

# Emissions of volatile organic compounds from wood and wood-based materials

Katja Butter, Martin Ohlmeyer

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# Emissions of volatile organic compounds from wood and wood-based materials

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Thünen Report 86a

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## Foreword

This review is part of the literature study *“Emissions from wood and wood products and their significance for health”* (DE: *“Emissionen aus Holz und Holzprodukten und ihre gesundheitliche Bedeutung“*), which was produced as part of the joint project *“Health assessment of emissions from wood and wood products in indoor spaces by means of experimental toxicological studies”* (DE: *“Gesundheitliche Bewertung von Emissionen aus Holz und Holzprodukten in Innenräumen mittels experimenteller toxikologischer Untersuchungen“*). This project was funded by the Federal Ministry of Food and Agriculture (BMEL) via the Agency for Renewable Resources (FNR). The literature study sheds light on possible adverse and also health-promoting effects of typical wood emissions. The composition of the emissions and the exposure concentrations in wooden houses are presented. Regulatory aspects for the assessment of building product emissions and indoor pollution are also discussed. A further point presented in this report deals with the characterisation of the emission behaviour of wood and wood products. Of particular interest here is the quantitative and qualitative composition of emissions from individual types of wood and wood-based materials. It also looks at the factors that influence this emission behaviour.

The final report on the joint project, including the complete literature study, can be downloaded via the following link:

<https://www.fnr-server.de/ftp/pdf/berichte/22011015.pdf>

### Special remarks for the English version

The present Thünen Report No. 86 was published in German in June 2021. This English version is a direct translation of the German original. No adjustments or updates have been made to the content to reflect the current state of scientific literature. The report therefore reflects the state of knowledge as of the aforementioned publication date.

## Zusammenfassung

Verschiedene Hölzer und Holzwerkstoffe werden aufgrund zahlreicher vorteilhafter Eigenschaften als Bauprodukt in Gebäuden verwendet. Als potenzielle Emissionsquelle können sie damit, wie auch andere im Innenraum verbaute Bauprodukte, eine Auswirkung auf die Innenraumluftqualität haben. Aus diesem Grund ist ein Verständnis der Emissionen holzhaltiger Produkte von Bedeutung. Daher wird in folgender Abhandlung das Emissionsverhalten von Vollhölzern sowie daraus hergestellten Holzwerkstoffen dargestellt. Dabei werden die wesentlichen flüchtigen organischen Verbindungen (VOC) und deren flächenspezifischen Emissionsraten aufgezeigt sowie die das Emissionsverhalten beeinflussenden Faktoren erörtert. Zudem wird die zeitliche Dynamik der Emissionen beleuchtet, die zur Folge hat, dass die Emissionsraten immer nur eine Momentaufnahme zu einem bestimmten Messzeitpunkt darstellen.

Eine vergleichende Betrachtung der einzelnen Holzprodukte bezüglich ihres Emissionsverhaltens wird damit ermöglicht. Hingegen erlaubt dies nicht zwingend, den tatsächlichen Beitrag der Holzproduktemissionen auf die VOC-Konzentrationen im Innenraum zu beurteilen. Dies wird entscheidend von der realen Einbausituation des Produktes sowie den Materialinteraktionen und klimatischen Gegebenheiten innerhalb des Raumes bedingt.

Schlagwörter: flüchtige organische Verbindungen (VOC), Emissionsraten, Vollholz, Holzwerkstoffe

## Abstract

Various woods and wood-based materials are used as building products in constructions due to their numerous beneficial properties. As a potential source of emissions, they can thus have an impact on indoor air quality, such as other building products used indoors. For this reason, an understanding of the emissions of wood-containing products is important. Therefore, the following document describes the emission behaviour of solid wood and wood-based materials made from it. The main volatile organic compounds (VOCs) and their area-specific emission rates are shown and the factors influencing the emission behaviour are discussed. In addition, the temporal dynamics of the emissions are highlighted, which means that the emission rates always only represent a snapshot at a specific point in time.

This makes it possible to compare the emission behaviour of the individual wood products. However, this does not necessarily allow the actual contribution of wood product emissions to indoor VOC concentrations to be assessed. This is decisively conditioned by the real installation situation of the product as well as the material interactions and climatic conditions within the room.

Keywords: volatile organic compounds (VOC), emission rates, solid wood, engineered wood

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## 1 Introduction

According to a guideline issued by the Association of German Engineers (VDI), health-compatible indoor air quality is given,

*"if indoor air and indoor climate make occupants feel pleasant and comfortable, if no negative odours are perceived and all physical, chemical and biological measurands lie within the ranges defined in this guideline" (VDI 6022-Part 3, 2011).*

In addition to measurable criteria (e.g. temperature, humidity, pollution), psychological factors also influence how the indoor air is perceived by residents. Accordingly, the assessment of indoor air quality always includes a subjective, individually varying component. The need to ensure a certain indoor air quality arises from the endeavour to protect the health and well-being of the population. In view of the fact that people in Germany spend a large proportion of their time indoors, indoor air quality requires special focus. A study by JANTUNEN et al. (1999), for example, found that the inhabitants of six European cities spend on average only one hour per day outdoors. As already noted by SALTHAMMER (2011), it is important to bear in mind that daily routines differ greatly between individuals, as do the locations they occupy and the time spent in them. The quality of indoor air needs to be examined in particular with regard to the elderly, ill individuals and children, as they are more vulnerable due to their age or health status. In addition, these groups are also the ones who spend a large amount of their time at home. According to BRASCHE & BISCHOF (2005), preschool children and elderly individuals spend 17.6 and 19.5 hours per day, respectively, well above the average of 15.7 hours. Taking into account all indoor spaces, including those outside the home, the German Environmental Survey for Children conducted between 2003 and 2006 found that 3 to 14-year-old children spend on average 22 hours indoors on a working day in winter (SCHULZ et al., 2007b). Consequently, the quality of indoor air in both private and public spaces needs to be analysed.

In addition to the two main components nitrogen and oxygen, natural air contains a number of other gases that occur in very low concentrations. Indoors, the chemical composition is influenced by emissions of organic and inorganic substances. Initially, however, the carbon dioxide content of the air was primarily considered as an indicator of indoor air quality within the research community. Among the organic substances, formaldehyde first came into focus after increased concentrations were detected indoors (SALTHAMMER, 2011). Wood-based materials, particularly particleboards, were identified as a source of emissions (alongside construction foams), with formaldehyde primarily originating from the adhesives (HORN et al., 2007; SALTHAMMER, 2011). As early as 1977, the German Federal Health Office recommended an indoor guide value of 0.1 ppm for this aldehyde. In the following decades, other volatile organic compounds also attracted attention, with the World Health Organisation (WHO) finally defining the individual substance classes according to their boiling points as very volatile<sup>1</sup>, volatile<sup>2</sup> and semi volatile<sup>3</sup> organic compounds in 1987 (SALTHAMMER, 2011). Human exposure to these substances can result in impairment due to an odour nuisance or harmful effects on health (SEIFERT, 1999). On the other hand, individual substances can also have a positive effect on well-being and health (MERCIER, PROST & PROST 2009). The decisive factors are the concentration levels of the substances in the air and the duration of exposure for the inhabitants. As a wide range of different emission sources interact indoors, from the occupants themselves to building materials, furniture, household items, etc. (ROßKAMP, 2012), the assessment of the resulting mixture of substances is correspondingly complex.

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<sup>1</sup> VVOC (very volatile organic compounds) – boiling point in the range of < 0 °C to 50 - 100 °C

<sup>2</sup> VOC (volatile organic compounds) – boiling point in the range of 50 - 100 °C to 240 - 260 °C

<sup>3</sup> SVOC (semi volatile organic compounds) – boiling point in the range of 240 - 260 °C to 380 - 400 °C

Wood and wood-based materials used in furnishings or as a construction product can affect indoor air. According to the European Construction Products Regulation (EU CPR, No. 305/2011), the term “*construction product*” refers to any product or kit,

*“which is produced and placed on the market for incorporation in a permanent manner in construction works or parts thereof and the performance of which has an effect on the performance of the construction works with respect to the basic requirements for construction works”.*

Building products are therefore of particular importance as a source of emissions in indoor spaces, as they are installed in the building over the long term and present a relatively large, potentially emitting surface area. In contrast to short-term emission peaks caused by human activities, which often subside quickly (HÖLLBACHER, 2016), they can have a continuous impact on indoor air quality (MØLHAVE & NIELSEN, 1992). In this context, the EU CPR (No. 305/2011) explicitly states that one of the basic requirements for buildings is to ensure that the health and safety of the occupants are not compromised during the entire lifecycle of the building. In order to assess the influence of wood and wood-based materials on indoor air quality, it is therefore necessary to outline their range of applications as construction products.

The total construction volume<sup>4</sup> in the building sector (€243 billion in residential construction, €109 billion in non-residential construction<sup>5</sup>) in Germany in 2019 was largely divided between modernisation (including refurbishment and conversion measures) as well as maintenance and repair work. Although the volume of new residential construction has increased significantly since 2009, when it reached its lowest level since reunification, it accounted only for 31 % of the construction volume in 2019 (40 % in non-residential construction). Energy refurbishment measures<sup>6</sup> in particular make up a significant proportion of construction work on existing buildings – around 27 % of the total construction volume in the building stock falls within this category (BBSR, 2020). The most recent study on the use of wood in construction by MANTAU, DÖRING & HILLER (2013) refers to the year 2012. 13.4 million m<sup>3</sup> (b)<sup>7</sup> of wood was used in construction at that time, with 80 % being used in residential construction and 20 % in non-residential construction. 85.6 % of the wood used was softwood, 12.6 % was hardwood<sup>8</sup> and 1.8 % was tropical wood. Hardwood is primarily used for flooring, followed by thermal insulation and exterior applications. Its relevance in structural applications (exterior walls, roofs, façades) is still low – this sector continues to be dominated by softwood. The majority of the wood used is sawn timber (60 %). Wood-based materials account for 28 %, with wood fibre insulation boards making up over half of the volume. The main areas of application for wood products are in structural applications (33 %), thermal insulation (22 %), exteriors and flooring (12 % each). Wood and wood-based materials are mainly used in building renovation, modernisation and conversion work, i.e. for applications in existing buildings (MANTAU, DÖRING & HILLER, 2013). The remaining 36 % of the wood volume is used in the realisation of new construction projects. The proportion of new buildings in which wood is the predominant building material in the load-bearing structure is currently around 19 % (Table 1).

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<sup>4</sup> Sum of all activities aimed at the construction or maintenance of buildings and structures (BBSR, 2020: 6)

<sup>5</sup> Commercial and public construction

<sup>6</sup> Measures include thermal insulation (on the roof, façade, etc.), replacement of windows and exterior doors, renewal of heating system and installation of solar thermal or photovoltaics (BBSR, 2020: 28)

<sup>7</sup> m<sup>3</sup> (b) = cubic metres that a product occupies in the building

<sup>8</sup> Hardwood without tropical wood content

**Table 1: Development of the number of approved new construction projects**

	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
<b>Residential buildings</b>	89,509	94,602	112,698	109,128	113,291	111,610	120,771	125,157	119,060	117,869	119,457
of which in timber construction	13,032	14,666	17,074	16,545	17,039	16,823	19,295	20,219	21,018	20,947	22,284
<b>Non-residential buildings</b>	29,517	31,059	32,099	30,364	28,611	26,765	26,533	29,101	26,952	27,147	27,036
of which in timber construction	5,878	6,085	5,824	5,652	5,035	4,713	4,453	5,625	4,605	4,832	5,261

Source: DESTATIS (2020)

As a natural organic product, wood contains a variety of organic substances that can be emitted, depending on the type of wood. In addition, secondary emissions can occur as a result of reaction processes of the wood components. The emission behaviour of wood-based materials is also influenced by the adhesives and additives used as well as the manufacturing process. An increased influence on indoor air quality is mainly to be expected from wood products whose surface is in direct contact with the interior. This primarily concerns flooring, wall and ceiling panelling. However, if the surfaces are treated with varnishes, oils or waxes, the emission behaviour is different to that of untreated wood (YRIEIX, MAUPETIT & RAMALHO, 2004; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011; STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNIK, 2014). The influence of substructures made of wood and wood-based materials, as used in timber-frame construction and drywall applications, on the VOC content of indoor air is difficult to estimate. Cladding, for example with gypsum boards, as well as plaster or wallpaper applications, can act as emission barriers. Consequently, the actual VOC concentrations in indoor air cannot therefore be derived solely from the sum of the surface-specific emission rates (determined in accordance with ISO standard 16000) of the materials used. Studies on the emission behaviour of material combinations (DÄUMLING et al., 2009; SENITKOVA, 2014; WEIGL et al., 2014) and on the sorption and diffusion behaviour of building materials (NIEDERMAYER et al., 2013) can provide initial indications of the extent to which different materials and their emissions interact with each other. Ultimately, even more meaningful results can only be obtained through VOC measurements in real living spaces in which wood products have been installed. It should be borne in mind that due to the large number of construction methods, materials, climatic conditions (especially air change rate) and different user behaviour, it is difficult to make generally valid statements. Further information can be found, for example, in the research project by OHLMEYER, MENNICKE & POTH (2020). In this project, four model houses with different timber construction methods were built and their indoor air was monitored for more than a year after completion of construction.

## 2 Objective

Information on the qualitative and quantitative composition of volatile organic compounds in indoor spaces can be obtained from the German Environmental Surveys<sup>9</sup>, which, among other things, analyse the pollution levels of the German population. The standardised national indoor air guide values (Guide Values I and II) can be used to assess the VOC concentrations determined. These values are developed by the German Committee for Indoor Air Guide Values (AIR, formerly the Ad-hoc Working Group for Indoor Air Guide Values) at the Federal Environment Agency for a range of substances on the basis of toxicological and epidemiological knowledge of the effect threshold of each substance (AD-HOC-ARBEITSGRUPPE IRW, 2012). A key challenge regarding the VOC concentrations measured indoors is that the substances cannot be clearly assigned to the individual sources due to the large number of emission sources. In order to protect the health of consumers and improve the quality of living, efforts are being made to label building products – including wood and wood-based materials – with regard to their emission behaviour in the future.

This compilation of literature provides an overview of the emission characteristics of the main types of wood and wood-based materials used in the German construction sector. It presents the key emitted substances and their respective concentration ranges. The focus here is on the pure emission behaviour of the materials without taking into account the fact that various interactions with the surrounding air and other materials can take place in real interior spaces. The main factors influencing the emission behaviour of the individual materials are also outlined. The data makes it possible to compare wood products with each other and with other material groups in terms of their emission behaviour. However, the emission data can only be evaluated in conjunction with the health assessment of the individual emitted substances, as provided by human and animal studies. In addition, studies investigating mixtures of substances must also be considered in order to assess whether such mixtures behave differently from the sum of the individual substance effects ( $\text{effect}_{\text{MIX}} = \text{effect}_A + \text{effect}_B + \dots$ ). Synergistic ( $\text{effect}_{\text{mix}} > \text{effect}_A + \text{effect}_B + \dots$ ) and antagonistic ( $\text{effect}_{\text{mix}} < \text{effect}_A + \text{effect}_B + \dots$ ) interactions are also conceivable (ECA, 1997b). A purely quantitative assessment of the total volatile organic compounds (TVOC<sup>10</sup>) is therefore of little relevance, as it does not allow any conclusion to be drawn about whether a product has an adverse effect on health.

The emission data can provide insights into the potential impact of wood products on indoor air quality. However, they do not indicate which VOC concentrations will actually occur when the products are used in real indoor environments. In addition to the loading factor, the air change rate and the climatic conditions, this depends primarily on how the product is incorporated into the space – whether as a directly emitting surface or concealed within the wall structure. This raises the question of how the different materials interact, which, due to the wide range of materials used and their differing properties, has so far been difficult to answer adequately.

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<sup>9</sup> Survey periods: 1985-86, 1990-92, 1997-99, 2003-06, 2014-17 (SCHULZ et al., 2007a; UBA, 2017)

<sup>10</sup> Sum of volatile organic compounds, sampled on Tenax® TA, which elute between and including n-hexane and n-hexadecane on a capillary column of a gas chromatograph (DIN ISO 16000-6:2012)

### 3 Material and methods

As part of the literature review, relevant studies on the emission behaviour of wood and wood-based materials are evaluated. The literature considered includes peer-reviewed publications and research reports from universities and authorities (up to and including 2018). Further data on the emission behaviour of other materials and products can also be taken from corresponding databases (Figure 1). Databases of certification systems for building products are not included, as they only cover information on low-emitting products.

**Figure 1: Further databases with emission data on various materials**

With access:

- France PANDORA database: <http://lasie.univ-larochelle.fr/PANDORA-A-compilation-of-iNDoOR>  
(Download: **PANDORA** software)
- Canada National Research Council (CNRC) materials emission database:  
<https://nrc.canada.ca/en/research-development/products-services/software-applications/ia-quest-indoor-air-quality-emission-simulation-tool>  
(Download: **IA-QUEST** software)
- National Institute of Standards and Technology (NIST) CONTAM software:  
<https://www.nist.gov/el/energy-and-environment-division-73200/nist-multizone-modeling/software/contam> (Download: **CONTAM** software)
- United States Environmental Protection Agency (EPA) Source Ranking Database (SRD) for Indoor Air Pollutants:  
<https://www.epa.gov/tsca-screening-tools/forms/srd-source-ranking-database-indoor-air-pollutants-download-and-install>

No access:

- EPA Source of Indoor Air Emissions (SIAE) database
- European Union Building Materials (BUMA) database
- Sources of Pollution for a Healthy and comfortable Indoor Environment (SOPHIE) database

Source: WILLEM & SINGER (2010), ABADIE & BLONDEAU (2011)

For wood species and wood-based materials, the focus is limited to those that are mainly used in the construction sector in Germany. For solid wood, the main emphasis is therefore on Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* (L.) H. KARST.) and European beech (*Fagus sylvatica* L.). Emission data from other native hardwood and softwood species are also recorded as a secondary priority, provided that publications are available. Particular attention is paid to pine, as it is one of the higher emitting wood species and is therefore likely to have a greater impact on indoor air quality. Among wood-based materials, OSB (*oriented strand board*) takes this place due to its aldehyde emissions. In the entire building construction sector, however, fibreboards (LDF, MDF, HDF<sup>11</sup>) are used to a greater extent in terms of quantity. The emission behaviour of particleboard and plywood (including LVL<sup>12</sup>), whose use in the construction sector is comparatively low (MANTAU, DÖRING & HILLER, 2013), is also being investigated. In the individual studies on the emission behaviour of wood products, the full species name (e.g. *Pinus sylvestris*) was not always given; sometimes only the genus (e.g. *Pinus*) was

<sup>11</sup> LDF = low density fibreboard or wood fibre insulation board, MDF = medium density fibreboard, HDF = high density fibreboard

<sup>12</sup> LVL = laminated veneer lumber

indicated. In European studies, it is assumed that pine, spruce and larch refer to Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* (L.) H. KARST.) and European larch (*Larix decidua* MILL.), respectively. This assumption cannot be made for non-European studies; in such cases, appropriate clarifications are provided within the text.

The entire spectrum of emissions from a material comprises a large number of individual substances. In some studies, the emission data of all individual substances are listed, in others they are summarised into compound groups or in the form of TVOC. In order to illustrate the emission characteristics of the individual wood species and wood-based materials, it was decided to analyse only a selection of individual substances in more detail. Substances were considered either if they make up a high proportion of the total emissions of a material or if they are relevant due to their potential adverse health effects. The latter includes, in particular, VOCs that have a low EU-LCI value (*lowest concentration of interest*) (Table 2). LCI values are derived on the basis of epidemiological and toxicological data and serve as a reference concentration for assessing the emissions of a building product following 28 days of emission testing. This approach is intended to ensure that long-term exposure to certain VOCs or products does not pose a health risk to the population (ECA 2013).

**Table 2: Selection of VOCs with EU-LCI values**

Substance	EU-LCI ( $\mu\text{g m}^{-3}$ )
$\alpha$ -Pinene	2,500
3-Carene	1,500
$\beta$ -Pinene	1,400
Limonene	5,000
Acetic acid	1,200
Hexanoic acid	2,100
Hexanal	900
Pentanal	800
2-Octenal	7
2-Heptenal	7
Furfural	10

Source: EU-LCI WORKING GROUP (2020)

In addition, TVOC values are reported to compare the overall emissions of materials. It should be borne in mind that such a comparison is generally only appropriate for similar products or materials. The potential harmful effects of compounds vary over a wide range of concentrations (Table 2). For some substances, even very low concentrations are sufficient to have an adverse effect on the organism, while others are completely harmless even in large quantities. As a result, the concentrations of individual relevant VOCs are more meaningful than the sum of all VOCs, meaning that the TVOC is not a sufficiently reliable indicator with regard to the health assessment of different materials (WOOLFENDEN, 2009). The emitted VOCs of a material are stated in the publications either as the concentration in the test chamber or test cell air (C, typically in  $\mu\text{g m}^{-3}$ ) or as area-specific emission rates (SER, typically in  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). To enable the comparison of the emission data from different studies, the air concentrations are converted into area-specific emission factors. This requires that the relevant studies provide information on the product loading factor (l) and the air change rate (n), or alternatively on the area-specific air flow rate (q) (Formula 1).

**Formula 1: Relationship between the area-specific emission rate of VOC emitted from a material and the VOC concentration in the test chamber or test cell air**

$$\text{SER } (\mu\text{g m}^{-2} \text{ h}^{-1}) = C (\mu\text{g m}^{-3}) * q (\text{m}^3 \text{ m}^{-2} \text{ h}^{-1})$$

$$q (\text{m}^3 \text{ m}^{-2} \text{ h}^{-1}) = \frac{n (\text{h}^{-1})}{l (\text{m}^2 \text{ m}^{-3})}$$

Both emission data in numerical form and those presented graphically are taken into account, whereby only rough estimates of the emission values can be made for the latter. The temporal variation of a material's emissions is captured by reporting the 3-day and 28-day values in accordance with DIN EN ISO 16000-9:2008 and 16000-10:2006. The 3-day value is intended to reflect the short-term and the 28-day value the long-term behaviour of a product. However, due to different objectives in the individual studies, the sampling times of the air measurements in the test chambers or cells differ in some cases, ranging from a single measurement to several measurements spread over several months. Accordingly, the assumptions listed in Table 3 are made in order to standardise the results.

**Table 3: Criteria for the allocation of area-specific emission rates to various sampling time points**

	Sampling time
<b>Short-term (TVOC and individual compounds)</b>	on the 3 <sup>rd</sup> day after the start of the emissions test → if not specified, then the value from the closest measurement day (min: 0 <sup>th</sup> day, max: 7 <sup>th</sup> day)
<b>Long-term (TVOC and individual compounds)</b>	on the 28 <sup>th</sup> day after the start of the emissions test → if not specified, then the value from the closest measurement day (min: 14 <sup>th</sup> day, max: 40 <sup>th</sup> day)

Source: own representation

### 3.1 Comparability of the results of different studies

Characterizing the emission behaviour of individual wood species is partly challenging due to the limited number of available studies. In addition, emission measurements are relatively time-consuming, meaning that data is often available with small sample sizes (usually single measurements without repetitions). In contrast to the formaldehyde measurement routinely used in the wood-based materials sector, VOC measurement is much more demanding. A number of substances must be identified and accurately quantified, often at very low concentrations in the  $\mu\text{g m}^{-3}$  range (HAGUE et al., 2009). When considering different studies, it should be noted that each study has different objectives and therefore the methodology may differ (Table 4, Table 5). In this context, ENGLUND (1999) points out that emission measurements do not provide any information about the VOC concentrations present in the wood itself, but only about the VOC quantities emitted under given conditions. As the emission rates of VOCs are dependent on a range of test conditions (temperature, relative humidity, air change rate, product loading factor, air velocity), the comparability of the results across studies is limited. Rather, this only allows a comparative analysis of different materials within a single study.

The determination of emissions from building products is a relatively complex, multi-stage process, which can therefore lead to deviating results for individual testing laboratories. Random and systematic errors must be taken into account for each individual step of the emission measurement. This includes the sample preparation, the test chamber or test cell examination with the air sampling and the subsequent analysis (MAKOWSKI, 2007). Inter-laboratory tests offer the opportunity to ensure the quality of the measurement methods and testing laboratories. The Federal Institute for Materials Research and Testing (BAM) has carried out several round-robin tests in which numerous laboratories throughout Europe have participated. Emission measurements of the same

reference material (acrylic sealant with seven main substances) showed that all participants (29 European testing institutes) achieved deviations of less than 20 % for four substances – higher deviations occurred for substances present at very low concentrations or for highly polar substances (WILKE et al., 2009). YRIEIX et al. (2010) carried out a round-robin test (6 participants) with a particleboard made of maritime pine wood. Due to material inhomogeneity, relative standard deviations of around 20 % for  $\alpha$ -pinene already existed within the institute performing the test. However, the inter-laboratory deviation was greater than the intra-laboratory deviation and ranged from 28 to 46 %. The authors concluded from the results of their round-robin test and those of BAM that a relative standard deviation of 20 % can be expected for homogeneous materials and around 40 % for heterogeneous materials and low VOC concentrations (below  $10 \mu\text{g m}^{-3}$ ). These measurement inaccuracies should therefore be taken into account in all results and assessments of building products (WOOLFENDEN, 2009).

### 3.1.1 Sample preparation

The emission behaviour of solid wood and wood-based materials is influenced by various factors. These arise, on the one hand, from the material itself (wood species, type and duration of drying, moisture content, manufacturing processes and composition of the wood-based materials, etc.). On the other hand, aspects related to sampling and sample preparation (transport conditions, type and duration of storage, etc.) also have a significant impact. Some publications contain insufficient information in this regard, which makes it difficult to assess the study results. Particularly in the case of products that were not purchased directly from the manufacturer but, for example, from hardware stores, there is generally little knowledge of the manufacturing and storage conditions.

### 3.1.2 Test chamber/test cell examination

Priority will be given to publications that have determined emission data using the emission test chamber (DIN EN ISO 16000-9:2008) or emission test cell (DIN EN ISO 16000-10:2006) methods. Most of the studies refer to the relevant test standards or contain information on the test conditions, albeit incomplete in some cases. In particular, temperature, humidity, air velocity, air change rate and product loading factor are significant factors that can influence the emission behaviour of a material. The **temperature** has an influence on the vapour pressure and the diffusion coefficient of the compounds, whereby an increase in emission rates, but also a faster decay of emissions, can be expected with increasing temperature (ZELLWEGER et al., 1997; WOLKOFF, 1998; FECHTER, ENGLUND & LUNDIN, 2006; OHLMEYER & STECKEL, 2012). In addition, STECKEL (2011) demonstrated that elevated test temperatures accelerate the formation of secondary emissions (e.g. aldehyde emissions from pine wood). The effect of **humidity** depends on the substance, whereby it is assumed that increasing humidity leads to higher emissions of polar substances, whereas it has no significant influence on non-polar substances (FECHTER, ENGLUND & LUNDIN, 2006; STECKEL, 2011). WILKE, SCHULZ & RICHTER (2012) showed that despite the variations in **air change rate** and **product loading factor** in  $1 \text{ m}^3$  emission chambers, comparable area-specific emission rates of a material (wooden board coated with polyurethane lacquer) result. The relative standard deviation of the five substances analysed was less than 6 % in two test series and a maximum of 24 % in the third test series. JANN, WILKE & BRÖDNER (1999) also determined in their experiments that there is an inverse proportionality between air change rate and air concentration for most of the substances analysed – however, this is not the case for all substances. STECKEL (2011) also observed differing effects of the test conditions. In pine sapwood, both the aldehyde and terpene emissions showed a direct or indirect proportionality to the loading factor and air change rate, respectively. OHLMEYER & STECKEL (2012) found the same behaviour in OSB. However, these proportional relationships were not found for heartwood, which STECKEL (2011) attributed to the fact that the emissions of the heartwood are possibly essentially controlled by evaporation, whereas the emissions of the sapwood are mainly diffusion-controlled. According to SALTHAMMER (2009), emission measurements in which emissions are primarily released by diffusion are less susceptible to changes in air

velocity and turbulence above the sample surface. MAKOWSKI (2007) demonstrated using OSB that the **size of the emission chamber** (1 m<sup>3</sup> chamber and 23 L glass desiccator) has no significant influence on the emission rates determined as long as the sample material is largely homogeneous. JANN, WILKE & BRÖDNER (1999) also determined comparable results for emission measurements of coated wood-based materials in 1 m<sup>3</sup> and 0.02 m<sup>3</sup> chambers and FLEC cells (field and laboratory emission cell). However, measurements in 1 m<sup>3</sup> and 20 m<sup>3</sup> chambers showed clear differences for individual compounds (lower concentration levels in the 20 m<sup>3</sup> chambers), which is attributed to the more pronounced wall effect in the larger chambers. In the studies by HÖLLBACHER et al. (2014), the determination of the emission behaviour in the emission chamber (0.225 m<sup>3</sup>), micro-chamber (48 mL) and FLEC cell (35 mL) led to different outcomes, whereby no general correlation could be established between the results for all materials. The choice of chamber size ultimately depends on the homogeneity of the material. With heterogeneous materials, test methods that only examine a small material surface (micro-chamber, FLEC cell) can lead to a misjudgement of the emission behaviour (SALTHAMMER & FURHMANN, 1996). Unless larger chambers with higher material loadings can be used, the number of samples must at least be increased in such cases.

### 3.1.3 Analytics

The **analytical method** for determining the VOCs in the test chamber or test cell air has a significant influence on the emission results. The VOCs from building products are usually adsorbed onto Tenax® TA<sup>13</sup>. After thermal desorption (TD), the mixture is then separated into the individual chemical compounds in a gas chromatograph (GC) and qualitatively and quantitatively determined in a mass spectrometer (MS) (if necessary, in combination with a flame ionisation detector) (DIN ISO 16000-6:2012). As Tenax® TA only has a low adsorption capacity for VVOCs (WIEGNER et al., 2012), the determination of very volatile and volatile aldehydes and ketones should be carried out in accordance with DIN ISO 16000-3:2013. In this method, the sample air is passed through a cartridge containing silica gel coated with DNPH (2,4-dinitrophenylhydrazine). The carbonyl compounds form stable derivatives with DNPH in the acidic medium, which are analysed using HPLC (high-performance liquid chromatography). Acetic acid, which is also classified as a VOC, cannot be detected with sufficient accuracy using Tenax® TA (RISHOLM-SUNDMAN et al., 1998; WIEGNER et al., 2012). Integration of the acetic acid peaks is challenging because they have a small area (response) and an asymmetrical shape (ideal form: Gaussian distribution). In addition, the calibration function is not linear, which increases the inaccuracy of the results (WIEGNER et al., 2012). Various other measurement methods for determining acetic acid are described in the literature (RISHOLM-SUNDMAN et al., 1998; OHLMEYER & STECKEL, 2012; WIEGNER et al., 2012) and in the VDI guideline VDI 4301-Part 7:2017.

**VOCs are quantified** using compound-specific response factors, which are determined by calibrating the analytical system with standard solutions. This approach provides the most reliable results. However, some studies employ a less labour-intensive method, in which all substances are evaluated using a single response factor, typically that of the internal standard<sup>14</sup>. Depending on the extent to which the individual response factors of the substances deviate from that of the internal standard, the two methods lead to significantly different results. YRIEIX et al. (2010) showed that the air concentrations of individual compounds are reduced by 6 % ( $\alpha$ -pinene) to 52 % (hexanal) when evaluated as toluene equivalents. This quantification method appears to be sufficient if, for example, decay rates of individual VOCs over a certain period of time are of interest. However, an exact determination of VOC concentrations is not possible with this method. Consequently, this also has an effect on the TVOC concentration – it can be determined either as the sum of all signals between n-hexane and

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<sup>13</sup> Porous, polymeric adsorber based on 2,6-diphenylene oxide

<sup>14</sup> Compound that is spiked in a consistent amount onto each sorbent tube prior to air sampling to control the performance of sampling and analysis (DIN ISO 16000-6:2012)

n-hexadecane in toluene equivalents, or as the sum of all identified and calibrated compounds, as well as non-identified substances expressed as toluene equivalents, in the range C<sub>6</sub> to C<sub>16</sub>. Depending on the composition of the emissions, the results of these two approaches can differ considerably (JANN, WILKE & BRÖDNER, 1999). WENSING (2017), for example, reported deviations of 2 % and 46 % for two product examples. HORN et al. (2007) found that for an OSB, over a measurement period of 1 to 81 days (five measurement points), the TVOC concentration determined as toluene equivalent accounted for only 46 to 88 % of the actual TVOC concentration. The reliability of the TVOC value as a comparative parameter for assessing different materials is further weakened by such analytical uncertainties. Apart from the analytical evaluation method, it is also challenging to compare different studies, as in some cases detailed tables with all detected individual substances are given, whereas others report only selected substances, substance classes or solely the TVOC.

### 3.2 Studies considered

The studies considered in this review are listed in Table 4. In addition to the analysed wood products, information on the test conditions of the emission measurements and the analytical methods used are also presented.

**Table 4: Studies considered on the emission behaviour of wood and wood-based materials**

Sources	Wood species and wood-based materials examined	Material	Testing Method	Test conditions	Testing time	Evaluation	VOC list	Unit	TVOC
BAUMANN, BATTERMAN & ZHANG (1999)	Particleboard and MDF from softwood and hardwood	•	K	•	○	•	•	SER	•
BAUMANN et al. (2000)	Particleboard and MDF from softwood and hardwood	•	K	•	○	•	•	SER	•
ČECH & TESAŘOVÁ (2015)	Poplar	•	K	•	•	○	○	C	•
CZAJKA & DZIEWANOWSKA-PUDLISZAK (2011)	Pine, oak, ash, alder, beech, larch	•	K	•	○	•	•	C	•
CZAJKIA & FABISIĄK (2012)	Pine	•	K	•	•	•	○	C	•
CZAJKIA & FABISIĄK (2013)	Spruce	•	K	•	•	•	○	C	•
CZAJKIA & FABISIĄK (2014)	Fir	•	K	•	•	○	○	C	•
DÄUMLING et al. (2009)	OSB	•	F	•	•	•	○	C	•
ENGLUND (1999)	Pine, spruce, beech, oak, birch	•	F	•	•	•	•	SER	•
ENGLUND (2010)	Pine, spruce, beech, oak, birch, ash, alder, poplar	d	F	d	•	d	○	SER	•
GACA & DZIEWANOWSKA-PUDLISZAK (2005)	Pine, oak, ash, alder, beech, larch, particleboard, plywood, particleboard with birch veneer	○	K	•	○	•	○	C	•
HASEGAWA et al. (2006)	Spruce	•	K	•	○	○	•	SER	•
HORN et al. (2007)	OSB, pine, particleboard, beech	○	K	•	•	•/•	•	C	•
HYTTINEN et al. (2010)	Pine, spruce, poplar	•	K	•	•	•	•	SER	•
KRANIOTIS et al. (2015)	Pine	•	F	•	○	○	○	SER	•
LARSEN et al. (1998)	Pine, spruce, birch, beech, birch plywood, oak parquet	•	K/F	•	•	•	•	SER	•
LARSEN, FROST & WINTHER FUNCH (2000)	Pine, spruce, beech, ash, particleboard from pine and spruce, particleboard with beech veneer, MDF and OSB from softwood, birch plywood	•/•	K	•	•	•	○ <sup>a</sup>	SER	•
MAKOWSKI (2007)	OSB	•	K	•	•	•/•	•	C	•
MANNINEN, PASANEN & HOLOPAINEN (2002)	Pine	•	K	•	○	○	•	%	○

Sources	Wood species and wood-based materials examined	Material	Testing Method	Test conditions	Testing time	Evaluation	VOC list	Unit	TVOC
OHLMEYER et al. (2008a)	OSB	●	K	●	●	●	●	C	●
OHLMEYER & STECKEL (2012)	Pine, beech, OSB	●/●	K	●	●	●	○	C	●
QUE et al. (2013)	Hardboard, plywood, MDF, pine, particleboard	●	S	●	○	●	○	C/SER <sup>c</sup>	●
RISHOLM-SUNDMAN (2002)	Particleboard, MDF, OSB, plywood, hardboard	●	F	●	●	○	○	SER	●
RISHOLM-SUNDMAN et al. (1998)	Ash, beech, sugar maple, oak, black cherry, spruce, pine, rubberwood	●	F	○	○	●	○	SER	●
SAARELA et al. (1994) in ECA (1997a)	Spruce, pine	a	a	a	●	a	●	SER	●
SALTHAMMER et al. (2003)	OSB	●	K	●	●	○	●	C	○/●
STACHOWIAK-WENCEK, PRĄDZYŃSKI & KRZYWOŚIŃSKA (2011)	Particleboard	●	K	●	○	●	●	C	●
STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATENIKO-NOŻEWNIK (2014)	Pine, beech, oak	●	K	●	○	●	●	C	●
STACHOWIAK-WENCEK et al. (2015)	Pine	●	K	●	●	●	●	C	●
STECKEL (2011)	Pine, spruce	●	K	●	●	●	●	C	●
STECKEL, WELLING & OHLMEYER (2010, 2013)	Spruce	●	K	●	●	●	●	C	●
TURTOLA et al. (2002)	Pine	●	K	●	○	○	●	SER <sup>b</sup>	○
VOLKMER et al. (2014)	Oak	●	K	●	●	○	○	C	●
WILKE et al. (2012)	Pine, OSB, spruce	●/●	K	●	●	●	●	C	●
YRIEIX, MAUPETIT & RAMALHO (2004)	Maritime pine, oak, pine, particleboard, plywood	●	K	●	●	●	○	C	●

<sup>a</sup> The study is not fully available, so not all parameters can be assessed.

<sup>b</sup>  $\mu\text{g kg}^{-1} \text{h}^{-1}$

<sup>c</sup> Area-specific air flow rate and area-specific emission rates were recalculated due to calculation errors within the study.

<sup>d</sup> Unknown, as the publication is in Swedish.

Source: own representation according to the given sources

**Explanations of the criteria shown in Table 4:**

Material	Criteria for solid wood:	
	<ul style="list-style-type: none"> <li>- Species name (i.e. scientific name, e.g. <i>Pinus sylvestris</i>)</li> <li>- Wood tissue (heartwood and/or sapwood)</li> <li>- Information on the storage of the wood</li> <li>- Moisture content of the wood</li> </ul>	
	Criteria for wood-based materials:	
	<ul style="list-style-type: none"> <li>- Species name (i.e. scientific name) of the wood used</li> <li>- Production conditions</li> <li>- Information on the storage of the wood-based material</li> <li>- Type of binder</li> </ul>	
	●	at least 3 out of 4 criteria specified
	◐	2 of 4 criteria specified
	○	only specification of the wood species or wood-based material type
Testing method	K	emission test chamber
	F	emission test cell
	S	other
Test conditions	●	complete information, i.e.: <ul style="list-style-type: none"> <li>- specification of temperature, relative humidity and surface-specific air flow rate (or air change rate and product loading factor) <b>or</b></li> <li>- reference to DIN EN ISO 16000-9:2008 or DIN EN ISO 16000-10:2006, along with the specification of the area-specific air flow rate (or air change rate and product loading factor)</li> </ul>
	◐	incomplete information
	○	not specified
Testing time	●	> 30 days
	◐	3 < x ≤ 30 days
	○	≤ 3 days
Evaluation	●	quantification using compound-specific response factors (unknown VOC: quantification as equivalent of the internal standard)
	◐	quantification as the equivalent of the internal standard
	○	deviating from ● and ◐, or no information provided
VOC list	●	almost complete listing of individual compounds
	◐	partial listing of individual compounds
	○	no detailed listing of individual compounds
Unit	C	air concentration (typically in $\mu\text{g m}^{-3}$ )
	SER	area-specific emission rate (typically in $\mu\text{g m}^{-2} \text{h}^{-1}$ )
	%	percentage
TVOC	●	reported
	◐	calculated from data on individual compounds or estimated from a graph
	○	not reported

Source: own representation

**Table 5: Objectives of the analyses in the individual studies**

Sources	Wood species and wood-based materials examined	Objective of the study
BAUMANN, BATTERMAN & ZHANG (1999)	Particleboard and MDF made from softwood and hardwood	- Differences due to wood species and material type
BAUMANN et al. (2000)	Particleboard and MDF made from coniferous and deciduous woods	- Differences due to wood species and material type
ČECH & TESAŘOVÁ (2015)	Poplar	- Influence of wood modification
CZAJKA & DZIEWANOWSKA-PUDLISZAK (2011)	Pine, oak, ash, alder, beech, larch	- Influence of different coating systems - Differences due to wood species
CZAJKA & FABISIAK (2012)	Pine	- Differences between sapwood and heartwood
CZAJKA & FABISIAK (2013)	Spruce	- Differences between juvenile and mature wood
CZAJKA & FABISIAK (2014)	Fir	- Differences between sapwood and heartwood
DÁUMLING et al. (2009)	OSB	- Influence of plasterboard on OSB emissions
ENGLUND (1999)	Pine, spruce, beech, oak, birch	- Differences due to wood species - Influence of wood moisture content, sapwood/heartwood, manufacturer, knots, drying conditions - Long-term behaviour
ENGLUND (2010)	Pine, spruce, beech, birch, ash, oak, alder, poplar	- Differences due to wood species - Differences between sapwood and heartwood
GACA & DZIEWANOWSKA-PUDLISZAK (2005)	Pine, oak, ash, alder, beech, larch, particleboard, plywood, particleboard with birch veneer	- Differences due to wood species and material type - Influence of varnish
HASEGAWA et al. (2006)	Spruce	- Influence of drying conditions
HORN et al. (2007)	OSB, pine, particleboard, beech	- Differences due to wood species and material type - Influence of different OSB manufacturers
HYTTINEN et al. (2010)	Pine, spruce, poplar	- Influence of thermal treatment
KRANIOTIS et al. (2015)	Pine	- Influence of air humidity
LARSEN et al. (1998)	Pine, spruce, birch, beech, birch plywood, oak parquet	- Differences due to wood species and material type - Influence of different manufacturers and coatings

Sources	Wood species and wood-based materials examined	Objective of the study
LARSEN, FROST & WINTHER FUNCH (2000)	Pine, spruce, beech, ash, pine and spruce particleboard, particleboard with beech veneer, MDF and OSB made from softwood, birch plywood	<ul style="list-style-type: none"> <li>- Differences due to wood species and material type</li> <li>- Influence of sapwood/heartwood, region of origin and glaze</li> <li>- Influence of binder type and coating systems</li> </ul>
MAKOWSKI (2007)	OSB	<ul style="list-style-type: none"> <li>- Long-term behaviour</li> <li>- Influence of manufacturing parameters and storage conditions</li> <li>- Influence of test conditions (chamber size)</li> </ul>
MANNINEN, PASANEN & HOLOPAINEN (2002)	Pine	<ul style="list-style-type: none"> <li>- Influence of thermal treatment</li> <li>- Influence of region of origin</li> </ul>
OHLMEYER et al. (2008a)	OSB	<ul style="list-style-type: none"> <li>- Influence of board thickness</li> </ul>
OHLMEYER & STECKEL (2012)	Pine, beech, OSB	<ul style="list-style-type: none"> <li>- see STECKEL (2011)</li> </ul>
QUE et al. (2013)	Hardboard, plywood, MDF, pine, particleboard	<ul style="list-style-type: none"> <li>- Differences due to material type</li> <li>- Influence of storage and coating</li> </ul>
RISHOLM-SUNDMAN (2002)	Particleboard, MDF, OSB, plywood, hardboard	<ul style="list-style-type: none"> <li>- Differences due to material type</li> </ul>
RISHOLM-SUNDMAN et al. (1998)	Ash, beech, sugar maple, birch, oak, black cherry, spruce, pine, rubberwood	<ul style="list-style-type: none"> <li>- Differences due to wood species</li> </ul>
SAARELA et al. (1994) in ECA (1997a)	Spruce, pine	<ul style="list-style-type: none"> <li>- Differences due to wood species</li> </ul>
SALTHAMMER et al. (2003)	OSB	<ul style="list-style-type: none"> <li>- General emission behaviour</li> </ul>
STACHOWIAK-WENCEK, PRĄDZYŃSKI & KRZYWOSIŃSKA (2011)	Particleboard	<ul style="list-style-type: none"> <li>- Influence of coating</li> </ul>
STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATENKO-NOŻEWNIK (2014)	Pine, beech, oak	<ul style="list-style-type: none"> <li>- Differences due to wood species</li> <li>- Influence of varnish</li> </ul>
STACHOWIAK-WENCEK et al. (2015)	Pine	<ul style="list-style-type: none"> <li>- Influence of varnish</li> </ul>
STECKEL (2011)	Pine, spruce	<ul style="list-style-type: none"> <li>- Influence of wood drying</li> <li>- Influence of sapwood/heartwood and upper/lower trunk section</li> <li>- Long-term behaviour</li> <li>- Influence of test conditions (temperature, relative humidity, air velocity, loading factor, air change rate)</li> </ul>

Sources	Wood species and wood-based materials examined	Objective of the study
STECKEL, WELLING & OHLMEYER (2010, 2013)	Spruce	<ul style="list-style-type: none"> <li>- Influence of wood drying</li> <li>- Influence of sapwood/heartwood</li> </ul>
TURTOLA et al. (2002)	Pine	- Influence of region of origin and forest management (soil fertilisation)
VOLKMER et al. (2014)	Oak	- Influence of wood modification (steaming)
WILKE et al. (2012)	Pine, OSB, spruce	<ul style="list-style-type: none"> <li>- Influence of sapwood/heartwood and upper/lower trunk section (pine)</li> <li>- Influence of storage</li> <li>- Influence of different manufacturers and manufacturing parameters</li> <li>- Influence of the addition of antioxidants (OSB)</li> </ul>
YRIEIX, MAUPETIT & RAMALHO (2004)	Maritime pine, oak, pine, particleboard, plywood	<ul style="list-style-type: none"> <li>- Differences due to wood species and material type</li> <li>- Influence of different paint systems and coatings</li> </ul>

Source: own representation according to the given sources

## 4 Emission behaviour of solid wood

Comparatively few studies are available that deal with the emission characteristics of solid wood. This can be explained by the fact that emission behaviour is a relatively new field of research compared to other wood properties, such as mechanical strength. According to SALTHAMMER (2011), the topic of indoor air only became the focus of scientific activity in the mid-1970s. The very definition of volatile organic compounds and their categorisation according to their boiling points was not established by the WHO until the late 1980s (SALTHAMMER, 2011).

There is a lack of systematic and comprehensive studies on the individual wood species that examine the influence of wood origin and growth conditions on the emission behaviour of the wood. The variability of the emission rates and composition due to the variability of the material is therefore largely unexplored. Only the distinction between different wood tissues (sapwood/heartwood, juvenile/mature wood, wood with/without knots) has been investigated in more detail (Table 5). Previous studies have dealt much more with the influence of individual wood processing steps on the emission behaviour. The effects of various drying processes, storage conditions, wood modifications and surface treatments (coating, painting, staining) on VOC emissions have been discussed (Table 5). These factors are largely controllable by humans. In contrast, it is only possible to influence the tree and its wood to a very limited extent. The absence of systematic data on the diversity of the emission behaviour within a wood species as a result of intrinsic (tree-related) factors makes it difficult to assess the impact of individual wood species on indoor air quality.

### 4.1 Scots pine (*Pinus sylvestris* L.)

After Norway spruce, Scots pine is Germany's second most important coniferous tree species with a total timber stock of 768 million m<sup>3</sup>. Its main distribution area is in the North German Plain from Lower Saxony to Brandenburg and Saxony as well as in the Palatinate Forest, the Rhine-Main lowlands and the Upper Palatinate basin and hill regions (BMEL, 2016). According to the third National Forest Inventory, an average of 13.3 million m<sup>3</sup> of timber was harvested annually between 2002 and 2012, which corresponds to around 18 % of total timber use (THÜNEN-INSTITUT, 2014).

#### 4.1.1 Sum of volatile organic compounds (TVOC)

According to the studies by WILKE et al. (2012) and ENGLUND (1999), the sum of volatile organic compounds ranges from 16,000 to 22,000 µg m<sup>-2</sup> h<sup>-1</sup> in fresh sapwood and from 23,000 to 35,000 µg m<sup>-2</sup> h<sup>-1</sup> in fresh heartwood (Table 6).

**Table 6: TVOC emission rates of fresh/moist pine wood**

Tissue	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
Sapwood	16,228 – 22,469 <sup>a</sup>	1,309 – 1,536 <sup>a</sup>	Test chamber	WILKE et al., 2012 <sup>b</sup>
	23,062 – 34,823 <sup>a</sup>	3,587 – 10,760 <sup>a</sup>	Test chamber	WILKE et al., 2012 <sup>b</sup>
Heartwood	29,087 (MC: 85 %) – 10,465 (MC: 19 %) <sup>c</sup>	No data	Test cell	ENGLUND, 1999

<sup>a</sup> Variation within a tree depending on the height

<sup>b</sup> The authors provide no information on the actual wood moisture content. However, they refer to "fresh pine wood" (WILKE et al., 2012:46).

<sup>c</sup> MC = wood moisture content

Source: own representation according to the given sources

The moisture content of wood at the time of installation should generally be below 20 %. The wood drying required for this causes a considerable proportion of the VOCs to volatilise during the drying process. ENGLUND (1999) demonstrated, based on studies of Scots pine wood from wet to dry conditions, that drying from an initial moisture content of 85 % to 19 % or 10 % results in a reduction of TVOC by 64 % and 98 %, respectively. Using extractive measurements, ENGLUND & NUSSBAUM (2000) determined that the original monoterpene content (heartwood: 0.8 - 1.1 %, sapwood: 0.4 - 0.5 %, based on oven-dry wood) is reduced by 25 to 50 % in heartwood and 20 to 40 % in sapwood due to drying. Several other studies (SHMULSKY, 2000a, 2000b; BANERJEE, 2001; CONNERS, YAN & BANERJEE, 2002; GRANSTRÖM, 2005) describe the VOC release during drying and the parameters that influence it. Clausthaler Umwelttechnik-Institut GmbH, for example, developed a process to recover the terpenes produced during the drying of pine wood chips and thus utilise them (BORMANN & SIEVERS, 2003).

The emission data of dry pine wood show a wide range of TVOC values and can reach the double-digit  $\text{mg m}^{-2} \text{h}^{-1}$  range. The area-specific emission rates range from 300 to 19,500  $\mu\text{g m}^{-2} \text{h}^{-1}$  for heartwood and 100 to 6,700  $\mu\text{g m}^{-2} \text{h}^{-1}$  for sapwood. In studies that do not provide any information about the analysed wood tissue, comparatively low emission rates of between 200 and 3,800  $\mu\text{g m}^{-2} \text{h}^{-1}$  were determined, which suggests a higher proportion of sapwood in the tested material or a longer storage period. The VOC emissions decay rapidly in the first weeks, so that the 28-day value is reduced on average by around half of the 3-day value. The decay rates are between 12 and 83 %, whereby a stronger reduction cannot be determined for initially high-emitting material. After about one month, this results in TVOC emission rates of 200 to 9,900  $\mu\text{g m}^{-2} \text{h}^{-1}$  for heartwood and 100 to 3,200  $\mu\text{g m}^{-2} \text{h}^{-1}$  for sapwood (Table 7).

Table 7: TVOC emission rates of dry pine wood

Tissue	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	short-term	long-term		
Predominantly heartwood	334 <sup>a</sup>	187 <sup>a</sup>	Test chamber	WILKE et al., 2012
	458	No data	Test cell	ENGLUND, 1999
	966	486	Test cell	ENGLUND, 1999
	1,858	607	Test cell	ENGLUND, 1999
	2,093 – 5,691 <sup>b</sup>	1,327 – 2,770 <sup>b</sup>	Test chamber	STECKEL, 2011
	No data	4,550	Test cell	ENGLUND, 2010
	6,050	5,310	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
	8,099	4,600	Test chamber	STECKEL, 2011; OHLMEYER & STECKEL, 2012
	8,251	3,934	Test cell	ENGLUND, 1999
	10,100	5,050	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
	12,152	No data	Test cell	ENGLUND, 1999
	12,975	7,357	Test chamber	CZAJKA & FABISIAK, 2012 <sup>c</sup>
	19,540	9,862	Test chamber	STECKEL, 2011; OHLMEYER & STECKEL, 2012
Predominantly sapwood	70 <sup>a</sup>	61 <sup>a</sup>	Test chamber	WILKE et al., 2012
	280	125	Test cell	ENGLUND, 1999
	470 – 2,139 <sup>b</sup>	249 – 657 <sup>b</sup>	Test chamber	STECKEL, 2011
	830	348	Test cell	ENGLUND, 1999
	1,090	701	Test chamber	STECKEL, 2011; OHLMEYER & STECKEL, 2012
	1,582	980	Test chamber	CZAJKA & FABISIAK, 2012 <sup>c</sup>
	1,600	1,070	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
	2,616	1,331	Test cell	ENGLUND, 1999
	No data	1,381	Test cell	ENGLUND, 2010
	3,142	750	Test chamber	STECKEL, 2011; OHLMEYER & STECKEL, 2012
6,650	3,180	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000	
Not specified	215	No data	Test chamber	STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNIAK, 2014; STACHOWIAK-WENCEK et al., 2015
	240 – 300 <sup>d</sup>	No data	Test cell	KRANIOTIS et al., 2015
	361	61	Test chamber	LARSEN et al., 1998
	361	69	Test chamber	SAARELA et al. 1994 in ECA, 1997a
	390 – 480 <sup>d</sup>	No data	Test cell	KRANIOTIS et al., 2015
	1,463	909	Test chamber	HYTTINEN et al., 2010
	1,552	No data	Test chamber	GACA & DZIEWANOWSKA-PUDLISZAK, 2005; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011
3,700	No data	Test cell	RISHOLM-SUNDMANN et al., 1998	

<sup>a</sup> Stored for 14 months

<sup>b</sup> Variations due to different drying methods

<sup>c</sup> No information on wood moisture content

<sup>d</sup> Different humidity conditions between 30 and 80 % relative humidity

Source: own representation according to the given sources

ENGLUND (1999), STECKEL (2011), CZAJKA & FABISIAK (2012) and WILKE et al. (2012) directly compared the emission behaviour of heartwood and sapwood from the same tree. This showed that the emission level of the heartwood is generally significantly higher than that of the associated sapwood. However, there are no fixed ratios – in some samples the TVOC of the sapwood corresponds to only 3 %, in others 88 % of the heartwood. In one pair of samples, the sapwood emissions were significantly higher than those of heartwood due to very high aldehyde emissions. In general, it can be assumed that a higher proportion of sapwood in the wood leads to reduced overall emissions. The differences in the emission rates of sapwood and heartwood are ultimately due to the different chemical composition of the wood tissues (Chapter 4.1.2). Long-term studies by ENGLUND (1999, 2010) and STECKEL (2011) show that the emissions of pine wood continue to decrease even beyond 28 days, albeit at a significantly lower decay rate (STECKEL, 2011). SALTHAMMER & FUHRMANN (1996) refer to a state of equilibrium that occurs after a certain period of time. In their measurement series of commercially purchased pine shelving boards, this equilibrium was reached after seven months. However, the exact time at which this state was achieved could not be determined in the study, as emission data were only collected up to day seven and then again after seven months. The standard 28-day value determined in accordance with DIN EN ISO 16000-9:2008 and 16000-10:2006 does not necessarily reflect a state of equilibrium and therefore has only limited information regarding the long-term behaviour of the pine wood.

Table 8 presents the TVOC emission rates of commercially purchased pine wood (boards, planks, plywood). Information on sapwood/heartwood content, manufacturing conditions and time, as well as storage conditions of the material are therefore only partially known. Nevertheless, the data can provide important indications of the emission rates that consumers can expect when using pine wood. Comparatively high emissions of 3,500 to 12,500  $\mu\text{g m}^{-2} \text{h}^{-1}$  (short-term) and 2,300 to 7,000  $\mu\text{g m}^{-2} \text{h}^{-1}$  (long-term behaviour) are achieved with a predominant proportion of heartwood in the products. In the samples without specification of the sapwood/heartwood content, significantly lower TVOCs values were determined with a maximum of 1,600  $\mu\text{g m}^{-2} \text{h}^{-1}$  (both short-term and long-term behaviour). This may indicate a higher proportion of sapwood or a longer storage period for the material. Another reason could be the measurement method used (emission chamber versus emission cell). The typical decay behaviour of the emissions could not be observed by WILKE et al. (2012) in all of the investigated pine plywood samples. In two of the five analysed panels, the concentrations increased during the course of the measurement, which is unusual for pine wood. However, the substances primarily relevant for the TVOC,  $\alpha$ -pinene and 3-carene, were outside the calibration range at the 3-day values, so this was more likely due to an analytical problem.

**Table 8: TVOC emission rates of purchased pine plywood or sawn timber**

Heartwood content	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
Not specified	290	220	Test chamber	HORN et al., 2007
	304	No data	Test cell	QUE et al., 2013 <sup>a</sup>
	313	399	Test chamber	WILKE et al., 2012
	678	390	Test chamber	WILKE et al., 2012
	1,075	445	Test chamber	WILKE et al., 2012
	1,102	1,628	Test chamber	WILKE et al., 2012
	1,500	625	Test chamber	YRIEIX, MAUPETIT & RAMALHO, 2004
	1,596	814	Test chamber	WILKE et al., 2012
Predominantly heartwood	3,456	2,307	Test cell	ENGLUND, 1999
	3,656	3,090	Test cell	LARSEN et al., 1998
	4,485	2,830	Test cell	ENGLUND, 1999
	4,659	3,330	Test cell	ENGLUND, 1999
	5,209	3,157	Test cell	LARSEN et al., 1998
	6,254	5,810	Test cell	ENGLUND, 1999
	6,857	3,638	Test cell	ENGLUND, 1999
	7,220	6,044	Test cell	ENGLUND, 1999
	8,450	6,063	Test cell	ENGLUND, 1999
12,486	6,960	Test cell	ENGLUND, 1999	

<sup>a</sup> No exact species given (*Pinus spec.*)

Source: own representation according to the given sources

### 4.1.2 Significant individual substances

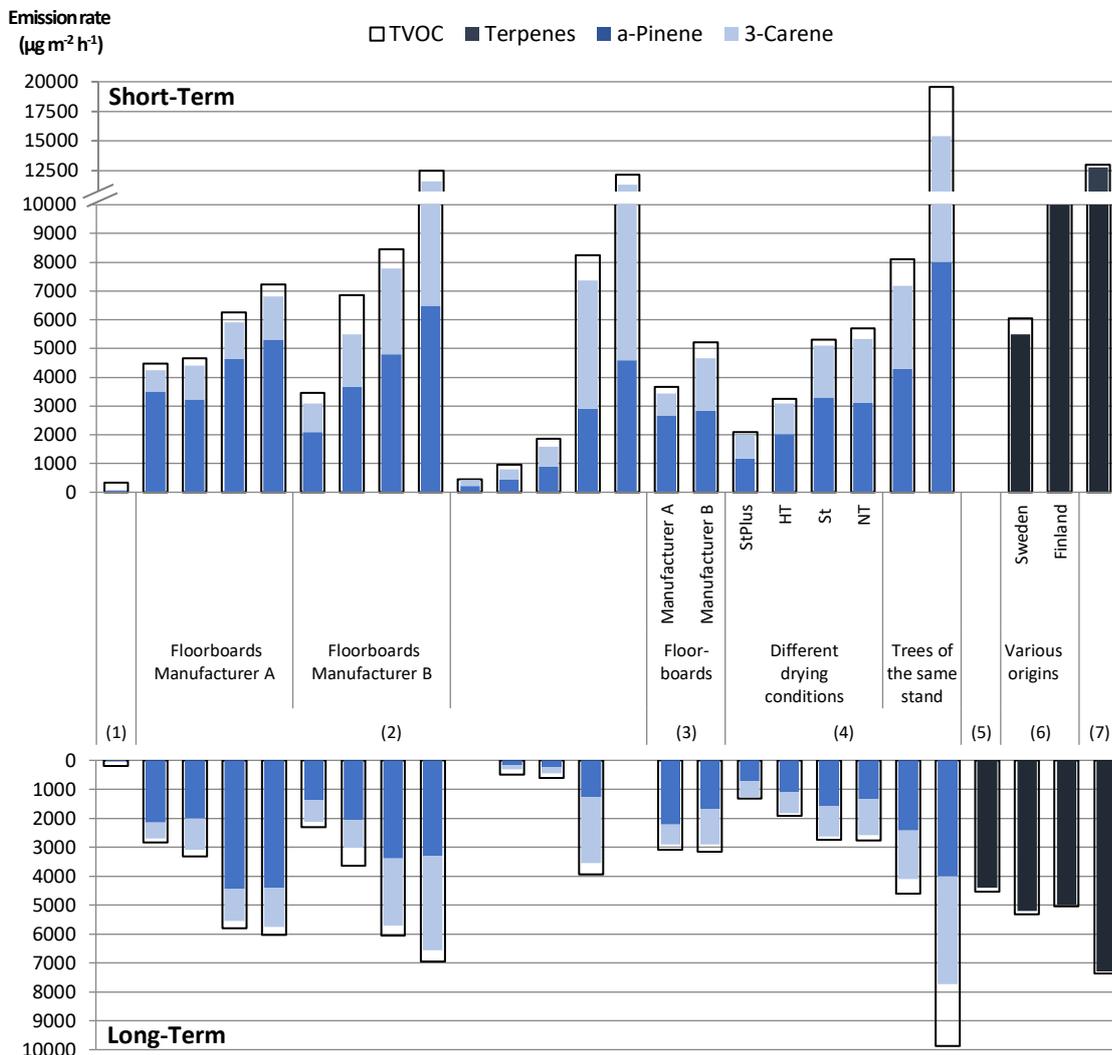
Pine wood mainly emits monoterpenes. Aldehydes (especially hexanal and pentanal), terpenoids (e.g. terpineol) and organic acids (especially acetic and hexanoic acid) also occur in comparatively small proportions (ENGLUND, 1999, 2010; STECKEL, 2011).

#### Terpenes

The emissions from pine heartwood consist mainly of terpenes, which generally make up over 90 % of the TVOC (ENGLUND, 1999, 2010; LARSEN, FROST & WINTHER FUNCH, 2000; STECKEL, 2011; CZAJKA & FABISIAK, 2012; WILKE et al., 2012). The two monoterpenes<sup>15</sup>,  $\alpha$ -pinene and 3-carene, are the dominant emissions both initially and after one month (Figure 2). Another substance from the group of monoterpenes (such as terpinolene,  $\beta$ -pinene, limonene, camphene) often ranks third.

<sup>15</sup> Further literature on terpenes: RAMAWAT, K. G.; MÉRILLON, J.-M. (2013)

**Figure 2: Significant VOCs emitted from pine heartwood (heartwood content > 65 %, short- and long-term behaviour)**



St = standard drying, NT = low temperature drying, HT = high temperature drying, StPlus = standard drying with increased final temperature

- (1) WILKE et al., 2012      (4) STECKEL, 2011      (6) LARSEN, FROST & WINTHER FUNCH, 2000
- (2) ENGLUND, 1999      (5) ENGLUND, 2010      (7) CZAJKA & FABISIAK, 2012
- (3) LARSEN et al., 1998

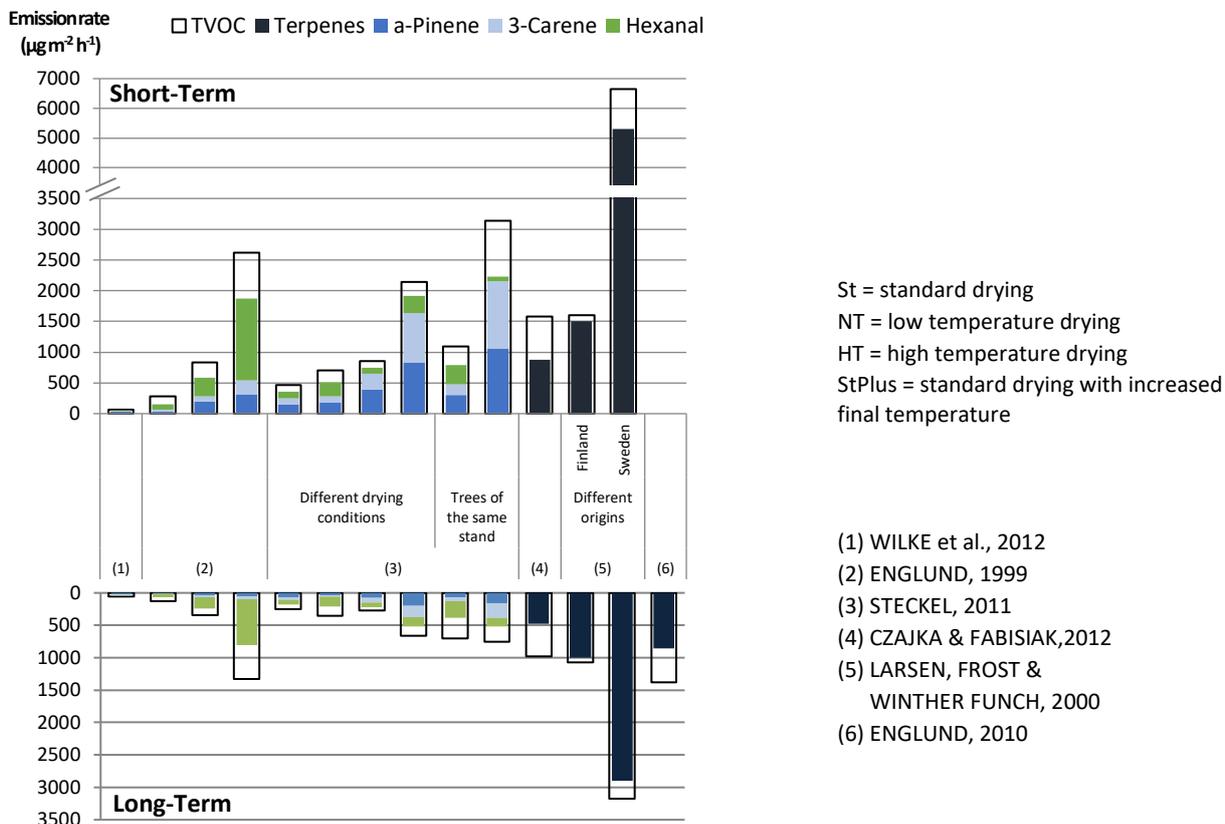
Source: own representation according to the given sources

Pine sapwood is mainly characterised by the emissions of monoterpenes (especially  $\alpha$ -pinene and 3-carene) and aldehydes (particularly hexanal and pentanal). If high terpene emissions occur, the proportion of aldehydes in the total emissions is relatively low (Figure 3).

Over the course of the emission measurement up to one month, an increase in the aldehyde content and a decrease in the terpene content of the TVOC can often be observed (ENGLUND, 1999; STECKEL, 2011; WILKE et al., 2012). The relative composition of the emissions is therefore subject to fluctuations and is less predictable compared to heartwood. Hexanal is the quantitatively dominant aldehyde, with emissions generally falling below  $300 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days. An exception with  $700 \mu\text{g m}^{-2} \text{h}^{-1}$  is a measurement by ENGLUND (1999). The terpene emissions - totalling less than  $1,000 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days - are considerably lower than in heartwood, but can

also reach quite high values (up to 3,000  $\mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days) in individual samples (LARSEN, FROST & WINTHER FUNCH, 2000).

**Figure 3: Significant VOCs emitted from pine sapwood (sapwood content > 96 %, short- and long-term behaviour)**



Source: own representation according to the given sources

Not only the level of total emissions differs between pine wood samples, but also the relative ratio of the individual compounds to each other. The emission rate of  $\alpha$ -pinene, for example, is 0.7 (ENGLUND, 1999) to 7.5 times (STACHOWIAKWENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNIK, 2014) greater than that of 3-carene. In most cases, however, the  $\alpha$ -pinene emissions are higher than those of 3-carene, typically by a factor of one to two.  $\beta$ -Pinene plays a role in some samples with a share of up to 22 % of the TVOC (STECKEL, 2011), but for the most part it is below 3 % of the TVOC. In addition to sesquiterpenes and neutral (diterpene aldehydes, alcohols, resin acid esters, etc.) and acidic (resin acids) diterpenoids, the monoterpenes mentioned are a component of the oleoresin found in the resin canals and resin pockets of pine (EKMAN & HOLMBOM, 2000). They serve as solvents for the non-volatile resin components and thus reduce the viscosity of the oleoresin. The resin fulfils the task of sealing tree injuries mechanically and water-repellent (BACK, 2000). Distillation of the oleoresin yields turpentine (about 16 to 19 % of the resin), whose main components are monoterpenes, while the non-volatile residue, rosin, remains (70 to 82 %, mainly resin acids) (several sources in HAFIZOĞLU, 1983). The level of terpene emissions is therefore dependent on the oleoresin content of the wood. According to BACK (2000), the genus *Pinus* has the best developed resin canal system consisting of interconnected axial and radial canals. The oleoresin is located within these canals and in the epithelial cells surrounding them. The radial resin canals run along the wood rays – they are generally more frequent but smaller in diameter than the axial resin canals. Axial resin canal formation occurs in the cambium and is initiated by mechanical stress (e.g. wind, injury), drought stress and insect infestation. TURTOLA et al. (2003), for example, showed that spruce and pine seedlings react to

severe drought stress with increased resin acid and monoterpene production. In addition to genetic predisposition, external factors thus play a role in the level of oleoresin content (BACK, 2000; TURTOLA et al., 2003; PANDA, 2008). Consequently, this varies both between the trees within a stand and within an individual tree (e.g. depending on the tree height) (BACK, 2000). As a result of heartwood formation, the oleoresin components are redistributed. The epithelial cells in the resin canals produce additional oleoresin, which is released into the surrounding tissue. The epithelial cells then lignify and die (BACK, 2000). In the sapwood, in contrast, the oleoresin is primarily located in the resin canals and possibly in resin pockets (EKMAN & HOLMBOM, 2000). Accordingly, the oleoresin content in the heartwood is significantly higher than in the sapwood (LANGE, KUBEL & WEIßMANN, 1989; EKMAN & HOLMBOM, 2000; PANDA, 2008; ARSHADI et al., 2013), which consequently also explains the higher terpene emissions of the heartwood (Figure 2, Figure 3). The composition and level of emissions cannot automatically be deduced from the extractives content alone, as the actual emission rates of the individual compounds depend on several factors. In addition to the material moisture content, the vapour pressure and the diffusion coefficient of the compounds as well as their distribution within the heterogeneous wood matrix are important (ENGLUND, 1999). Nevertheless, the content and composition of the extractives provide an important indication of the emission behaviour of wood.

While the composition of the resin acids is largely independent of the origin of the wood (LANGE & WEIßMANN, 1988; LANGE & STEVANOVIĆ JANEŽIĆ, 1993; EKMAN & HOLMBOM, 2000), the results of the monoterpene compositions indicate an influence of the provenance (several sources in HAFIZOĞLU, 1983; LANGE & STEVANOVIĆ JANEŽIĆ, 1993). Thus, not only the absolute monoterpene content of pine wood varies between different trees, but also the monoterpene composition, which ultimately affects the emission composition (Figure 2, Figure 3). Several studies suggest that the relative proportion of some monoterpenes is less dependent on environmental factors, but is mainly genetically controlled (THORIN & NOMMIK, 1974; BARADAT & YAZDANI, 1988; HANOVER, 1992; KIVIMÄENPÄÄ et al., 2012). Analyses of turpentine showed that a distinction can be made between pines with high (Central European and Scottish turpentine) and with little to no 3-carene content (Turkish and Spanish turpentine) (several sources in HAFIZOĞLU, 1983; LANGE & WEIßMANN, 1988; PARDOS, LANGE & WEIßMANN, 1990; TÜMEN & REUNANEN, 2010). Extract analyses by SJÖDIN, PERSSON & NORIN (1992) also show a wide range of 3-carene and  $\alpha$ -pinene proportions in pine wood. HANOVER (1992) summarises that there appears to be a north-south decrease in the frequency of the 3-carene gene – although there are also major fluctuations within countries. STECKEL (2011) confirmed this on the basis of emission measurements, with considerable variability occurring even within a pine stand (Figure 2, Figure 3) – both in terms of the quantity and composition of emissions.

### Aldehydes

Alongside terpenes, aldehydes are the second main class of compounds emitted by pine wood. They are not direct components of wood, but are formed from unsaturated fatty acids and their esters (triglycerides and sterol esters) as a result of autoxidation processes. Like resin acids, these have a low vapour pressure at room temperature and therefore hardly emit themselves. Initially, unstable lipid hydroperoxides are formed from the fatty acids, which can further decompose into aldehydes. Mainly saturated aldehydes in the chain length range from pentanal to decanal are formed. The fatty acid composition of the wood is decisive for the aldehyde composition of the wood emissions. According to analyses by HOLMBOM & EKMAN (1978), pine wood contains 3 % saturated, 37 % mono-unsaturated, 42 % di-unsaturated and 17 % tri-unsaturated fatty acids. Linoleic acid (9,12-18:2<sup>16</sup>) and oleic acid (9-18:1) make up the majority with a share of 41 % and 35 % respectively, followed by pinolenic acid (5,9,12-18:3) with 11 %. Pinolenic acid and taxoleic acid (5,9-18:2) are found exclusively in the

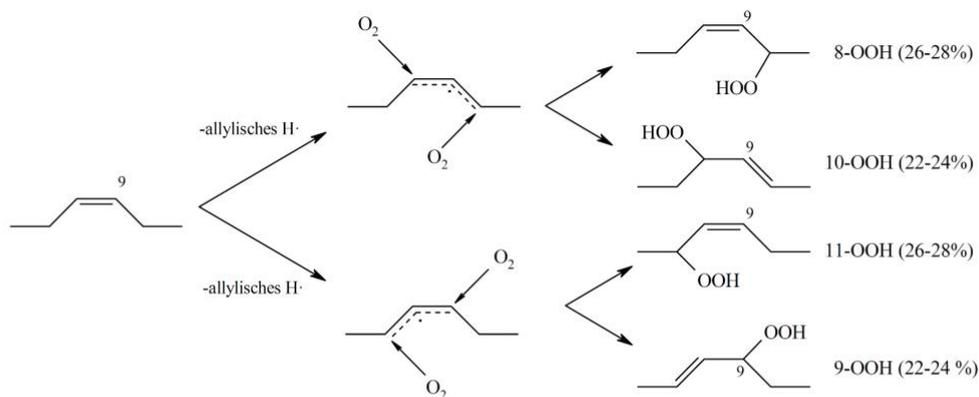
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<sup>16</sup> Structure of fatty acids: Position of the double bonds - Number of carbon atoms: Number of double bonds

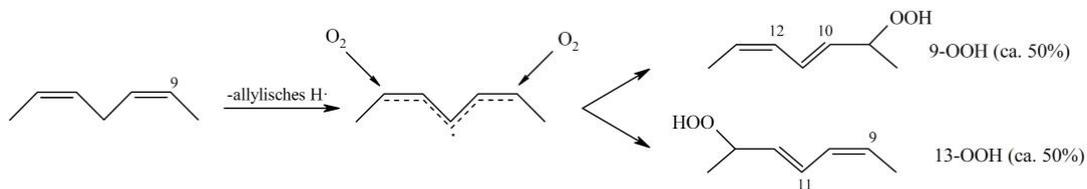
wood of the pine family<sup>17</sup>, but not in hardwoods, whose qualitative fatty acid composition is otherwise comparable (EKMAN & HOLMBOM, 2000). Each fatty acid can form its characteristic isomeric hydroperoxides (Figure 4), which in turn break down into corresponding reaction products. For example, linoleic acid, the most common fatty acid in pine wood (HOLMBOM & EKMAN, 1978), forms 9- and 13-lipid hydroperoxides. The main degradation product of the former is 2,4-decadienal, which in turn is further oxidised to hexanal, 2-octenal and other volatile compounds, while the latter mainly yields hexanal (NAWAR, 1984; GROSCH, 1987). Consequently, hexanal is mainly found among the aldehyde emissions.

**Figure 4: Formation of hydroperoxides during the autoxidation of oleic, linoleic and linolenic acid**

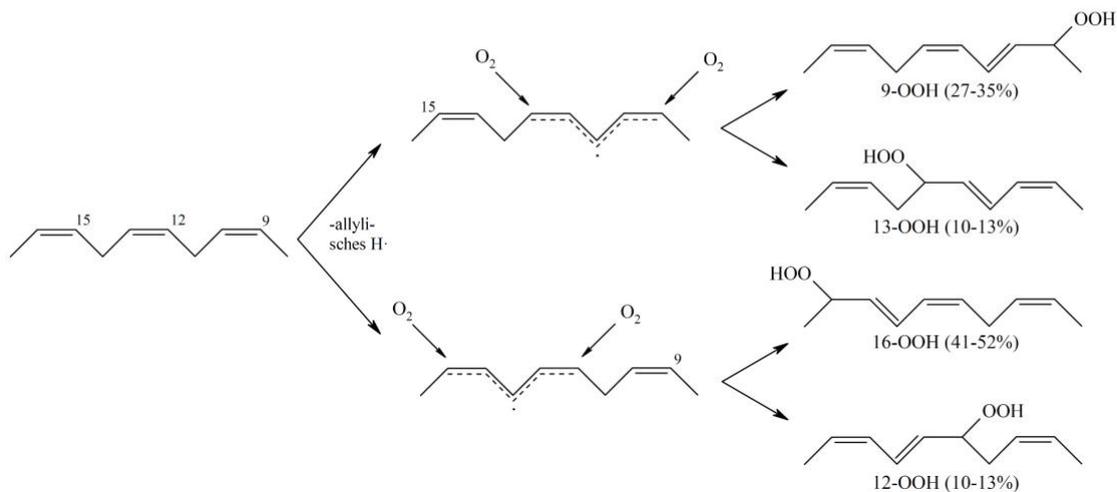
#### Oleic acid



#### Linoleic acid



#### Linolenic acid



Source: own representation according to HEITKAMP (1999)

<sup>17</sup> Includes the genera *Pinus*, *Abies*, *Larix* and *Picea*, among others

In addition to aldehydes, the main reaction products from oleic and linoleic acid, alcohols, alkanes, ketones and short-chain acids are also formed and emitted as a result of these reactions (GROSCH, 1987). Aldehydes can also be further oxidised to their corresponding carboxylic acids (NAWAR, 1984; GROSCH, 1987), so that the hexanoic acid detected in some studies of pine wood emissions (e.g. ENGLUND, 1999; STECKEL, 2011; WILKE et al., 2012) can presumably be attributed to this. Fatty acids and their esters are the extractives in the living cells (parenchyma cells). This parenchyma resin makes up about 75 % of the total resin in the sapwood of pine (BACK, 2000). While the proportion of resin acids (consequently also monoterpenes) and free fatty acids is lower in sapwood, it has a higher content of triglycerides compared to heartwood. During heartwood formation, the fats are hydrolysed so that only free fatty acids are present in the heartwood. However, the enzymatic hydrolysis of the triglycerides also begins in the sapwood after the tree has been felled and continues during the storage of the wood (ASSARSSON, CROON & DONETZHUER, 1963; ASSARSSON, 1966; EKMAN & HOLMBOM, 2000). The rate at which fatty acids undergo autoxidation depends on the availability of oxygen and on temperature. Metal ions (especially iron, manganese and copper), which are present in the parenchyma cells of the wood, and light (especially in the UV range) also act as catalysts. The oxidation rate is generally higher for fatty acids than for resin acids, for free fatty acids than for esterified fatty acids and for polyunsaturated compared to monounsaturated fatty acids. The level of aldehyde emissions is therefore dependent on the one hand on the properties of the wood (specific surface area, porosity, wood specie and composition and content of fatty acids and esters) and on the other hand on external influences, in particular the effect of temperature. While terpene emissions decrease continuously, aldehyde emissions initially reach a maximum and only then subside. Depending on the aldehyde, different emission levels are reached and at different times, which is related to the fact that the individual fatty acids of the woods have different oxidation rates and degradation products (BACK et al., 2000). Consequently, the time of the emission measurement has a decisive influence on the emission result. As already discussed, the resin acid content varies greatly between trees of the same species, whereas the fatty acid content is relatively constant (LANGE, KUBEL & WEIßMANN, 1989). According to EKMAN & HOLMBOM (2000), the proportion of unsaturated fatty acids is higher in sapwood than in heartwood, which would explain why aldehyde emissions from sapwood are generally higher than from heartwood (ENGLUND, 1999; STECKEL, 2011; CZAJKA & FABISIAK, 2012; WILKE et al., 2012). Another reason for the higher aldehyde emissions of sapwood could be that heartwood appears to have a stronger ability to act as a radical scavenger compared to sapwood, which BELT, HÄNNINEN & RAUTKARI (2017) were able to prove, at least for the ABTS<sup>18</sup> radical. The radicals formed from the fatty acids are the prerequisite for the formation of hydroperoxides, which in turn decompose into aldehydes (HEITKAMP, 1999). Furthermore, sapwood has a higher gas permeability than heartwood (COMSTOCK, 1970), which presumably favours the oxygen supply necessary for the autoxidation of the fatty acids.

### 4.1.3 Enantiomer composition

Chemical compounds that have the same constitution but exist as non-superimposable mirror images of each other are referred to as enantiomers. If chiral reaction partners are present, the enantiomers of a compound can show considerable differences with regard to their physiological and pharmacological effect. It is therefore possible that only one enantiomer, both enantiomers to the same extent or both to different extents produce an effect (JUNGE, 2004). According to KASANEN et al. (1998), for example, the (+)-enantiomers of  $\alpha$ - and  $\beta$ -pinene cause a stronger sensory irritation than the (-)-enantiomers. Knowledge of the enantiomer composition is therefore important in order to assess the emissions of pine wood and other wood species with regard to their health-related effects. By using a suitable chromatography column, enantioselective gas chromatography can be carried out in principle. Chiral stationary phases (e.g. cyclodextrins and their derivatives) are used in the separation column allowing the analysis of enantiomer mixtures (JUNGE, 2004; RICHTER, 2008). However, studies

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<sup>18</sup> 2,2'-Azino-di-(3-ethylbenzthiazoline-6-sulfonic acid)

that have carried out such analyses on pine wood emissions are not known. Instead, literature is available that examines what the living tree emits and the composition of the essential oils from various tree tissues (SADOF & GRANT, 1997; MACIAG et al., 2007; OCHOCKA et al., 2008). These findings play a role in the interaction between insects and their host trees and in the use of essential oils in the pharmaceutical, food and cosmetics industries (OCHOCKA et al., 2008).

The analyses of wood extractives carried out by SJÖDIN, PERSSON & NORIN (1992) and SJÖDIN et al. (1996, 2000) can provide initial indications of the enantiomer composition to be expected from wood emissions. The studies showed that the composition of monoterpene enantiomers varies both between trees of the same species and between different tree tissues (needles, wood, roots, cones, etc.). Consequently, studies analysing the composition in needles, for example, cannot be used to draw conclusions about the composition in wood. The results (Table 9) also indicate that only the (+)-enantiomer of 3-carene occurs in pine wood, whereas the (-)-enantiomer dominates in  $\beta$ -pinene and  $\beta$ -phellandrene. At least 70 % of  $\alpha$ -pinene is composed of the (+)-enantiomer, although SJÖDIN, PERSSON & NORIN (1992) point out that the great variability in the enantiomer composition observed in spruce monoterpenes, especially in  $\alpha$ -pinene (PERSSON et al., 1996), is probably also to be expected in pine.

**Table 9: Composition of the monoterpene enantiomers of wood extracts**

Source	Material (number of samples)	Proportion of the (-)-enantiomer (%)						
		3-Carene	$\alpha$ -Pinene	Limonene	$\beta$ -Pinene	Camphene	$\beta$ -Phellandrene	Sabinene
(1)	Pine Logs incl. bark (6)	0	4 – 29	36 – 91	62 – 85	18 – 49	92 – 100	84 – 98
(2)	Pine Logs (4)	0	5 – 23	45 – 90	60 – 100	20 – 50	90 – 100	80 – 90
(3)	Pine Logs (18)	0	5 – 30	25 – 90	60 – 100	20 – 50	90 – 100	25 – 100
(3)	Spruce Logs (7)	No data	20 – 80	No data	No data	No data	No data	No data
(4)	Spruce Branch wood (41)	0	24 – 88	23 – 90	87 – 99	50 – 94	77 – 99	5 – 81

(1) SJÖDIN, PERSSON & NORIN, 1992

(2) SJÖDIN et al., 1996

(3) SJÖDIN et al., 2000

(4) PERSSON et al., 1996

Source: own representation according to the given sources

In contrast to the wood extractives measurements of Scandinavian pine wood (SJÖDIN, PERSSON & NORIN, 1992; SJÖDIN et al., 1996, 2000), LANGE & STEVANOVIĆ JANEŽIĆ (1993) analysed turpentine from pine trees in south-eastern Europe. Two of the four samples analysed consisted mainly of  $\alpha$ -pinene, but were classified as levorotatory<sup>19</sup> with regard to their optical activity. LANGE & STEVANOVIĆ JANEŽIĆ (1993) concluded that the turpentine oils must therefore consist of at least 80 % (-)- $\alpha$ -pinene. Based on the available data, it can therefore be assumed that pine wood consists exclusively of (+)-3-carene, whereas  $\alpha$ -pinene occurs as a mixture of both enantiomers, with the ratio varying widely across the species' distribution range.

## 4.2 Norway spruce (*Picea abies* (L.) H. KARST.)

Norway spruce is Germany's most important and most common tree species with a total timber stock of 1,206 million m<sup>3</sup> (BMEL, 2016) and an average use of 39.7 million m<sup>3</sup> of harvested timber per year between 2002 and 2012, accounting for around 52 % of total harvesting volumes (THÜNEN-INSTITUT, 2014). However, as a result of

<sup>19</sup> Optical activity: Property of a substance to rotate the plane of polarised light

the silvicultural and forest policy objectives aimed at converting non-site-adopted pure spruce stands into stable and close-to-nature mixed stands or deciduous forests, both forest area and timber stock have decreased in recent years. The natural distribution area comprises mountainous regions, but beyond this, many areas have been afforested with spruce in the past, most recently on a large scale after the Second World War (BMEL, 2016). Spruce is widely used in the construction sector – it accounts for a significant proportion of the 86 % of softwood used in construction (MANTAU, DÖRING & HILLER, 2013).

#### 4.2.1 Sum of volatile organic compounds (TVOC)

According to studies by ENGLUND (1999), the sum of volatile organic compounds is around  $18,000 \mu\text{g m}^{-2} \text{h}^{-1}$  for fresh wood. As the wood dries from an initial moisture content of 65 % down to 20 % or 10 %, the TVOC decreases by more than 95 % to about 700 and  $50 \mu\text{g m}^{-2} \text{h}^{-1}$ , respectively (ENGLUND, 1999). The application of different drying conditions, which differ mainly in terms of drying temperature, has an influence on the emission level of the wood (HASEGAWA et al., 2006; STECKEL, WELLING & OHLMEYER, 2010). In principle, it can be assumed that higher emission quantities, especially terpenes, are already emitted at high temperatures during drying. Higher drying temperatures can affect the aldehyde emissions in two ways. On the one hand, it is assumed that aldehydes already present in the wood are emitted more quickly, and on the other hand, the higher temperature also promotes further aldehyde formation from the fatty acids. STECKEL, WELLING & OHLMEYER (2010) and HASEGAWA et al. (2006) determined that higher drying temperatures lead to lower overall emissions (affecting both terpene and aldehyde emissions) from spruce during subsequent use as a building product. This was particularly noticeable in the first days of emission measurement, after one month the differences are only slight. However, studies by ENGLUND (1999) showed that this is not always the case – in two out of three pairs of samples, the total emissions of wood dried at a higher temperature ( $108^\circ\text{C}$ ) were greater than those of wood dried at a normal temperature ( $69^\circ\text{C}$ ) (Figure 5).

The comparison of the TVOC emission rates of pine (Table 7) and spruce (Table 10) shows that spruce wood generally has significantly lower emissions. Spruce is also traversed by a vertical and horizontal resin canal system. However, no or hardly any additional oleoresin is formed during heartwood formation (BACK, 2000), so that the resin content and thus the terpene emissions of spruce are generally lower than those of pine. Extractive measurements by ENGLUND & NUSSBAUM (2000) confirm this – the original terpene content of spruce is significantly lower than that of pine (heartwood: 0.8 - 1.1 %, sapwood: 0.4 - 0.5 % based on oven-dry wood), at only 0.02 - 0.08 %. The TVOC emission rate is generally below 1,000 (short-term) or  $500 \mu\text{g m}^{-2} \text{h}^{-1}$  (long-term), with higher values observed only occasionally. Like pine, spruce can react to stress events (mechanical injury, drought, insect infestation, etc.) with increased resin formation. TEMNERUD (1999) found that spruce trees have a higher tendency to form resin pockets with increasing tree height. The thinning intensity also seems to have an influence (SCHUMACHER, TRATAMILLER & GROSSER, 1997). The main causes of increased resin pocket formation also appear to be drought stress (TEMNERUD, 1999; SEIFERT et al., 2010) and mechanical stress caused by wind (TEMNERUD, VALINGER & SUNDBERG, 1999; HERB & BECKER, 2006). STECKEL, WELLING & OHLMEYER (2010, 2013) showed that a comparatively highly emitting sapwood sample ( $\text{TVOC}_{3\text{d}} = 2,200 \mu\text{g m}^{-2} \text{h}^{-1}$ ) was characterised by an increased number of resin canals and oleoresin that had penetrated into the surrounding tissue. Another cause of strongly emitting spruce wood can be the presence of knots. Studies by ENGLUND (1999) indicate that the influence varies depending on the type of knot. In wood with completely sound knots, no effect on the emission level was found, whereas an oblong splay knot with a hint of pocketed bark led to strongly increased emissions ( $\text{TVOC}_{3\text{d}} = 5,200 \mu\text{g m}^{-2} \text{h}^{-1}$ ). Very low emission rates were observed by WILKE et al. (2012) on spruce plywood, whereby the production times and conditions were not known, so that the low emissions

are probably due to a longer storage period. Emission measurements on juvenile and mature wood<sup>20</sup> suggest that the oleoresin content increases with distance from the pith – CZAJKA & FABISIAK (2013) determined about 2.5 times higher total emissions in mature wood.

**Table 10: TVOC emission rates of dry spruce wood**

TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
Short-term	Long-term		
17 <sup>a</sup>	12 <sup>a</sup>	Test chamber	WILKE et al., 2012
22 <sup>a</sup>	11 <sup>a</sup>	Test chamber	WILKE et al., 2012
41	No data	Test cell	ENGLUND, 1999
No data	30	Test cell	ENGLUND, 2010
75	< 50	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
100 – 175 <sup>b</sup>	10 – 20 <sup>b</sup>	Test chamber	STECKEL, WELLING & OHLMEYER, 2010, 2013; STECKEL, 2011
120 – 265 <sup>b</sup>	15 – 60 <sup>b</sup>	Test chamber	STECKEL, WELLING & OHLMEYER, 2010, 2013; STECKEL, 2011
156 – 119 <sup>b</sup>	26 – 18 <sup>b</sup>	Test cell	ENGLUND, 1999
165	52	Test chamber	LARSEN et al., 1998
206 – 2,645 <sup>b</sup>	No data	Test chamber	HASEGAWA et al. 2006
226	72	Test chamber	SAARELA et al., 1994 in ECA, 1997a
241 – 377 <sup>b</sup>	50 – 78 <sup>b</sup>	Test cell	ENGLUND, 1999
329 – 251 <sup>c</sup>	112 – 81 <sup>c</sup>	Test cell	ENGLUND, 1999
332 – 5,240 <sup>c</sup>	84 – 564 <sup>c</sup>	Test cell	ENGLUND, 1999
366	196	Test chamber	HYTTINEN et al., 2010
380	154	Test cell	LARSEN et al., 1998
394 – 480 <sup>b</sup>	122 – 171 <sup>b</sup>	Test cell	ENGLUND, 1999
448 – 325 <sup>c</sup>	111 – 115 <sup>c</sup>	Test cell	ENGLUND, 1999
575	331	Test cell	ENGLUND, 1999
587 – 2,228 <sup>d</sup>	226 – 618 <sup>d</sup>	Test chamber	STECKEL, WELLING & OHLMEYER, 2010, 2013
603	160	Test cell	ENGLUND, 1999
652 – 1,730 <sup>e</sup>	224 – 525 <sup>e</sup>	Test chamber	CZAJKA & FABISIAK, 2013
655	No data	Test cell	ENGLUND, 1999
1,400	No data	Test cell	RISHOLM-SUNDMAN et al., 1998

<sup>a</sup> Purchased spruce plywood

<sup>b</sup> Variations due to different drying processes

<sup>c</sup> Differentiation between knot-free (1<sup>st</sup> value) and knotty wood (2<sup>nd</sup> value)

<sup>d</sup> Differentiation between heartwood (1<sup>st</sup> value) and sapwood (2<sup>nd</sup> value)

<sup>e</sup> Differentiation between juvenile (1<sup>st</sup> value) and mature wood (2<sup>nd</sup> value)

Source: own representation according to the given sources

The resin acid content (and thus indirectly the oleoresin and terpene content) is the same or even lower in heartwood than in sapwood. In spruce, sapwood and heartwood differ mainly with regard to the content of free and esterified fatty acids, which are significantly more abundant in the sapwood than in the heartwood and decrease further toward the pith (EKMAN et al., 1979; WILLFÖR et al., 2003). In spruce sapwood, as in pine, triglycerides dominate, which are hydrolysed and degraded in the transition to the heartwood (EKMAN et al.,

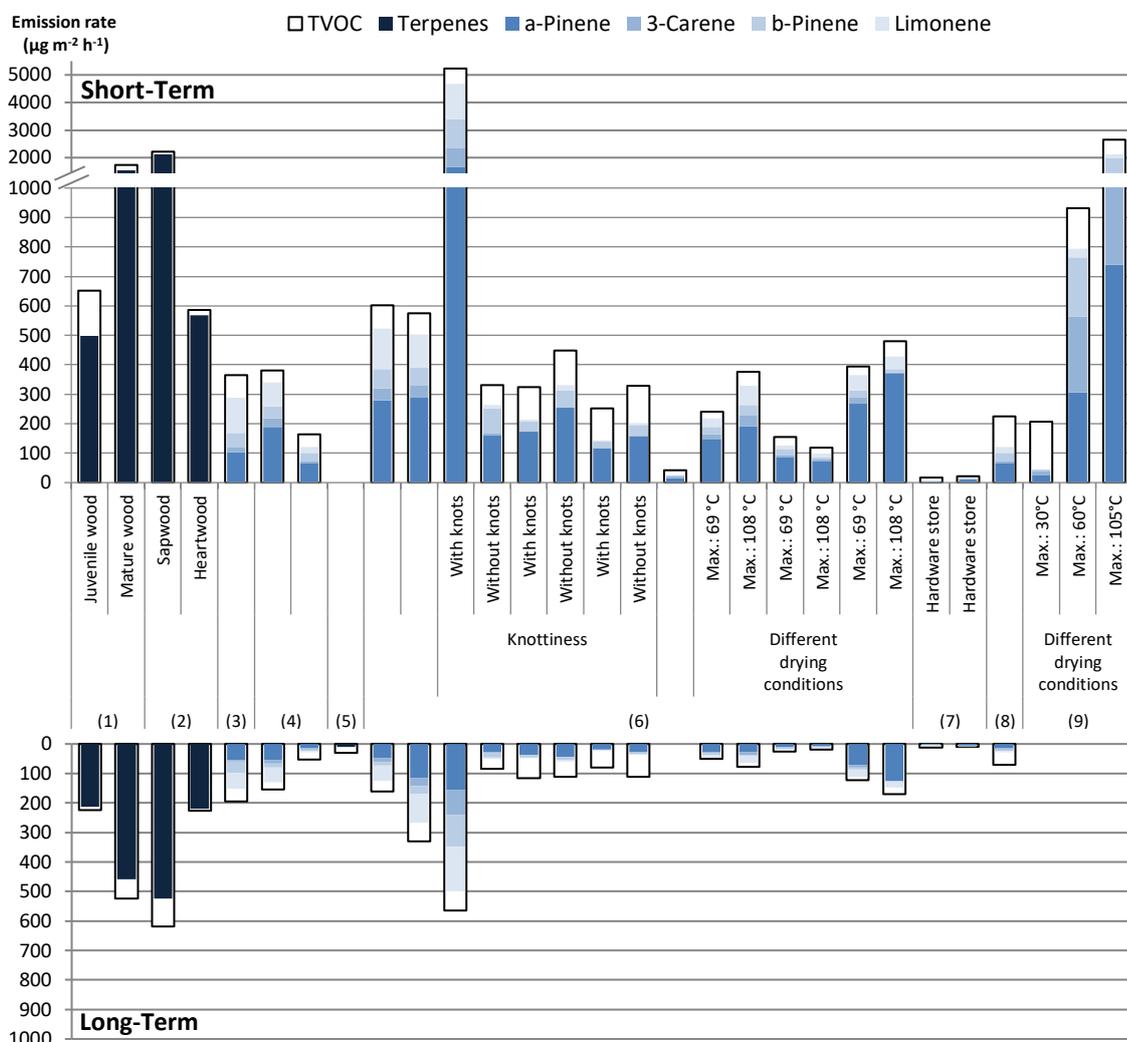
<sup>20</sup> Juvenile wood comprises the first growth rings, which are formed directly around the pith. Mature wood follows and forms the outer zone. The tissues differ in terms of their (sub)microscopic and macroscopic structure as well as in their chemical composition (CZAJKA & FABISIAK, 2013).

1979; WILLFÖR et al., 2003). In contrast to the radial distribution of the extractive content, EKMAN et al. (1979) found hardly any clear differences with increasing tree height. Spruce contains 10 % saturated, 29 % mono-unsaturated, 38 % di-unsaturated and 20 % tri-unsaturated fatty acids. The majority is made up of linoleic acid (36 %), oleic acid (25 %) and pinolenic acid (15 %) (HOLMBOM & EKMAN 1978). The composition of the fatty acids is thus basically comparable to that of pine, although spruce wood has lower amounts of bound and free fatty acids and a higher proportion of saturated compared to unsaturated fatty acids (ASSARSSON & ÅKERLUND, 1967; HOLMBOM & EKMAN, 1978). Lower aldehyde emissions are therefore generally expected from spruce.

### 4.2.2 Significant individual substances

The emissions are essentially composed of monoterpenes with a proportion of generally over 75 % (LARSEN et al., 1998; ENGLUND, 1999; HASEGAWA et al., 2006; HYTTINEN et al., 2010; STECKEL, 2011; CZAJKA & FABISIAK, 2013; Figure 5).

**Figure 5: Main VOCs emitted from spruce wood (short- and long-term behaviour)**



- (1) CZAJKA & FABISIAK, 2013
- (2) STECKEL et al., 2010, 2013
- (3) HYTTINEN et al., 2010
- (4) LARSEN et al., 1998
- (5) ENGLUND, 2010
- (6) ENGLUND, 1999
- (7) WILKE et al., 2012
- (8) SAARELA et al., 1994 in ECA, 1997a
- (9) HASEGAWA et al., 2006

Source: own representation according to the given sources

$\alpha$ -Pinene is the most important representative in terms of quantity - in addition,  $\beta$ -pinene and 3-carene are other important spruce monoterpenes. Compared to pine, limonene appears to play a greater role in some samples (Figure 5). In spruce with a low oleoresin content, the aldehydes are more prominent in percentage terms. As with pine, the di-unsaturated linoleic acid is one of the most common fatty acids, so that hexanal in particular, the main degradation product of this acid (EKMAN & HOLMBOM, 2000), is present in the emissions. However, both short- and long-term measurements revealed low values, below  $100 \mu\text{g m}^{-2} \text{h}^{-1}$  and  $50 \mu\text{g m}^{-2} \text{h}^{-1}$ , respectively (see Table 10 for sources).

### 4.3 Other conifers

European Larch (*Larix decidua* MILL., 102 million  $\text{m}^3$ ), silver fir (*Abies alba* MILL., 93 million  $\text{m}^3$ ) and Douglas fir (*Pseudotsuga menziesii* (MIRB.) FRANCO, 72 million  $\text{m}^3$ ) have a comparatively low timber stock in Germany (BMEL, 2016). According to the third National Forest Inventory, they accounted for around 6 % (4.2 million  $\text{m}^3$  harvested timber per year) of total timber use in the period from 2002 to 2012 (THÜNEN-INSTITUT, 2014).

There is hardly any emission data available for the wood species mentioned (Table 11). Both larch and Douglas fir have a vertical and horizontal resin canal system running through the tree (BACK et al., 2000). Fir, on the other hand, only forms traumatic (facultative) resin canals, which generally run axially. Resin canals in fir are therefore found only after stress events during tree growth (mechanical injuries or other external factors). Consequently, fir wood generally has a lower oleoresin content (BACK et al., 2000) and thus lower terpene emissions than the other coniferous wood species mentioned so far. The comparatively very low terpene emissions found by CZAJKA & FABISIAK (2014) (Table 11) are therefore attributed to the formation of traumatic resin canals.

**Table 11: TVOC emission rates of dry softwood**

Wood species	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
Fir	519 – 126 <sup>a</sup>	90 – 68 <sup>a</sup>	Test chamber	CZAJKA & FABISIAK, 2014
Larch	81	No data	Test chamber	CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011

<sup>a</sup> Differentiation between heartwood (1<sup>st</sup> value) and sapwood (2<sup>nd</sup> value)

Source: own representation according to the given sources

### 4.4 Hardwood

Hardwoods account for around 40 % of the total wood supply in German forests, with European beech (*Fagus sylvatica* L., 635 million  $\text{m}^3$ ) and pedunculate and sessile oak (*Quercus robur* L. and *Quercus petraea* (MATT.) LIEBL., 361 million  $\text{m}^3$ ) as the most common tree species. All other hardwoods (including maple, ash, birch and poplar) form a considerable total timber stock of 424 million  $\text{m}^3$  (BMEL, 2016), although the quantitative significance in relation to the individual tree species is low. On average, around 18.5 million  $\text{m}^3$  of hardwood were harvested annually between 2002 and 2012, which corresponds to around 24 % (beech: 14 %, oak: 4 %, other hardwoods: 6 %) of total wood use (THÜNEN-INSTITUT, 2014). However, native hardwoods only account for 13 % of the wood used in the construction sector. They are mainly used in flooring (42 % of hardwood use), in thermal insulation (17 %), in outdoor applications (12 %) and for doors (10 %), whereas their importance in structural applications has so far been rather low (MANTAU, DÖRING & HILLER, 2013). The silvicultural and forest policy objective of converting non-site-adapted pure coniferous stands into structurally rich deciduous and mixed forests will lead to an increase in both the forest area and the timber stock of deciduous trees in the future (BMEL, 2016). Despite increased efforts to develop new applications and markets for hardwood, coniferous wood will initially continue to play the dominant role in construction (MANTAU, DÖRING & HILLER, 2013).

#### 4.4.1 Sum of volatile organic compounds (TVOC)

Comparatively little data is available on the emission behaviour of hardwoods (Table 12).

**Table 12:** TVOC emission rates of dry hardwood

Wood species	TVOC ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
<i>Fagus sylvatica</i>	No data	20	Test cell	ENGLUND, 2010
<i>Fagus spec.</i>	30	29	Test cell	LARSEN et al., 1998
<i>Fagus sylvatica</i>	30	No data	Test cell	RISHOLM-SUNDMAN et al., 1998
<i>Fagus spec.</i>	45	11	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
<i>Fagus spec.</i>	86	No data	Test chamber	STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNIK, 2014
<i>Fagus sylvatica</i>	103 – 138 <sup>a</sup>	62 – 77 <sup>a</sup>	Test cell	ENGLUND, 1999
<i>Fagus spec.</i>	140	62	Test chamber	HORN et al., 2007
<i>Fagus spec.</i>	175	No data	Test chamber	GACA & DZIEWANOWSKA-PUDLISZAK, 2005; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011
<sup>a</sup> three test specimens from two stems				
<i>Quercus robur</i>	No data	26	Test cell	ENGLUND, 2010
<i>Quercus robur</i>	4 – 123 <sup>a</sup>	2 – 9 <sup>a</sup>	Test cell	ENGLUND, 1999
<i>Quercus spec.</i>	19	7	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
<i>Quercus robur</i>	24	No data	Test cell	ENGLUND, 1999
<i>Quercus spec.</i>	40	No data	Test chamber	GACA & DZIEWANOWSKA-PUDLISZAK, 2005; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011
<i>Quercus spec.</i>	96	No data	Test chamber	STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNIK, 2014
<i>Quercus spec.</i>	188	125	Test chamber	YRIEIX, MAUPETIT & RAMALHO, 2004
<i>Quercus robur</i>	203	16	Test chamber	VOLKMER et al., 2014
<i>Quercus robur</i>	210	No data	Test cell	RISHOLM-SUNDMAN et al., 1998
<sup>a</sup> two test specimens from one plank				
<i>Fraxinus excelsior</i>	No data	19	Test cell	ENGLUND, 2010
<i>Fraxinus spec.</i>	30	No data	Test chamber	GACA & DZIEWANOWSKA-PUDLISZAK, 2005; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011
<i>Fraxinus excelsior</i>	30	No data	Test cell	RISHOLM-SUNDMAN et al., 1998
<i>Fraxinus spec.</i>	30	6	Test chamber	LARSEN, FROST & WINTHER FUNCH, 2000
<i>Betula spec.</i>	65	54	Test chamber	LARSEN et al., 1998
<i>Betula spec.</i>	104	47	Test cell	SAARELA et al., 1994 in ECA, 1997a
<i>Betula pubescens</i>	110	No data	Test cell	RISHOLM-SUNDMAN et al., 1998
<i>Betula pendula</i>	378	No data	Test cell	ENGLUND, 1999
<i>Betula spec.</i>	No data	140	Test cell	ENGLUND, 2010
<i>Betula pendula</i>	482 – 908 <sup>a</sup>	No data – 591 <sup>a</sup>	Test cell	ENGLUND, 1999
<sup>a</sup> two test specimens from one plank				
<i>Populus alba</i>	20	17	Test chamber	ČECH & TESAŘOVÁ, 2015
<i>Populus tremula</i>	311	137	Test chamber	HYTTINEN et al., 2010
<i>Populus tremula</i>	No data	209	Test cell	ENGLUND, 2010

Wood species	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
<i>Alnus spec.</i>	50	No data	Test chamber	GACA & DZIEWANOWSKA-PUDLISZAK, 2005; CZAJKA & DZIEWANOWSKA-PUDLISZAK, 2011
<i>Alnus spec.</i>	No data	47	Test chamber	ENGLUND, 2010

Source: own representation according to the given sources

Native deciduous trees do not produce oleoresin and therefore do not emit terpenes. The total emissions are therefore qualitatively composed of fewer substances and are quantitatively very low, usually less than  $100 \mu\text{g m}^{-2} \text{h}^{-1}$  in long-term behaviour. Occasionally, higher emission rates have been observed in the fat-rich wood species birch and poplar (Chapter 4.4.2). According to the studies by RISHOLM-SUNDMAN et al. (1998), acetic acid emissions play an important role in hardwood. However, as these can only be inadequately captured using the standard method of VOC determination (Tenax/TD-GC-MS in accordance with DIN ISO 16000-6:2012) (RISHOLM-SUNDMAN et al., 1998; WIEGNER et al., 2012), the emission rates listed in Table 12, which were determined using this method, presumably underestimate the total emissions.

#### 4.4.2 Significant individual substances

##### Acetic acid

VOCs emitted from hardwood are characterised mainly by carbonyl compounds, in particular acetic acid and hexanal, as well as alcohols (RISHOLM-SUNDMAN et al., 1998). As already stated (Chapter 3.1.3), acetic acid cannot be detected with sufficient accuracy using Tenax<sup>®</sup> TA as a sorbent. RISHOLM-SUNDMAN et al. (1998), on the other hand, suggest the use of silica gel as the collection medium. The acetic acid is desorbed from it with sodium hydroxide and then analysed using ion chromatography. The comparison of the emission rates determined with both methods (Table 13) clearly shows that with Tenax adsorption and subsequent gas chromatographic analysis on a low/medium-polarity column, the acetic acid can be detected qualitatively, but not completely quantified. A large proportion of the emitted acetic acid was not determined. The study by ENGLUND (2010) provides comparable findings.

**Table 13: Acetic acid and TVOC emission rates of various wood species**

	Acetic acid emission rates in $\mu\text{g m}^{-2} \text{h}^{-1}$ (FLEC-cell, Silica gel <sup>a</sup> )	TVOC emission rates in $\mu\text{g m}^{-2} \text{h}^{-1}$ (FLEC-cell, Tenax <sup>b</sup> )
<i>Quercus robur</i>	2,800	210
<i>Prunus serotina</i>	2,000	60
<i>Hevea brasiliensis</i>	640	50
<i>Fraxinus excelsior</i>	370	30
<i>Acer saccharum</i>	270	20
<i>Fagus sylvatica</i>	250	30
<i>Pinus silvestris</i>	120	3,700
<i>Picea abies</i>	190	1,400
<i>Betula pubescens</i>	< 10	110

Material: dry, freshly planed wood

Time of measurement: no exact information (0.5 to 8 hours after the test cell was set up)

<sup>a</sup> Emission test cell method, adsorbent: Silica gel

<sup>b</sup> Emission test cell method, adsorbent: Tenax; no detailed listing of acetic acid emissions

Source: own representation according to RISHOLM-SUNDMAN et al. (1998)

OHLMEYER & STECKEL (2012) also compared two different measurement methods for determining acetic acid emissions from beech. On the one hand, they carried out the VOC measurement according to DIN ISO 16000-6:2012, on the other hand, they passed the test chamber air through sodium hydroxide solution and analysed it using ion chromatography (Table 14). Compared to the results of RISHOLM-SUNDMAN et al. (1998) and the emission data listed in Table 12, they determined significantly higher acetic acid emissions from beech. This can only partly be explained by different test conditions and measurement times. In addition, they observed only minor differences between the two measurement methods used.

**Table 14: Acetic acid emission rates from beech wood**

	Acetic acid emissions in $\mu\text{g m}^{-2} \text{h}^{-1}$	
	Chamber test, IC <sup>a</sup>	Chamber test, Tenax <sup>b</sup>
<b>3-day value</b>	1,300 – 1,900	1,500 – 2,000
<b>28-day value</b>	400 – 500	500

Material: dry, freshly planed beech plank stored for 3 years

<sup>a</sup> Emission test chamber method, absorbent: sodium hydroxide solution

<sup>b</sup> Emission test chamber method, adsorbent: Tenax

Source: own representation according to OHLMEYER & STECKEL (2012)

A number of scientists have explicitly focussed on the release of acetic acid from wood. Apart from the influence of acetic acid on indoor air quality and thus potentially on the human organism, it can also cause undesirable reactions with some materials. Several studies examined the extent to which, and under which conditions, acetic acid emissions can cause damage to exhibits in museums (e.g. in display cases, showcases and storage cabinets or rooms) (GRZYWACZ, 2006; SCHIEWECK & SALTHAMMER, 2009; GIBSON & WATT, 2010). FARMER (1962a, b), for example, reports on the corrosive effect of wood on metals, which occurs particularly in warm and humid climatic conditions. The metal does not have to be in direct contact with the wood, but it is also sufficient if it is in the vapour space of the wood.

Acetic acid is formed in wood in the presence of water as a result of the hydrolysis of the acetyl groups of the hemicelluloses (PACKMAN, 1960; RÜCKERT, 1986; GIBSON & WATT, 2010). PACKMAN (1960) showed that the amount of acetic acid formed corresponds well with the decrease in bound acetyl groups. According to

GRZYWACZ (2006), acetic acid can also be formed during the oxidation of acetaldehyde, provided that an oxidising agent (e.g. peroxide or ozone) is available. Acetaldehyde (ethanal) is a very volatile organic compound (VOC) and is also emitted by wood (LARSEN et al., 1998; LARSEN, FROST & WINTHER FUNCH, 2000; HASEGAWA et al., 2006; HORN et al., 2007). However, it is not known to what extent this formation pathway plays a role in wood products. Therefore, it is assumed that the formation from acetyl groups of hemicelluloses represents the main source of acetic acid emissions. After cellulose, hemicelluloses are the second most important polysaccharide group in the structural substance of wood. In contrast to cellulose, they are made up of different sugar molecules, have a lower degree of polymerisation and form side branches on their main molecular chains. Their proportion in softwood is around 15 - 20 %, whereas in hardwood it is 25 - 35 %. In addition to this difference in the total content, the composition of the hemicelluloses also differs between hardwoods and softwoods. The former consists mainly of pentosans (above all xylans), while the latter are composed of hexosans (above all mannans). Some of the hydroxyl groups of hemicelluloses are substituted with acetyl groups (FENGEL & WEGENER, 1989). The content of acetyl groups is species-specific and ranges from 1 to 6 % of the dry weight of the wood (RÜCKERT, 1986). However, the results of PACKMAN (1960), ROFFAEL (1989) and several sources in FENGEL & WEGENER (1989) show that the content is often higher in hardwood than in softwood (Table 15). Acetic acid emissions can therefore be expected from all woods. PACKMAN (1960) assumes that the acetic acid present in the wood is formed during industrial drying. He sees this theory confirmed by the fact that no free acetic acid could be detected in either fresh or air-dried birch wood. In addition, according to FARMER (1962a), kiln-dried wood has a stronger corrosive effect on metals than air-dried wood. He assumes that some of the acetic acid formed from the split acetyl groups at elevated temperatures remains in the wood, especially in material with larger diameter. In air-dried wood, on the other hand, the formation of acetic acid is comparatively low, so that the rates of formation and emission are roughly balanced (FARMER, 1962a; PACKMAN, 1960).

**Table 15: Acetyl group content of native wood species**

Wood species	Acetyl group content (%)	Source
<i>Pinus sylvestris</i>	1.6	several sources in FENGEL & WEGENER,1989
Pine wood	1.4	ROFFAEL, 1989
Spruce wood	1.3	ROFFAEL, 1989
<i>Pseudotsuga menziesii</i>	1.1	PACKMAN, 1960
Beech wood	4.2	ROFFAEL, 1989
Oak wood	3.8	ROFFAEL, 1989
<i>Betula spec.</i>	3.6	PACKMAN, 1960
<i>Fagus sylvatica</i>	3.2	PACKMAN, 1960
<i>Robinia pseudoacacia</i>	2.7	several sources in FENGEL & WEGENER,1989
<i>Quercus spec.</i>	2.6	PACKMAN, 1960

Source: own representation according to the given sources

GIBSON & WATT (2010) found in 14 analysed wood species that hardwoods generally emit more acetic acid than softwoods, which can be explained, among other things, by the lower acetyl group content of the latter. However, larch<sup>21</sup> and red pine emitted at about the same level as oak<sup>21</sup> and beech<sup>21</sup> (Table 16). Thus, there is not necessarily a direct correlation between the acetyl group content of the wood and the amount of volatile acids emitted (PACKMAN, 1960; ROFFAEL, 1989). The content of acetyl groups only represents the total amount of acetic acid that can theoretically be formed (FARMER, 1962a). Apart from this, the main influencing factors are the rate of hydrolysis and release of acetic acid, factors which are also specific to the wood species and can also be influenced by various parameters (PACKMAN, 1960; RÜCKERT, 1986). Increased temperature and relative

<sup>21</sup> Exact species is missing in the source, only the genus is provided.

humidity (RH) accelerate the release of the acetic acid already present in the wood and also promote its further formation (PACKMAN, 1960; RÜCKERT, 1986; ROFFAEL, 1989; GIBSON & WATT, 2010). OHLMEYER & STECKEL (2012) determined that, in particular with increasing humidity (25, 50 and 75 % RH), there is a significant increase in acetic acid emissions from beech, which they attributed to the interaction of the polar substances water and acetic acid. Comparable results were also obtained by GIBSON & WATT (2010) for a number of wood species when the relative humidity was increased from 6 to 54 % RH. However, a further increase to 100 % RH did not lead to a further rise in acetic acid emissions in all the wood species analysed (especially not in the conifers) (Table 16). The wood density and the pH value of the wood extract, on the other hand, did not correlate with the level of acetic acid emissions (GIBSON & WATT, 2010).

**Table 16: Acetic acid emissions of native wood species at 20 °C and various relative humidity (RH) levels**

Wood species	Acetic acid concentration ( $\mu\text{g m}^{-3}$ )		
	6 % RH	54 % RH	100 % RH
<i>Taxus spec.</i>	<10	132 ± 139	115 ± 2
<i>Pinus spec.</i>	<10	447 ± 90	390 ± 180
<i>Pseudotsuga menziesii</i>	<10	495 ± 32	415 ± 59
<i>Betula spec.</i>	32 ± 25	698 ± 38	1,650 ± 249
<i>Fraxinus spec.</i>	25 ± 22	883 ± 37	2,610 ± 200
<i>Prunus spec.</i>	13 ± 5	952 ± 48	1,280 ± 91
<i>Fagus spec.</i>	37 ± 4	1,500 ± 27	2,080 ± 58
<i>Quercus spec.</i>	253 ± 70	1,740 ± 15	841 ± 361
<i>Larix spec.</i>	<10	1,890 ± 297	1,040 ± 267

**Method: Wood samples were placed in a desiccator without air supply (static conditions). The acetic acid concentration of the vapour space above the wood sample was measured after reaching the equilibrium concentration (7 days) with a passive sampler and analysed using ion chromatography.**

Source: own representation modified according to GIBSON & WATT (2010)

A conclusive assessment of the individual wood species with regard to their acetic acid emissions is hardly possible due to the insufficient data available. However, several studies (PACKMAN, 1960; ROFFAEL, 1989; RISHOLM-SUNDMAN et al., 1998; ENGLUND, 2010; GIBSON & WATT, 2010), albeit with very different methodologies, indicate that oak wood emits comparatively high levels of acetic acid. Area-specific emission rates of several hundred (ENGLUND, 2010) to several thousand  $\mu\text{g m}^{-2} \text{h}^{-1}$  (RISHOLM-SUNDMAN et al., 1998, Table 13) were observed. OHLMEYER & STECKEL (2012) also determined quite high emission rates for beech wood with up to 2,000  $\mu\text{g m}^{-2} \text{h}^{-1}$  after 3 days and up to 500  $\mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days of testing. In comparison, beech wood analysed by AKRAMI (2014) emitted lower quantities of acetic acid (day 3 = 300  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). However, the emission rates were significantly higher than for poplar wood (day 3 = 80  $\mu\text{g m}^{-2} \text{h}^{-1}$ ).

### Aldehydes

Native hardwoods do not have a resin canal system, but have a comparable parenchyma resin composition to conifers, consisting of free and esterified fatty acids (triglycerides), sterols and their esters as well as free and ester-bound triterpene alcohols (EKMAN & HOLMBOM, 2000). In addition to acetic acid, aldehyde emissions are also to be expected in hardwoods, which are formed from the fatty acids as a result of autoxidation processes (Chapter 4.1.2). As with softwoods, the fatty acids are located in the parenchyma tissue of the sapwood (especially in the radial rays), but are subject to redistribution in the heartwood. The qualitative composition of the fatty acids does not differ significantly from that of conifers, except for the absence of pinolenic acid and taxoleic acid. The quantity of unsaturated fatty acids, which are preferentially autoxidised, also dominates over saturated fatty acids. Oleic, linoleic and linolenic acids are often among the most important substances (EKMAN

& HOLMBOM, 2000). However, the content and percentage composition of the fatty acids are species-specific. Birch and poplar in particular are among the more fat-rich deciduous trees. Accordingly, significant aldehyde emissions were only found in these two wood species (compared to the other species listed in Table 12). Birch wood (*Betula pendula*) contains mainly linoleic acid (approx. 60 %), followed by palmitic acid (16:0, approx. 10 %) and oleic acid (approx. 8 %). In addition to linoleic acid (approx. 50 %), linolenic acid (approx. 40 %) plays a greater role in poplar (*Populus tremula*) (several sources in EKMAN & HOLMBOM, 2000). The aldehydes determined in emission measurements are mainly composed of hexanal and pentanal, with the former usually dominating (LARSEN et al., 1998; RISHOLM-SUNDMAN et al., 1998; ENGLUND, 1999; HYTTINEN et al., 2010). The hexanal emission rates from birch and poplar wood can reach several hundred  $\mu\text{g m}^{-2} \text{h}^{-1}$ <sup>22</sup>, whereas they are generally below 100  $\mu\text{g m}^{-2} \text{h}^{-1}$  for the other hardwoods (see Table 12 for sources).

In addition to acetic acid and aldehydes, other substances (including alcohols, alkanes, alkenes and other acids) were detected in some studies (ENGLUND, 1999; HYTTINEN et al., 2010; OHLMEYER & STECKEL, 2012), but these do not play a major role due to their low emission rates.

## 4.5 Summary

Compared to other wood properties, the amount of available data on the emission behaviour of different wood species is limited. Scots pine, which has received particular attention within the context of VOC emissions due to its sometimes rather high terpene emissions, has been analysed the most extensively. A major issue with the existing data is that it is only partially comparable. On the one hand, the emission behaviour of a material depends on numerous factors related both to the material itself and to its manufacturing conditions. In the case of solid wood, these are primarily the chemical composition of the wood (main components and extractives) as well as the drying and storage conditions. On the other hand, the magnitude of VOC emission rates is also strongly influenced by the emission testing procedure itself, so that identical material does not necessarily lead to identical emission rates across different research institutions. In contrast to mechanical properties of wood, for example, the area-specific emission rates of individual VOCs are not fixed, stable values, but rather represent a snapshot at a specific point in time (Figure 6). The starting point of the emission test therefore has a decisive influence on the result.

The entire spectrum of VOCs from wood can comprise several dozen substances. The most relevant compound classes include terpenes, aldehydes and organic acids. In contrast to hardwood, softwood is primarily characterised by its terpene emissions. Scots pine is considered a particularly strong terpene emitter – especially of  $\alpha$ -pinene and 3-carene – with the heartwood showing the highest levels. All other native softwoods generally emit significantly lower quantities of terpenes. However, exceptions prove the rule – certain events (mechanical damage, storms, drought, insect infestation, etc.) during the lifetime of the trees can also initiate increased oleoresin formation and thus lead to elevated terpene emissions during wood utilisation. Due to the sometimes high terpene emissions from pine, the TVOC of unseasoned, freshly planed pine heartwood can be in the lower double-digit  $\text{mg m}^{-2} \text{h}^{-1}$  range. After one month of emission testing, a TVOC in the maximum single-digit  $\text{mg m}^{-2} \text{h}^{-1}$  range can be expected. However, generalisations in the form of an average value with regard to pine emissions can hardly be made. On the one hand, the available data and the large natural variability of wood do not support such an approach; on the other hand, it would not be meaningful. The same applies to the enantiomer composition of terpenes emitted from pine wood. While 3-carene appears to be present almost exclusively in the (+)-form,  $\alpha$ -pinene occurs in various mixtures of (+)- and (-)-enantiomers. However, corresponding studies based on wood emissions are lacking – the composition has so far only been determined

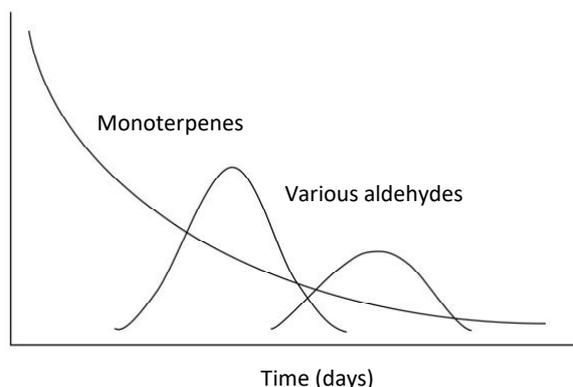
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<sup>22</sup> Maximum values determined: birch = 660  $\mu\text{g m}^{-2} \text{h}^{-1}$  (day 3) and 450  $\mu\text{g m}^{-2} \text{h}^{-1}$  (day 28), poplar = 220  $\mu\text{g m}^{-2} \text{h}^{-1}$  (day 3) (ENGLUND, 1999; HYTTINEN et al., 2010)

in wood extractives or turpentine oils. Nevertheless, knowledge of the chirality is of great importance for the health assessment of pine wood insofar as the enantiomers can often cause different effects.

**Figure 6: Different emission profiles of primary emissions, e.g. monoterpenes, and secondary emissions, e.g. aldehydes formed through autoxidation processes of unsaturated fatty acids**

Area-specific emission rate



Source: own representation modified according to Back et al. (2000)

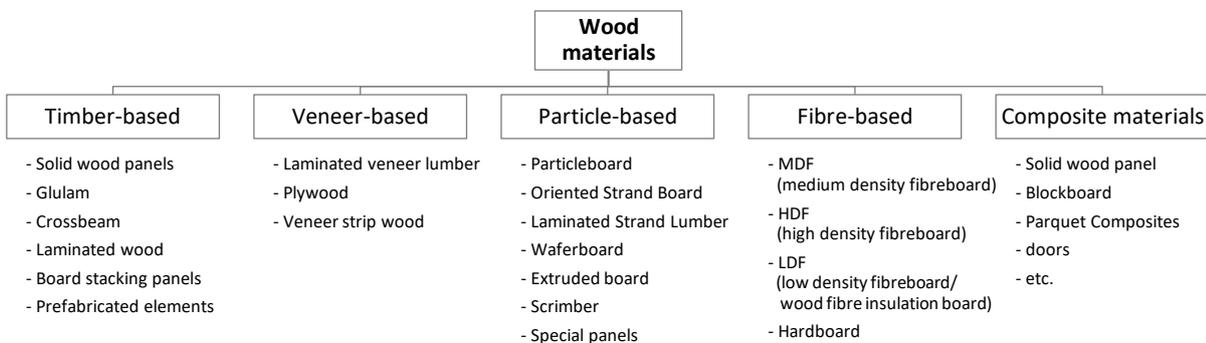
In contrast to terpenes, aldehydes are not inherent components of wood, but are only formed as a result of autoxidation processes from unsaturated fatty acids – a process that is quite complex due to the large number of involved fatty acids and reaction pathways. In contrast to terpenes, which are classified as primary emissions, these are therefore considered secondary emissions. As fats and fatty acids are present in all native wood species, aldehyde emissions can be expected, albeit in very small quantities. Comparatively high aldehyde emissions of several hundred  $\mu\text{g m}^{-2} \text{h}^{-1}$  are only to be expected from wood species rich in fats (pine, birch and poplar) and when conditions favouring autoxidation (especially high temperatures) prevail. Within this substances group, hexanal predominates, as it is the main degradation product of linoleic acid, which in turn is the most frequently occurring unsaturated fatty acid found in these woods.

Acetic acid is another substance that is emitted by all woods. Its formation primarily results from the hydrolytic cleavage of the acetyl groups from the hemicelluloses, so it is also categorised as a secondary emission. The concentration of acetic acid in the air cannot be adequately quantified using the standard method for determining VOCs (DIN ISO 16000-6:2012). The results from studies that have worked exclusively according to this method are therefore presumably not sufficiently reliable for this compound. However, few alternative studies are available. Oak wood and to some extent also beech wood appears to be characterised by comparatively high acetic acid emissions (emission rates up to the lower single-digit  $\text{mg m}^{-2} \text{h}^{-1}$  range possible). A conclusive assessment of other wood species cannot be made due to the insufficient data available. It is generally assumed that most softwoods emit lower levels of acetic acid compared to hardwoods – although exceptions certainly confirm the rule here too.

## 5 Emission behaviour of wood-based materials

Wood-based materials consist of wood elements that have been broken down and then reassembled, usually using adhesives as binding agents (NIEMZ & WAGENFÜHR, 2012). Depending on the wood-based material, the degree of decomposition of the wood ranges from fibres, chips and veneers to solid wood (Figure 7). Urea resins (UF<sup>23</sup> and MUF<sup>24</sup>) are mainly used as adhesives for particleboard and MDF. European OSB production, on the other hand, relies primarily on polymeric diphenylmethane diisocyanate (PMDI). Phenol-formaldehyde resins (PF) are mainly employed for plywood and laminated veneer lumber (MANTANIS et al., 2018). In addition, additives (e.g. hardeners<sup>25</sup>, formaldehyde scavengers<sup>26</sup>, paraffins<sup>27</sup>, flame retardants<sup>28</sup>) can be added to the materials, which are process-related or necessary to achieve certain product properties (NIEMZ & WAGENFÜHR, 2012; MANTANIS et al., 2018).

**Figure 7: Categorisation of wood-based materials**



Source: own representation modified according to NIEMZ & WAGENFÜHR (2012)

The total production capacity of the German engineered wood industry in 2015 was 12.5 million m<sup>3</sup>. Despite the significant reduction in particleboard production over the last 20 years, this product group is still the most important wood-based material in terms of volume at 5.7 million m<sup>3</sup>. A structural change has taken place, which has manifested itself in increased capacity expansion for OSB, MDF/HDF and LDF (Table 17, DÖRING, GLASENAPP & MANTAU, 2017).

Together with Romania and Poland, Germany is the main producer of OSB in Europe (MANTANIS et al., 2018). OSB production has been established in Germany since 2001 and currently has three production sites in the north-east of the country. In 2015, the production capacity was around 1.3 million m<sup>3</sup> with a capacity utilisation of 92 %. 1.6 million m<sup>3</sup> wood were used for this, with DÖRING, GLASENAPP & MANTAU (2017) assuming that this was industrial softwood<sup>29</sup>. In contrast, lower-cost assortments are generally used for particleboard (sawmill by-

<sup>23</sup> Urea-formaldehyde resin

<sup>24</sup> Melamine-urea-formaldehyde resin

<sup>25</sup> Accelerate the polymerisation reaction of the adhesive

<sup>26</sup> Reduce formaldehyde emissions by binding free formaldehyde

<sup>27</sup> Serve as hydrophobic agents

<sup>28</sup> Impart fire-retardant properties to the material

<sup>29</sup> Industrial wood: Raw wood (round wood) that is to be mechanically, mechanically-chemically or chemically broken down

products<sup>30</sup>, waste wood<sup>31</sup>, other industrial wood residues<sup>32</sup>, bark) – the industrial wood share of softwood and hardwood is only around 25 % in total. LDF production capacities in particular have risen sharply in recent years to 1.4 million m<sup>3</sup>, although only 67 % of capacity was utilised in 2015, meaning that total production has fallen slightly compared to 2010. Sawmill by-products are primarily used for this product group. In comparison, industrial wood played a larger role for MDF/HDF with a share of 56 % (Figure 8, DÖRING, GLASENAPP & MANTAU, 2017).

**Table 17: Development of the production capacity of various wood-based materials in Germany**

Year	Particleboard (million m <sup>3</sup> )	OSB (million m <sup>3</sup> )	MDF/HDF (million m <sup>3</sup> )	LDF (million m <sup>3</sup> )	Total (million m <sup>3</sup> )
1999	9.735	0.000	3.190	0.250	13.175
2001	8.974	0.650	2.990	0.250	12.864
2003	8.758	1.105	3.550	0.250	13.663
2005	8.062	1.235	4.018	0.495	13.810
2010	7.519	1.207	4.432	1.054	14.213
2015	5.683	1.328	4.153	1.368	12.531

Source: own representation according to DÖRING, GLASENAPP & MANTAU (2017)

In the construction sector, wood-based materials account for 28 % of the semi-finished wood products used, with wood fibre insulation boards making up over half of this. OSB, with a share of 4 %, is primarily used in structural applications (usually as a substructure in walls and floors), but is also employed in visible areas as cladding or flooring. Particleboards have a comparable range of applications, but play a smaller role in construction than OSB - its main area of use is in furniture manufacturing. MDF and HDF account for 6 % of the semi-finished wood products used in the construction sector and are primarily employed in roof construction and flooring (Figure 8).

The following chapters describe the emission behaviour of veneer-, particle- and fibre-based panels, i.e. wood-based materials in the narrower sense. For solid wood materials, particularly those mechanically joined (nails, dowels, screws, etc.), emissions are generally expected to be comparable to those of solid wood. The adhesives used in the wood-based panel industry (UF, MF, PF and PMDI) are generally not a source of VOCs<sup>33</sup>. Emissions are attributed to the wood itself and its processing (RISHOLM-SUNDMAN, 2002). The individual process steps from raw material to wood-based material can be traced using the diagrams shown in Figure 10. The raw material composition of the wood-based materials (Figure 8), in conjunction with the wood species-specific emission behaviour (Chapter 4), provides initial insights into the emissions to be expected from each panel type. However, the degree of wood disintegration (usually mechanical, sometimes following hydrothermal pre-treatment of the raw material), which increases the specific wood surface area, as well as the thermal exposure during wood drying and hot pressing, are important steps that can influence the subsequent emission behaviour of the wood-based material.

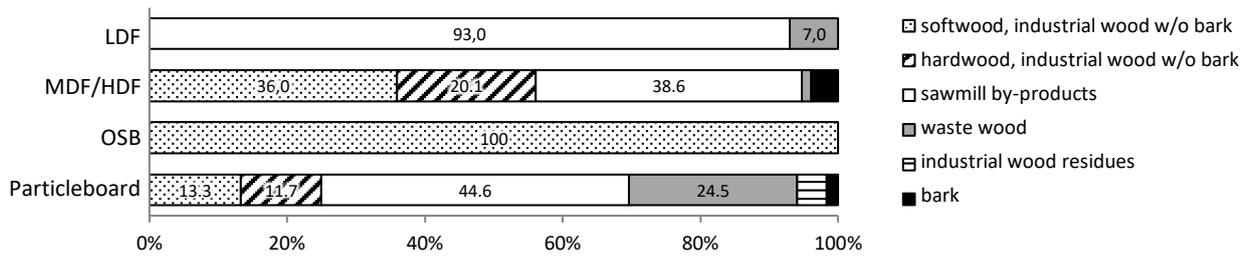
<sup>30</sup> Wood residues generated in sawmills during the processing of round timber into sawn timber or during the further processing of sawn timber (sawdust, slabs/splinters, wood chips, shavings, etc.)

<sup>31</sup> Industrial wood residues or used wood that arises as waste in accordance with §2 of the German Waste Wood Ordinance (DE: *Altholzverordnung*), provided it has already been used at least once as a final product.

<sup>32</sup> Residual wood from other sectors of the wood processing industry or trade

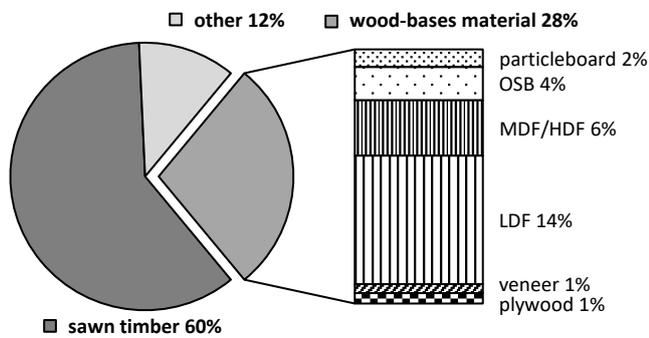
<sup>33</sup> According to the WHO definition (Chapter 1) and DIN ISO 16000-6:2012, formaldehyde is not a VOC but a VVOC.

**Figure 8: Wood consumption in 2015 by product group and type of raw material**



Source: own representation according to DÖRING, GLASENAPP & MANTAU (2017)

**Figure 9: Semi-finished wood products used in construction in 2012**



100 % = 13.4 million m<sup>3</sup> (volume occupied by the products in the building)

Source: own representation according to MANTAU, DÖRING & HILLER (2013)



## 5.1 OSB (Oriented Strand Board)

OSB belong to the particle-based materials which, in contrast to particleboard, are made up of strands (large, elongated wood particles). Compared with conventional particles, these strands have a greater length (75 - 150 mm), width (15 - 25 mm) and thickness (0.3 - 0.7 mm). OSB are characterised by an oriented alignment of the strands – in the surface layer parallel to the production direction, in the middle layer perpendicular to the surface layer or randomly distributed (IRLE & BARBU, 2010). Compared to particleboard, OSB of the same density achieves higher strength values (NIEMZ & WAGENFÜHR, 2012). OSB produced in Europe consists mainly of softwood, mostly pine. Spruce, Douglas fir and some hardwood species are probably also used to a lesser extent. In North America, on the other hand, poplar (northern production sites) and various hardwoods and softwoods (south-eastern production sites) are mainly employed (PICHELIN, 2002). Another difference concerns the adhesives used: North American producers largely rely on PF, whereas PMDI dominates in Europe (MANTANIS et al., 2018). In the past, combinations with PMDI in the middle layer and MUPF in the top layer were also frequently used (PICHELIN, 2002). In Europe, OSB is predominantly manufactured for load-bearing applications intended for use in humid conditions (type OSB/3<sup>34</sup>) (MANTANIS et al., 2018).

### 5.1.1 Emission behaviour

Pine-based OSB supplied directly after production emit relatively large quantities of VOCs due to the comparatively high terpene emissions of pine. Various factors, such as board thickness, process parameters and storage conditions, influence the total emissions (Table 18). Apart from this, the considerable range of the emission behaviour of pine also has an effect on the emission level of OSB. As expected, there is a clear decrease in emission rates with increasing storage time (Table 18). For OSB, it is also evident that, at least for freshly manufactured boards, a measurement period of only 28 days does not reflect the long-term behaviour, as a further, significant reduction in emissions was also observed beyond this period (HORN et al., 2007; MAKOWSKI, 2007; OHLMEYER et al., 2008a; DÄUMLING et al., 2009; OHLMEYER & STECKEL, 2012; WILKE et al., 2012). MAKOWSKI (2007) and WILKE et al. (2012), for example, determined, using fresh or recently produced boards, that the total emissions are reduced to a third of the 28-day value after around eight weeks. The TVOC range shown in Table 18 as a function of the storage period expresses this fact.

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<sup>34</sup> In accordance with DIN EN 300:2006, OSB are categorised into four quality classes according to their area of application.

**Table 18: TVOC emission rates of pine-based OSB as a function of storage duration (days after manufacture)**

Storage time	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Influencing factors	Source
	Short-term	Long-term			
1	11,172 – 7,420	1,328 – 933	Test chamber	Pressing time factor (1 <sup>st</sup> value: 8 s/mm, 2 <sup>nd</sup> value: 12 s/mm), T <sub>strand</sub> : 120 °C	(1)
	9,825 – 8,401	1,596 – 1,368	Test chamber	Pressing time factor (1 <sup>st</sup> value: 8 s/mm, 2 <sup>nd</sup> value: 12 s/mm), T <sub>strand</sub> : 170 °C	(1)
	7,290 – 6,649	1,458 – 1,405	Test chamber	Chamber size (1 <sup>st</sup> value: 0,023 m <sup>3</sup> , 2 <sup>nd</sup> value: 1 m <sup>3</sup> )	(1)
	6,888 – 2,959	1,236 – 603	Test chamber	Pressing temperature (1 <sup>st</sup> value: 180 °C, 2 <sup>nd</sup> value: 260 °C)	(1)
	6,630	1,935	Test chamber		(1)
	6,363 – 3,795	1,144 – 854	Test chamber	Panel thickness (1 <sup>st</sup> value: 26 mm, 2 <sup>nd</sup> value: 16 mm)	(2)
	4,870 – 3,198	985 – 697	Test chamber	Particle size in face layer (1 <sup>st</sup> value: strands, 2 <sup>nd</sup> value: particles)	(1)
3	10,692	1,350	Test chamber	Storage in stack	(1)
5	6,631 – 2,513	2,155 – 861	Test chamber	Pine from lower (1 <sup>st</sup> value) and upper (2 <sup>nd</sup> value) trunk section, T <sub>strand</sub> : 400 °C	(3)
	3,596 – 2,210	1,423 – 962	Test chamber	Pine from lower (1 <sup>st</sup> value) and upper (2 <sup>nd</sup> value) trunk section, T <sub>strand</sub> : 250 °C	(3)
7	5,456	871	Test chamber		(3)
19	4,767 – 3,896	No data – 1,189	Test chamber	T <sub>strand</sub> (1 <sup>st</sup> value: 250 °C, 2 <sup>nd</sup> value: 400 °C), PT: 190 °C	(3)
	3,250 – 2,525	925 – 921	Test chamber	T <sub>strand</sub> (1 <sup>st</sup> value: 400 °C, 2 <sup>nd</sup> value: 250 °C), PT: 250 °C	(3)
68	742 – 633	525 – 502	Test chamber	Pine from lower (1 <sup>st</sup> value) and upper (2 <sup>nd</sup> value) trunk section, T <sub>strand</sub> : 250 °C	(3)
	668 – 407	527 – 364	Test chamber	Pine from lower (1 <sup>st</sup> value) and upper (2 <sup>nd</sup> value) trunk section, T <sub>strand</sub> : 400 °C	(3)
180	2,595	819	Test chamber	Storage in stack	(1)
309	83	30	Test chamber		(3)

T<sub>strand</sub> = drying temperature of strands

(1) MAKOWSKI (2007)

(2) OHLMEYER et al. (2008a)

(3) WILKE et al. (2012)

Source: own representation according to the given sources

In practice, OSB is trimmed after the pressing process, cooled and stored in stacks (OHLMEYER et al., 2008b). Accordingly, commercially purchased OSB<sup>35</sup>, which can be assumed to have been stored for a certain period of time after production, have a lower emission level compared to freshly manufactured boards, whereby the difference is particularly noticeable during the first few days of emission measurement (Table 19).

The qualitative and quantitative composition of the emissions is a consequence of the emission behaviour of the raw material used, in this case pine (Chapter 4.1), as well as the influence of the individual manufacturing steps leading to the wood-based material (Figure 10). The emissions from OSB are therefore mainly composed of terpenes, in particular  $\alpha$ -pinene and 3-carene, and aldehydes, in particular hexanal (Figure 11, Figure 12). The main differences compared to solid wood are the increased (un)saturated aldehyde emissions and the reduced terpene emissions. According to MAKOWSKI (2007), both effects are related to the higher specific surface area of the particles compared to solid wood as well as the increased thermal exposure during drying of the strands and the hot-pressing process. After the wood has been chipped, the strands are dried to a moisture content of around 2 % at temperatures ranging from 100 °C (dryer outlet) to 250 °C (dryer inlet). During pressing, the mat is subjected to a temperature of around 250 °C in the surface layer (OHLMEYER et al., 2008b). This means that

<sup>35</sup> In studies of the emission behaviour of OSB purchased from the trade (Table 19), it is assumed that these are also OSB made of pine wood, provided they are European studies.

some of the terpenes already emit during OSB production. However, the strands also offer a larger surface area for the parameters that favour the autoxidation of the fatty acids, so that in combination with the high temperatures, increased fatty acid degradation is initiated. In contrast to the terpene emissions, which subside comparatively quickly, the aldehyde emissions from freshly produced boards initially rise and then fall again after a certain point in time (MAKOWSKI, 2007). This process is the result of the radical chain reaction, which takes place autocatalytically. The hydroperoxides formed from the fatty acids decompose into radicals, promoting further hydroperoxide formation. The hydroperoxide concentration consequently increases rapidly, reaches a maximum and then slowly decreases again as a result of its degradation to stable end products, in particular aldehydes (NAWAR, 1984; GROSCH, 1987). According to studies by MAKOWSKI (2007) and OHLMEYER et al. (2008a, b), the peak in aldehyde emissions from OSB is reached in the first few weeks after production. Since the fatty acids present in pine have different oxidation rates and degradation products (Chapter 4.1.2), the timing and magnitude of the emission peak differ among individual aldehydes (Figure 6). Consequently, freshly produced pine-based OSB shows a change from terpene-dominated emissions (aldehyde content of only a few percent) to emissions with a higher aldehyde content (around 40 %) within the first four weeks (MAKOWSKI, 2007; OHLMEYER et al., 2008a, b; WILKE et al., 2012; Figure 11).

**Table 19: TVOC emission rates from OSB, commercially available products**

Storage time	TVOC ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		Test procedure	Source
	Short-term	Long-term		
No data	2,741	877	Test chamber	WILKE et al. (2012)
	2,424	934	Test chamber	WILKE et al. (2012)
	2,200	500	Test chamber	OHLMEYER & STECKEL (2012)
	2,100	400	Test chamber	OHLMEYER & STECKEL (2012)
	1,900	550	Test chamber	HORN et al. (2007)
	1,820 – 1,515 <sup>a</sup>	497 – 507 <sup>a</sup>	Test chamber	WILKE et al. (2012)
	1,558	544	Test chamber	WILKE et al. (2012)
	1,480	430	Test chamber	HORN et al. (2007)
	1,476	921	Test chamber	WILKE et al. (2012)
	1,400	480	Test chamber	HORN et al. (2007)
	1,400	300	Test chamber	HORN et al. (2007)
	1,252	680	Test chamber	HORN et al. (2007)
	1,250	500	Test cell	DÄUMLING et al. (2009)
	990	180	Test cell	RISHOLM-SUNDMAN (2002)
	982	493	Test chamber	WILKE et al. (2012)
	900	400	Test chamber	HORN et al. (2007)
	450	170	Test chamber	HORN et al. (2007)
	214 <sup>b</sup>	83 <sup>b</sup>	Test chamber	WILKE et al. (2012)
	184	144	Test chamber	WILKE et al. (2012)

<sup>a</sup> sanded (1<sup>st</sup> value) and unsanded (2<sup>nd</sup> value)

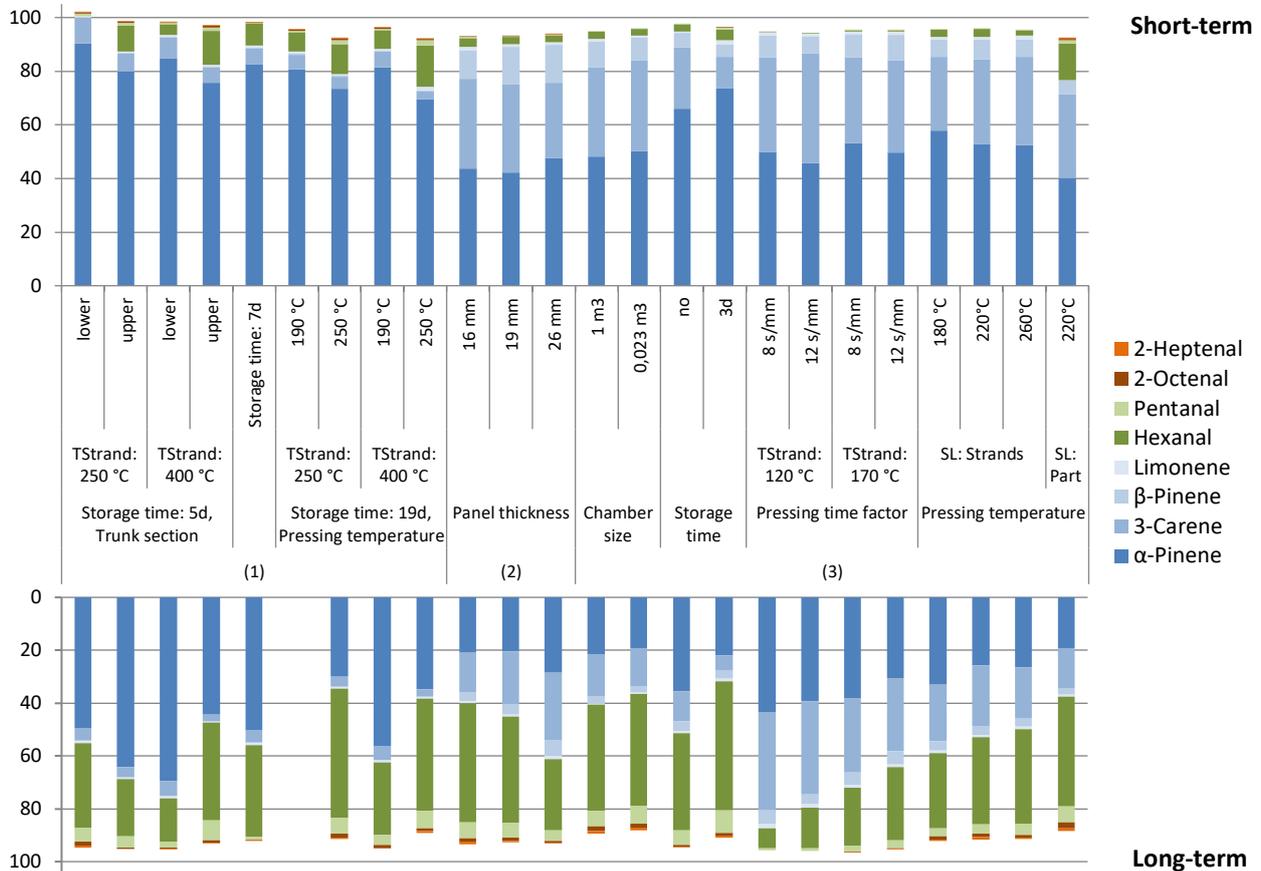
<sup>b</sup> stored for 12 months after purchase

Source: own representation according to the given sources

In other studies (LARSEN, FROST & WINTHER FUNCH, 2000; HORN et al., 2007; DÄUMLING et al., 2009; OHLMEYER & STECKEL, 2012), decreasing aldehyde emissions were determined over the entire course of the emission measurement. The emissions of these panels had therefore already passed through their aldehyde peak

before the start of the emission measurement. This indicates that the production of the boards and the emission measurement took place several weeks or even months apart. In this case, the emissions from the panels were already composed of higher aldehyde and acid proportions during the short-term behaviour (Figure 12).

**Figure 11: Composition (%) of emissions from freshly manufactured OSB**



TStrand = drying temperature of the strands, SL = surface layer, Part = Particles

WILKE et al. (2012) quantified VOCs using compound-specific response factors, but reported TVOC values as toluene equivalents. This leads to an underestimation of the TVOC, which in turn causes the summed percentages of the individual compounds to occasionally exceed 100 %.

(1) WILKE et al., 2012

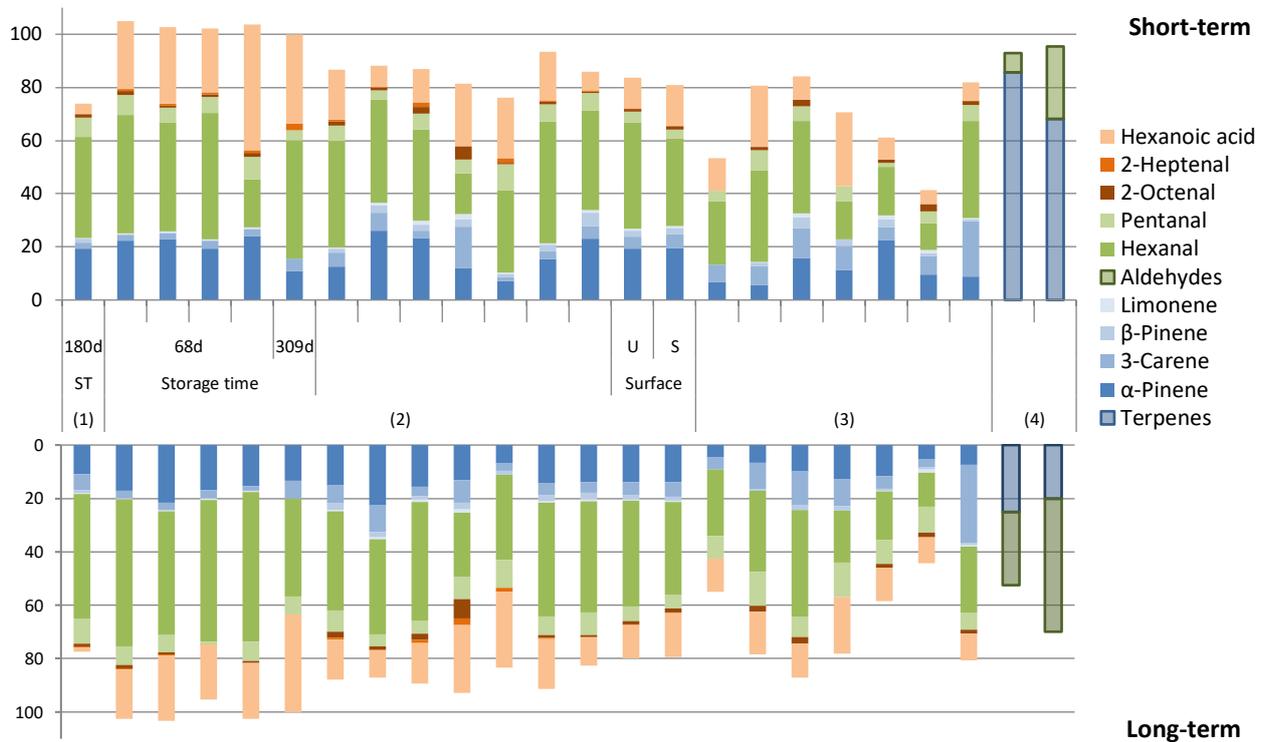
(2) OHLMEYER et al., 2008a

(3) MAKOWSKI, 2007

Source: own representation according to the given sources

The dominant terpenes are  $\alpha$ -pinene and 3-carene. The wide range of relative proportions of these two monoterpenes found in pine wood is also reflected in the emissions of OSB. For example, the OSB produced by WILKE et al. (2012) and MAKOWSKI (2007) have significantly different  $\alpha$ -pinene/3-carene ratios (Figure 11). Together with the data shown in Figure 12, however, it can be assumed that more  $\alpha$ -pinene is usually emitted. The emission rates after 28 days are below  $500 \mu\text{g m}^{-2} \text{h}^{-1}$  for 3-carene, even for freshly produced boards. For  $\alpha$ -pinene, somewhat higher values were occasionally observed: a maximum of  $1,500 \mu\text{g m}^{-2} \text{h}^{-1}$  for freshly produced boards (Makowski 2007) or boards stored for only a few days or weeks (WILKE et al., 2012).

**Figure 12: Composition (%) of emissions from OSB stored for at least 2 months or purchased from retailers**



ST = Storage time, U = Unsanded, S = Sanded;

WILKE et al. (2012) quantified VOCs using compound-specific response factors, but reported TVOC values as toluene equivalents. This leads to an underestimation of the TVOC, which in turn causes the summed percentages of the individual compounds to occasionally exceed 100 %.

(1) MAKOWSKI, 2007

(2) WILKE et al., 2012

(3) HORN et al., 2007

(4) OHLMEYER & STECKEL, 2012

Source: own representation according to the given sources

The saturated aldehydes are mainly characterised by hexanal – followed by pentanal, whose emission rate corresponds to about one eighth of that of hexanal. As a rule, the boards emit less than  $500 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days – only freshly produced pine-based OSB occasionally reached values of up to  $700 \mu\text{g m}^{-2} \text{h}^{-1}$  (MAKOWSKI, 2007). Other aldehydes (butanal, heptanal, octanal, nonanal, benzaldehyde) were detected in the individual studies, albeit with very low emission rates (HORN et al., 2007; MAKOWSKI, 2007; OHLMEYER et al., 2008a, b; WILKE et al., 2012). In contrast to solid pine wood, OSB also emit unsaturated aldehydes, primarily 2-octenal and 2-heptenal (HORN et al., 2007; MAKOWSKI, 2007; OHLMEYER et al., 2008a, b; WILKE et al., 2012). These are of particular relevance as they have a very low EU-LCI (*lowest concentration of interest*) value (Table 2). The emission rates for 2-octenal range from 0 to  $25 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days (HORN et al., 2007; MAKOWSKI, 2007; OHLMEYER et al., 2008a, b; WILKE et al., 2012), whereby the higher values were achieved in particular for the boards stored for a few days. 2-Heptenal is generally emitted in smaller quantities than 2-octenal. Depending on the time at which the OSB emissions were measured, the determined emission rates of the individual aldehydes correspond to either the rising, peak, or declining phase of their respective emission curves (Figure 6). It must be taken into account that each aldehyde develops its maximum emission rate at a different time, which is related to the different oxidation rates and degradation products of the individual fatty acids in the wood (BACK et al., 2000). It should also be noted that the low air concentrations of these compounds are often close to the

quantification limit. For some OSB samples, HORN et al. (2007) listed 2-octenal emission rates determined according to both DIN ISO 16000-6:2012 (Tenax/TD-GC-MS) and DIN ISO 16000-3:2013 (DNPH/HPLC), with substantial deviations observed in some cases between the two analytical methods.

Apart from terpenes and aldehydes, pine-based OSB emit a number of other compounds. In terms of quantity, organic acids play the largest role, although in this context it must be pointed out once again that they are insufficiently analysed using the standard method of VOC determination (Tenax/TD-GC-MS in accordance with DIN ISO 16000-6:2012) (Chapter 3.1.3 and 4.4.2). Hexanoic acid was mainly detected, followed by acetic acid (RISHOLM-SUNDMAN, 2002; HORN et al, 2007; MAKOWSKI, 2007; OHLMEYER et al, 2008a, b; DÄUMLING et al, 2009; OHLMEYER & STECKEL, 2012; WILKE et al, 2012). The maximum hexanoic acid emissions determined amount to  $440 \mu\text{g m}^{-2} \text{h}^{-1}$  after 3 days and  $170 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days for an OSB sample acquired by WILKE et al. (2012). In general, however, the emission rates were significantly lower. Acetic acid emissions were only sporadically reported in the studies – a maximum of  $200 \mu\text{g m}^{-2} \text{h}^{-1}$  on the 3rd and  $70 \mu\text{g m}^{-2} \text{h}^{-1}$  on the 28th day of measurement was determined for an OSB purchased and analysed by HORN et al. (2007). HORN et al. (2007) also detected pentanoic, heptanoic, octanoic and nonanoic acid in some OSB. While acetic acid is presumably formed through the hydrolytic cleavage of hemicellulose acetyl groups (Chapter 4.4.2), the other acids mentioned are probably oxidation products of the aldehydes (MAKOWSKI 2007).

### 5.1.2 Factors influencing the emission behaviour

In the past, technological modifications to wood-based panels to reduce formaldehyde emissions were necessary in order to comply with the required formaldehyde limit values. This primarily involves adjusting the composition of adhesives and additives. Additional drivers for technological improvements in the product itself or in the manufacturing process include reducing production costs or developing new products with higher added value (MANTANIS et al., 2018). Various patent applications from wood-based panel manufacturers and research institutions have focussed on reducing VOC emissions from OSB in recent years. This reflects the increasing attention given to the emission behaviour of building products. According to SALTHAMMER et al. (2003), the main strategies for reducing VOC emissions from OSB include selecting appropriate wood species, pre-treating the raw materials, optimising process conditions (drying/hot pressing), sanding the boards and prolonging the storage period between production and use.

#### Wood species

One approach to reducing VOC emissions from pine-based OSB is the complete or partial substitution of the raw material with wood species containing lower extractives. A corresponding patent (WO 2005042218A1) for the use of various hardwoods in the top and/or middle layer was published by Fritz Egger GmbH & Co in 2005. Studies by OHLMEYER et al. (2008b) show that OSB made of hardwoods have very low total emissions with a maximum of  $310 \mu\text{g m}^{-2} \text{h}^{-1}$  (1-day value) and  $50 \mu\text{g m}^{-2} \text{h}^{-1}$  (28-day value), whereby these are essentially acetic acid emissions (accounting for > 74 % of TVOC). AKRAMI (2014) also determined low VOC emission rates for OSB made from poplar and beech wood, which are largely made up of acetic acid. As hardwoods generally emit more acetic acid than softwoods (Chapter 4.4.2), this behaviour can also be expected for OSB made from them. OSB produced from oleoresin-poorer or oleoresin-free softwood species such as spruce, larch, Douglas fir and fir also lead to a significant reduction in VOC emissions (Table 20). Aldehyde emissions only occurred with spruce-based OSB, albeit with very low emission rates (28-day value <  $10 \mu\text{g m}^{-2} \text{h}^{-1}$ ) (OHLMEYER et al., 2008b).

**Table 20: Emission rates of OSB made from hardwood and softwood**

	TVOC	Acetic acid	Terpenes	TVOC	Acetic acid	Terpenes
	1-day value ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )			28-day value ( $\mu\text{g m}^{-2} \text{h}^{-1}$ )		
Beech ( <i>Fagus sylvatica</i> )	307	294	0	47	42	0
Birch ( <i>Betula spec.</i> )	177	152		42	31	
Maple ( <i>Acer pseudoplatanus</i> )	228	209		38	36	
Poplar ( <i>Populus spec.</i> )	295	263		35	28	
Spruce <sup>a</sup> ( <i>Picea abies</i> )	< 330	< 40	280	< 70	< 50	< 30
Larch <sup>a</sup> ( <i>Larix decidua</i> )			250			
Douglas fir ( <i>Pseudotsuga menziesii</i> )			230			
Fir <sup>a</sup> ( <i>Abies alba</i> )	30	30	0	40	40	0

<sup>a</sup> estimated emission rates based on several diagrams

Source: own representation modified according to OHLMEYER et al. (2008b)

In addition to a complete substitution of the pine strands, a partial substitution is also possible, for example by using lower-emitting wood species only for the top layers of the OSB. However, such an OSB with beech in the top layer and pine in the middle layer resulted in an emission composition comparable to that of a pure pine-based OSB<sup>36</sup>, consisting mainly of terpenes and aldehydes (OHLMEYER et al., 2008b). Accordingly, the beech top layer is permeable to these VOCs, which diffuse from the middle layer to the panel surface. According to OHLMEYER et al. (2008b), this process is presumably favoured by both the concentration<sup>37</sup> and temperature gradients<sup>38</sup> within the panel. An increase in the proportion of the top layer (TL) made of beech at least initially results in lower terpene emissions (2-day value; TL = 30 %: 2,100  $\mu\text{g m}^{-2} \text{h}^{-1}$ , TL = 50 %: 1,500  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). After two weeks, however, there are no longer any significant differences between the two variants, and after 28 days the terpene emissions are below 200  $\mu\text{g m}^{-2} \text{h}^{-1}$ . Increasing the beech content also reduces the aldehyde emissions – in contrast to the terpenes, the emission rates do not equalise even after two months. The maximum aldehyde emissions were determined after 14 days of testing at 110  $\mu\text{g m}^{-2} \text{h}^{-1}$  (TL = 30 %) and 80  $\mu\text{g m}^{-2} \text{h}^{-1}$  (TL = 50 %) (OHLMEYER et al., 2008b). THOLE (2009) also found a reduction in total emissions with increasing proportion of top layer made of beech. He additionally analysed OSB that consisted exclusively of pine strands in the middle layer and a mixture of pine and beech strands in the top layer. As expected, the total emissions of the boards increased with higher pine strand content in the top layer.

Terpene emissions from pine-based OSB can be reduced by specifically using pine wood with a low oleoresin content (e.g. higher proportion of sapwood) or by storing the strands for longer before pressing (SCHNEIDER & DIX, 2006). WILKE et al. (2012) also showed that OSB emits fewer terpenes when pine wood from the upper instead of the lower trunk section is used. However, the aldehyde emissions are not significantly affected. This illustrates once again the large fluctuations in the oleoresin content even within a single tree trunk, whereas the fatty acid content is largely evenly distributed. WILKE et al. (2012) further note that such pre-sorting of the pine raw material is hardly feasible in industrial practice.

According to MAKOWSKI (2007), the structure of the top layer strands also influences the emission behaviour of the OSB. In pine-based OSB with particles in the top layers, he observed markedly increased aldehyde emissions in the first two weeks of measurement as well as reduced terpene emissions. This is explained by the larger

<sup>36</sup> The pine strands used for the three-layer OSB were not freshly chipped and dried, but several weeks old. However, a pure pine-based OSB was not produced with these strands, so a direct comparison is not possible. The absolute emission level of the three-layer OSB in comparison to a pure pine-based OSB can therefore not be assessed.

<sup>37</sup> Terpene-rich core layer made of pine wood, terpene-free top layers made of beech wood.

<sup>38</sup> After hot-pressing, the top layer cools faster than the core layer.

specific wood surface area, which allows terpenes to escape more quickly while making the fatty acids more available for autoxidation processes, thereby enhancing aldehyde formation.

### Panel thickness

Emission rates describe the mass of volatile organic compounds emitted by a product per time at a given time from the start of the emission test (DIN EN ISO 16000-9:2008). Since building products, in particular panel materials, are generally exposed via their surface, the rates are expressed in relation to the material surface area<sup>39</sup> (unit:  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). OHLMEYER et al. (2008a) examined the emission behaviour of pine-based OSB of different thicknesses (panel thickness: 16, 19 and 26 mm). They observed that increasing board thickness led to higher area-specific terpene emission rates. This is due to the greater amount of wood in thicker boards, which provides more terpenes that can diffuse from the interior to the surface. This is favoured by the porous and capillary structure of the wood and in particular the wood-based materials. In contrast, the emission rates of aldehydes decreased with increasing board thickness. This is presumably due to the longer pressing time required for thicker boards (pressing time factor of all tested boards:  $12 \text{ s mm}^{-1}$ ) in combination with hotplate temperatures of over 200 °C. This may result in polymerisation of the fatty acids, making them less easily cleavable by autoxidation. Apart from this, oxygen availability at the board surface is greater, so that autoxidation is likely a surface-near process that is less influenced by board thickness (OHLMEYER et al., 2008a).

### Raw material modification and addition of additives

Various patents (WO 2006032267 A1, WO 2009156258 A1, WO 2006039914 A1, DE 102007055415 A1, DE 102008020642 A1) have been filed that describe VOC reduction by adding additives to the wood strands. The methods aim to influence the autoxidation process of the fatty acids using antioxidants, alkaline compounds, oxidising or reducing agents. One mechanism is to prevent the cleavage of the fatty acids. Another approach is to break down the unsaturated fatty acids during the production process and either removing the resulting aldehydes or converting them into less toxic or non-toxic compounds. ROFFAEL, SCHNEIDER & DIX (2015), for example, investigated the influence of treating pine strands with an oxidising agent (hydrogen peroxide) or reducing agent (sodium sulphite) on the emission behaviour of the strands. As expected, the oxidising agent leads to a marked increase in aldehyde emissions. The reducing agent had no recognisable effect on the aldehyde emissions of the strands, but the strands were dried at room temperature, so the energy required for autoxidation was missing. SCHNEIDER & DIX (2006) showed that treating strands with sodium hydroxide reduced their terpene emissions while increasing their aldehyde emissions. In contrast, an OSB made from sodium hydroxide-treated pine industrial strands exhibited both lower terpene and aldehyde emissions. WILKE et al. (2012) attempted to reduce the aldehyde emissions of pine-based OSB by adding antioxidants or preservatives during production to prevent or at least slow down the autoxidation process. Aldehyde emissions were permanently reduced as a result, but terpene emissions increased.

WIDHALM et al. (2017) reduced the terpene emissions of OSB by first exposing the pine strands to bacterial cultures (genus: *Pseudomonas*) that utilise the terpenes as a carbon source. Resin and fatty acids can also be reduced by a pretreatment of pine chips (*Pinus taeda* and *P. virginiana*) with bacteria (BURNES, BLANCHETTE & FARRELL, 2000), which could possibly reduce the aldehyde emissions of the subsequent wood product. STRATEV et al. (2011) further showed that treating pine strands (*Pinus sylvestris*) with fungi (*Ophiostoma piliferum*) led to reduced aldehyde emissions from the OSB produced. A similar effect was observed when the strands were treated with wastewater from the wood-based panel industry (STRATEV et al., 2015). The biofilm present in the wastewater, consisting of various microorganisms with different nutrient preferences and degradation

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<sup>39</sup> For other products mass-specific ( $\mu\text{g kg}^{-1} \text{h}^{-1}$ ), volume-specific ( $\mu\text{g m}^{-3} \text{h}^{-1}$ ) or unit-specific emission rates ( $\mu\text{g piece}^{-1} \text{h}^{-1}$ ) may be more suitable for characterising the emission behaviour.

capabilities, metabolises the free fatty acids, reducing their content by more than 80 %. According to STRATEV et al. (2015), this biotechnological treatment could be applied to freshly cut chips and strands during industrial panel production, directly in the respective storage bunkers. A corresponding patent application has also been filed (WO 2014064209 A1).

The binders used in OSB production are not themselves sources of VOC emissions. However, due to their properties, they can influence the emission behaviour of the wood, in particular aldehyde emissions (OHLMEYER et al., 2008b). OHLMEYER et al. (2008b) were unable to establish any clear correlations, as various influencing factors (pH value of the adhesive mix, adhesive content of the board, type and quantity of binder) can act simultaneously. However, they assume that autoxidation is accelerated in an alkaline environment, which is the case with PF-bonded OSB. Phenol-bonded boards, which harden in a neutral to acidic environment, exhibited lower aldehyde emissions than UF-bonded boards, which in turn is attributed to an antioxidant effect of phenol. THOLE (2009) also found differences in the emission behaviour of OSB depending on whether PMDI or MUPF was used as the adhesive in the top layer – however, only the 2-day values were reported.

HASCH & BOROWKA (WO 2014072304 A1) reduced the emissions of OSB by adding porous carbon (in particular activated carbon) to the boards, which can bind some of the VOCs due to its high adsorption capacity. In addition, KALWA, MÜLLER & THIELECKE (WO 2017050949 A1) incorporate hydrogen sulphite salts into the boards, which is intended to reduce the emissions of saturated and unsaturated aldehydes in particular.

### Process conditions

The individual production steps are responsible for the different emission behaviour of pine-based OSB compared to solid pine wood. It is therefore obvious that the VOC emissions of OSB can be influenced by changing the process conditions. BARRY & CORNEAU (1999) showed on poplar-based OSB that increasing the pressing time and the binder content reduces the TVOC, aldehyde and ketone emissions. However, only a single measurement time was analysed and, according to OHLMEYER et al. (2008b), poplar-based OSB has only low total emissions anyway. MAKOWSKI (2007) analysed the influence of various process parameters (drying temperature of the strands, pressing temperature, pressing time factor) on the emissions of pine-based OSB. The study shows that the terpene emissions of OSB can be reduced by higher and longer temperature exposures, because a large proportion of the terpenes evaporate during the drying of the strands and the pressing process. Studies by THOLE (2009) and SALTHAMMER et al. (2003) confirm these results. The latter demonstrated using pine particles that an increase in the drying temperature (variants: 30 °C – 24 h, 120 °C – 6 h, 220 °C – 2 h) led to over 40 % (120 °C) and 90 % (220 °C) reduced terpene emissions in the particles (1-day value), whereby hexanal was also detected at a temperature of 220 °C. In contrast, pine strands dried at 250 °C showed lower terpene emissions (1-day value) than strands dried at 400 °C in the investigations by WILKE et al. (2012). In addition, they were only able to detect aldehyde emissions in the strands dried at 400 °C.

MAKOWSKI (2007) showed that the process parameters affect different substance classes in distinct ways. An increase in the strand drying temperature promotes autoxidation due to the higher energy input. As a result, the aldehyde emissions of the OSB increase more strongly, exhibit a higher maximum, but also decline more quickly. This means that it is primarily the course of aldehyde emissions that changes – the extent to which the total quantity of aldehydes emitted changes was not clarified. In contrast, increasing the pressing temperature (variants: 180, 220 and 260 °C) had the opposite effect – the highest aldehyde emissions were achieved at the lowest pressing temperature, whereby the emission rates of all variants equalised after six weeks. MAKOWSKI (2007) attributes the lower aldehyde emissions at the high pressing temperatures to a possible polymerisation of the fatty acids, making them less prone to autoxidative cleavage. However, in studies by THOLE (2009), the strand drying temperature had no significant influence on the level of hexanal emissions. Furthermore, an increase in the pressing temperature was associated with higher hexanal emissions. Since only the 2-day value

was determined, the results are hardly comparable with those of MAKOWSKI (2007), who presented emission trends over several weeks to months.

According to MAKOWSKI (2007), it is only possible to a limited extent to reduce the VOC emissions of OSB in the long term by changing manufacturing conditions alone. In addition, the individual factors sometimes have different effects on primary and secondary emissions. WILKE et al. (2012) also concluded, based on investigations of the influence of various process parameters (strand drying temperature, hot pressing temperature), that no sufficient VOC reduction can be achieved. The results showed no clear correlation between the parameters studied and the emission behaviour of the pine-based OSB.

### **Storage conditions of the panels**

Industrially produced OSB are first cooled after hot pressing and then stored in stacks (OHLMEYER et al., 2008b). WILKE et al. (2012) analysed both commercially purchased and self-produced OSB immediately after purchase or production as well as after several months of storage (in stacks). As expected, both primary and secondary emissions were significantly reduced during storage. However, investigations by JANN, WILKE & BRÖDNER (1999) showed that a particleboard (veneered with oak and lacquered) still exhibited approximately the same total emissions after about one month of stacked storage as a directly tested board. The duration of storage is decisive. While MAKOWSKI (2007) found a significant reduction in terpene emissions from pine-based OSB stored in stacks for six months, boards stored for a short time (three days) initially (1<sup>st</sup> week of measurement) had the highest terpene emissions compared to boards tested directly after manufacture. When stored, the OSB had a surface temperature of 50 °C, which remained around 35 °C even after three days in the stack. As a result, there is only a delayed cooling of the boards in the stack. The elevated temperature generally promotes increased terpene emissions (STECKEL, 2011), but the emitting surface is limited to the panel edges during stacked storage. Consequently, terpenes presumably accumulate at the board surface, resulting in high initial emission rates at the beginning of the emission measurement (MAKOWSKI, 2007). With regard to aldehyde emissions, in the measurements by MAKOWSKI (2007), the six-month-stored OSB exhibited the highest emission rates until the second week after the start of the test. Apparently, there was an accumulation of aldehydes, which could only escape insufficiently via the side surfaces of the board. In the further course of the emission test (weeks 2 to 4), however, freshly manufactured and only briefly stored OSB had higher aldehyde emission rates. Accordingly, isolated board storage with continuous air exchange seems most suitable for achieving low VOC emission rates.

## **5.2 Particleboard**

Particleboards differ from OSB primarily in terms of the raw materials used and the particle size. Both factors have an influence on the emission behaviour of the product, making a separate consideration of particleboards necessary. While mainly coniferous industrial wood is used for OSB, various raw material assortments (sawmill by-products, waste wood, industrial wood residues, bark) are employed for particleboard. Approximately one quarter of the wood used is waste wood (Figure 8), which may introduce contaminants into the board and thus lead to unexpected emissions (YU & KIM, 2012; ROFFAEL, SCHNEIDER & DIX, 2015). As already discussed for OSB, the composition and level of VOC emissions from a wood-based material are strongly determined by the wood species used with its characteristic emission behaviour (Chapter 4). Due to the diverse raw material compositions, no generally statements can be made regarding the emission behaviour of particleboards. This is particularly emphasised by the studies of BAUMANN, BATTERMAN & ZHANG (1999) and BAUMANN et al. (2000). The authors determined the emission rates (2-day value) of particleboards and MDF from various North American manufacturers as a function of the wood species used. Despite the different range of wood species compared to the German particleboard market, these studies provide important insights into the influence of the wood species and the board type and thus the manufacturing conditions on the emission behaviour. In addition, the comparatively large sample size provides a good overview of the variability within a given board

type. The highest total emissions were determined for particleboards made of pine, whereas the boards made of Douglas fir had the lowest emissions. The decisive factor is again the content and composition of extractives in the wood raw material (Table 21).

**Table 21: Emission composition of particleboard (2-day value)**

Wood species (number of samples)	Emission figures – mean value (standard deviation)				
	TVOC ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	Hexanal ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	2-Octenal ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	Aldehyde content in TVOC (%)	Terpenes ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )
Southern pine <sup>a</sup> (22)	2,066 (604)	981 (314)	33 (12)	68 (10)	95 (-)
Other pine <sup>b</sup> (8)	1,939 (1,465)	851 (804)	41 (42)	58 (12)	284 (-)
Douglas-fir <sup>c</sup> (4)	262 (147)	106 (57)	4 (5)	49 (30)	31 (-)
Hardwood <sup>d</sup> (3)	1,567 (1,605)	1,245 (1,279)	36 (36)	90 (4)	15 (-)
Other <sup>e</sup> (2)	1,613 (1,812)	1,089 (1,462)	49 (53)	80 (41)	99 (-)

<sup>a</sup> *Pinus palustris*, *P. echinata*, *P. taeda*, *P. elliotii*

<sup>b</sup> *Pinus contorta*, *P. ponderosa*, *P. monticola*

<sup>c</sup> *Pseudotsuga menziesii*

<sup>d</sup> *Quercus spec.*, *Acer spec.*, *Populus spec.*, *Alnus spec.*, *Tilia spec.*

<sup>e</sup> *Abies spec.*, *Sequoia sempervirens*, softwood mix

Source: BAUMANN, BATTERMAN & ZHANG (1999); BAUMANN et al. (2000)

According to BAUMANN, BATTERMAN & ZHANG (1999) and BAUMANN et al. (2000), emissions from softwood-based particleboards consist mainly of aldehydes and terpenes – small amounts of alcohols and ketones were also detected. The substance spectrum essentially corresponds to that of OSB. The aldehyde content, with hexanal as the main representative, accounted for between 49 and 90 % of the TVOC, depending on the wood species. Compared with OSB, lower terpene emission rates were determined for particleboards, even when pine wood was used (Table 21). However, it should be noted that the boards were temporarily stored at 2 °C for 8 to 10 weeks after receipt of the samples from the manufacturers. High terpene emissions from OSB were primarily measured when the boards were tested directly after production (Chapter 5.1). Apart from this, the pine species used in the study were North American pines rather than *Pinus sylvestris*. According to the authors, the large standard deviation within the emission results is presumably related to the broad range of wood species aggregated under a single category. More detailed information on the wood used and the manufacturing conditions would be required to explain this variability (BAUMANN, BATTERMAN & ZHANG, 1999; BAUMANN et al., 2000). Outliers with particularly high aldehyde emissions in the hardwood and “other pine” particleboard groups may indicate the use of wood species with a high content of unsaturated fatty acids.

A number of studies (JANN, WILKE & BRÖDNER, 1999; LARSEN, FROST & WINTHER FUNCH, 2000; RISHOLM-SUNDMAN, 2002; GACA & DZIEWANOWSKA-PUDLISZAK, 2005; HORN et al, 2007; STACHOWIAK-WENCEK, PRĄDZYŃSKI & KRZYWOSIŃSKA, 2011) investigated particleboards from Europe. However, there is often no information available regarding the composition of the raw materials, the age of the boards or the storage conditions. Consequently, the results are only of limited usefulness for deriving a typical emission level. The terpene fraction of the total emissions is also quite low for these boards, with the aldehyde – and in some cases acetic acid – emissions dominating. As with OSB production, the wood is exposed to high temperatures during particle drying and hot pressing. Due to the higher specific surface area of the particles compared to OSB strands, a large proportion of the terpenes is probably already emitted during panel production. In the short-term behaviour, the boards exhibited a TVOC of 300 - 2,700  $\mu\text{g m}^{-2}\text{h}^{-1}$  (1-day value, STACHOWIAK-WENCEK, PRĄDZYŃSKI & KRZYWOSIŃSKA, 2011), 900  $\mu\text{g m}^{-2}\text{h}^{-1}$  (1-day value, RISHOLM-SUNDMAN, 2002), 900 - 1,100  $\mu\text{g m}^{-2}\text{h}^{-1}$  (2-day value, GACA & DZIEWANOWSKA-PUDLISZAK, 2005), 1,600  $\mu\text{g m}^{-2}\text{h}^{-1}$  (3-day value, HORN et al, 2007) and 900  $\mu\text{g m}^{-2}\text{h}^{-1}$  (3-day value, JANN, WILKE & BRÖDNER, 1999). Comparatively high acetic acid

emissions were determined by HORN et al. (2007) of around  $1,100 \mu\text{g m}^{-2} \text{h}^{-1}$  and  $800 \mu\text{g m}^{-2} \text{h}^{-1}$  (3- and 28-day values). They assume that the proportion of hardwood in the board was increased or that an alkaline curing binder was used.

### 5.3 Fibreboard

Fibreboards consist of fibres, fibre bundles or fibre fragments of wood or wood-containing materials. There are two different manufacturing processes (Figure 10). The majority of fibreboards, including MDF and HDF, are manufactured using a dry process, whereby the fibre mat is formed mechanically or pneumatically with dry fibre material. Insulation boards (porous fibreboards, LDF) and hardboards, on the other hand, are usually produced using the wet process, whereby the fibre mat is formed in an aqueous medium by sedimentation of fibres from the fibre suspension. However, insulation boards produced using the dry process are also available on the market (NIEMZ & WAGENFÜHR, 2012). In contrast to particleboards, waste wood is hardly used in fibreboards – accounting for only 7 % and 1 % of the wood input for MDF/HDF and LDF, respectively. While LDF is mainly produced from sawmill by-products, more than 50 % of the raw material for MDF/HDF consists of industrial roundwood (softwood: 36 %, hardwood: 20 %), supplemented by 39 % sawmill by-products (Figure 8, DÖRING, GLASENAPP & MANTAU, 2017). These by-products generally originate from softwood, as this type of wood is mainly sawn in Germany (DÖRING & MANTAU, 2012). Consequently, fibreboards usually consist primarily of softwoods. The binder predominantly used for fibreboards is urea-formaldehyde-based resin (UF, MUF). PMDI is used in small quantities, usually for special applications such as moisture-resistant and formaldehyde-free boards (MANTANIS et al., 2018). In the construction sector, fibreboards are mainly applied as insulation boards – this product group represents the largest market share among wood-based materials in the construction sector. MDF/HDF are used in roof constructions and flooring and roughly match the consumption of particle-based wood materials in the construction sector (Figure 9, MANTAU, DÖRING & HILLER, 2013). Another important area of application is the furniture industry (MANTANIS et al., 2018).

According to OHLMEYER et al. (2012), MDF has low TVOC values compared to other wood-based materials, typically below  $500 \mu\text{g m}^{-2} \text{h}^{-1}$  (28-day value). They tested laboratory-produced MDF made from pine, spruce, fir and a mixture of pine and beech wood. The industrially manufactured pine-based MDF they analysed showed slightly higher emission rates. BAUMANN, BATTERMAN & ZHANG (1999) and BAUMANN et al. (2000) also concluded that MDF generally release lower total emissions than particleboards. This is reflected in the fact that, despite the use of extractive-rich wood species such as pine, there are no significant terpene emissions from MDF (BAUMANN, BATTERMAN & ZHANG, 1999). In addition, MDF has lower aldehyde emissions than particle-based materials. An exception was MDF made from Southern Pine, which had markedly higher aldehyde and thus also total emissions than particleboards manufactured from the same raw material (Table 21, Table 22).

**Table 22: Emission composition of MDF (2-day value)**

Wood species (number of samples)	Emission figures – mean value (standard deviation)				
	TVOC ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	Hexanal ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	2-Octenal ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )	Aldehyde content in TVOC (%)	Terpenes ( $\mu\text{g m}^{-2}\text{h}^{-1}$ )
Southern pine <sup>a</sup> (6)	3,087 (1,649)	1,781 (902)	60 (37)	81 (7)	2 (-)
Other pine <sup>b</sup> (5)	602 (673)	285 (544)	4 (6)	60 (30)	12 (-)
Hardwood <sup>c</sup> (5)	162 (108)	54 (36)	3 (8)	40 (26)	7 (-)
Other <sup>d</sup> (2)	213 (65)	30 (43)	0 (0)	16 (18)	4 (-)

<sup>a</sup> *Pinus palustris*, *P. echinata*, *P. taeda*, *P. elliotii*

<sup>b</sup> *Pinus contorta*, *P. ponderosa*, *P. monticola*

<sup>c</sup> *Quercus spec.*, *Acer spec.*, *Populus spec.*, *Alnus spec.*, *Tilia spec.*

<sup>d</sup> *Abies spec.*, *Sequoia sempervirens*, softwood mix

Source: BAUMANN, BATTERMAN & ZHANG (1999); BAUMANN et al. (2000)

However, when considering the studies by BAUMANN et al. (2000), it must be borne in mind that Southern Pine comprises a group of several pine species with different extractive compositions and contents. Therefore, the comparison between the two wood-based panel groups is only possible to a limited extent. Apart from this, variations in emission behaviour also occur within a species, which explains the high standard deviation in the emission results (Table 22).

BAUMANN, BATTERMAN & ZHANG (1999), BAUMANN et al. (2000) and OHLMEYER et al. (2012) attribute the generally lower overall emissions of MDF compared to particle-based wood materials to the different manufacturing processes used for the two board types. The main process that influences the subsequent product emissions is the pulping of the wood into fibres. The wood chips are first pre-steamed at 60 to 80 °C and then enter the digester, where they are exposed to a temperature of 175 to 195 °C at a pressure of 6 to 10 bar for three to seven minutes (DIX & SCHNEIDER, 2006; IRLE & BARBU, 2010). This softens the lignin-rich middle lamella between the cells, enabling gentler mechanical separation of the fibres in the refiner (defibrator). For the fibreboards produced using the dry process, the hot and moist fibres are then blended with adhesives, dried at up to 200 °C, formed into a mat and hot-pressed at 180 to 210 °C (IRLE & BARBU, 2010). During fibre pulping and subsequent drying, a large proportion of the terpenes is already released. Consequently, MDF emissions typically contain rather monoterpenes with lower vapour pressure (e.g. limonene) (BAUMANN, BATTERMAN & ZHANG, 1999). The VOC emissions from MDF are therefore dominated by aldehydes and acids, both secondary emissions originating, as already discussed, from fatty acids and hemicelluloses. Studies with different grinding gaps in the refiner showed that the aldehyde emissions from MDF increase with increasing grinding gap and thus larger fibre size (OHLMEYER et al., 2012). For smaller fibres, some of the aldehydes presumably already escape during the drying and pressing process due to the greater specific surface area. If pine is used, the proportion of aldehydes in the total emissions is expected to play a more significant role than with other softwoods and hardwoods (BAUMANN et al., 2000; OHLMEYER et al., 2012). Depending on the raw material used and the age of the boards, the VOC emissions consist almost entirely or at least to a large extent of acetic acid (RISHOLM-SUNDMAN, 2002; OHLMEYER et al., 2012). As previously discussed, acetic acid can only be insufficiently quantified using the standard method of VOC determination (Tenax/TD-GC-MS in accordance with DIN ISO 16000-6:2012). RISHOLM-SUNDMAN (2002) suspects that the emission rates are underestimated as a result. The investigations by OHLMEYER et al. (2012) also show partially implausible trends in acetic acid concentrations over the test period of the MDF. In some MDF emission tests, they determined the emission rates of acetic acid using a further quantification method (sodium hydroxide solution/ion exchange chromatography) in addition to the Tenax/TD-GC-MS method. Significant deviations were recorded between the results, although there were no clear correlations between the methods – in one sample the Tenax/TD-GC-MS value was considerably higher, while in

the remaining samples it was significantly lower than the concentrations determined using ion exchange chromatography.

Apart from aliphatic (un)saturated aldehydes, which are also emitted by OSB and particleboards, the heterocyclic aldehyde furfural occurs among the VOC emissions in MDF (OHLMEYER et al., 2012). Furfural is formed as a result of hemicellulose degradation processes, which are initiated by thermal stress (SALTHAMMER & FUHRMANN, 2000). The pressure and temperature applied during wood chip plasticisation are the main factors influencing furfural formation. The hemicelluloses are first hydrolysed into simple sugars (pentoses, hexoses, uronic acids). Acid-catalysed dehydration from the pentoses and uronic acids then produces furfural or hydroxymethylfurfural from the hexoses. In particular, the acetic and formic acids naturally present in the wood act as catalysts (AEHLIG & FISCHER, 2009). In an alkaline environment, however, furfural formation is significantly reduced (AEHLIG & FISCHER, 2009; VOLKMER et al., 2014). Occasionally, furfural emissions were also detected from solid wood. Englund (1999) determined a maximum of 33 and 4  $\mu\text{g m}^{-2} \text{h}^{-1}$  (3- and 28-day values) from oak and 6 and 2  $\mu\text{g m}^{-2} \text{h}^{-1}$  (3- and 28-day values) from spruce dried at high temperatures. STACHOWIAK-WENCEK, PRĄDZYŃSKI & MATEŃKO-NOŻEWNİK (2014) measured 23  $\mu\text{g m}^{-2} \text{h}^{-1}$  for pine and 28  $\mu\text{g m}^{-2} \text{h}^{-1}$  for beech and oak (3-day value). HORN et al. (2007) determined maximum emission rates of 7  $\mu\text{g m}^{-2} \text{h}^{-1}$  for a beech board. Thermally treated wood in particular shows significantly higher furfural emissions than untreated wood (MANNINEN, PASANEN & HOLOPAINEN, 2002; PETERS, FISCHER & FISCHER, 2008; HYTTINEN et al., 2010; VOLKMER et al., 2014; ČECH & TESAŘOVÁ, 2015). For example, heat-pressure steamed oak emitted 460 and 190  $\mu\text{g m}^{-2} \text{h}^{-1}$  furfural (3- and 28-day values), whereas untreated wood released less than 30  $\mu\text{g m}^{-2} \text{h}^{-1}$  (VOLKMER et al. 2014). In studies by HYTTINEN et al. (2010), furfural emissions after 28 days were 23  $\mu\text{g m}^{-2} \text{h}^{-1}$  for heat-treated spruce, 18  $\mu\text{g m}^{-2} \text{h}^{-1}$  for pine and 11  $\mu\text{g m}^{-2} \text{h}^{-1}$  for poplar, while the untreated samples emitted only single-digit  $\mu\text{g m}^{-2} \text{h}^{-1}$  values. According to OHLMEYER et al. (2012), the furfural emission rates from MDF were at a maximum of around 30  $\mu\text{g m}^{-2} \text{h}^{-1}$ , but mostly below 10  $\mu\text{g m}^{-2} \text{h}^{-1}$  (28-day value). Rather higher values occurred with pine-based MDF with a high board thickness (38 mm) and with boards produced from fibers obtained under high refining pressures or temperatures (11 bar) or prolonged chip cooking times (6 min). MDF made from fir or spruce showed lower emission rates than those made of pine or pine-beech blends. In contrast, MAKOWSKI (2007) determined no furfural emissions from OSB, except for one panel pressed at elevated temperature (260 °C) with 2  $\mu\text{g m}^{-2} \text{h}^{-1}$  (3-day value). HORN et al. (2007) also detected small quantities of furfural emissions (max. 6  $\mu\text{g m}^{-2} \text{h}^{-1}$  on day 1) in a particleboard.

In addition to the wood species, the pulping process<sup>40</sup>, the pulping conditions and the type of binder are the main factors influencing the composition and quantity of VOCs from fibreboards. Additionally, the factors already discussed for OSB, such as board thickness, pressing temperature, pressing time factor and storage duration/conditions, also play a role. It has been observed that these individual factors can have different effects on the various substances or substance groups (DIX & SCHNEIDER, 2006; OHLMEYER et al., 2012).

Compared to MDF, HDF is characterised by a higher bulk density with a board thickness of only a few millimetres. MAKOWSKI (2007) and OHLMEYER et al. (2012) have shown that the total emissions of OSB and fibre materials decrease with decreasing board thickness. The fact that less raw material is used reduces the amount of volatile extractives and precursors that can react to form secondary emissions. Consequently, lower emissions can be expected from HDF than from MDF, which usually has a board thickness of 16 to 19 mm (MANTANIS et al., 2018). Pine-based HDF examined by OHLMEYER et al. (2012) with a thickness of 3 and 7 mm emitted only 90 and 140  $\mu\text{g m}^{-2} \text{h}^{-1}$ , respectively, after 28 days, whereby the emissions consisted mainly of acetic acid. RISHOLM-SUNDMAN (2002) obtained similarly low values of 30  $\mu\text{g m}^{-2} \text{h}^{-1}$  (28-day value).

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<sup>40</sup> The wood fibres are usually produced via thermomechanical pulping (TMP process). Another variant of this is the chemothermomechanical pulping process, in which the wood chips are chemically pre-treated with sodium hydroxide or a combination of sodium hydroxide and sodium sulphide (DIX & SCHNEIDER, 2006).

A significant proportion of the wood-based materials used in the construction industry are wood fibre insulation boards, which are characterised by a low bulk density and usually greater thickness than MDF. SCHERER (2011) investigated the emission behaviour of insulation materials made from conventional and renewable raw materials. The total emissions of the wood fibreboards were in the range of around 100 to 950  $\mu\text{g m}^{-2} \text{h}^{-1}$  (3-day value) and 50 to 750  $\mu\text{g m}^{-2} \text{h}^{-1}$  (28-day value), with acetic acid and furfural being detected. The lower values were achieved with wood fibre acoustic panels, which, in contrast to conventional insulation materials, can also be installed in such a way that their surface freely emits into the interior space (SCHERER, 2011).

## 5.4 Veneer-based products

The wood for veneer-based materials (plywood and laminated veneer lumber being the main representatives) is exposed to various thermal stresses during the manufacturing process. These include the steaming of the logs before peeling, the drying of the veneers and the pressing of the veneers into the final panel. As a result, the emission behaviour of the solid wood used is modified. The chemical reactions already mentioned take place during these processes, which lead to the formation of secondary emissions. Spruce-based plywood, for example, is characterised by higher aldehyde emissions than solid spruce wood (RISHOLM-SUNDMAN, 2002). While WILKE et al. (2011) detected almost no VOC emissions from purchased spruce-based plywood after 28 days, a panel tested by RISHOLM-SUNDMAN (2002) still had a TVOC of 700  $\mu\text{g m}^{-2} \text{h}^{-1}$ . WILKE et al. (2011) also determined higher emission values in some cases on pine-based plywood – the terpene emissions of a panel were over 1,600  $\mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days. The authors concluded that after one month of testing, pine-based plywood had lower aldehyde emissions, but higher terpene emissions than OSB. In contrast, YRIEIX, MAUPETIT & RAMALHO (2004) detected mainly acetic acid and furfural on a plywood panel after 28 days of testing, with total emissions below 50  $\mu\text{g m}^{-2} \text{h}^{-1}$ , indicating it was likely a hardwood panel. Likewise, a birch-based plywood panel analysed by LARSEN et al. (1998) emitted primarily acetic acid (130  $\mu\text{g m}^{-2} \text{h}^{-1}$ ), hexanal (38  $\mu\text{g m}^{-2} \text{h}^{-1}$ ) and pentanal (8  $\mu\text{g m}^{-2} \text{h}^{-1}$ ) after 28 days.

## 5.5 Summary

In principle, the same VOCs can be expected to be emitted from wood-based materials as from solid wood. The differences in emission behaviour between the individual wood species are therefore also reflected in the wood-based materials made from them. However, the relative composition of the emitted compounds changes as a result of the wood decomposition with the increase in the specific wood surface area and the overall (hydro)thermal stresses (steaming or cooking of the wood, wood drying, hot-pressing of the wood) during the manufacturing process. Furthermore, some substances are only formed under increased thermal treatment. The binders used in the wood-based materials are not a source of VOC emissions, although their properties can influence the relative composition of emissions. The waste wood used in particleboards can theoretically lead to contaminations with other VOCs, but the studies analysed did not provide any evidence for this.

VOC emissions from wood and wood-based materials originate from both volatile extractives (primary emissions) and non-volatile wood components, which can react through various processes to form volatile compounds (secondary emissions). While primary emissions are generally reduced in wood-based materials, the proportion of secondary emissions is increased compared to solid wood. Aldehydes, which are formed through autoxidation of unsaturated fatty acids and by dehydration of the hemicellulose monomers, are the most important substance class in this regard. Acetic acid emissions also play a role, which arise through hydrolytic cleavage of the acetyl groups from the hemicelluloses. Wood-based materials are characterised by greater homogeneity compared to solid wood as the degree of disintegration of the wood increases. Terpene emissions from pine in particular are subject to large fluctuations in the emission rate depending on the tree. In contrast, the area-specific terpene emission rates of OSB made of pine wood are lower and vary less between the boards. The reason for this lies in the increased process-related emissions and in the mixing of the raw material during the manufacturing process.

As the degree of wood disintegration increases, primary emissions are reduced, meaning that fibreboards made from softwood only have low terpene emissions. This can be attributed to the larger specific wood surface area of the fibres and the additional process step – compared to particle-based materials – of steaming or cooking the wood chips before they are defibrated. The wood-based materials analysed have aldehyde emissions in common, which is related to the increased thermal energy input during production. Furfural is emitted in very small quantities, tending to occur more from MDF than from OSB or particleboard. The emission rates of (un)saturated aliphatic aldehydes, on the other hand, appear to be higher for particle-based materials than for fibreboards. However, it is not possible to make generalised statements, as the amount of aldehyde is strongly dependent on the wood species used and its content of unsaturated fatty acids. While pine wood, which is naturally rich in fatty acids, is mainly used for OSB, different raw materials are used for particleboard and fibreboard. Using OSB as an example, it was additionally illustrated how various parameters, e.g. process conditions, board thickness, raw material modification and storage conditions, influence the emission behaviour. This showed that the individual factors can have different effects on the emission rates of the individual substances.

## 6 Conclusions

In Germany, various wood species and wood-based materials are used as construction products in buildings due to their numerous favourable properties. As potential sources of emissions, they can therefore have an impact on indoor air quality, just like other building products used indoors. However, the emission behaviour of a single product does not necessarily indicate how much it will contribute to the VOC mixture in indoor air. This depends primarily on the actual installation situation of the product as well as the climatic conditions and material interactions within the room. The cause of VOC emissions from wood and wood-based materials is the chemical composition of the raw material. This is characterised by the cell wall-forming main constituents – cellulose, hemicellulose and lignin – as well as by the extractives. The wood species differ in terms of the content and composition of these components, which also has an effect on their emission behaviour. Volatile extractives, such as terpenes, are referred to as primary emissions. Secondary emissions, on the other hand, are only formed through chemical reactions of wood components. Fatty acids present as non-volatile extractives can autoxidise to (particularly aliphatic) aldehydes, which are then emitted. Furfural, another volatile aldehyde, is formed through the dehydration of the hemicellulose monomers. Acetic acid emissions are in turn the result of the hydrolytic cleavage of the acetyl groups from the hemicelluloses. These reactions take place to varying degrees in the individual wood products, so that even when using the same wood species, a different emission pattern results depending on the wood-based material produced from it. The level of the area-specific emission rates and the composition of the VOC emissions from wood products are therefore influenced not only by the wood species but also by the conditions prevailing during the manufacturing process.

In principle, the main compound groups and individual compounds emitted by the different wood species and wood-based materials are well known. However, it is not practical to specify an exact value for the area-specific emission rates of individual VOCs or TVOCs for wood products. On the basis of the available data, emission ranges can rather be defined. On the one hand, this is due to the great variability of emission behaviour within some wood species (e.g. pine). On the other hand, the area-specific emission rates of individual VOCs are not a fixed value, but are subject to a temporal progression. Although it can be assumed that a state of equilibrium is almost reached after a certain period of time, this is often not yet achieved after a test period of four weeks. Primary emissions are generally subject to a decay behaviour, with a rapid decrease typically observed at the beginning of the emission test. In contrast, some secondary emissions initially show an increase in emission rates. Consequently, the emission behaviour always depends on the time of measurement and the storage duration and conditions.

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