

## **Emissions of volatile organic compounds from convection dried Norway spruce timber**

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### **Abstract**

Building products and furnishing are, among others, sources that emit volatile organic compounds (VOCs). The Construction Products Directive of the European Union (CPD) establishes requirements for building products, including demands regarding emission properties. Also harmonised standardisation related to the CPD is under way, implying that VOC emissions will be a relevant property of building products in the future. Objective of this study was to look into the effect of the drying schedule on VOC emissions from convection dried Norway spruce timber. Four different drying schedules, mainly varying in temperature, were employed. Samples dried at high temperature emitted noticeably less VOCs compared to samples dried at low temperature. With less varying temperatures, no clear effects were observed. Further, the amount of moisture lost during drying seems to affect product emissions. The composition of compounds was not influenced by the drying schedule and no harmful compounds could be detected. Emissions did not exceed concentrations of 300 µg m<sup>-3</sup>. In contrast, preliminary tests on other Norway spruce samples showed emissions up to one order of magnitude higher, due to stress-induced high extractives content of the wood. This example illustrates that spruce wood, being a natural material, may vary considerably regarding its emissions.

### **1 Introduction**

The term volatile organic compounds (VOCs) indicates a very diverse group of hydrocarbons that are volatile at ambient temperature. VOCs are emitted into the air by, e.g., plants, animals, microorganisms and most production processes and products. Human exposure to VOCs is regarded as a public health issue. Major sources influencing indoor air quality are polluted outdoor air, human activities such as cooking, use of cleaning agents or smoking as well as building products and furnishing. For reduction and control of potential indoor emission sources several national and European activities like health-related assessment schemes or eco-labelling were started. The European Construction Products Directive (CPD, 89/106/EEC) established a framework of requirements for building products to ensure their free circulation and unrestricted use in the European single market. The essential requirement No. 3 of the CPD states that hygiene and health of occupants must not be put at risk by dangerous substances emitted from the building. In 2008, the European Commission

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proposed substituting the CPD by an EU regulation with the same objective (Anonymous 2008). Concurrently, harmonised standardisation related to the specifications of the CPD is under way (Anonymous 2005). These developments show that VOC emissions will be a relevant property of building products in the future.

Kiln dried timber of Norway spruce (*Picea abies*) is commonly used in Central and Northern Europe for manufacturing building products and furniture. However, softwoods are usually rich in extractives and therefore can release substantial amounts of VOCs, mainly terpenes and aldehydes. While terpenes are constituent parts of softwood resin and responsible for the typical smell of softwood timber, aldehydes are secondary emissions that are formed by oxidation of fatty acids in the wood. They mostly smell dull and rancid. The amount of both terpenes and fatty acids varies from tree to tree and also depends on the location in the tree (Back and Allan 2000). Besides, stress of the tree can result in an increased extractives content. In contrast to synthetic products, emissions of timber cannot be influenced by changing its composition. Instead manipulation of the drying schedule might be a tool to affect subsequent emissions, assuming that compounds that are emitted in the drying process cannot contribute to emissions of the dried timber later.

Wu and Milota (1999) showed that the cumulated emissions of a drying run were increased at higher drying temperatures compared to lower temperatures. If the target moisture content (MC) was constant, higher initial MC resulted in higher absolute process emissions. The authors concluded that increasing the temperature results in increased volatilisation of compounds. Further, higher initial MC means that more water and, correspondingly, more volatile compounds are transported to the wood surface and evaporate. Also drying time is prolonged with higher green MC, resulting in a longer impact of temperature. Bengtsson and Sanati (2004) compared the total hydrocarbon emissions of Scots pine sapwood and heartwood during drying at different temperatures. An increase in temperature resulted in higher process emissions, especially of terpenes. Comparing drying runs at 90 °C with runs at 50 °C, the total amount of emitted  $\alpha$ -pinene (a terpene) increased threefold. Using a laboratory drier allowed for exact control of exhaust air streams. Englund and Nussbaum (2000) studied the terpene content of Norway spruce timber before and after drying at 60 °C or 110 °C. The depletion of the wood was higher after high-temperature drying than after drying at normal temperature.

The aim of the study presented was to look into the effect of drying temperature on the emissions of convection dried Norway spruce timber. Four different schedules, varying chiefly in temperature, were applied. Samples taken from the dried boards were emission tested. The results were compared regarding amount and composition of the released compounds. Besides, the results were compared to preliminary emission tests of other dried Norway spruce samples.

## 2 Material

Six logs of Norway spruce (*Picea abies*) with a length of three meters and a mid-diameter of 30 to 35 cm were selected for cutting sample boards. The logs had been harvested two months before drying in the Black Forest (south-western Germany). From each log, four to six sample boards were prepared with the dimensions 2000 x 100 x 27 mm<sup>3</sup>. The quarter sawn boards contained both sapwood and heartwood in similar amounts. Additional logs of the same origin were cut for being used for protection during transport and as filling material in the kiln. The material was grouped in four packages according to the four drying schedules. The objective was to prevent effects of the drying schedule being biased due to differences in the extractives content of the wood. Extractives content can vary considerably between trees. Therefore, by distributing the boards from each log evenly on the four packages, comparable material was provided to go into each drying run. Until drying, the wrapped packages were stored in a refrigerated storage house. A lab convection kiln was used for drying. One charge contained 0.85 m<sup>3</sup> of material in total, with the sample boards stacked in the middle of the pile. The following drying schedules were applied:

- Normal temperature schedule (NT): Starting at a dry bulb temperature (DBT) of 55 °C and an equilibrium moisture content (EMC) of 12 %. The temperature was raised to a maximum of 65 °C while the EMC was decreased to 5 %
- Normal temperature schedule with increased final temperature (NTplus): See NT schedule, but with a final temperature of 85 °C instead of 65 °C.
- High temperature schedule (HT): Starting at a DBT of 85 °C and an EMC of 12 %. The temperature was increased to 115 °C while the EMC was decreased to 4 %.
- Low temperature schedule (LT): On the basis of the System 603 by Muehlboeck GmbH (Eberschwang, Austria). At minimum EMCs, increased air flow, and 45 °C DBT, the timber is dried until fiber saturation. Then temperature and EMC are increased to 65 °C and 12 %, respectively. An additional fan was installed in a side opening of the pilot kiln, and the exhaust air-outlet was opened permanently during the first part of the schedule to ensure increased airflow. Minimum EMC was 3.75 %.

Each drying schedule ended with a conditioning phase of three hours at 12 % EMC. An equalisation phase at 20 to 25 °C with slowly rotating fans followed for at least three days. Since wood MC influences emissions, very homogeneous MCs were essential. Prior to and after drying, a bar (cross section) was cut from each sample board to determine the MC. Immediately after unstacking, the sample boards were cut to prepare specimens for emission testing. Knots, cracks, resin pockets as well as material from the ends of the boards were excluded. The area of a single emission test specimen was 200 x 60.2 mm<sup>2</sup>, with a thickness of 25 to 26.5 mm. The edges of the specimens were sealed with self-adhesive aluminium foil to test faces exclusively. Emission testing was accomplished using test chambers with controlled climatic parameters. The total

sample area required for a single test chamber was 722 cm<sup>2</sup>. Thus, three specimens were combined according to the logs they originated from. After each drying session, two emission tests were run in parallel. Table 1 shows sample composition. The parallel samples A and B are composed from timber of different trees. Thus, deviations of the results are expected due to varying extractives content of the trees.

Table 1: Sample composition and occupation of test chambers

Test chamber 1 with sample A	Test chamber 2 with sample B
Specimen log 1 - area: 240.8 cm <sup>2</sup>	Specimen log 1 - area: 240.8 cm <sup>2</sup>
Specimen log 2 - area: 240.8 cm <sup>2</sup>	Specimen log 2 - area: 240.8 cm <sup>2</sup>
Specimen log 3 - area: 240.8 cm <sup>2</sup>	Specimen log 3 - area: 240.8 cm <sup>2</sup>
Total sample area A : 722.4 cm <sup>2</sup>	Total sample area B: 722.4 cm <sup>2</sup>

### 3 Methods

Equipment, sampling, as well as analytical procedures, were in accordance with ISO 16 000 part 6 (2004) and part 9 (2008). The emission test chambers consisted of glass desiccators with a volume of 23 litres (see Figure 1).



Figure 1: Emission test chamber

In the chamber, 23 °C, 50 % relative humidity, and an air exchange rate of 3.1 h<sup>-1</sup> were maintained constantly. The loading factor, i.e., sample area related to chamber volume, was 3.1 m<sup>2</sup> m<sup>-3</sup>, and the air velocity above the sample surface was 0.15 m s<sup>-1</sup>. The emission tests were run for 28 days. In weekly intervals, air samples of 1 to 9 l were pumped from the test chambers. The compounds were trapped on adsorbent filled tubes. For identification and quantification of the compounds, a gas chromatograph, coupled with a thermo desorption unit and a mass sensitive detector were used. The emission results are presented as concentration C of compounds in chamber air (in µg m<sup>-3</sup>). Due to the selected loading factor and air exchange rate, as well as the relatively slow decay of the concentration over time, the results can also be read directly

as area-specific emission rates (emission factors,  $SE R_a$  in  $\mu\text{g m}^{-2} \text{h}^{-1}$ ), according to ECA report no. 18 (1997). This is very convenient when comparing results with those of other studies. "Sum of VOCs" means total VOC emissions in the respective air sample. It is calculated by adding the concentrations of all single compounds detected in that air sample.

## 4 Results and Discussion

### 4.1 MCs and development of VOC emissions over time

The green MC of boards from logs 1, 2 and 3 was 68.4 % with a coefficient of variance (CoV) of 40.1 % and a sample size of 14. Green boards of the logs 4, 5 and 6 had an MC of 54.4 % with a CoV of 30.5 % at a sample size of 16. The MC of the dried sample boards ranged between 11 to 14 % (see Table 2).

Table 2: Moisture contents after drying

	LT- A	LT-B	NT- A	NT-B	Ntplus-A	Ntplus-B	HT- A	HT-B
Mean in %	13.7	13.5	11.2	10.7	10.6	10.7	12.2	12.3
CoV in %	3.9	4.3	4.6	4.3	5.4	4.8	7.9	8.3

Figure 2 shows the development of the sum of VOCs emitted over time. A-samples released between 119 and 265  $\mu\text{g m}^{-3}$  on day 1 and 15 to 56  $\mu\text{g m}^{-3}$  on day 28. B-samples emitted generally less compounds with 103 to 175  $\mu\text{g m}^{-3}$  on day 1 and 9 to 22  $\mu\text{g m}^{-3}$  on day 28.

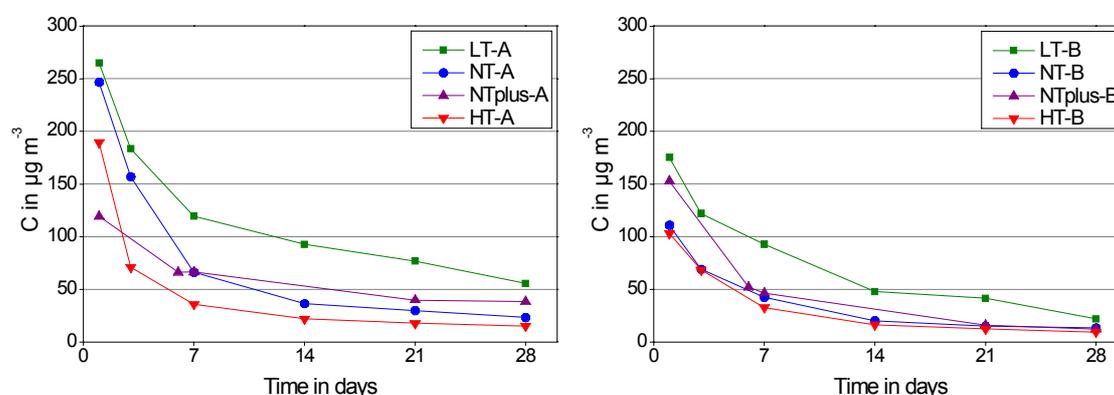


Figure 2: Sum of VOCs emitted over time by samples A and B

The results are in a similar range as those found in the literature regarding kiln dried Norway spruce timber. Larsen *et al.* (2000) determined less than  $100 \mu\text{g m}^{-2} \text{h}^{-1}$  on day 4 and about  $30 \mu\text{g m}^{-2} \text{h}^{-1}$  after 28 days of testing. Englund (1999) found emission rates of samples dried at high and normal temperature between 120 and  $480 \mu\text{g m}^{-2} \text{h}^{-1}$  on day 3 and 20 to  $170 \mu\text{g m}^{-2} \text{h}^{-1}$  on day 28. However, lower terpene emissions could not always be attributed to the higher drying temperature. In the study at hand, LT-A and LT-B samples released higher concentrations of VOCs over the entire time of testing than the samples dried with other schedules. Besides, HT-A and HT-B samples showed

the lowest emissions from day 3 on. NT-A and NTplus-A samples generally emitted VOC concentrations between those of the LT-A-samples and the HT-A-samples. However, the curves are not clearly separated from each other. Due to a defect in sample introduction to the analytical system, no data exists for all NTplus samples on day 3 and 14. From day 3 on emissions of the NT-B sample and the NTplus-B sample gave off concentrations similar to those of the HT-B sample.

The results suggest that an effect of the drying schedule on the product emissions exists, since samples dried at higher temperatures released less VOCs. While the effect is articulate when comparing HT samples with LT samples, results were not consistent regarding NT samples and NTplus samples. Maybe differences between the NT and NTplus schedules were not large enough to account for noticeable differences in the product emissions. Furthermore, varying final MCs may have influenced emissions. Higher loss of moisture results in a higher loss of VOCs (see 1). The MCs (after drying) of the NT and NTplus samples were lower than those of the HT samples (see Table 2), which may have decreased emissions disproportionately. Also, the final MCs of the HT samples were lower than those of the LT samples. Probably, the differences in moisture loss added to the differences in released VOC concentrations due to drying temperature.

#### 4.2 Terpenes

Emissions were composed of 85 % terpenes on average. Thus, the terpene emission curves are very similar to the sum of VOCs emission curves. Generally, over 90 % of the terpene emissions consisted of  $\alpha$ - and  $\beta$ -pinene in a ratio of 2...4 to 1. Also limonene, phellandrene, camphene, 3-carene and tricyclene were found, all common monoterpenes in softwoods (Englund 1999 and Englund 2000). Differences in the composition of terpene emissions between samples from the different drying schemes could not be detected.

#### 4.3 Aldehydes and other compounds

Figure 3 shows aldehyde emissions. A-samples released 8 to 25  $\mu\text{g m}^{-3}$  on day 1 and 4 to 10  $\mu\text{g m}^{-3}$  on day 28 of testing.

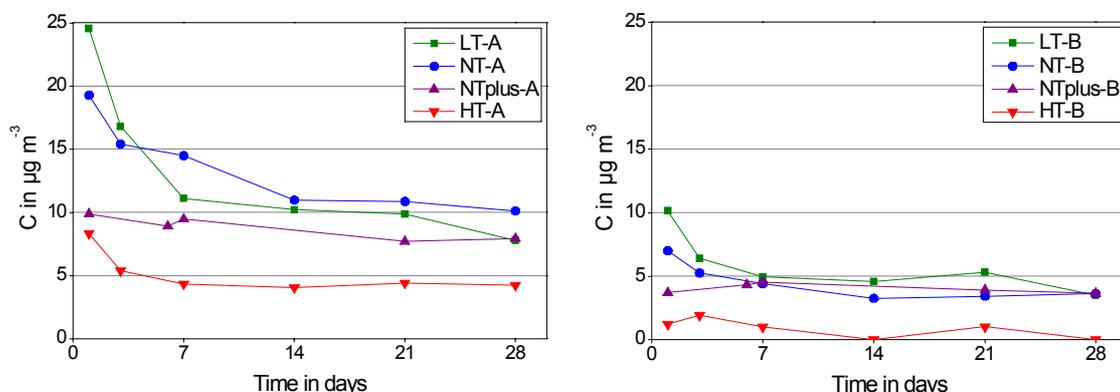


Figure 3: Aldehyde emissions over time of samples A and B

Similar to the terpene emissions, the B-samples emitted less aldehydes than the A-samples with 1 to 10  $\mu\text{g m}^{-3}$  on day 1 and 0 to 4  $\mu\text{g m}^{-3}$  on day 28. Aldehyde emissions of the samples HT-A and HT-B were always lower than those of the corresponding samples from the other drying schedules. This observation, as well as total amounts, are in accordance with the findings of Englund (1999). Samples from LT, NT and NTplus drying released rather similar concentrations in the second half of testing. The effect of high drying temperature on aldehyde emissions probably is manifold. Besides of volatilising more aldehydes already present in the wood, it can possibly accelerate aldehyde formation. Typically regarding wood, aldehyde emissions were always composed of hexanal and, to a lesser extent, pentanal. Toluene and butanol occurred in some samples with 8  $\mu\text{g m}^{-3}$  at most.

#### 4.4 Comparison with results of preliminary tests

A single log of Norway spruce of about 1.2 m length and 35 cm diameter was obtained about one week after felling. The tree had grown in a forest close to Hamburg (Germany). The log was cut into boards of 25 mm thickness and 10 cm width that were dried in a drying cabinet at 65 °C with air circulation. Two samples were prepared consisting either of sapwood, or, respectively, heartwood. Visible knots and resin pockets were excluded. Sample preparation, loading factor of the emission chambers as well as test parameters were the same as applied in the drying study presented above (see 3). The MC of the green heartwood was about 30 %, while that of green sapwood amounted to 150 %. After drying, heartwood had an MC of 10 %, and sapwood of 3 %. Figure 4 shows terpene and aldehyde emissions of the samples. Most striking are the high terpene concentrations. On the third day of testing, heartwood emitted 567  $\mu\text{g m}^{-3}$  and sapwood 2129  $\mu\text{g m}^{-3}$ . On day 28, heartwood terpene emissions had decreased to 219  $\mu\text{g m}^{-3}$ , while sapwood gave off 524  $\mu\text{g m}^{-3}$ . These values are much higher than terpene emissions found in the drying study. However, Risholm-Sundman *et al.* (1998) determined a total emission factor of 1400  $\mu\text{g m}^{-2} \text{h}^{-1}$  for Norway spruce.

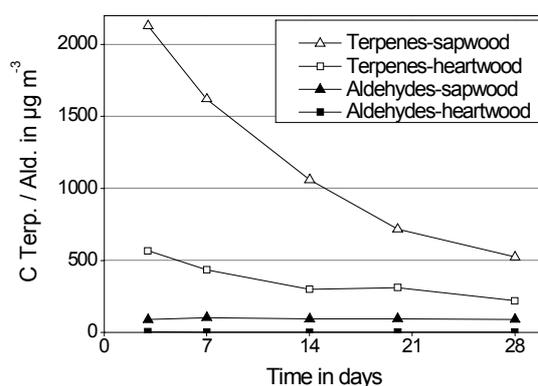


Figure 4: Terpene and aldehyde emissions of sapwood and heartwood

Also, the differences between sapwood and heartwood were unexpected since resin contents should be equal (Back and Allan 2000). Aldehyde emissions were composed of hexanal, pentanal, and traces of a few other aldehydes. They ranged from  $4 \mu\text{g m}^{-3}$  from the heartwood sample to about  $100 \mu\text{g m}^{-3}$  from sapwood. The latter emitted rather constant concentrations, thus probably showing the peaking of aldehyde formation. Oxidative degradation of fatty acids is triggered by increased temperature and includes a series of reactions, thus, the appearance of degradation products, *i.e.*, aldehydes, is delayed (Makowski *et al.* 2005). Due to the long equalisation phase applied in the drying study, peaking was probably over when emission testing started, therefore only decreasing aldehyde emissions could be observed.

Figure 5 shows microphotographs of transverse sections of the emission tested spruce sapwood and regular spruce wood. In contrast to the regular sample, the sapwood sample contained much more resin ducts and resin that migrated into the surrounding tissue, accounting for the high terpene emissions. These observations show that the tree was highly exposed to stress. It should be noticed that the macroscopic appearance of the timber did not differ from the samples of the drying study.



Figure 5: Emission tested sapwood sample (left) and regular sample (right)

## 5 Conclusion

The results of the drying study indicate that drying temperature impacts product emissions of Norway spruce timber. High drying temperature resulted in lower terpene and aldehyde emissions compared to the low temperature schedule. Emissions of samples dried with the NT and NTplus programmes were alike, suggesting that differences in the drying schedules were not distinct enough to affect product emissions noticeably. The composition of emissions was not influenced by the drying schedule. At no time during testing, the sum of detected VOCs exceeded  $300 \mu\text{g m}^{-3}$ . Further testing with more samples is needed to arrive at detailed and profound conclusions. Moreover, the effect of moisture loss on product emissions should be explored. Preliminary tests on spruce sapwood and heartwood resulted in emissions about an order of magnitude higher than from samples of the drying study, caused by increased extractives content due to stress of the tree. The example shows that spruce wood as a natural material can emit highly variable amounts of VOCs.

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