µg m^{-3} to 13 µg m^{-3}. However, the lowest concentration of Δ-3-carene was reached in week 19 with a value of 6 µg m^{-3}, which would equal a reduction of 70 %, but emissions increased again during the last month of the testing period. In fact, the increase of terpene emissions at the last measurement was caused by both α-pinene as well as Δ-3-carene (see Figure 9).

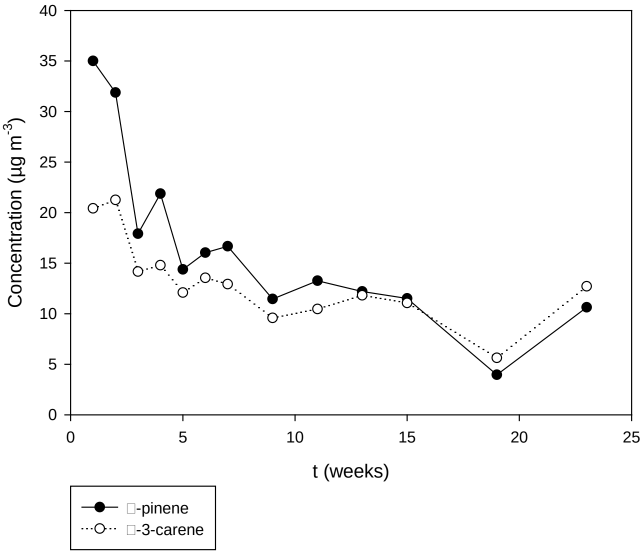

Figure 5: Terpene concentrations in the OSB model room

Five calibrated aldehydes could be identified and quantified in the OSB model room. Their emission rates are shown in Figure 10. Hexanal was the most abundant aldehyde with a starting value of 163 µg m^{-3}. Over the whole test period hexanal emissions were reduced by 80 % to 32 µg m^{-3}. Hexanal accounted for 66 % of the total aldehyde emissions at the beginning of the measurements and 63 % after 23 weeks. Therefore, emission rate of the sum of aldehydes reflected mainly the development of hexanal emissions, which also showed their steepest decline during the first five weeks when a concentration of 63 µg m^{-3} was reached. Pentanal showed the second highest concentration of aldehydes in the OSB model room, though they were much lower than hexanal emissions. In week one, a concentration of 51 µg m^{-3} was measured, followed by a decline to a value of 11 µg m^{-3} in week 23. Other aldehydes, such as octanal, 2-octenal and nonanal, were even less concentrated. Starting values ranged between 6 µg m^{-3} and 17 µg m^{-3}. After 15 weeks, octanal was no longer detected, whereas 2-octenal and nonanal were detected during the whole testing period at low concentrations. Both substances showed fluctuations. 2-octenal emissions decreased slightly, while nonanal emissions were detected in a concentration range of 4 to 15 µg m^{-3} and showed no significant reduction.

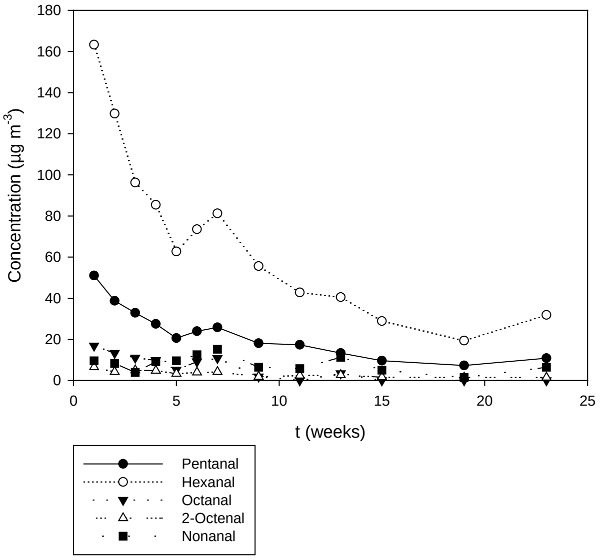

Figure 6: Aldehyde concentrations in the OSB model room

The composition of the terpenes and aldehydes emitted from CLT was consistent with previous findings for Norway spruce (Risholm-Sundman et al., 1998, Englund and Nussbaum, 2000, Steckel et al., 2010).

Terpene emissions measured in the OSB model room showed a similar composition as terpene emissions from solid pine wood (Englund and Nussbaum, 2000, Risholm-Sundman et al., 1998), yet, aldehyde emissions were much higher This is maybe due to the production process of OSB panels which supports the formation of aldehydes (Wilke et al., 2013). Other investigations on OSB made from Scots pine showed a similar ratio of aldehydes and terpenes taking into account that panels were not freshly produced (Wilke et al., 2013, Makowski et al., 2005).

Conclusions

The aim of this study was the determination of indoor air quality in timber constructions that more closely resembles the real situation than established small-scale methods do.

The composition of the identified VOC is similar to findings of other authors and the emitted substances show a clear decline over time although fluctuations could be observed. These fluctuations occur probably due to the fact that wood as a natural building material does not show homogenous emission rates. CLT emissions are comparable to emissions from Norway spruce. However, emissions from OSB do not fully reflect the emissions from its raw material, Scots pine. This may be explained by the fact that OSB panels pass a production process which has a severe influence on their emission profile, as aldehyde emissions are increased when compared to terpene emissions. Nevertheless, VOC emitted by OSB panels are comparable to other studies.

The concept of the model room presented here appears to be suitable as a large-scale testing method though further improvements are possible. According to the results of the study, the model room concept provides the possibility to construct a room similar to a real building and thereby enables the determination of VOC emissions from wood-based building products in a real room scenario.

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Influences of interior fitting processes on the indoor air quality of two model rooms

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Abstract

Interior fitting was performed stepwise in two model rooms built from OSB (Oriented Strand Board) and X-lam (Cross Laminated Timber), respectively. The walls and ceiling of the OSB model room were covered with gypsum plasterboard, parquet flooring was laid and the room was finally equipped with office furniture and textiles. In the X-lam model room an acoustical ceiling and parquet were installed. The walls were painted with wood stain and the room was adapted as bed room and equipped with textiles. After each fitting step VOC (volatile organic compounds) samples were taken and analyzed. In the OSB model room the TVOC (total volatile organic compound) concentration decreased throughout the whole fitting process from 119 µg m^{-3} prior to the interior fitting to a final value of 26 µg m^{-3}. In the X-lam model room, a change of the VOC profile and/or concentration could be observed after each fitting step. The highest TVOC concentration (225 µg m^{-3}) was reached seven days after the installation of the acoustical ceiling. Terpenes, aldehydes and acetic acid were the most abundant substances identified. It was shown for both model rooms that some of the applied building materials acted as diffusion barriers or as emission sinks.

Introduction

Since people in industrial countries nowadays spend up to 90 % of their time indoors and low air exchange rates in modern energy efficient buildings support the accumulation of possible pollutants in indoor air, indoor air quality has become an issue with regard to occupants' health (Zhang and Smith, 2003, Jones, 1999). Rising health awareness during the last decades led to an improved knowledge about indoor air quality and indoor air pollutants (Weschler, 2009). In the indoor environment volatile organic compounds (VOC) represent an important influencing factor concerning air quality. Generally, the term VOC comprises substances with a low boiling point or a high vapor pressure, respectively. VOC emit from various natural or anthropogenic sources. Beside others, building products are said to be long-lasting sources of VOC in the indoor environment (Kostiainen, 1995, Salthammer and Bahadir, 2009, Wolkoff and Nielsen, 2001). The rising awareness for VOC emissions and their possible health effects led to increased research on indoor air quality (Jones, 1999). Due to these research activities broad knowledge was gained about VOC emissions from building products, which led to the development of standardized testing procedures like the ISO 16000 series (DIN, 2011, DIN, 2008) and various regulation and classification schemes (Salthammer, 2011) such as the German AgBB-scheme (AgBB, 2015). Additionally, several environmental labels such as "The Blue Angel" (RALgGmbH, 2011) or "Natureplus" (natureplus, 2008) include threshold values for VOC emissions in their requirements. Further research concentrated on indoor air quality in real buildings and possible influencing factors (Edwards et al., 2001, Geiss et al., 2011, Derbez et al., 2014, Guo, 2011, Nyrud et al., 2012). Scientific studies showed that building materials, installed in a room together, may influence their mutual VOC emissions (Uhde and Salthammer, 2007, Niedermayer et al., 2013). However, only little attention was paid so far to the development of VOC emissions during an interior fitting process, which lasts over a considerable length of time and gradually combines different construction and furnishing materials. Earlier studies observed that in addition to the impact of the surrounding materials, VOC emission from a building product are also influenced by environmental effects like temperature or humidity (Liang et al., 2014, Järnström et al., 2006, Järnström et al., 2007). To avoid such unwanted environmental effects in the present study, two model rooms with defined climate conditions and air exchange rate were constructed. These rooms were then fitted with various building materials. The basic construction of the model rooms was made of common wood-based construction products, which are frequently used in the construction of prefabricated houses. Materials frequently used for interior fitting were chosen. Therefore, a high share of wood-based materials, which are known sources of VOC, was applied (Hodgson et al., 2000). Softwoods, which are often used for producing building and furnishing materials, emit mainly aldehydes and terpenes (Risholm-Sundman et al., 1998, Roffael, 2006) while other wood species may also set free other substances (Jensen et al., 2001, Roffael, 2006). Furthermore, the emissions from wood-based building products also depend on the relevant production process (Makowski et al., 2005, Guo et al., 2002, Kim, 2010).

The aim of this study was the simulation of an interior fitting process under conditions as close as possible to reality but to the exclusion of unwanted environmental effects in order to gain further knowledge about the influence of building activities on indoor air quality.

Materials and Methods

Model rooms

Fitting steps were performed in two model rooms with dimensions of 3x4x2.5 m^3. The basic construction of the model rooms was finished nine months prior to the start of the interior fittings and was completely made from oriented strand board (OSB) and cross laminated timber (X-lam), respectively. Each model room was equipped with three measuring sites for indoor air sampling. The model rooms were supplied with cleaned and conditioned air from an external air conditioning system at an air exchange rate of 0.5 h^{-1}. Temperature was set to 23 ± 2 °C and relative humidity to 50 ± 5 % according to ISO 16000-9 (DIN, 2008). The setup and operating mode of the model rooms and the air conditioning system has been described in detail in (Höllbacher et al., 2015).

Fitting steps

Both model rooms were furnished with different interior fittings. The installation of the materials was performed successively. After each installation step, VOC samples were drawn and analyzed. Thereby, the development and change of the VOC concentrations and the VOC emission profile during the interior fitting process was examined.

All fitting steps were performed over a time period of 15 weeks from July until November 2013. The individual fitting steps, the week of their performance and the installed materials are shown in Table 1. The timeline starts with the first interior construction step which is referred to as week 1. However, when comparing the VOC concentration at different construction steps, one has to keep in mind that the basic construction of the model rooms was finished nine months (36 weeks) prior to the installation of the interior fittings.

Table 1: Fitting steps performed in each model room, calendar week of performance, and specification of the applied materials

	week	fitting step	product	description
OSB model room	1	dry lining	vapor barrier sheet	polyamide reinforced with fleece
			gypsum plasterboard	gypsum plasterboard
			wall paint	limewash
	5	flooring	solid parquet	larch wood, oil-based coating, UV cured
	11	furniture	writing desk	laminated particleboard, metal
			3 chairs	synthetic material, metal
			tall table	laminated particleboard, metal
			bookshelf	laminated particleboard
	15	textiles	pin board	cork, pine wood
			blinds	cotton
			doormat	polypropylene, synthetic latex backing
X-lam model room	1	ceiling	insulation	wood-fiber insulation board, softwood
			acoustical ceiling	solid, untreated larch wood
	5	flooring	solid parquet	oak wood, oil-based coating, UV cured
	7	wall paint	wood stain	water-based, UV stable, for indoor use
	11	furniture	bed	solid, untreated pine wood
			2 stools	solid, untreated pinewood
			2 closets	solid, untreated pinewood; hardboard
	15	textiles	carpet	polypropylene, synthetic latex backing
			curtain	polyester
			mattress	polyester, polyether filling
			pillow	polyester, polyester filling
			blanket	cotton, polyester, polyester filling

In the OSB model room, a dry lining was performed in the beginning. Subsequently, parquet flooring was laid. The room was then furnished with office furniture and finally equipped with textiles, including a pin board which covered an area of 0.23 m² and had a weight of 0.9 kg and blinds with an area of 3.2 m² and a weight of 0.6 kg. The X-lam model room was equipped with an acoustical ceiling. In the next step, parquet flooring was laid. Afterwards, the wooden walls of the X-lam construction were painted with a wood stain for indoor use. Then, the X-lam model room was furnished with bedroom furniture and equipped with textiles, including a carpet with an area of 2.8 m² and a weight of 5.1 kg and curtains with an area of 8.4 m² and a weight of 0.4 kg, in a final step. All materials were purchased from retailers, except the acoustical ceiling which was delivered by the manufacturer. The dry lining and wood stain and parquet floorings were purchased from retailers specialized on building materials and floorings, respectively. Bedroom furniture and textiles were purchased from furniture stores, while office furniture was given on loan basis from the shop of a producer and retailer of office furniture. They were delivered to the construction site of the model room maximum one week prior to their installation and stored packed under constant climate conditions comparable to those in the model room to avoid any unwanted contamination with environmental VOC.

Air samples were taken from the empty model room prior to the start of the interior fittings as well as seven and 14 days after each fitting step. The results of both measurement dates are presented for all fitting steps except for the last two steps “furniture” and “textiles”. In this case, only the results 14 days after installation are presented in this article. Earlier samples from these two fitting steps showed an untypical emission profile which was presumably caused by the packaging of the materials. Therefore, it seemed necessary to exclude the results from day seven to avoid any influence of the packaging material and to guarantee a steady state of the VOC emissions.

µ-CTE™ tests

All building materials installed in the model rooms were additionally and individually tested in a Micro-Chamber/Thermal Extractor (µ-CTE™; Markes International Ltd, Llantrisant, Wales, UK). These tests were performed simultaneously to the fitting steps in order to gain information about the substances emitted by the individual material and identify possible VOC sources. Further details on the setup and operation mode of the µ-CTE™ have been described in (Schripp et al., 2007)

Two circular specimens with a diameter of 43 mm were cut out from each building material. The wood stain was applied on an X-lam specimen already examined in the course of an earlier test series. Each specimen was placed in an individual cell of the µ-CTE™ and kept there over the whole measurement period. The climate parameters were set to the same values as in the model room (23 ± 2 °C, 50 ± 5 % rH (relative humidity)). The air exchange rate was set to 40 h^{-1}. Air samples were taken three, seven and 14 days after the specimens were placed in the µ-CTE™. These measurement dates were chosen to guarantee results comparable to those in the model rooms 7 days after installation of a new material. However, only results from day 3 are presented in this paper as no new substances could be identified after seven and 14 days. As furniture and textiles were partially supplied on a loan basis and non-destructive sampling was not possible, furniture and textiles were not examined in the µ-CTE™.

VOC sampling and analysis

Samples of indoor air were taken according to ISO 16000-6 (DIN, 2011). Sorption tubes made of glass and filled with Tenax TA® (Sigma-Aldrich, St. Louis, Missouri, USA) as adsorbent were used. Prior to the start of the interior fitting, a total of four samples (n=4) was taken from each model room using a FLEC® pump (Chematec, Roskilde, Sjælland, Denmark). During interior fitting, two consecutive samples of indoor air were drawn at each of the three measuring sites (n=6) in both model rooms. Sampling was performed over 60 minutes at an air flow rate of 100 mL min^{-1}. Additionally, one sorption tube served as field blank to detect possible contaminations at the measuring site. The field blank was handled in the same way as the other sorption tubes, although not loaded with indoor air. In the µ-CTE™ sampling was done at an air flow rate of 7 mL min^{-1} using the volume flow rate through the µ-CTE™. One sample was taken from each cell. Before sampling, each tube was spiked with 0.37 µg toluene-d8 dissolved in methanol as internal standard.

Analytes were thermally desorbed from the adsorbent at a temperature of 250 °C for five minutes using a thermal desorption unit (TDAS 2000; Chromtech, Idstein, Hesse, Germany) connected to the programmed temperature vaporizing (PTV) injector of a GC/MS system (7890A/5975C; Agilent Technologies, Santa Clara, California, USA). The desorbed substances were cryo-focused in the PTV injector at a temperature of -15 °C and abruptly heated to 280 °C at a heating-up rate of 720 °C min^{-1}. The PTV injector was operated in split mode with a split of 30:1. The analytes were separated on a dimethylpolysiloxane column (HP-PONA, 50 m x 0.2 mm x 0.5 mm; Agilent Technologies, Santa Clara, California, USA) using helium as a carrier gas. The following temperature program was used: Three minutes at 35 °C, then ramp to 160 °C at 10 °C min^{-1}, and ramp up to 310 °C at 20 °C min^{-1}. The MS detector was set to scan over an m/z range of 12–400. Detected compounds were identified by comparison of the mass spectra to commercial mass spectral libraries (John Wiley & sons, Hoboken, New Jersey, USA). Analytes were quantified using authentic calibration standards (Sigma Aldrich Co. LLC, St.Louis, Missouri, USA). Substances without existing calibration were quantified as toluene equivalents. These substances are marked with the abbreviation TE. The total VOC (TVOC) concentration was calculated for each sample as the sum of all substances with a concentration ≥ 1 µg m^{-3} including acetic acid. The arithmetic mean and standard deviation were calculated from all samples drawn in the model room (n=4, n=6) for further analysis. Samples from the µ-CTE™ were only analyzed qualitatively.

Results and Discussion

OSB model room

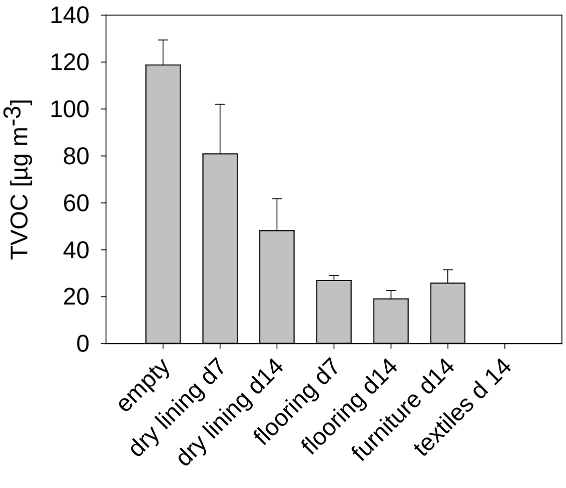

Figure 1: TVOC concentration in the OSB model room showing mean value (grey bar) and standard deviation. d7 and d14 denote the TVOC measurement seven and 14 days after the construction step, respectively. After the installation of textiles all identified substan

Table 2: Concentration of the major VOC after each individual construction step in the OSB model room. Standard deviation is shown in parentheses. d7 and d14 denote TVOC measurement seven and 14 days after each construction step, respectively

VOC [µg m^{-3}]	empty	dry lining		flooring		furniture	textiles
		d7	d14	d7	d14	d14	d14
α-pinene	14 (4)	18 (9)	11 (7)	3 (0)	2 (0)	5 (0)	b.q.
3-carene	18 (2)	13 (4)	10 (2)	b.q.	b.q.	b.q.	b.q.
cymene	7 (0)	b.q.	b.q.	b.q.	b.q.	b.q.	-
verbenone	10 (0)	-	-	-	-	-	-
pentanal	18 (2)	10 (4)	5 (1)	3 (0)	b.q.	b.q.	-
hexanal	31 (4)	19 (6)	12 (2)	10 (1)	7 (1)	9 (4)	b.q.
benzaldehyde	11 (0)	11 (1)	10 (1)	10 (1)	10 (1)	11 (0)	b.q.
nonanal	11 (1)	10 (1)	b.q.	b.q.	b.q.	-	-

Figure 1 shows the TVOC concentration in the OSB model room before as well as seven and 14 days after each fitting step. The highest TVOC concentration of 119 µg m^{-3} was measured in the empty model room. Hexanal was the most abundant substance with a concentration of 31 µg m^{-3} as shown in Table 2. Several other aldehydes and terpenes as well as organic acids have been identified. All substances identified in the model room were also detected in the microchamber (see Table 4) and can therefore be clearly assigned to OSB. Seven days after installation of the dry lining the TVOC concentration decreased by 32 % to 81 µg m^{-3}, followed by a further decrease to 48 µg m^{-3} after 14 days. Most of the identified substances also showed a clear decrease. The share of terpenes (32 %) and aldehydes (62 %) remained in a range comparable to that of the empty model room. Additionally, it can be stated that the emission of both, aldehydes and terpenes, though they are chemically different substance groups, were lowered to similar extents after installation of the dry lining. It can thus be concluded that the barrier effect of the dry lining was responsible for the observed decrease of the TVOC concentration. This is supported by the fact that the model room was constructed nine months prior to the start of the interior fitting process, and emission rates therefore had already reached a steady state level. This was shown in an earlier study on the model rooms, where the natural decrease of emission rates in the model rooms after 6 months was very slow (Höllbacher et al., 2015). Both vapor barrier sheet and gypsum plasterboard act as diffusion barriers for VOC emissions of underlying materials (Niedermayer et al., 2013) The VOC profile did not change after the installation of the dry lining (Table 1), which is strong evidence that the used materials did not emit any new substances. This was confirmed by the µ-CTE™ tests showing that neither gypsum plasterboard nor the vapor barrier sheet emitted any VOC (Table 4). Seven days after the parquet flooring was laid, the TVOC concentration decreased again to a value of 27 µg m^{-3}. Although nine substances were identified in the model room, only four of them were above the limit of quantification of 1 µg m^{-3} and could thus be quantified. 14 days after the installation, a TVOC concentration of 19 µg m^{-3} was observed. No change of the VOC profile could be observed for both measurements. Eleven substances were detected in the air samples taken from the µ-CTE™ (Table 4). Nine of these substances were also detected in the model room. However, the same substances also emitted from the OSB panels that had been used as construction material. Earlier studies on parquet flooring with UV cured coatings showed that these coatings act as efficient diffusion barriers for VOC emissions (Kim, 2010). It can be assumed that the contribution of the VOC emissions from the flooring to the TVOC is too small to counteract the decrease of VOC emissions from other products previously installed in the model room.

In the next fitting step the OSB model room was equipped with customary office furniture. After this step, the TVOC concentration slightly increased to a value of 26 µg m^{-3} after 14 days. However, no change in the VOC profile could be observed. Only three substances could be identified and quantified. It is not clear from the data whether the TVOC increase was caused by the VOC emission from the furniture itself or by other sources which were already present in the model room. Possibly, the increase was due to the delayed diffusion of VOC from the construction material through the applied coverings.

As a last fitting step, the room was equipped with textiles. After this step, the concentration of all identified VOC dropped below quantification limits. The installed textiles obviously acted as a strong VOC sink (Jorgensen and Bjorseth, 1999).

X-lam model room

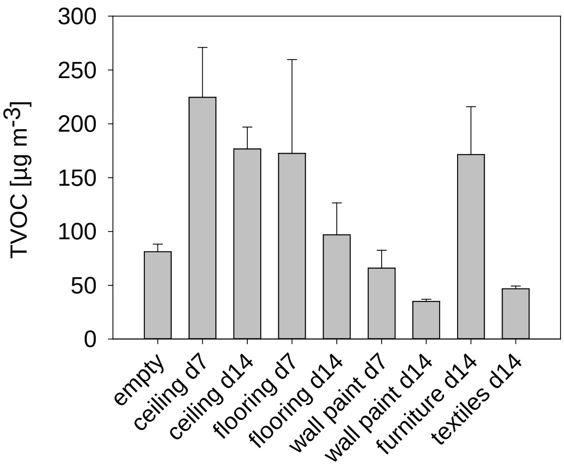

Figure 2: TVOC concentration in the X-lam model room showing mean value (grey bar) and standard deviation. d7 and d14 denote TVOC measurement seven and 14 days after the construction step, respectively.

Table 3: Concentration of the main VOC after each individual construction step in the X-lam model room. Standard deviation is shown in parentheses. d7 and d14 denote TVOC measurement seven and 14 days after each construction step, respectively

VOC [µg m^{-3}]	empty	ceiling		flooring		wood stain		furniture	textiles
		d7	d14	d7	d14	d7	d14	d14	d14
α-pinene	8 (1)	20 (3)	20 (7)	7 (5)	5 (1)	2 (1)	2 (1)	65 (26)	12 (1)
β-pinene	b.q.	2 (1)	b.q.	b.q.	b.q.	b.q.	b.q.	5 (3)	b.q.
3-carene	9 (1)	15 (1)	12 (2)	9 (1)	8 (0)	8 (0)	8 (0)	40 (11)	13 (0)
cymene	7 (0)	7 (0)	7 (0)	7 (0)	b.q.	b.q.	b.q.	b.q.	b.q.
limonene	17 (1)	18 (0)	17 (1)	b.q.	b.q.	b.q.	b.q.	20 (1)	b.q.
pentanal	8 (1)	10 (2)	7 (1)	5 (4)	b.q.	-	-	9 (7)	2 (1)
hexanal	13 (1)	19 (2)	13 (2)	10 (2)	9 (1)	8 (0)	9 (1)	22 (4)	10 (1)
furfural	-	14 (1)	13 (0)	13 (0)	-	-	-	-	-
benzaldehyde	11 (1)	11 (1)	10 (1)	10 (1)	10 (1)	11 (1)	11 (1)	11 (0)	10 (1)
nonanal	11 (1)	10 (1)	10 (1)	b.q.	b.q.	b.q.	b.q.	-	-
acetic acid	-	100 (13)	68 (9)	112 (74)	64 (14)	25 (13)	-	-	-
2-butoxyethanol TE	-	-	-	-	-	5 (2)	1 (1)	-	-
propylene glycol TE	-	-	-	-	-	8 (2)	3 (1)	-	-

Figure 2 shows the TVOC concentration in the X-lam model room before as well as seven and 14 days after each fitting step. In the empty model room a TVOC concentration of 81 µg m^{-3} was measured. Both aldehydes and terpenes accounted for 50 % of all detected substances. The concentration of the main VOC identified in the empty X-lam model room and after each fitting step is shown in Table 3.

In the first interior fitting step the ceiling was installed. Seven days after the installation the TVOC concentration reached a value of 225 µg m^{-3}. Acetic acid and furfural were detected for the first time. Acetic acid showed a high abundance and accounted for 44 % of the TVOC reaching a concentration of 100 µg m^{-3}. The concentration of α-pinene and 3-carene increased considerably from 8 µg m^{-3} to 20 µg m^{-3} and 9 to 15 µg m^{-3}, respectively, compared to the empty model room. For aldehydes, a significant increase of pentanal and hexanal was observed. The findings from the model room were confirmed by µ-CTE™ analysis as shown in Table 4. Here, acetic acid and furfural were detected in addition to other substances which mainly belonged to the groups of aldehydes and terpenes. Additionally, both substances were also detected in specimens of the insulating material in the µ-CTE™ investigations. The fairly high concentration of acetic acid in the model room was thus caused by the emissions of both the acoustical ceiling and the insulating material. 14 days after installation of the ceiling a TVOC concentration of 177 µg m^{-3} was measured. The decrease of the TVOC concentration was mainly due to a decrease of the acetic acid concentration. The pattern and the concentration of all other identified VOC were comparable to those which had been measured seven days after this fitting step.

Seven days after the oak wood flooring had been laid, the TVOC concentration amounted to 173 µg m^{-3}. New substances were not detected. Except for acetic acid, all detected substances showed a decrease compared to the prior fitting step. This decrease was substantial for some substances as in case of α-pinene (65 %). Limonene was only detected in traces and could no longer be quantified correctly. This decrease is due to the natural decay of the emissions from the acoustical ceiling and the insulating material which were installed four weeks prior to the installation of the flooring. On the other hand, acetic acid increased to a concentration of 112 µg m^{-3}. Acetic acid was also detected in the µ-CTE.

In the following seven days the TVOC concentration declined distinctly to a value of 97 µg m^{-3}. This decrease occurred mainly due to a decrease of acetic acid which was reduced by 43 % within seven days. In the next fitting step, the wooden surface of the walls was painted with a wood stain. In the first measurement seven days after painting, 2-butoxyethanol and propylene glycol were detected. 14 days after the application of the wood stain, both substances were still present, although their concentration had decreased. The µ-CTE™ measurements clearly showed that propylene glycol and 2-butoxyethanol emissions originated from the wood stain. Despite the appearance of these new substances, the TVOC concentration decreased to 66 µg m^{-3} and 35 µg m^{-3} seven days and 14 days after the walls had been painted, respectively. 14 days after the model room was equipped with pine wood furniture, the TVOC concentration strongly increased to a value of 172 µg m^{-3}. A-pinene, 3-carene, and hexanal showed the highest increase. Limonene reached a concentration of 20 µg m^{-3}. Although no samples from the furniture were tested in the µ-CTE™, earlier studies showed that this VOC profile as determined here is typical for solid pine wood (Englund and Nussbaum, 2000, Paczkowski et al., 2013) In the last fitting step the model room was equipped with textiles. 14 days after this step the TVOC concentration amounted to only 47 µg m^{-3}. The VOC profile did not change, but the concentration of the detected substances decreased. Again, as in the OSB model room, textiles acted as a sink for VOC. However, it is known that this sink effect is not permanent and VOC emission may only be delayed. Studies showed that substances initially adsorbed to textiles are desorbed when they are subsequently exposed to clean air (Jorgensen and Bjorseth, 1999, Jorgensen et al., 1999).

Table 4: VOC profile of the applied building materials as identified in the µ-CTETM at day 3 and comparison to the VOC profile detected in the model room after installation of the materials. X = identified, — = not identfied

	OSB		**X-lam**		**acoustical ceiling**		**insulating material**		**oak flooring**		**larch flooring**		**vapour barrier**		**gypsum plasterboard**		**wood stain**	
	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR	µ-CTE	MR
organic acids																		
acetic acid	X	X	—	—	X	X	X	X	X	X	X	—	—	—	—	—	—	X
hexanoic acid	X	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
aldehydes																		
pentanal	X	X	X	X	—	X	—	X	—	X	X	X	—	X	—	X	—	—
hexanal	X	X	X	X	X	X	—	X	—	X	X	X	—	X	—	X	—	X
heptanal	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
furfural	—	—	—	—	X	X	X	X	X	X	—	—	—	—	—	—	—	—
benzaldehyde	X	X	X	X	X	X	—	X	—	X	X	X	—	X	—	X	—	X
octanal	X	—	—	—	—	—	—	—	—	—	X	—	—	—	—	—	—	—
2-octenal	X	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
nonanal	X	X	X	X	—	X	—	X	—	X	X	X	—	X	—	X	—	X
terpenes																		
α-pinene	X	X	X	X	X	X	—	X	—	X	X	X	—	X	—	X	—	X
β-pinene	X	X	X	X	X	X	—	X	—	X	X	X	—	X	—	X	—	X
3-carene	X	X	X	X	X	X	—	X	—	X	X	X	—	X	—	X	—	X
p-cymene	X	X	X	X	—	X	—	X	—	X	X	X	—	X	—	X	—	X
limonene	X	X	X	X	—	X	—	X	—	X	X	X	—	X	—	X	—	X
verbenone	X	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
isolongifolene	X	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
others																		
2-heptanone	X	X	X	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
2-butoxyethanol	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	X	X
Propylene glycol	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	X	X
sum of detected substances	17	15	10	9	7	11	2	11	2	11	11	9	0	9	0	9	2	11

Conclusion

The aim of this study was to show the various influences on indoor air quality during an interior fitting process, including the interaction between different materials but to the exclusion of environmental effects. We were able to demonstrate that each interior fitting step, performed within a room, can influence indoor air quality. However, the amount and the development of VOC emissions in indoor air proved to be very complex. Barrier or sink effects of certain materials may lead to a reduction or time-delayed emission of VOC, just as the combination of materials may lead to an increased VOC concentration. Considering all observed effects it may be stated that the examination of the VOC profile and VOC concentration related to a single construction or furnishing product does not necessarily reflect its actual influence on indoor air quality. Furthermore, source apportionment through laboratory testing methods is hindered by the fact that many building and furnishing materials emit similar VOC. The results of this study clearly show that the individual emissions from a construction or furnishing material can be strongly altered by the surrounding materials, resulting in a VOC profile and/or concentration which is significantly different from what is expected from the individually tested material.

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Emissions of indoor pollutants from six user scenarios in a model room

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Abstract

In this study six common user scenarios putatively influencing indoor air quality were performed in a model room constructed according to the specifications of the European Reference Room given in the new horizontal technical specification CEN/TS 16516 to gain further information about the influence of user activities on indoor air quality. These scenarios included the use of cleaning agent, an electric air freshener, an ethanol fireplace and cosmetics as well as cigarette smoking and peeling of oranges. Four common indoor air pollutants were monitored: volatile organic compounds (VOC), particulate matter (PM), carbonyl compounds and CO_2. The development of all parameters was determined during and after the test performance. For each measured parameter, well-defined maximum values could be assigned to one or more of the individual user scenarios. The highest VOC concentration was measured during orange-peeling reaching a maximum value of 3547 µg m^{-3}. Carbonyl compounds and PM were strongly elevated while cigarette smoking. Here, a maximum formaldehyde concentration of 76 µg m^{-3} and PM concentration of 378 µg m^{-3} were measured. CO_2 was only slightly affected by most of the tests except the use of the ethanol fireplace where a maximum concentration of 1612 ppm was reached. Generally, the user scenarios resulted in a distinct increase of several indoor pollutants that usually decreased rapidly after the removal of the source.

Introduction

According to the National Human Activity Pattern Survey of the USA adults are spending an average of 87% of their time in enclosed buildings and about 6% of their time in enclosed vehicles (Klepeis et al., 2001). As to improve energy efficiency, building design has changed leading to more airtight structures and to the accumulation of air pollutants in indoor air (Jones, 1999). Therefore, the exposure of humans to the indoor environment is of high concern. Indoor air quality in living or working spaces is influenced by a large number of possible pollution sources (Kostiainen, 1995, Parra et al., 2008, Wolkoff, 2013). These sources include outdoor air, building and furnishing materials as well as user activities such as cleaning, cooking, or smoking (Salthammer and Bahadir, 2009, Wolkoff and Nielsen, 2001). The strong influence of user behavior on indoor air quality is known from various studies (Steinemann et al., 2011, Nazaroff and Weschler, 2004, Petry et al., 2014). However, for a long time research activities mainly focused on continuous, persistent sources of indoor pollutants. An example are VOC and formaldehyde emissions from building products like floorings, wall coverings or paints and varnishes which have been studied thoroughly during the last decades (Bluyssen et al., 1997, Gehrig et al., 1994, Lin et al., 2009, Risholm-Sundman and Wallin, 1999, Kim, 2010, Guo et al., 2004). Based on this research accredited test procedures for the emissions of building products and furniture were developed and defined in international standards such as the ISO 16000 series (DIN, 2011b, DIN, 2008). Furthermore, various national and international regulation and classification schemes such as the German “AgBB-scheme” (AgBB, 2015) define threshold values for VOC emissions from building products to guarantee a minor impairment of indoor air quality. Until recently, no comparable effort has been made to investigate the possible influence of user activities on indoor air quality, although they are manifold. They vary from terpene emissions from the use of fragranced products (Steinemann, 2015) to increased particulate matter concentration due to, for instance, environmental tobacco smoke (Sahu et al., 2013). Distinct changes in both the concentration and the types of indoor pollutants were identified after the occupation of newly constructed buildings (Järnström et al., 2006, Derbez et al., 2014, Yamaguchi et al., 2006). Nowadays, more and more attention is paid to user activities and their role regarding indoor air quality (Bartzis et al., 2015, Nazaroff and Weschler, 2004, Steinemann, 2015). This even lead to first attempts to develop reliable and reproducible testing procedures for various indoor pollutants emitted by consumer products (Bartzis et al., 2015). However, most investigations on emissions from consumer products are carried out in test chambers. The comparability of these chambers to the real room situation is limited, due to the fact that they usually do not resemble real rooms in terms of size and configuration. Furthermore, examining consumer products and user activities as possible indoor emissions sources is difficult. Building products, for instance, are continuous emission sources and may therefore be examined under steady conditions for variable time periods (De Bortoli et al., 1999, Wolkoff, 1998, Wolkoff, 1999). In constrast, user activities are temporary emission sources (Brown et al., 1994, Bartzis et al., 2015). Laboratory studies mainly focused on the emission of indoor pollutants during the use of a special consumer product or device (Derudi et al., 2012, Schripp et al., 2014), whereas measurements in real buildings also displayed the presence of indoor pollutants before and after a certain user activity is finished. However, tests in real indoor settings could neither exclude nor control any influences of environmental effects such as outdoor emissions or climate parameters (Järnström et al., 2007, Guo, 2011). Thus, an approach to combine both laboratory and real life measurements was chosen for the present study. Precisely defined user scenarios were performed in a model room simulating a real room in terms of size and configuration, but under controlled environmental conditions. It was the aim of this study to gain further knowledge about the effect of user behavior on indoor air quality. Therefore, four parameters of indoor air quality including VOC, carbonyl compounds, particulate matter, and CO_2 were investigated before, during and after each scenario.

Material and Methods

Six selected scenarios were performed in two model rooms. The construction of the model rooms according to CEN/TS 16516 is described in detail in (Höllbacher et al., 2015). The rooms had a volume of 30 m^3 and were equipped with interior fittings such as a carpet, curtains and furniture to resemble real rooms both in terms of size and configuration. The rooms were supplied with purified and conditioned air at an air exchange rate of 0.5 h^{-1}. Indoor climate parameters were set to 23 ± 2 °C and 50 ± 5 % relative humidity. Indoor air was well mixed by using a fan. The performed tests as well as the applied materials and the test procedures are listed in Table 1. Each test is marked with an individual identification code (T1-T6). All materials used for the tests were customary consumer or household products and purchased from retailers. The liquid fuel used for the ethanol fireplace was purchased together with the fireplace according to the recommendation given by the retailer.

Four common pollutants having an influence on indoor air quality were investigated for each user scenario: VOC, carbonyl compounds (including formaldehyde), PM and CO_2. PM and CO_2 were measured continuously. Sampling of VOC and carbonyl compounds was started simultaneously at the start of the test procedure for tests T2, T3, T4 and T6. For T1 and T5 the measurements were started immediately after performing the relevant user activities. This approach was chosen to keep the influence of the person performing the activities as small as possible.

Table 1: Test and material specifications as performed in the model room

ID	Test	Material	Applied quantities	Test procedure
T1	Cleaning agent	Liquid cleaning agent	25 ml Dilution in water: 1:10	10 min cleaning
T2	Electric air freshener	Scent oil in a glass flask Magnolia and cherry blossom scent Electric vaporizer Adjustable intensity	0.03 g scent oil	30 min operating time at highest intensity, left in the room deactivated
T3	Cigarette smoke	Filter tipped cigarette	2 pieces	Smoked within 30 min
T4	Peeling oranges	Oranges, pesticide-free	3 pieces	Peeling within 10 min, peels left in the room
T5	Cosmetics	Hair spray, Perfume Sprayed together	5 sprays each 0.795 g hair spray 0.185 g perfume	Sprayed within 2 min
T6	Ethanol fireplace	Decorative ethanol fireplace, Liquid fuel (96,6 % Ethanol, contains 2-Butanone)	100 ml liquid fuel	Extinguished manually after 30 min burning time, left in the room extinguished

VOC

Volatile organic compounds were sampled on desorption tubes filled with Tenax® TA using calibrated air sampling pumps (Chematec ApS, Roskilde, Sjælland, Denmark; Markes International Ltd., Llantrisant, Wales, UK) at a flow rate of 100 mL min^{-1}. VOC sampling was always performed in duplicate in the same order:

Empty – one hour sampling prior to the test

Sample 1 – 30 minutes sampling simultaneously with or immediately after the test

Sample 2 – 30 minutes sampling immediately after Sample 1

Sample 3 – 30 minutes sampling subsequent to Sample 2

Desorption tubes were analyzed according to ISO 16000-6 (DIN, 2011b) using a thermodesorption unit (TDAS 2000; Chromtech GmbH, Idstein, Hesse, Germany) linked to a gas chromatograph with mass spectrometric detection (7890A/5975C; Agilent Technologies, Santa Clara, California, USA) equipped with a HP-Pona column (Agilent Technologies; 50 m x 0.2 mm x 0.5 µm). Toluene-d8 and cyclodecane were used as internal standards. The detected substances were identified by comparison of the mass spectra to commercial spectral libraries (John Wiley & sons, Hoboken, New Jersey, USA) and quantified using authentic calibration standards (Sigma Aldrich Co. LLC, St.Louis, Missouri, USA). Substances which were not calibrated were quantified as equivalents of toluene-D8 and are marked with TE. The total VOC concentration (TVOC) was calculated for each sample as the sum of all identified substances with a concentration ≥ 1 µg m^{-3}.

Carbonyl compounds

Carbonyl compounds including formaldehyde were sampled on DNPH cartridges (Sigma Aldrich Co. LLC, St.Louis, Missouri, USA) according to ISO 16000-3 (DIN, 2011a) using a linear pump (Gardner Denver Inc., Milwaukee, Wisconsin, USA) at an air flow rate of 70 L h^{-1}. Carbonyl compound sampling was always performed in the same order:

Empty – one hour sampling prior to the test

Sample 1 – 30 minutes sampling simultaneously with or immediately after the test

Sample 2 – one hour sampling immediately after Sample 1

Loaded DNPH cartridges were analyzed according to ISO 16000-3 (DIN, 2011a). The cartridges were eluted against the loading direction using 5 mL acetonitrile. The hydrazones in the eluate were subsequently identified using HPLC-DAD (1200 series; Agilent Technologies, Santa Clara, California, USA) equipped with a SB - C18 column (3.5 µm, 4.6 x 250 mm, Agilent Technologies, Santa Clara, California, USA) and quantified using an external standard (CARB-Method 1004 DNPH mixture 2; Sigma Aldrich Co. LLC, St.Louis, Missouri, USA).

Particulate matter

PM concentration was determined continuously by an aerosol spectrometer (Model 1.108; Grimm Aerosol Technik GmbH & Co. KG, Ainring, Bavaria, Germany). The data acquisition covered 15 size ranges from 0.3 to 25 µm. The results for the classes PM10 (aerodynamic diameter ≤ 10 µm), PM2.5 (aerodynamic diameter ≤ 2.5 µm) and PM1.0 (aerodynamic diameter ≤ 1 µm) in µg m^{-3} were automatically calculated from the number of particles and the process-specific C-coefficient. The default value of the C-coefficient was 1. The aerosol spectrometer was placed on a table in the middle of the model room.

CO_2

CO_2 concentration was determined continuously according to VDI 4300/9 (VDI, 2005) with a multifunction analyzer (Mosway Electronics GmbH, Wald, Zurich, Switzerland) using non-dispersive infrared spectroscopy (NDIR). The CO_2 analyzer was placed in the middle of the model room.

Results and Discussion

Table 2: Measured values for TVOC, carbonyl compounds, PM10, and CO_2 for six (T1-T6) user scenarios. For TVOC and carbonyl compounds the concentrations in the empty model room prior to the tests as well as during/immediately after the test (sample 1) and in the following hour (sample 2 and 3) are shown. For PM10 and CO_2 minimum, maximum, and mean/median values over a period of 90 min starting with the test procedure are shown.

[a] No data due to technical problems

Parameter		T1	T2	T3	T4	T5	T6
TVOC [µg m^{-3}]	Empty	50	113	43	20	93	96
	Sample 1	1230	310	210	3547	478	121
	Sample 2	559	328	161	1853	253	547
	Sample 3	187	188	144	1636	212	301
Carbonyl compounds [µg m^{-3}]	Empty	97	97	12	97	97	12
	Sample 1	278	154	523	128	127	310
	Sample 2	167	247	221	142	114	689
PM10 [µg m^{-3}]	Min	0.04	-[a]	0.14	0.01	-[a]	0.14
	Max	73	-	378	7.1	-	12
	Mean	13	-	109	0.99	-	1.3
	Median	4.8	-	67	0.22	-	0.57
CO_2 [ppm]	Min	487	461	502	464	-[a]	497
	Max	655	496	694	588	-	1612
	Mean	579	485	605	521	-	1082

VOC

Table 3: Most abundant VOC identified during each test scenario. The share given for each substance is calculated from the TVOC at sample 1 minus the blank value measured in the empty model room.

T1	T2	T3	T4	T5	T6
Limonene (59%)	Linalool (36%)	Limonene (20%)	Limonene (94%)	Linalool (37%)	2-Butanone (90%)
3-Carene (22%)	Dipropylene gylcole methyl ether (16%)	Furfural (18%)	β-Myrcene (4%)	Linalyl acetate (21%)	
α-Pinene (7%)	Ethly 2-methylpentanoate (9%)	γ-Terpinene (14%)	α-Pinene (1%)	Limonene (15%)	
β-Pinene (5%)	Limonene (8%)	Toluene (6%)	Sabinene (1%)	γ-Terpinene (7%)	
γ-Terpinene (2%)	Benzyl acetate (7%)	Nicotine (6%)		3-Carene (3%)	
Dihydromyrcenol (2%)	1-(2-methoxy-1-methylethoxy)-2-propanol (7%)	Syrene (6%)		Seychellene (2%)	
p-Cymene (1%)	α-Terpineol (4%)	Benzene (4%)		Hexanal (2%)	
Hexanal (1%)	Hexanal (4%)	Benzofuran (4%)		Benzyl acetate (2%)	
Pentanal (1%)	3-Carene (3%)	Ethylbenzene (3%)		β-Ionone (2%)	
	α-Pinene (3%)	Acetophenone (2%)		α-Patchoulene (2%)	

The TVOC of all tests is shown in Table 2. Table 3 additionally gives the ten most abundant substances identified during each test and their relevant share of the TVOC. Each user scenario provoked an increase of TVOC. In most cases the maximum TVOC was reached at sample 1 which was taken simultaneously to or immediately after the test procedure. Exempted hereof are T2 and T6. For T2 the

TVOC of sample 1 and sample 2 were at a similar level, while sample 3 showed a moderate decline. T2 included the operation of an electric air freshener which was deactivated prior to the start of sample 2 but left in the room. Obviously, a considerable amount of scent oil was still dispensed by the warm vaporizer although the device had already been turned off. Overall, the operation of the air freshener led to increased emissions of nine substances of which linalool showed the highest concentration reaching a share of 25 % of the TVOC. For T6 a considerable increase of TVOC from sample 1 to sample 2 was observed. This increase was caused by strong emissions of 2-butanone which accounted for 90 % of the TVOC. The source of 2-butanone most likely was excess unburnt liquid fuel in the ethanol fireplace after it was extinguished (sample 2). This assumption was confirmed by an additional test carried out in the model room. During this test the fireplace was not extinguished but operated until the fuel was completely burnt. The results obtained in this additional test did not show such high emissions of 2-butanone. It is very likely that the emissions of 2-butanone were accompanied by high amounts of ethanol in the model room. However, the analytical methods chosen for this study did not allow the detection of ethanol. An increase of TVOC by the factor 4.9 was measured in the model room during smoking (T3). The most abundant substance was found to be limonene which is used as an additive in cigarettes to reduce unpleasant odor of sidestream smoke (Fowles and Noiton, 2001), followed by furfural which is naturally present in tobacco leaves and released during smoking (Rice, 1954).

The use of cosmetics (T5) also caused increased TVOC in indoor air. The maximum value was reached at sample 1, immediately after spraying of cosmetic products. Here, TVOC was more than 5 times higher than in the empty model room. Despite the subsequent decline of TVOC, the concentration at sample 3 was still more than twice as high as in the empty model room. Linalool and linalyl acetate were the most abundant substances. The by far strongest increase of TVOC was measured for peeling oranges (T4), followed by cleaning (T1). For T1 the TVOC increased by a factor of 24.5 after the application of the cleaning agent (sample 1). Subsequently, TVOC declined rapidly to a value of 187 µg m^{-3} one hour after the test procedure was performed. The most concentrated substances were limonene and 3-carene reaching a share of 59 % and 22 % of the maximum TVOC, respectively.

T4 led to an increase of the TVOC by a factor of 177 from the empty model room to the maximum value at sample 1. The TVOC declined during the following two measurements (sample 2 and 3) but remained at an elevated concentration due to the orange peels which were left in the model room. Limonene was the most concentrated substance accounting for 94 % of the TVOC at all samples.

The decline of VOC concentration observed in the tests might be attributed not only to the ventilation of the model rooms but also to sink effects inside the room. As mentioned above, the rooms were furnished like real rooms including textiles. It is known that especially textiles act as potent emission sinks (Jorgensen et al., 1999). The decay curve is slower for T2, T4 and T6 than for the other tests. This was probably due to the fact that the emission source (orange peel, air freshener, ethanol fireplace with unburnt fuel) remained inside the model room.

Carbonyl compounds

Table 4: Most abundant carbonyl compounds identified during each test scenario. The share given for each substance is calculated from the sum of carbonyl compounds calculated for sample 1 minus the blank value measured in the empty model room. Exempted hereof is T6 where the share was calculated from the sum of carbonyl compounds calculated for sample 2.

T1	T2	T3	T4	T5	T6
Acetone (52%)	Acetone (95%)	Acetaldehyde (35%)	Acetaldehyde (67%)	Acetaldehyde (57%)	Butanone (54%)
Hexanal (23%)	Crotonaldehyde (4%)	Acetone (32%)	Acetone (22%)	Acetone (24%)	Acetaldehyde (21%)
Pentanal (10%)		Formaldehyde (14%)	Formaldehyde (8%)	Formaldehyde (9%)	Methacrolein (11%)
Benzaldehyde (5%)		Butanone (5%)			Formaldehyde (5%)
Butanone (3%)		Crotonaldehyde (4%)			Acetone (3%)

The sum of all measured carbonyl compounds is shown in Table 2. Additionally, the five most abundant carbonyl compounds are shown in Table 4. The strongest increase of carbonyl compounds was measured during cigarette smoking (T3). The carbonyl compound concentration was clearly increased compared to the empty model room. High amounts of acetaldehyde, acetone and formaldehyde were emitted. These substances are known components of cigarette smoke (Izu et al., 2014). After the test was finished still an elevated concentration of all three substances could be measured in the indoor air of the model room (sample 2 and 3).

After burning of liquid fuel in an ethanol fireplace 2-butanone and acetaldehyde were the most concentrated carbonyl compounds in indoor air. During the burning phase increased emissions of formaldehyde (62 µg m^{-3}) and acetaldehyde (108 µg m^{-3}) could be measured. The formation of these two substances is typical for burning ethanol (Tanner et al., 1988, Correa et al., 2003). Increased formaldehyde concentration during the use of an ethanol fireplace was already stated by Schripp et al. (2014). After the fire had been extinguished 2-butanone was the highest concentrated substance due to the reasons already explained above. Additionally, significantly elevated emissions of acetaldehyde were identified. These emissions can most likely be attributed to the incomplete combustion of the liquid fuel (Correa et al., 2003).

The other tests showed a lower increase of carbonyl compounds compared to T3 and T6. For T1 the most abundant substances were acetone, hexanal and pentanal. T2 led to a rising acetone concentration. For T4 and T5 the most abundant carbonyl compound was acetaldehyde.

Particulate matter

Particulate matter is of concern due to its negative effects on human health. Inhalable particles with an aerodynamical diameter below 2.5 µm may penetrate the thoracic region of the respiratory system and cause severe respiratory and cardiovascular diseases and even lung cancer (WHO, 2013). Slightly elevated PM concentrations were determined for all tests. Low concentrations like those for T4 and T6 were most likely caused by the presence of a person in the room who dispersed dust by walking (Tian et al., 2014). This assumption is supported by the fact that the maximum values were measured at the beginning of the measurement period when someone was present in the model room to perform the test procedure. The same applies for T1 where dust was dispersed by cleaning activities. Although cleaning did not directly contribute to the formation of PM, it can be stated that due to an elevated concentration it affected indoor air quality with regard to PM.

The by far highest PM concentration was measured while cigarette smoking (T3). Environmental tobacco smoke is a known source of particulate matter in indoor air (Martin et al., 1997, Sahu et al., 2013). The results are displayed in more detail in Figure 1. A very high share of the measured PM is classified as PM1. This result is comparable to the findings of other studies (Sahu et al., 2013, Invernizzi et al., 2004). The maximum value of PM is reached in the middle of the test procedure, approximately 15 min after the test was started. It amounts to 378 µg m^{-3} PM10, 374 µg m^{-3} PM2.5 and 342 µg m^{-3} PM1 compared to concentrations of 0.14 µg m^{-3} PM2.5 and 0.11 µg m^{-3} PM1 prior to smoking.

The rapid decrease of the PM concentration after the cigarette was finished can be attributed to both the discharge from the room through the ventilation system and sedimentation within the room. Comparable concentrations and decay curves of PM have been identified by other authors (Invernizzi et al., 2004).

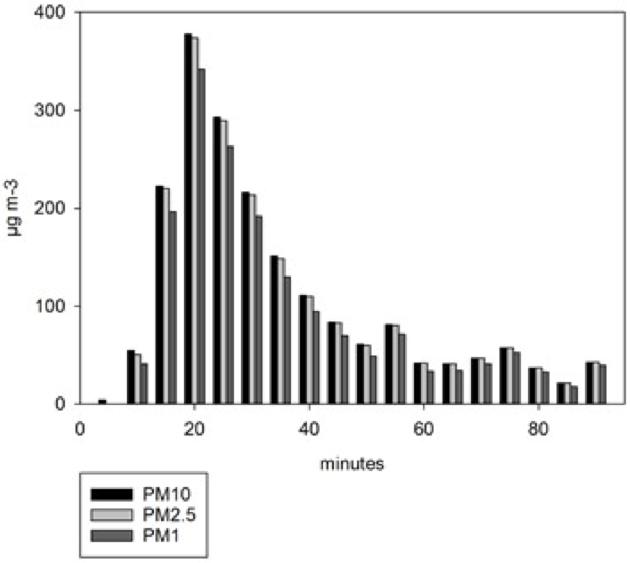

Figure 1: PM concentration of the classes PM10, PM2.5 and PM 1 during and after T3. 90 min of continuous measurement are shown. The test was performed between minute 10 and 40.

CO_2

T1 and T4 caused an increase of CO_2 in the indoor air of the model room which was mainly caused by the exhalation of the person in the room who was performing the test procedures. A similar CO_2 level was measured for T3. The maximum value was measured during the smoking process and is therefore most likely caused by it. The highest CO_2 level was measured during the use of an ethanol fireplace in the model room. Here, a maximum value of 1612 ppm CO_2 was determined in indoor air. As the ethanol fireplace used in the test scenario was non-vented, elevated concentrations of CO_2 were to be expected (Schripp et al., 2014). However, a clear negative effect on indoor air quality can be observed as all CO_2 concentrations above 1000 ppm are considered low indoor air quality in Europe (DIN, 2007).

Conclusion

In the course of this study, six user scenarios supposed to influence indoor air quality were performed within the controllable environment of a model room. For determining indoor air quality, four parameters were investigated. It can be concluded from the obtained results that user activities influence indoor air quality in various ways. For each measured parameter, well-defined maximum values can be assigned to one or more individual user scenarios. In most cases, the values clearly exceed those in the empty room. However, tests also indicate that changes in indoor air quality depend on the user behavior. Not every action changes all four parameters. In fact, some parameters show distinct changes, whereas others remain on a level comparable to those in the empty room. Moreover, the results clearly show that, individual actions result in a strong increase of emissions of indoor pollutants, but these emissions subsequently decrease rapidly. However, one must consider that the user scenarios were examined under laboratory conditions while possible interactions with further user activities or external influences were not evaluated. However, the chosen test set-up would allow for non-interacting as well as interacting investigation of emissions and emission scenarios in a standardized real room with the possibility to control environmental effects. The fast decay of pollutants like VOC or PM in the air of the model room after the investigated user scenarios suggests that air exchange is not the sole driving force but that there are various modes of interaction between, for example, the furniture and the indoor air which could also be evaluated in future studies with this test set-up.

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Publications

Publications	Höllbacher, E., Rieder-Gradinger, C., Stratev, D., Srebotnik, E. (2014): Measuring VOC emissions from wood-based building products under real room conditions in idealised model rooms. *In: Proceedings of the International Panel Products Symposium 2013 (IPPS 2013)*, Llandudno, Wales, UK, 9-10 October 2013
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	Indoor Air 2014 – 13th International Conference on Indoor Air Quality and Climate,
	7.-12.Juli 2014, Hong Kong, China
	PTF BPI 2014 – The 3rd International Conference on Processing Technologies for the Forest and Biobased Products Industry 2014, 24.-26.September 2014, Kuchl, Österreich
Lectures	Annual meeting of the Ecology Committee, Fachverband der Holzindustrie, Vienna, 2012
	Umdasch Forschungsdialog, Vienna, 2013
	Annual meeting of the Ecology Committee, Fachverband der Holzindustrie, Vienna, 2013
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	Meeting Working Group Construction, CEI-Bois, Brussels, 2015