

Determination of emissions from solid wood elements

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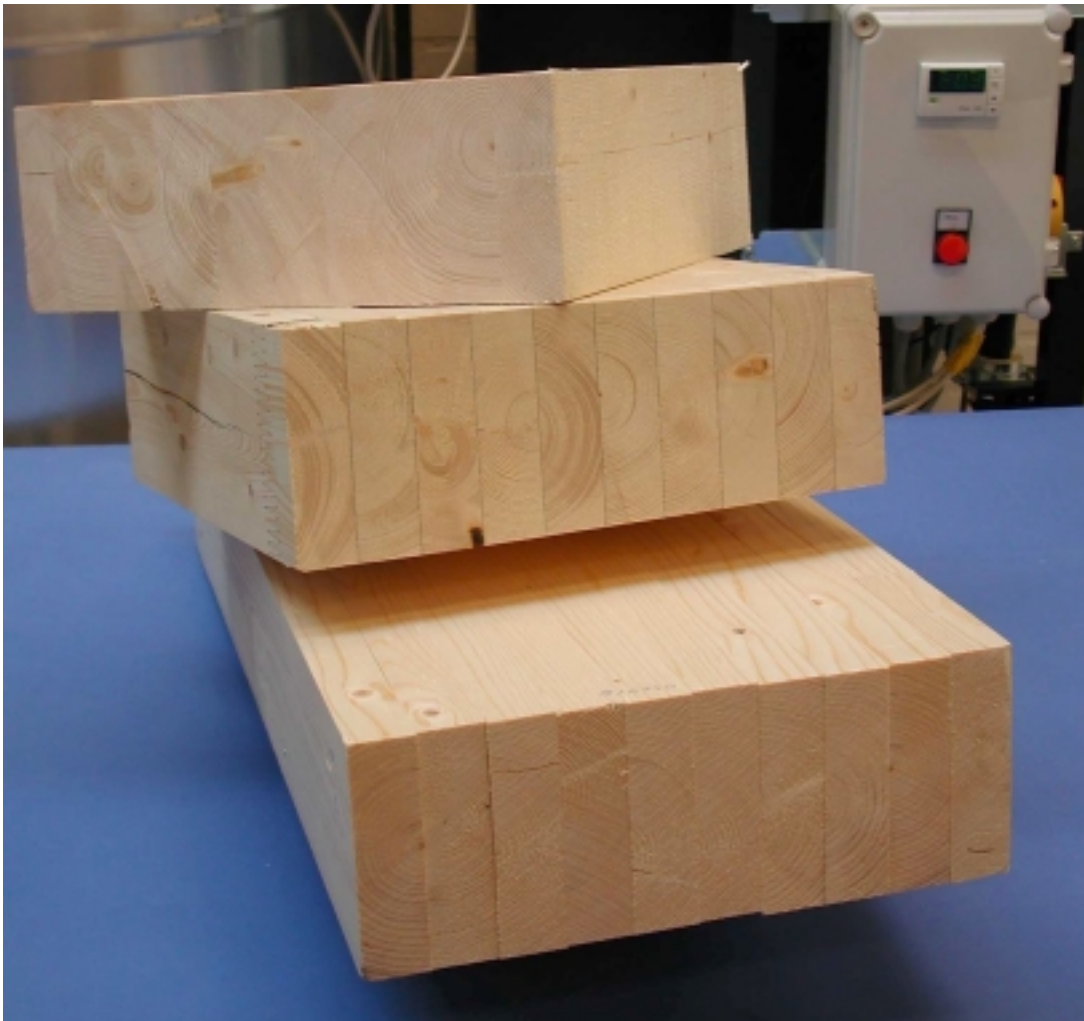


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Preface

Solid wood elements are a relatively novel type of products on the market for building construction materials. Hence, there has been a need on behalf of the whole chain in the building material industry to develop, research in and provide documentation for the properties of the products. This need is formulated as a quest for development, research and subsequent documentation with regard to marketing on behalf of the producers, a desire for accredited documentation on behalf of the counselling engineers with regard to the physical and mechanical design, a wish for product standards on behalf of the technical laboratories involved in the certification process and a general quest for documentation and answers to basic questions from the end consumers. In sum, this documentation targets the six essential requirements of the European Construction Products Directive (CPD).

This report contributes to the documentation for the requirement relating to hygiene, health and the environment. The report contains results from laboratory determination of the emissions from solid wood elements. The report is supplemented by a study of in-situ emissions measured in a single-family house constructed by use of glued solid wood elements (Funch & Clorius, 2002). These results enter the package of documentation that shall be used for establishing product standards for solid wood elements.

The work has been carried out at Danish Technological Institute, Wood Technology, in the period July 2001 to May 2002. The work is part of a Nordic research and development project for solid wood elements in building constructions "Massivtræelement". This project is co-funded by the Nordic Industrial Fund through the Nordic Wood Programme (Project P00023), the national Danish activities have been co-funded by the Danish Ministry of the Environment, through the sector support given by "Produktudviklingsordningen for Skov- og Træindustri".

Summary

Laboratory testing of emissions of formaldehyde and volatile organic compounds from three different types of solid wood elements is reported. The testing for formaldehyde emission is performed by the chamber method according to the requirements in (EN 717-1,1998).

The main findings are that emissions of formaldehyde are below the E1 requirement of (prEN 14080 (DRAFT), 2000) ($\leq 0,124 \text{ mg/m}^3 \text{ air}$) for both nailed, Phenol-Resorcinol-Formaldehyde glued, and Melamine-Urea-Formaldehyde glued solid wood elements.

Regarding Volatile Organic Compounds the testing shows that besides acetaldehyde emitting from the Phenol-Resorcinol-Formaldehyde glued element all the measured concentrations are below the odour threshold taken from (Jensen & Wolkoff, 1996).

The results serve both generally to enhance the knowledge of emissions from wood-based products used in building constructions and shall specifically be used to establish a basis for validating the use of the formaldehyde emissions requirements for glued laminated timber in Annex B of (prEN 14080 (DRAFT), 2000) in a proposal for a product standard for glued solid wood elements (Clorius & Adelhøj, 2002).

Resumé

Der rapporteres laboratoriebestemmelse af emissionerne af formaldehyd og flygtige organiske forbindelser (VOC'er) fra tre forskellige typer af massive træelementer. Bestemmelsen af formaldehydemissionen er foretaget ved kammermetoden i overensstemmelse med kravene i (EN 717-1,1998).

Hovedobservationen er, at emissionerne af formaldehyd er under E1 grænseværdien i (prEN 14080 (DRAFT), 2000) ($\leq 0,124 \text{ mg/m}^3 \text{ air}$) for såvel sømmede, phenol-recorsinol-formaldehyd limede som melamin-urea-formaldehyd limede massive træelementer.

Med hensyn til flygtige organiske forbindelser viser undersøgelsen, at bortset fra acatataldehyd, som afgasser fra det phenol-recorsinol-formaldehyd limede element, er alle de målte koncentrationer under lugtgrænsen som angivet i (Jensen & Wolkoff, 1996).

Resultatet tjener både til at øge den generelle viden om emissionerne fra træbaserede produkter, der bliver anvendt i bygningskonstruktioner, og vil herudover specifikt blive anvendt til at validere brugen af formaldehyd emissionskravene for limtræ, som de fremgår af Annex B til (prEN 14080 (DRAFT), 2000), i et forslag til en produktstandard for limede massive træelementer (Clorius & Adelhøj, 2002).

1. Background and scope

Wood is known to contain and emit Volatile Organic Compounds (VOCs) which can be demonstrated by the fact that wood smells. Wood products for the building and furniture industry are often a combination of wood and the materials added e.g. adhesive. Emissions from solid spruce are mostly terpenes and aldehydes. When glue is added to the wood material, other compounds such as formaldehyde and aromatic hydrocarbons may emit from the wood-based material.

Where formaldehyde-containing materials, particularly, aminoplastic resins have been added to wood-based materials as a part of the production process, the product shall be tested and classified into one of the two emission classes: E1 or E2 according to (prEN 13986, 2001) for wood-based panels and (prEN 14080 (DRAFT), 2000) for glued laminated timber. These classes are part of the normative requirements stated in Annex B of both standards. When tested in accordance with (EN 717-1,1998), the limit value for class E1 is $\leq 0,124 \text{ mg/m}^3$ air, and the limit value for class E2 is $\geq 0,124 \text{ mg/m}^3$ air according to (prEN 13986, 2001) and (prEN 14080 (DRAFT), 2000).

In the latest version of the harmonised standard for glued laminated timber (prEN 14080, 2002) deviations from (EN 717-1,1998) regarding test criteria are required. These deviations correspond to a lower material load. To maintain consistency with the body of standards the testing has been made corresponding to the requirement of (prEN 14080 (DRAFT), 2000).

The Danish building regulation (Bygningsregelement, 1995) states general performance requirement with respect to emissions: Building materials must not emit vapours and gasses, which can give an unsatisfactory indoor environment. For formaldehyde emissions quantified requirements similar to those in (prEN 14080 (DRAFT), 2000) and (prEN 13986, 2001) are given.

The use of glued solid wood elements in constructions entails that the volume of wood present in the construction is larger compared to traditional post and beam timber constructions. Where the solid wood elements have freely exposed surfaces, these can contribute considerably with emissions to the indoor air in both offices and dwellings. Therefore, the overall purpose of the project is to document the influence of large volumes and/or large exposed areas of solid wood elements on the indoor environment.

This report contains the results of a laboratory analysis of chemical emission from solid wood elements. In (Funch & Clorius, 2002) the laboratory results of the present report are used to calculate a theoretical estimate of the expected emission level in a specific single family house, and these theoretical results are compared to in-situ measurements of the same dwelling.

The present report and the results in (Funch & Clorius, 2002) establish the background for the recommendations with regard to emission class for glued solid wood elements that are given in the proposal for a product standard by (Clorius & Adelhøj, 2002).

2. Scope of testing

The scope of the testing is to evaluate the emission of formaldehyde and VOCs from three types of solid wood elements in a laboratory analysis of chemical emissions by use of the method given in (EN 717-1,1998). The determinations were carried out in emission tests at chamber conditions typical for building interiors.

The results shall be used in a general context of assessing new product types with respect to the third of the six essential requirements of the CPD (BEK nr 118 af 16/02/1998). In a more specific context the scope is to assess the validity of transferring the requirements for emission from glued laminated timber of Annex B in (prEN 14080 (DRAFT), 2000) to the proposed product standard for glued solid wood elements (Clorius & Adelhøj, 2002). These requirements will then be consistent with the requirements for wood-based panels (prEN 13986, 2001).

The emission from the three types of solid wood elements are related to VOCs i.e. terpenes and aldehydes that primarily originate from the wood itself, and formaldehyde and other aromatic hydrocarbons that stem from the used adhesive.

The evaluation of the emission of formaldehyde is related to the two classes, E1 and E2 of (prEN 14080 (DRAFT), 2000) and similar in (prEN 13986, 2001).

The evaluation of the emission of volatile organic compounds is related to the threshold values for odour and mucous membrane irritation, (Jensen & Wolkoff, 1996). Odour threshold concentrations are generally 1-3 orders of magnitude lower than those for mucous membrane irritation. Concentrations below the odour threshold values do most often not give reason to irritation.

3. Test material

3.1 General

The three types of solid wood elements used tested were glued or assembled with respectively:

- Nails; Figure 1
- Phenol-Resorcinol-Formaldehyde (PRF) glue; Figure 2
- Melamine-Urea-Formaldehyde (MUF) glue; Figure 3

The nailed elements represent an element type where all emissions originate from the wood itself. The elements glued with PRF glue will probably show a very low formaldehyde release, as PRF-glulam is known to have low emissions of formaldehyde, see e.g. (Bygningsregelement, 1995). Of the three element types tested, the elements glued with MUF represent the type with the previously known highest emission of formaldehyde, amongst the three types tested. Furthermore, this type of element is the most interesting as it, due to the light gluelines, is known to be the consumers' choice when selecting glued solid wood elements.

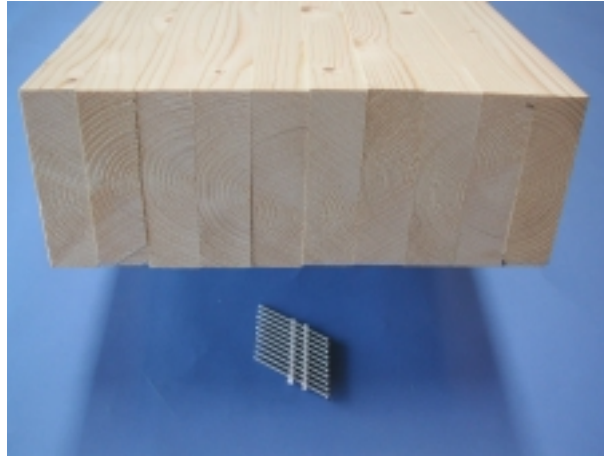


Figure 1: Nailed element specimen – i.e. a specimen not containing any adhesive components.



Figure 2: Phenol-Resorcinol-Formaldehyde (PRF) glued specimen.



Figure 3: Melamine-Urea-Formaldehyde (MUF) glued specimen.

All elements were produced by Limtræ Danmark A/S at the factory Lilleheden. The MUF glued element type was double tested: one specimen was taken from the material delivered at the construction site of the single-family house, for which in-situ measurements are reported in (Funch & Clorius, 2002). Another specimen was delivered directly from the factory together with the nailed and the PRF glued sample. The

materials used for the laboratory testing have been selected randomly and shall thus be considered as representative examples of the tested products.

3.2 Wood and glue

3.2.1 Wood material

The wood material used in the samples originates from the production stock of *picea abies* of Nordic origin, used in the production of glued laminated timber at Lilleheden.

3.2.2 Phenol-Resorcinol-Formaldehyde (PRF) glue

PRF adhesives are classified as thermosetting polymers and are produced by a condensation polymerisation between formaldehyde, phenol and resorcinol. PRF adhesives are cold-setting with very high weather and water resistance as well as heat resistance to be used in products for exterior (service class 3), humid (service class 2) and interior (service class 1) conditions.

The specific glue used in the tested PRF glued specimen was a DYNOSOL S-205 used with hardener H-622 or H-625, more specifications for the used glue are given in (Dyno Industrier, 1994). PRF glue is related to class I of (EN 301,1992).

3.2.3 Melamine-Urea-Formaldehyde (MUF) glue

MUF adhesives are classified as thermosetting polymers and are produced by a condensation reaction between melamine, urea and formaldehyde. These MUF polymers can be formulated to provide various degrees of water and weather resistance for use in exterior (service class 3), humid (service class 2) and interior (service class 1) conditions.

The glue used in the two tested MUF glued specimens was a DYNOMEL L-435 used with hardener H-469, more specifications for the used glue are given in (Dynea Oy, 1999). MUF glue is related to class I of (EN 301,1992).

3.3 Specimen geometry and preparation

All four test specimens consist of 10 lamellas each yielding nine glue-lines or lamella interstices in the glued and nailed specimens, respectively. The lamellas had the standard measure of 33.3 mm, hence the width of the specimens was 333 mm, the specimen length was 350 mm for the glued specimens and 700 mm for the nailed. The specimen thickness was 120 mm.

The glued specimens were sealed with aluminium foil on the small faces. Hence, only the wide faces with glue lines were open for emission. This gave a total emission surface of 231.000 mm² (2 x 330 mm x 350 mm) for the glued specimens.

The nailed specimen was sealed with aluminium foil on the small faces as well as on one of the wide faces. This gave a total emission surface of 233.100 mm² (1 x 333 mm x 700 mm) with a corresponding “inner surface” from the nine lamella interstices with 120 mm depth. A one-sided emission surface was chosen for the nailed elements in order to simulate the possible emission from the lamella interstices as a one sided phenomenon. This gives no undue contribution from airflow through the lamella interstices and neither a too little contribution from only half depth interstices that would have been the consequence of a double-sided specimen.

4. Test method

4.1 Test chamber conditions

The tests are carried out in accordance with (EN 717-1,1998). It is noted that the requirements in Annex B of (prEN 14080, 2002) are less severe than the standard test conditions specified in (EN 717-1,1998). The latter requires material load of $1 \text{ m}^2/\text{m}^3$ and 1 air change per hour, whereas (prEN 14080, 2002) requires $0.3 \text{ m}^2/\text{m}^3$ tested in a 1 m^3 chamber, and no specification of the air change. The more severe conditions of (EN 717-1,1998) have been used, this conforms to the requirements of (prEN 14080 (DRAFT), 2000) and (prEN 13986, 2001). The used climate chamber conditions are summarised in Table 1.

The glued elements were placed in the climate chamber standing on the sealed small face with the airflow parallel to the glue joints. The nailed element was standing on the sealed wide face with the airflow parallel to the lamella interstices.

Table 1: Summarised chamber conditions.

Climate chamber	225 l polished stainless steel
Temperature	$23^\circ \text{ C} \pm 0.5^\circ \text{ C}$
Relative humidity	$45\% \pm 3\%$
Ventilation	$1 \text{ h}^{-1} \pm 0.05 \text{ h}^{-1}$
Air velocity	$0.1 - 0.3 \text{ m/s}$
Material load	$1,03 \text{ m}^2/\text{m}^3$ (0,231/0,225 m^2/m^3)

4.2 Quantitative analysis

The chamber measurements of the element delivered at the construction site of the single-family house were carried out in July 2001. The measurements of the elements delivered directly from the factory Lilleheden were carried out in May 2002.

Air samples were taken from the chamber after 3 and 10 days. Furthermore, formaldehyde emission from a MUF-glued element was analysed after 28 and 56 days. Formaldehyde was sampled in distilled water and determined fluoremetrically by the acetylacetone method according to (EN 717-1,1998).

Other aldehydes were sampled on Sep-pack-tubes (C_{18} polymers, coated with 2,4-dinitrophenylhydrazin), and after elution with acetonitrile analysed by HPLC (high-pressure-liquid-chromatography) with UV detection. VOCs were sampled on Tenax, thermally desorbed and analysed by GC-MS (gas-chromatography with mass spectrometric detection) after addition of internal standards.

5. Results

The results from the chamber measurements are given in Table 2 for nailed specimen, Table 3 for PRF-glued specimen, Table 4 for MUF-glued specimen from factory and in Table 5 for MUF-glued specimen taken from building site.

Table 2: Emissions from nailed solid wood element. At the given test conditions the concentration in $\mu\text{g}/\text{m}^3$ equals the emission rate in $\mu\text{g}/\text{m}^2\text{h}$. All threshold values are from the VOCBase (Jensen & Wolkoff, 1996); # = threshold value for irritation according to the VOCBase. • = no measurement made at 28 days.

Chemical compound	3 days [$\mu\text{g}/\text{m}^3$]	10 days [$\mu\text{g}/\text{m}^3$]	28 days [$\mu\text{g}/\text{m}^3$]	Odour threshold [$\mu\text{g}/\text{m}^3$]
<i>Aldehydes</i>				
Formaldehyde	< 10	< 10	•	150 # (E1=124)
Acetaldehyde	47	23	•	340
Propanal	6	4	•	14
Butanal	4	2	•	28
Pentanal	18	11	•	22
Hexanal	43	29	•	58
Benzaldehyde	3	-	•	186
<i>Sum of aldehydes</i>	122	69	•	
<i>Ketones</i>				
Acetone	61	33	•	14000
<i>Alifatic Hydrocarb.</i>				
C10-C16	33	4	•	7760 (C-11)
<i>Aromatic hydrocarb.</i>				
Toluene	4	<1	•	644
Xylene/ethylbenzene	1	<1	•	2140
<i>Sum of aromatic hydrocarb.</i>	5	<1	•	
<i>Monoterpenes</i>				
α -Pinene	190	60	•	3890
β -Pinene	22	9	•	36000
3-Carene	9	4	•	2450
Limonene	19	10	•	2450
<i>Sum of monoterpenes</i>	240	83	•	

Table 3: Emissions from solid wood element glued with Phenol-Resorcinol-Formaldehyde. At the given test conditions the concentration in $\mu\text{g}/\text{m}^3$ equals the emission rate in $\mu\text{g}/\text{m}^2\text{h}$. All threshold values are from the VOCBase (Jensen & Wolkoff, 1996); # = threshold value for irritation according to the VOCBase. • = no measurement made at 28 days.

Chemical compound	3 days [$\mu\text{g}/\text{m}^3$]	10 days [$\mu\text{g}/\text{m}^3$]	28 days [$\mu\text{g}/\text{m}^3$]	Odour threshold [$\mu\text{g}/\text{m}^3$]
<i>Aldehydes</i>				
Formaldehyde	27	15	13	150 # (E1=124)
Acetaldehyde	672	404	•	340
Propanal	12	7	•	14
Butanal	5	2	•	28
Pentanal	18	12	•	22
Hexanal	27	20	•	58
Benzaldehyde	5	3	•	186
Decanal	17	6	•	6
<i>Sum of aldehydes</i>	783	469	•	
<i>Ketones</i>				
MIBK/Hexanone	6	4	•	-
Acetone	45	61	•	14000
<i>Sum of ketones</i>	51	65	•	
<i>Alifatic Hydrocarb.</i>				
C10-C16	52	15	•	7760 (C-11)
<i>Aromatic hydrocarb.</i>				
Toluene	6	2	•	644
Xylene/ethylbenzene	13	6	•	2140
<i>Sum of aromatic hydrocarb.</i>	19	8	•	
<i>Monoterpenes</i>				
α -Pinene	310	100	•	3890
β -Pinene	77	40	•	36000
3-Carene	45	20	•	2450
Limonene	75	35	•	2450
b-myrcene	6	2	•	
Phellendrene	8	3	•	
<i>Sum of monoterpenes</i>	522	200	•	

Table 4: Emissions from solid wood element glued with Melamine-Urea-Formaldehyde, specimen 1 from factory. At the given test conditions the concentration in $\mu\text{g}/\text{m}^3$ equals the emission rate in $\mu\text{g}/\text{m}^2\text{h}$. All threshold values are from the VOCBase (Jensen & Wolkoff, 1996); # = threshold value for irritation according to the VOCBase. • = no measurement made at 28 days.

Chemical compound	3 days [$\mu\text{g}/\text{m}^3$]	10 days [$\mu\text{g}/\text{m}^3$]	28/56 days [$\mu\text{g}/\text{m}^3$]	Odour threshold [$\mu\text{g}/\text{m}^3$]
<i>Aldehydes</i>				
Formaldehyde	107	69	73/50	150 # (E1=124)
Acetaldehyde	75	43	•	340
Propanal	13	8	•	14
Butanal	3	<1	•	28
Pentanal	18	14	•	22
Hexanal	35	25	•	58
Benzaldehyde	8	4	•	186
<i>Sum of aldehydes</i>	259	163	•	
<i>Ketones</i>				
Acetone	53	35	•	14000
<i>Alifatic Hydrocarb.</i>				
C10-C16	34	16	•	7760 (C-11)
<i>Aromatic hydrocarb.</i>				
Toluene	6	2	•	644
Xylene/ethylbenzene	5	<1	•	2140
<i>Sum of aromatic hydrocarb.</i>	11	2	•	
<i>Monoterpenes</i>				
α -Pinene	245	90	•	3890
β -Pinene	27	13	•	36000
3-Carene	7	4	•	2450
Limonene	17	12	•	2450
<i>Sum of monoterpenes</i>	296	119	•	

Table 5: Emissions from solid wood element glued with Melamine-Urea-Formaldehyde, specimen 2 from building site. At the given test conditions the concentration in $\mu\text{g}/\text{m}^3$ equals the emission rate in $\mu\text{g}/\text{m}^2\text{h}$. All threshold values are from the VOCBase (Jensen & Wolkoff, 1996); # = threshold value for irritation according to the VOCBase. • = no measurement made at 28 days.

Chemical compound	3 days [$\mu\text{g}/\text{m}^3$]	10 days [$\mu\text{g}/\text{m}^3$]	28 days [$\mu\text{g}/\text{m}^3$]	Odour threshold [$\mu\text{g}/\text{m}^3$]
<i>Aldehydes</i>				
Formaldehyde	96	64	52	150# (E1=124)
Acetaldehyde	67	35	20	340
Propanal	4	5	3	14
Butanal	6	4	1	28
Pentanal	27	19	11	22
Hexanal	56	40	25	58
Benzaldehyde	2	2	3	186
<i>Sum of aldehydes</i>	258	169	115	
<i>Ketones</i>				
MIBK/Hexanone	Failing	5	5	-
Acetone	56	44	29	14000
<i>Sum of ketones</i>	56	49	34	
<i>Alifatic Hydrocarb.</i>				
C10-C16	Failing	8	8	7760 (C-11)
<i>Aromatic hydrocarb.</i>				
Toluene	Failing	26	20	2140
Xylene/ethylbenzene	Failing	44	39	644
<i>Sum of aromatic hydrocarb.</i>	-	70	59	
<i>Monoterpenes</i>				
α -Pinene	Failing	77	42	3890
β -Pinene	Failing	14	10	36000
3-Carene	Failing	7	6	2450
Limonene	Failing	22	14	2450
<i>Sum of monoterpenes</i>	-	120	72	
<i>Others</i>				
Butyl acetate	Failing	6	5	47
Propanoic acid	Failing	5	3	110

6. Interpretation and discussion

The results of the formaldehyde concentration is evaluated in accordance with (prEN 14080 (DRAFT), 2000) for all elements. As seen from Tables 2-5 the concentrations of formaldehyde are below the E1-level for all types of solid wood elements. Regarding formaldehyde emission the observations are:

- The emission of formaldehyde from the nailed element Table 2 is as expected very low. Solid wood is known to have low emission of formaldehyde about 4-8 $\mu\text{g}/\text{m}^3$, (Boehme, 2000).
- The PRF glue has a low emission of formaldehyde 13 $\mu\text{g}/\text{m}^3$ after 28 days, Table 3, this is due to the chemical bonding between Phenol and Formaldehyde being both strong and stable.
- The emission of Formaldehyde from the element glued with Melamine-Urea-Formaldehyde in the order 70-50 $\mu\text{g}/\text{m}^3$ at the measurements after 10 and 28 days, Table 4 and Table 5. It is observed that the concentration is reduced to approximately 50% of the 3 days value after 28 days.

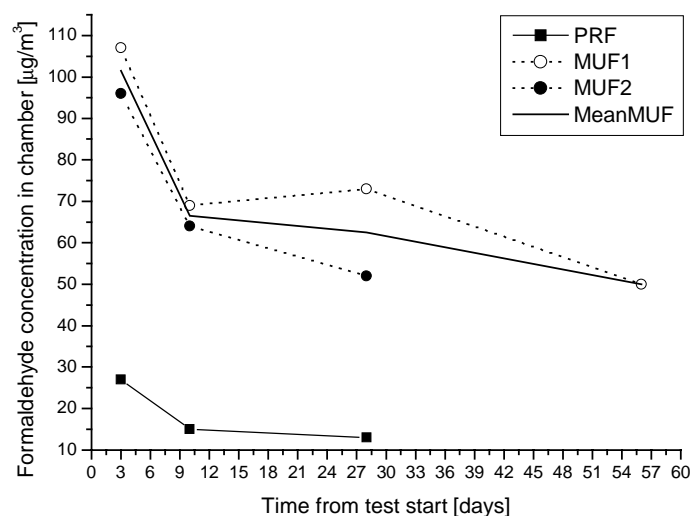


Figure 4: Formaldehyde concentrations in test chamber from MUF and PRF glued specimens at 3, 10, 28 days and for MUF specimen1 also at 56 days.

Unlike formaldehyde, no requirements exist regarding VOC emissions measured by the climate chamber method. Hence, the results of the VOC emissions are not evaluated against any standard requirements. In Tables 2 to 5 the odour threshold values according to (Jensen & Wolkoff, 1996) are given. The main observations regarding VOC emissions can be summarised:

- The measured VOCs are characterised by being dominated by aldehydes, mainly acetaldehyde and hexanal, and monoterpenes, mainly α -pinene, β -pinene, 3-carene and limonene.

- The results of the total amount of terpenes emitted after 10 days vary from 83 to 200 $\mu\text{g}/\text{m}^3$. According to (Miljøprojekt Nr. 501, 1999), this variation is considered normal for softwood, because the emission of terpenes is dependent on factors like growth conditions and origin.
- As the measured emission of acetaldehyde from the PRF-glued element was 10 times higher than the nailed and the MUF-glued element, it can be concluded that the PRF-glue is a source of acetaldehyde.
- Besides acetaldehyde emitting from the PRF-glued element, Table 3, all the measured concentrations are below the odour threshold.

A discussion of health- and comfort threshold values of some aldehydes and monoterpenes are given in (Funch & Clorius, 2002) with regard to the recommendations given for the indoor environment given in (Nielsen et al., 1996) and by WHO, (World Health Organisation, 2000).

7. Concluding remarks

Nailed, Phenol-Resorcinol-Formaldehyde glued, and Melamine-Urea-Formaldehyde glued solid wood elements have been tested for emissions by the climate chamber method, according to the requirements in (EN 717-1,1998). The emissions of formaldehyde are below the E1 requirement of (prEN 14080 (DRAFT), 2000), this threshold is the same as used for wood-based panels, (prEN 13986, 2001).

Regarding Volatile Organic Compounds the testing shows that besides acetaldehyde emitting from the PRF-glued element all the measured concentrations are below the odour threshold as given in (Jensen & Wolkoff, 1996).

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