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Emission of formaldehyde from indoor surface materials

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SUMMARY
Measurements of formaldehyde emission from wooden boards, insulation and paints were performed as a part of a project aiming to identify major sources of formaldehyde among construction and finishing products in Danish residential buildings. The methodology of measuring followed standard DS/EN 717-1:2004. The concentrations differed widely between tested materials. The initial concentration ranged from below 0.01 mg/m\textsuperscript{3} to 0.115 mg/m\textsuperscript{3} and it dropped below 0.01 mg/m\textsuperscript{3} for 7 out of 12 the investigated materials at the end of the measuring period, while for the remaining 5 materials it did not change noticeably. All measured concentrations were below Danish requirements, however performed calculations of formaldehyde concentration in a room finished with investigated materials suggest that indoor concentrations above 0.1 mg/m\textsuperscript{3} recommended by WHO are well-founded, and concentrations as high as 0.5 mg/m\textsuperscript{3} can be expected.

KEYWORDS
Formaldehyde, wooden boards, insulation, paint

INTRODUCTION
Building and finishing materials comprise some of the major sources of pollution in residential buildings. One common indoor pollutant with a wide range of sources is formaldehyde. This chemical compound has been known as a major irritant in indoor air for many years and in 2006 it was recognized to be carcinogenic to humans (IARC 2006). Recently, measurements of formaldehyde concentrations in 20 new houses in Denmark have been made (Gunnarsen et al. 2008). All of the houses were erected and occupied within the last seven years. The mean indoor air concentration was 0.05 mg/m\textsuperscript{3} with a standard deviation of 0.026 mg/m\textsuperscript{3}. The concentrations of formaldehyde were higher than the WHO guideline value of 0.1 mg/m\textsuperscript{3} (WHO, 2000) in two houses. The newer houses had the highest formaldehyde concentrations in this study. Relatively high concentrations in homes were found in several other studies. Concentrations of formaldehyde measured in 185 homes in Perth, Australia, were in the range 0.0006 – 0.11 mg/m\textsuperscript{3} with negative correlation between age of the house and formaldehyde levels (Dingle and Franklin, 2002). In a study by Marchand et al. (2006) conducted in 22 residential homes in Strasbourg, France, formaldehyde concentration was in the range 0.013 – 0.123 mg/m\textsuperscript{3}, and in 399 Turkish homes it was from 0 to 1.1 mg/m\textsuperscript{3} (Vaizoglu et al. 2003).

These results pointed out a need to look for sources of formaldehyde in newly constructed buildings. The current regulation in Denmark for formaldehyde emission from wood-based construction products indicates that the indoor air concentration in an emission chamber should not exceed 0.124 mg/m\textsuperscript{3} (DN/EN 13986:2004). This has probably induced a development where traditional sources such as wood based construction board less often emit
high amounts of formaldehyde. Instead, indoor concentrations of formaldehyde are probably affected by small contributions from many other sources, including formaldehyde releasers often used as antimicrobial preservatives. Thus the objective of this project is to identify major sources of formaldehyde among construction and finishing products in Danish residential buildings. The measurements included wooden boards, insulation products, commercially sold furniture and paints.

METHODS
The measurements included 9 wooden boards (used as roof, flooring or furniture materials), 2 commercially sold pieces of furniture (bookcase and chest of drawers), two types of insulation and two types of paints for indoor use (non-glossy and glossy, used e.g. in bathrooms or kitchens). The 9 boards included: 3 plywood boards, 2 chipboards, 2 medium density fibreboards (MDF) and 2 oriented strand boards (OSB). The two pieces of furniture were made of chipboard.

After purchasing the wooden boards from construction supply stores they were placed in a large ventilated room at 20-24 °C and 30-50 % RH for 2-3 weeks. Later they were cut to smaller pieces and wrapped in aluminium foil for transportation to the laboratory. Immediately before testing, they were unwrapped and cut to size (DS/EN ISO 16000-11:2006). The edges of wooden boards were partly sealed. The ratio of open (unsealed) edges to surface area of test piece was 1.5 m/m², which was in agreement with requirements (DS/EN 717-1:2004). Sealing of the edges was not possible for insulation materials, thus area of the edges was included in the calculation of the total area of each test piece. Paints were applied on gypsum board (once side), which was assumed to be formaldehyde free. The amount of paint applied to the board was calculated based on manufacturer instruction (amount of paint per square meter). Two coats were applied. Painted gypsum boards were placed in the chamber immediately after painting.

The set up was prepared according to standard DS/EN 717-1:2004, Annex A3. Measurements were done in polished stainless steel chambers with a volume of 0.225 m³. The temperature was kept at 23 °C (± 0.5 °C) and relative humidity at 45 % (± 3 %). The air exchange rate was 1 h⁻¹ (± 0.05 h⁻¹) and an area specific ventilation rate was 1 m³/(m²h). According to the standard (DS/EN 717-1:2004), the measurements shall be continued until a steady-state is obtained. However, in order to limit expenses, measurements were shortened to 5-6 days for samples with low initial concentrations. The concentration of formaldehyde was recorded continuously using a Skalar online process analyzer (model SA 9101) with a detection limit of 0.01 mg/m³. It was used to monitor concentration decay and determine the steady state. The final concentration of formaldehyde was also determined manually by using acetylacetone method as described in standard DS/EN 717-1:2004 and this value was taken as the final concentration.

RESULTS
The results are presented in Table 1. The concentrations differed widely between tested materials. The initial concentration ranged from below 0.01 mg/m³ for one type of insulation to 0.115 mg/m³ for MDF board and chipboard. At the end of measuring period, the concentration dropped below 0.01 mg/m³ for 7 out of the 12 investigated materials. The rest of the materials had a steady state concentration similar to the initial one. None of the tested materials exceeded the maximum concentration of formaldehyde permissible for E1 class materials (DN/EN 13986:2004). The measurements showed differences in formaldehyde emission behaviour across investigated materials (Figure 1). While exponential decay was observed for plywood boards and insulation materials, chipboard, MDF and OSB board showed relatively constant emission during the whole duration of the measuring period.
Table 1. Initial and steady state formaldehyde concentrations (mg/m$^3$) in emission chamber.

<table>
<thead>
<tr>
<th>Material</th>
<th>Initial concentration</th>
<th>Steady state concentration</th>
<th>Material</th>
<th>Initial concentration</th>
<th>Steady state concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plywood board 1</td>
<td>0.033</td>
<td>&lt;0.01 (0.006)</td>
<td>OSB board 2</td>
<td>0.068</td>
<td>0.060</td>
</tr>
<tr>
<td>Plywood board 2</td>
<td>0.014</td>
<td>&lt;0.01 (0.007)</td>
<td>Chipboard bookcase</td>
<td>0.045</td>
<td>0.040</td>
</tr>
<tr>
<td>Plywood board 3</td>
<td>&lt;0.01 (0.007)</td>
<td></td>
<td>Chipboard drawer</td>
<td>0.045</td>
<td>0.035</td>
</tr>
<tr>
<td>Chipboard 1</td>
<td>0.115</td>
<td>0.100</td>
<td>Insulation 1</td>
<td>0.045</td>
<td>0.010</td>
</tr>
<tr>
<td>Chipboard 2</td>
<td>0.048</td>
<td>0.045</td>
<td>Insulation 2</td>
<td>&lt;0.01 (0.006)</td>
<td>&lt;0.01 (0.004)</td>
</tr>
<tr>
<td>MDF board 1</td>
<td>0.115</td>
<td>0.100</td>
<td>Paint 1 (gloss 6)</td>
<td>0.025</td>
<td>0.01</td>
</tr>
<tr>
<td>MDF board 2</td>
<td>0.015</td>
<td>&lt;0.01 (0.008)</td>
<td>Paint 2 (gloss 30)</td>
<td>0.015</td>
<td>&lt;0.01 (0.006)</td>
</tr>
<tr>
<td>OSB board 1</td>
<td>0.010</td>
<td>&lt;0.01 (0.008)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*The steady state concentrations express the concentration obtained at the end of measuring period (for most of the materials after 10 days), even though the steady state was obtained already after 2-3 days for some materials. \( ^b \)Information not available due to problems with instrument

Figure 1. Formaldehyde emission from chipboard 1 (Left) and plywood 1 (Right).

DISCUSSION

The concentrations measured in this study are comparable or lower than concentrations measured in earlier studies. Kelly et al. (1999) investigated formaldehyde emission rates from a wide selection of products, including both dry and wet products. The concentrations varied from below 0.001 mg/m$^3$ for melamine laminated products up to 0.73 mg/m$^3$ for urea-formaldehyde bonded chipboard. For non-laminated products, the lowest emission was measured from plywood boards (4.1-170 µg/m$^2$h, which corresponds to 0.002-0.078 mg/m$^3$) and the highest from chipboard (104-1580 µg/m$^2$h, which corresponds to 0.048-0.727 mg/m$^3$), which was in agreement with the results from the present study.

Brown (1999) measured formaldehyde emission from MDF plates and chipboard plates. The measurements started 7 days after the materials were manufactured and continued for 20 days. After completion of the emission experiments, the plates were stored in controlled environment for several months, after which long-term formaldehyde emission was measured for 2 days. Initial concentration was higher for chipboard plates (0.159-0.219 mg/m$^3$) than MDF plates (0.131-0.156 mg/m$^3$). Formaldehyde emission in the first weeks after manufacture was well described by a first-order exponential decay model, but the decay was more pronounced for chipboard plates. When the results from the long-term measurements were included into the model, decay rates became much slower and emissions were consistent with a double exponential decay model, which were quick initially and much slower after some weeks.

The manufacture date of materials investigated in the present experiment is not known. The materials were not specially selected, but purchased as they would be by an ordinary
customer. The gentle slope of decay curve for some of the products could therefore probably be explained by long storage time.

Formaldehyde emission from all products investigated in the present study was below the limits that are mandatory in Denmark, thus the results did not give direct explanation for the relatively high concentrations measured in Danish homes during phase 1 of the project (Gunnarsen et al. 2008). It is expected that these high concentrations are not caused by one strong source but are result of a small contribution from many sources. Calculated formaldehyde concentration in the model room (floor area of 7 m², wall area of 24 m² and volume of 17.4 m³, ventilated by 0.5 h⁻¹ of outdoor air, DS/EN ISO 16000-9:2006) painted by paint 1 (formaldehyde emission 0.01 mg/m³·h) and with floor covered by chipboard 1 (formaldehyde emission 0.1 mg/m²·h) is 0.123 mg/m³, which is above the value recommended by WHO. Moreover, if this calculation is based on the highest possible emission for E1 products (0.124 mg/m³) and all surfaces (ceiling, walls and floor) are made of products emitting formaldehyde at the permissible rate, the concentration is estimated to be 0.54 mg/m³, which is five times the WHO limits. This calculation does not include emission from furniture, textiles, carpets, cleaning products etc., thus the real value could be even higher.

CONCLUSIONS
Formaldehyde emission was measured from several building and finishing products. Chipboard and MDF were the strongest sources, however both initial and steady state concentrations of formaldehyde were below limits that are mandatory in Denmark and EU. Calculations of formaldehyde concentration in a room finished with investigated materials resulted in values above the 0.1 mg/m³ recommended by WHO. As a consequence CE marking does not assure that concentrations in indoor air are below the WHO guideline value.

REFERENCES