Final report

Analysis of consumer exposure associated with the use of products and articles containing formaldehyde – based resins

Executive summary

Consumer exposure to formaldehyde in Europe has been assessed in the scope of the determination of the most appropriate Risk Management Option for formaldehyde. Main sources of formaldehyde in indoor situations include 'man made sources' like wood based materials, carpets, textiles, paints and mineral wools, as well as 'natural sources' like fire, smoke and air pollution. This study focusses on the first type of sources.

Firstly, average and reasonable worst case indoor air inhalation exposure have been assessed by use of measured data described in literature. Measurements were all taken indoors, with a varying description of determinants. Based on a data set of >2500 measurements, the central tendency of the data turned out to be around 0.025 mg/m^3, whereas the reasonable worst case level as determined from all data is 0.085 mg/m^3. Measurements in new homes do (probably temporarily) show slightly elevated levels of formaldehyde. Due to the rather limited information on determinants, it was not possible to draw relations between different determinants and indoor air exposure. As a consequence, conclusions on whether this assessment would be sufficiently worst case, could not be drawn.

Secondly, the emission rates of different product types have been compared to each other. Emission rates show a large range over the different product types and also within a product category. The testing method has a relatively large influence on the test result. The results indicate that the main sources of formaldehyde emission are the uncoated materials and plywood, whereas products like paints, mineral wools and foams have lower emissions. In general emissions decrease over time.

Finally, to investigate the reasonable worst case exposure levels in newly built houses with new materials like floorings and furniture, model estimates have been performed. The necessary assumptions were made in an expert panel, and include assumptions on emission rates, loading rates and influence of coating on the evaporation. Two scenarios were calculated based on the current industry standards for emission rates. The E1 criteria did lead to an estimated indoor air exposure of 0.093 mg/m^3, whereas the estimated exposure based on the E1+ criteria is 0.079 mg/m^3.

Taking all data and estimates into account, it can be concluded that the general concentration of formaldehyde in homes, as well as a reasonable worst case estimate of exposure in new homes, are below the reference value. The emission rate from materials is one of the key determinants in the exposure assessment and restriction of this emission rate appears to be technically feasible.

Contents

1. Introduction

Formaldehyde (CAS-number 50-00-0) is an important industrial chemical that is mainly used in the production of adhesives or binder resins. It has been registered under the REACH-regulation in 2010. In the Chemical Safety Report (CSR) of 2010 the exposure of consumers to formaldehyde was not yet described. Therefore available literature was reviewed for data on formaldehyde indoor air concentration. Both indoor air concentrations (mg/m^3) and emission data ($mg.m^{-2}.h^{-1}$) were reviewed, focussing primarily on wood based products containing formaldehyde. Data were compared to the DNEL for the general public of 0.1 mg/m^3 (0.081 ppm). This report describes the main findings.

A list of abbreviations is provided in Annex I.

2. Formaldehyde indoor air concentrations

According to Järnström (2007) the formaldehyde indoor air concentration is influenced by the:
- Outdoor pollutant concentration;
- Ventilation rate;
- Indoor source strength;
- Inside mixing conditions;
- Pollutant decay rate.

Formaldehyde indoor air concentrations are generally higher than outdoor air concentrations, with a higher ratio between indoor/outdoor concentration for urban areas (Salthammer, Mentese and Marutzky, 2010, Roda *et al.*, 2011, Santarsiero and Fuselli, 2008). Higher average values were found in Central and North European countries compared to Southern European countries (Sarigiannis *et al.*, 2011). The relative humidity in indoor air was significantly positively correlated with the concentration of formaldehyde. Higher concentrations were also found when temperatures increased (Boehme, 2000). Partharasathy *et al.* (2010) showed with experiments that the logarithm of the emission value increases linearly with temperature and that the emission is also higher for higher relative humidity. The influence of season, as seen by Raw *et al.* (2004), with the highest values in autumn and by Roda *et al.* (2011), with the highest values in the hot season, may also relate to these factors. Lower concentrations were found in buildings with mechanical supply air (Järnström, 2007). Usually higher values are found in new houses due to release of formaldehyde from construction materials and new furniture. Therefore a distinction is made between measurements data from older houses and new prefabricated houses.

2.1 Older houses

Different studies have investigated the indoor air concentration of formaldehyde in (randomly selected) houses. The sampled houses are mostly of varying ages.

For the selection of relevant literature studies the following criteria were used:
- Only relatively new measurement data, here defined as performed >1990, have been included. This is because the products used as construction material and for e.g. furniture have changed over the years. Data from more recent studies are considered to be more representative of the situation with present materials than older studies.
- Focus on measurement data from EU studies, as this is where the REACH regulation is in force and these measurement situations and the construction materials used reflect the current situation in Europe.
- Houses where people smoke are excluded as far as possible from this overview, because of the known emission of formaldehyde from smoking.

Figure 1 gives an overview of 13 recently performed studies, with >2500 measurement points. For each study the minimum, mean (in red) and maximum values are presented. The results indicate that a relatively conservative estimate of the central tendency of the concentration in older homes is approximately **0.025 mg/m^3** with almost all maximum values below the DNEL of 0.1 mg/m^3. The approximate 'reasonable worst case value' based on these 13 datasets is estimated by a value in the upper area of the total dataset. A rounded value of 0.085 mg/m^3 is considered to be the reasonable worst case. This is between the third and fourth highest maximum of the 13 datasets. In six homes (0.2%) the DNEL was exceeded by the 3-day average concentration in the study performed by Raw (2004). Of

these six homes five were built > 1995, that is, within about three years of the study. The study also included homes where smoking occurred. It is not clear whether in the homes where the limit was exceeded smoking occurred, but the authors do not indicate a significant effect of smoking on the concentrations of formaldehyde.

The value of 0.025 mg/m^3 is in line with other findings:
- Järnström (2007) mentions an average or geometric mean indoor air concentration of 0.012-0.022 mg/m^3 for established dwellings (>2 years old).
- Salthammer, Mentese and Marutzky (2010) indicate that the average exposure of the population to formaldehyde appears to lie between 0.020-0.040 mg/m^3
- Sarigiannis *et al.* (2011) indicate a typical indoor air concentration between 0.010-0.050 mg/m^3

However, outside the EU much higher values, exceeding the DNEL, are reported, specifically for Cairo, Tianjin and Bejing (Zhang *et al.*, 2009).

Figure 1. Overview of formaldehyde indoor air concentration (mg/m^3) of different EU studies
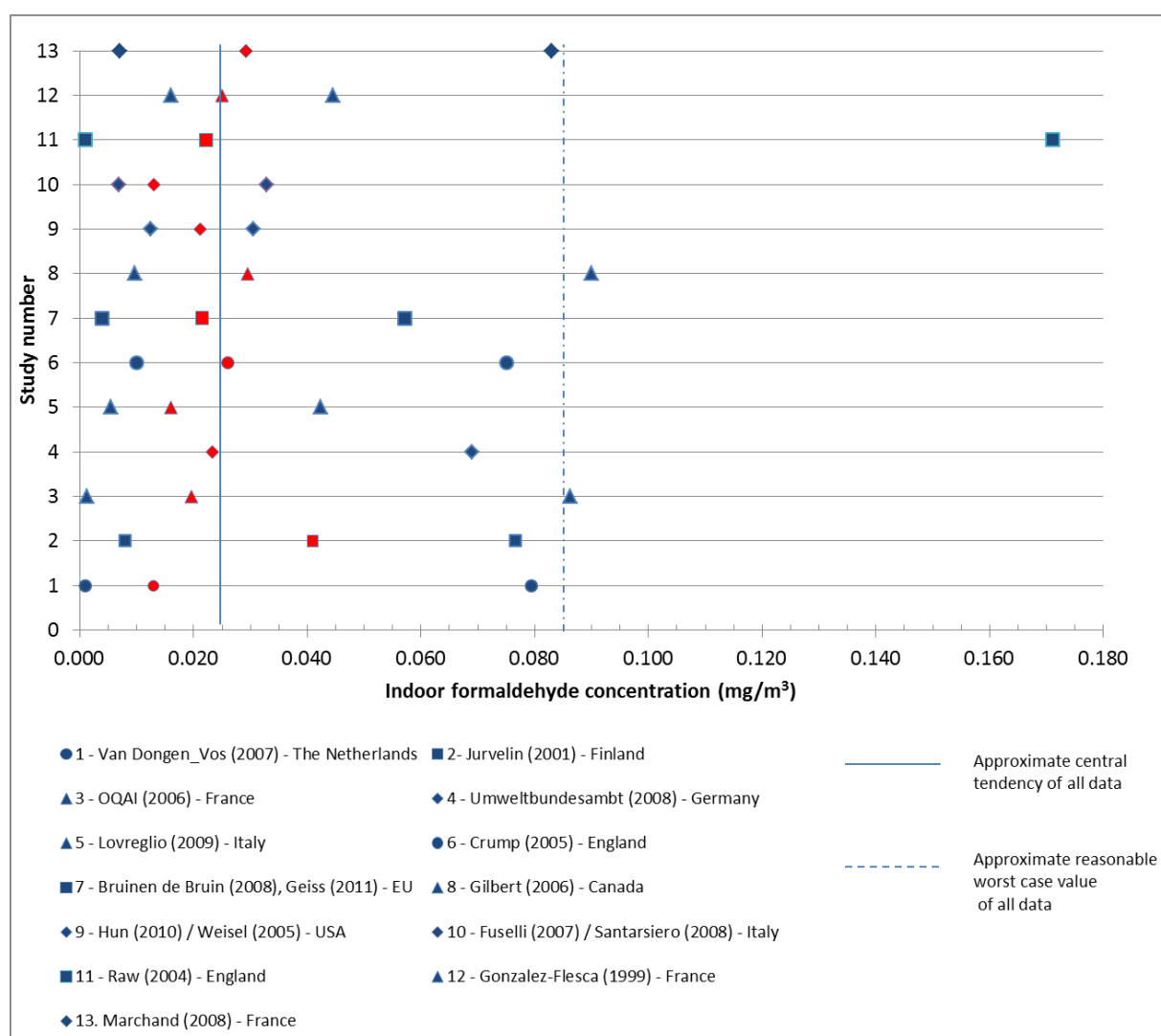
Some remarks can be given on these studies to put the results into perspective.

Van Dongen and Vos (2007) measured weekly average concentrations in a selection of 359 houses that was weighted to provide representative data on social housing, private rent houses and inhabitant owned houses. Measurements were done from October to March, *i.e.* in the season when houses are heated. Temperatures in the houses were on average around 20-21 °C and relative humidity was on average around 44-45%.

Jurvelin *et al.* (2001) measured 48 hour average concentrations in 15 houses in Helsinki from May to September. In one of the houses smoking occurred. Temperature and humidity were not recorded.

The French 'Observatoire de la Qualité de l'Air Interieur' (OQAI, 2006) measured weekly average concentrations of formaldehyde in 554 houses throughout France in a period of two years, reasonably spread over the seasons. The average temperatures were around 20-22 °C and the average relative humidity was between 45 and 55%.

The Umweltbundsamt in Germany performed a study in 586 houses, spread over Germany, between May 2003 and May 2006 (Schultz*et al.*, 2010). Formaldehyde concentrations were significantly different between the following situations:

- Renovation within the last 12 months (increased geometric mean if yes)
- Flooring or wall covering wood based (decreased geometric mean if yes)
- Particle board furniture in the room (increase in geometric mean from none via some to many pieces)
- Newer particle board furniture in the room (increased geometric mean if newer)
- Solid wooden furniture in the room (decrease geometric mean if yes)

Temperature and relative humidity were not recorded.

Lovreglio *et al.* (2009) did measurements of 24 hour average concentrations in 59 homes in southern Italy in the spring and subsequently in 20 homes of non-smokers between January and May. In the first study the concentrations appeared to increase with number of cigarettes smoked during the measurement. Mean concentration in 31 homes of non-smokers was significantly increased in homes with furniture that was bought or restored within a year before measurements. In the second study concentrations were higher in January and February, much lower in March and April and higher again in May.

The study of Crump *et al.* (2005) measured three day average concentrations in 37 houses in summer and winter (January to March). There was a higher geometric mean in summer, but a higher minimum and maximum value in winter. The measured mean air exchange rate in winter was 0.44 air exchanges per hour and in summer 0.62. The average relative humidity was 42% in winter and 52% in summer.

The Airmex study reported by Bruinen de Bruin *et al.* (2008) and Geiss *et al.* (2011) measured week average indoor air concentrations in 79 homes in 8 cities throughout Europe, both in cold and in warm months. The concentrations in Northern Europe appeared to be higher than in Southern Europe and the concentrations in colder months were slightly higher compared to warmer months.

Gilbert *et al.* (2006) measured 24 hour average concentrations in homes in Quebec, Canada in the period of January to April. The geometric mean temperature was just over 20 °C, while the relative humidity was on average rather low, with a geometric mean of 31%. The geometric mean of the air exchange rate was also low (0.2 per hour). In a multiple regression analysis, a positive relation with formaldehyde concentration was seen for:

- electrically heated houses, compared to oil, gas or other heated houses (very strong relation)
- new wooden furniture installed within a year of measurements
- the sampled room was painted or varnished within a year of the measurements (very strong relation)
- lower air change rates (very strong relation).

There was no significant effect of new 'melamine furniture' and relative humidity in this analysis.

Hun *et al.* (2010) report measurements in homes of one or two 48 hour periods (three months apart if there are two periods). Measurements were done in 179 homes in California, Texas and New Jersey (USA) and most homes were more than 5 year old. The first period was in summer and the second (where done) in autumn. Air change rates were relatively high (median of 0.8 per hour) and so was temperature (median 24 °C), while the median relative humidity was 45%. In this study no relation was found with temperature, nor with ventilation rate, building type or age of the building.

The study of Fuselli *et al.* (2007) and Santarsiero and Fuselli (2008) was on 10 apartments (wholly refurbished in the last ten years) in the Rome area (Italy). Ten day average values are reported for four periods (June, July, September and December). By means of Principal Component Analysis the authors conclude that the formaldehyde concentrations are mainly produced by indoor sources.

Raw *et al.* (2004) report three day average concentrations in 833 bedrooms in throughout England. They showed a significant effect of season, with a clear difference between autumn (high) and winter (low). The authors indicate that emissions from materials in new homes influence exposure levels. No other determinants for formaldehyde concentrations were found in this study.

Gonzalez-Flesca *et. Al.* (1999) measured indoor air concentrations as part of a pilot study on personal exposure to a number of components for 10 volunteers in Nancy, France. Indoor measurements were done in the bedroom of each volunteers during one working week. The average indoor temperature was 20 °C.

Marchand *et al.* (2008) studied 162 dwellings in Strasbourg, France. Measurements were done in the living room and the bedroom (though not always in both). Concentrations in the bedroom were slightly higher than in the living room with a median of 0.031 mg/m^3 for the bedroom and 0.027 mg/m^3 for the living room (n = 143 for both). All individual values were below 0.1 mg/m^3 and the averages of living room and bedroom ranged from 0.007 to 0.083 mg/m^3.

Most of these studies used either a passive badge method or an active sampling method with the DNPH (dinitrophenyl hydrazine) derivation (NIOSH 2016 method or EPA IP-6 method). Crump *et al.* (2005) did not report details of the analysis method, Hun *et al.* (2010) used derivatisation with dansylhydrazine (DNSH), Raw used acetonitril desorption (ISO/DIS 16,000-4 method).

2.2 New (prefabricated) houses

In new prefabricated houses indoor air concentrations of formaldehyde might be (temporarily) increased due to emission of formaldehyde from building materials and new articles like flooring and furniture.

- Measurements carried out in new prefabricated houses resulted in a median concentration of 0.049 mg/m^3 but with 14% of the measurements > 0.1 ppm (Salthammer, Mentese and Marutzky 2010). However, during the last series of measurements (2006) the DNEL was not exceeded.
- Järnström (2007) summarizes some studies in new prefabricated houses (< 2 years old). The more recent EU studies indicate that levels are generally below 0.1 mg/m^3, with a 95th percentile value of 0.103 reported by Thalmann and Coutalides (2006, referenced by Järnström, 2007) in a Swiss study in 19 dwellings.
- A study by Järnström (2006 & 2007) in newly constructed houses with low emitting materials (< 0.050 mg.m^{-2}.h^{-1} *i.e.* within the Finnish M1 criteria) showed average concentrations of 0.022 mg/m^3 (0.013 - 0.037 mg/m^3) with a slightly increasing trend during the first year from 0.019 mg/m^3 to 0.026 mg/m^3. However, measurement duration was only two hours in a closed room.
- Marchand *et al.* (2008) found a tendency that newer houses have higher formaldehyde indoor air concentrations and that furniture being added <1 year ago also results in higher concentrations. Older ceiling materials seem to result in lower indoor air concentrations. This is in line with findings of Clarisse (2003) who found higher levels of formaldehyde when floor coverings or wall coverings

were refurbished <1 respectively <6 months ago and with findings of Sakai *et al.* (2004) who found higher indoor levels in dwellings <10 years old compared with older dwellings.

- Hun *et al.* (2010) found no statistical correlation between the concentration of formaldehyde and air exchange rate, building type and age of the building in a study in the USA.
- In Austria a study is being performed on indoor air quality, measuring formaldehyde concentrations in approximately 60 new buildings. Measurements are done in living rooms and bedrooms of houses with and without modern ventilation systems 3 months after the inhabitants moved in and 12 months later. Houses are furnished, but no information is gathered on the area of furniture based on wood based panels in the measured rooms. The first sets of results show median values (from about 60 measurements) in conventionally ventilated buildings of approximately 40 µg/m³ and in modern energy efficient buildings of 20-30 µg/m³. In the conventionally ventilated buildings three measurements were at or above the reference value: 100 (n=2) and 110 µg/m³ and in energy efficient buildings two values were above the reference value: 110 and 140 µg/m³ (Tappler, undated presentation). Details have been made available from measurements in houses that have wood (or wood based material) as main material for walls as well as a flooring material made of wood or cork in the bedroom. These houses can be considered the worst case in relation to formaldehyde emission from construction materials. Results are presented in Table 1.

Table 1. Results of measurements in new houses with walls and floor covering in the bedrooms of wood or wood based material

Parameter \ Ventilation[1]	Natural	Mechanical [2]
Number of houses	20	33
# measurements living room and bedroom each 3 months	20	33
# measurements living room and bed room each15 months	7	26
GM formaldehyde concentration living room 3 months ($\mu g/m^3$)	37.4	34.6
GSD formaldehyde concentration living room 3 months	1.52	1.49
90th percentile formaldehyde concentration living room 3 months ($\mu g/m^3$)	64.1	57.5
GM formaldehyde concentration bedroom 3 months ($\mu g/m^3$)	37.3	28.3
GSD formaldehyde concentration bedroom 3 months	1.59	1.66
90th percentile formaldehyde concentration bedroom 3 months ($\mu g/m^3$)	67.3	54.2
GM formaldehyde concentration living room 15 months ($\mu g/m^3$)	23.8	28.9
GSD formaldehyde concentration living room 15 months	1.52	1.29
90th percentile formaldehyde concentration bedroom 15 months ($\mu g/m^3$)	40.6	40.0
GM formaldehyde concentration bedroom 15 months ($\mu g/m^3$)	22.5	22.6
GSD formaldehyde concentration bedroom 15 months	1.72	1.37
90th percentile formaldehyde concentration bedroom 15 months ($\mu g/m^3$)	45.2	33.8

[1] GM = Geometric mean, GSD = Geometric standard deviation
[2] These are houses with an energy efficient mechanical ventilation system

The 90th percentiles of mechanically ventilated houses appear to be lower than those of naturally ventilated houses, largely due to lower variation in concentrations. The geometric mean values and 90th percentiles of both types of houses appeared to have decreased between the measurements after 3 and 15 months. A precise factor for the decrease cannot yet be calculated because not all results after 15 months are already available. However, the decrease in 90th percentiles appears to be in the order of 30% (unpublished data provided by Tappler).

Salthammer, Mentese and Marutzky (2010) mentions other studies that describe a decrease in indoor air formaldehyde concentration as a function of time, with higher values (above the DNEL) following completion of the construction work. As these studies are often relatively old (< 1991), performed outside the EU (Australia, Korea) or are performed in only a small number of houses (Brown, 2002 and Wolkoff and Kjaergaard, 1991 (both referenced by Salthammer, Mentese and Marutzky, 2010)) it is unclear whether these results are representative for the current situation within the EU.

3. Emission rate data

Besides indoor air measurement data, also emission rate data of different products have been reviewed. In total > 15 studies were included that describe relevant emission data, both from experimental studies and from measurements in real houses. From the available data it is clear that there is a wide variation in the emission factors due to the many different materials that are on the market. These materials include wood based products used for flooring (plywood, particle board, parquet) but also ceiling materials, wall paper, furniture and paints.

According to Järnström (2007) in general, the emission from building materials is composed of several rate dependent steps like:
- organic vapour production;
- transport through the bulk phase;
- transport through a physical barrier;
- diffusional transport from the building material into the room air.

Factors that affect various steps include temperature, humidity, barrier thickness and permeability, face velocities, and room concentration of the pollutant. In addition, ageing or depletion of the source affects emissions over a period of time. The rate of release of a compound is mainly determined by the rate of diffusion of the compound in the material and the air circulation in the boundary layer just above the surface of the material. Evaporation controls the emissions of liquids, e.g. paints, in the drying phase (Järnström, 2007).

3.1 Wood based products

All wood based products contain at least some formaldehyde. The content and emission is determined by different factors, such as moisture content (recently cut wood, or already dried), with/without bark or coating, thickness of the material and resin of glue that is used. Table 2 presents the results for wood based products. In some studies only emission rates are described. Other studies also mention a concentration in mg/m^3, often measured under laboratory test conditions, sometimes measured in real houses.

Based on literature and on expert consultation the following observations can be made:
- Lowest formaldehyde emission rates are found for solid wood species with maximum concentrations of about 0.011 mg/m^3;
- In general emissions from coated materials are about 10% of these of uncoated materials;
- Urea-formaldehyde wood based products emit the highest formaldehyde concentrations;

The results indicate that the main sources of formaldehyde emission are the uncoated materials and plywood. Values can be compared with various emission category criteria, such as:
- E1 emission method (chamber testing) steady state concentration of 0.124 mg/m^3 (0.1 ppm); conditions: temperature 23 °C, relative humidity 45%, loading factor 1 m^2/m^3, air exchange rate 0.5 h^{-1} and air velocity at the test surfaces 0.1-0.3 m/s (DIN, 2005);
- Maximum allowed concentration set by the US Department of Housing and Urban Development (Environmental Health) of 0.37 mg/m^3;
- Indoor material emission labelling (France), ranging from category C (> 0.120 mg/m^3) to A^+ (< 0.010 mg/m^3; conditions (ISO 16000-9): 23°C, relative humidity 50%, loading factor 1 m^2/m^3 (for walls), air

exchange rate 0.5 h^{-1} and air exchange at the test surfaces 1.25 $m^3.m^{-2}.h^{-1}$ for ceiling and floor (Ministère de l'Écologie du Développement Durable, des Transports et du Logement, 2011);

- M1 / S1 classification by FiSIAQ (Finland) where M1 is < 0.05 $mg.m^{-2}.h^{-1}$ (conditions: 23°C, 50% relative humidity, air velocity near the sample 0.1-0.3 m/s) and S1 is 0.03 mg/m^3 (concentration in real rooms, only from building material) (Saarela *et al.*, 2004. Säteri, 2002).

Table 2. Emission rate and concentration for different groups of wood based products

Wood based products	Concentration (mg/m³)	Remark
Bare wood species (uncoated)	0.011	Maximum value (Boehme, 2000; Böhm, Salem and Srba, 2012) (*1m³ chamber, 30 °C, air changes 1/h*), Meyer and Boehme (1997) (*23 °C, 45% RH, air changes 1/h*)
Particleboard UF	0.06	Average (26 samples after 28 days) (Yrieix *et al.*, 2010) (*ISO 16000-9; 23 °C, 50% rel. humidity, 1.25 Area specific air flow rate (m³ . m⁻² . h⁻¹)*
Particleboard (PF, UF, PMDI)	<0.11	UF uncoated (Boehme, 2000, *1 m³ chamber, 30 °C*)

Emission rates ($mg.m^{-2}.h^{-1}$)

Wood based products	Emission rate (mg.m⁻².h⁻¹)	Remark
Bare wood species (uncoated)	<0.431	Maximum value (Boehme, 2000; Böhm, Salem and Srba, 2012, Meyer and Boehme, 1997); maximum from Meyer and Boehme, 1997 (*gas analysis method: maximum emission at 60 °C; acetylacetone derivatisation*)
Uncoated UF	0.164	Median value (Kelly, Smith and Satola, 1999) (*21-26°C, 50% relative humidity; DNPH derivatisation and continuous HCHO monitor*)
Coated UF	0.015	Median value (Kelly, Smith and Satola, 1999) (*see above*)
Uncoated PF	<0.016	Maximum value (Kelly, Smith and Satola, 1999) (*see above*)
Plywood	0.13-2.7	Range (Böhm, Salem and Srba, 2012) *gas analysis method: at 60 °C; DNPH derivatisation*)
Particleboard UF Particleboard (PF, UF, PMDI)	0.06	Average (26 samples after 28 days) (Yrieix *et al.*, 2010) (*ISO 16000-9; 23 °C, 50% rel. humidity, 1.25 Area specific air flow rate (m³ . m⁻² . h⁻¹)*)
Parquet	0.046	Maximum value indoors (Järnström, 2007)
Ceiling material (perforated compressed wood)	0.17	Crunaire *et al.* (2012) (*chamber test method, 23°C, 50% RH, 0.5 air changes/h*)

There are several methods of estimating the emission. These do <u>not</u> give the same results. The gas analysis method is a method aimed at providing a maximum emission in a short period by exposing the product to hot air (60 °C). Chamber test methods may provide results that are closer to real life emissions. The most realistic emissions result from chamber test methods with normal temperature (e.g. 21-23 °C), normal relative humidity (around 45%), a realistic ventilation (0.5 air changes per hour) and an air flow rate over the emitting surface that is close to realistic air flow rates over ceilings, walls and floors in buildings (1 – 1.3 $m^3.m^{-2}.h^{-1}$). Values outside of these conditions can only be used as an indication of relative emission and not as the basis for exposure estimates.

Figure 2 (from Boehme, 2000) shows how the concentration changes over time in experimental 1 m^3 testing chambers for Norway Spruce materials (wood particles with/without bark (rinde)).

Figure 2. Formaldehyde emission over time (ppb) in small chamber emission test
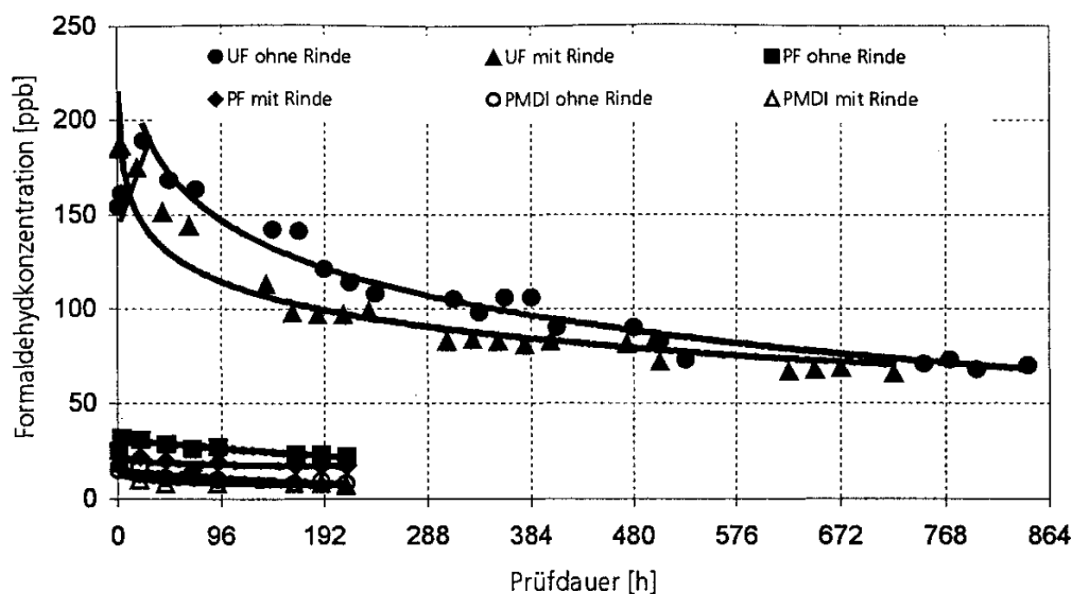

Bild 29: Verlauf der Formaldehydkonzentration in der 1 m³-Kammer von Laborspanplatten aus Fichtenspänen mit und ohne Rinde, hergestellt mit verschiedenen Bindemitteln

Explanation:
Original title: Trend in formaldehyde concentration in the 1 m^3 chamber from laboratory particleboard made from Spruce with and without bark, produced with different adhesives.
UF = urea formaldehyde, PF = phenol formaldehyde; PMDI = polymeric methylene diisocyanate;
Rinde = bark
Prüfdauer = duration of the experiment in hours, maximum duration is 36 days

3.2 Paint
Another source of formaldehyde release is the release of formaldehyde from paint. Salthammer (2010) mentions that coating materials, paints and lacquers might contain formaldehyde as preservative. Potential strong sources like acid-curing lacquers made of modified urea- and melamine-formaldehyde resins have almost completely been replaced. The same is true for formaldehyde and formaldehyde-releasing compounds that were often uses as biocides in water-based paints and fungicidal products Short-term formaldehyde emissions under test chamber conditions have been found with indoor wall paints equipped with modified TiO2 to serve as a catalyst under daylight or artificial light to reduce

indoor air pollutants (Salthammer, Mentese and Marutzky, 2010). Chang (2001) mentions that biocide replacement can reduce only the long-term emissions since other sources (e.g. additives and binders) are present in paints.

Different studies have measured exposure to formaldehyde as a result of applying paint, both in real houses and in laboratory settings. Data are presented in Table 3. Indoor air concentrations are below the DNEL of 0.1 mg/m^3, results from laboratory testing are sometimes above this value.

Table 3. Emission rate and concentration after the application of paint

Description	Emission rate (mg.m^{-2}.h^{-1})	Concentration (mg/m^3)	Reference
Real houses			
Mean value – painting <12 months ago		0.016-0.048	Gilbert *et al.*, 2006 (*concentrations in real houses, DNPH derivatisation method*)
Maximum values – painting <12 months ago		0.09	Gilbert *et al.*, 2006 (*see above*)
Newly build houses – painted walls (gypsum & concrete) – maximum emission	0.037		6 months old building - Järnström, 2007 (*Calculated from measurements in real houses, acetyl acetone method*)
Mean indoor concentrations - different rooms painted		0.016-0.032	Wieslander *et al.*, 1997 (*measurements in bedrooms in real houses*)
Laboratory setting			
Laboratory testing of two paints: Initial values Final values	0.01	0.023-0.043 0.01	Kolarik *et al.*, 2011 (*chamber 0.225 m^3, 23°C, 45% RH, 1 air change/h, area specific ventilation rate 1 m^3.m^{-2}.h^{-1}, continuous monitor Skalar and acetylacetone method*)
Laboratory testing – initial emission – maximum concentrations	0.33-0.65	0.16	Kelly, Smith and Satola, 1999 (*21-26°C, 50% RH; DNPH derivatisation*)
Laboratory testing – peak concentrations after applying 2 paints on a glass plate in order to maximize the VOC emissions		5.5	Chang, 2001 (*23°C, 50% RH, 0.5 air changes/h, loading factor 0.5 m^2/m^3, DNPH derivatisation*)

3.3 Mineral wool

Mineral wools used as insulation material are also known as a source of formaldehyde release. Initial emission after application of the materials might result in increased levels of formaldehyde. These levels rapidly decrease, average steady state values after 120-240 hours were <0.05 mg/m^3 (in large chamber experiments with 23°C, 45% RH, loading rate 1 m^2/m^3, air change rate 1 /h) with a 50 % further decrease of these values after 3-6 months (Marutzky *et al.*, 1993). These products are mostly applied behind

indoor walls. This strongly reduces the emission of formaldehyde from the insulation materials to the indoor environment.

3.4 Furniture

Yu and Kim (2012) describe a case were a new wardrobe is unpacked and installed into a bedroom. The steady-state emission rates were determined for coated and non-coated wardrobe materials. For coated materials the emission rates were 0.007 mg.m^{-2}.h^{-1} for MDF and 0.040 mg.m^{-2}.h^{-1} for melamine-formaldehyde particleboard. For uncoated materials the emission rates were 0.041 mg.m^{-2}.h^{-1} for raw particleboard and 0.096 mg.m^{-2}.h^{-1} for raw MDF. Measurements were done in a 18.9 m^3 bedroom of an energy efficient house. The bedroom contained a bed and built-in cupboards and the bedroom was carpeted.

3.5 Textile and Carpet

Formaldehyde emission from four new carpets has been measured by Katsoyiannis, Leva and Kotzias (2008) under small chamber testing conditions with 23 °C, 45% RH, an air change rate of 0.5 and a loading factor of 0.4 m^2/m^3. The highest measured value after 24 hours was 0.024 mg/m^3 and 0.014 mg/m^3 after 72 hours.

3.6 Foams

The emission of formaldehyde from foams depends on the thickness of the foam layer. In general a layer of 50 mm is common in consumer applications. The current generation of foams has a DIN-EN 717-1 test chamber measured concentration of 0.05 - 0.063 mg/m^3 which is well below the DNEL of 0.1 mg/m^3. It is expected that new foams, released on the market from 2014 onward, will have an even lower emission. Test results conform DIN-EN 717-1 performed by industry show a measured concentration of 0.0125 - 0.025 mg/m^3 (unpublished data provided by an industry expert).

3.7 Contribution of emission rates to indoor air concentration

The overall indoor air concentration is influenced by different sources. Not only wood based products contribute to the indoor air concentration, but the indoor air quality is also dependent on outdoor concentration (e.g. due to traffic) and indoor activities (e.g. cooking, smoking, burning incense).

- In new fabricated houses Järnström (2007) found that the contribution from surfaces decreased to less than 0.020 mg/m^3 throughout the first year, although they were responsible for about 65-75% of the indoor formaldehyde concentrations. The contribution of emission from the ceiling was the most significant, followed by emissions from walls. Crunaire *et al.* (2012) also found the highest emissions from ceiling plates.
- Raw *et al.* (2004) found a difference between homes with particleboard flooring and those without, the mean concentrations were 0.032 and 0.0203 mg/m^3 respectively.

3.8 Time dependent concentrations

Formaldehyde release from products is influenced by different variables such as temperature, relative humidity and air exchange rate. When houses are new or have new installed furniture the indoor air concentrations are generally higher. This concentration decreases in time. Some studies have been performed to determine this decline in indoor air concentration.

- In small chamber tests (at 23 °C and 45% RH with 1 air change/h and a surface velocity of approximately 0.3 m/s) performed by Brown (1999) the formaldehyde emission rate decreased from 0.38 mg.m^{-2}.h^{-1} to 0.17 mg.m^{-2}.h^{-1} after > 300 days for MDF and from 0.51 mg.m^{-2}.h^{-1} to 0.084 mg.m^{-2}.h^{-1} for particle board after > 200 days.

- Hare *et al.* (1996) conducted a study in a newly constructed house using different types of loading (medium and high loads of wood based products like flooring material). Measurements were done with an EPA method based on DNPH and a NIOSH method based on chromotropic acid. Concentrations increased during the first days for the high load and then started to descend (Figure 3). For the medium load it took longer to reach the maximum values. This was caused by an increased relative humidity of 70 % instead of the intended value of 50 %.
- Järnström (2007) performed measurements in 14 new houses, at 0, 6 and 12 months after completion. The results showed an average increase of the concentration going from 0.019 mg/m^3 (0-months) to 0.027 mg/m^3 (12-months). According to the author *'New formaldehyde sources, such as furniture, were common in the inhabited buildings. This caused a slightly increasing trend in the mean concentration and, especially, led to elevated maximum formaldehyde concentrations in the occupied apartments'*.
- The unfinished study on new houses in Austria also shows a decrease in concentrations from 3 to 15 months after inhabitants have moved in.

Figure 3. US-EPA (1996) - concentration of formaldehyde in time in a new house with high and medium loading of wood based products
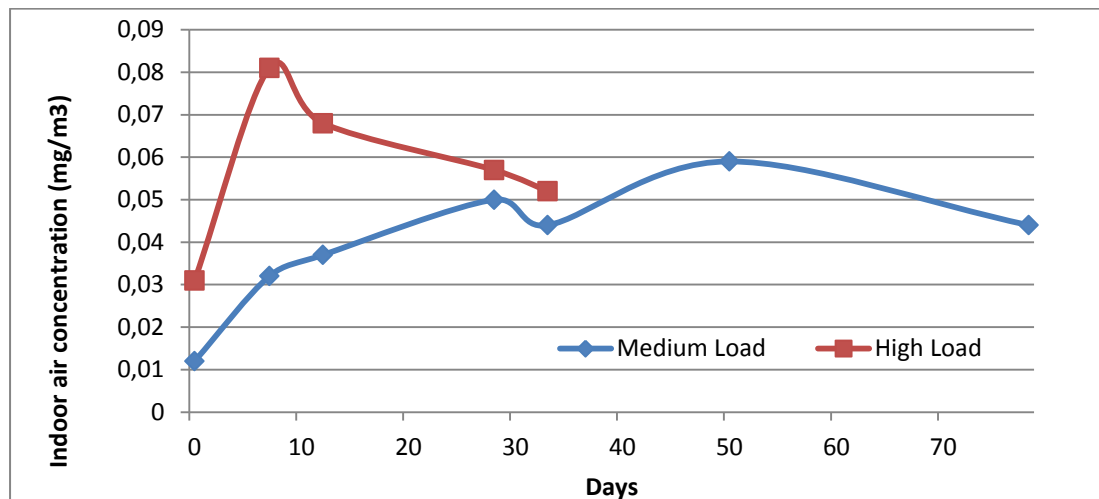
In general, it can be concluded that emissions and thereby concentrations are decreasing in time. However, it is expected that at least some emission of formaldehyde will occur throughout the service life period. The major reason for this is that chemical processes like hydrolysis and diffusion will constantly result in small amounts of additional free formaldehyde in the products. The effect of these chemical processes and its contributing factors (e.g. temperature, humidity) has been investigated by several institutes, and urea-formaldehyde resins appear to be most vulnerable for this kind of disintegration processes (Myers, 1985). Within the past decades a lot of effort was spend in increasing product sustainability (Dunky, 1998). Overall, the continuous small release of formaldehyde after the initial higher release is considered to be covered within the long-term indoor measurements as described in section 2.1.

4. Exposure Scenario

Based on the literature findings as well as a discussion between TNO Triskelion and industry experts, a reasonable worst case exposure scenario is generated. For this scenario two maximum exposure concentrations in a standard room are calculated, by use of two different emission rates (E1 and E1+). These emission rates are the current industry standards and apply to different types of wood based panels. As applying the E1 and E1+ criteria as a restriction or voluntary industry action may be suitable risk management options, both criteria are taken into account. The following parameters are used:

- The European Reference room (30 m^3, with air exchange rate of 0.5) – (Oppl, Neuhaus and Augustin, undated) with room dimensions of l x w x h = 4 x 3 x 2.5, one door and one window. Applying loading rates (surface of emitting material per volume of the room) of 1 m^2/m^3 for walls and 0.4 m^2/m^3 for flooring and ceiling, with a total loading rate of 1.8 m^2/m^3;
- Assuming that the ceiling and flooring are made up of wood based materials, where 90% of the materials are coated and maximal 10% of the total board surface is not sealed or covered (uncoated) due to e.g. drilling holes and uncoated edges. Applying specific emission rates for uncoated materials of 0.124 mg.m^{-2}.h^{-1} or 0.1 ppm (E1 emission criteria, EN 717-1; based on emission at 23°C and 45% RH, air change 1/h and a loading factor of 1 m^2/m^3) and 0.0124 mg.m^{-2}.h^{-1} for coated materials (10% of uncoated materials; based on the difference between coated and uncoated UF based material in Kelly, Smith and Satola, 1999).
- The room contains one large wardrobe (1.92 x 1.8 x 0.40 m – dimensions taken from an IKEA wardrobe, see Annex I). For this wardrobe emission rates are applied that are described by Yu (2012). The highest values for coated and uncoated materials are being used (0.096 for uncoated and 0.040 for coated materials). In this case the coated and uncoated materials are not the same materials: coated was MDF and uncoated was particleboard. Assumed is that 90% of the materials is coated.
- The total loading rate of building material (flooring and ceiling) and wardrobe together is 0.8+0.6 = 1.4 m^2/m^3.
- Using the following equation by Salthammer (2010):

Equation: SER = (n*C) / L (Salthammer, 2010) or

C = (SER * L) / n

Where:
C = concentration (mg/m^3)
SER = specific emission rate (mg.m^{-2}.h^{-1})
n = air exchange rate (1/h)
L = loading rate (m^2/m^3)

The inputs and results are presented in **Table 4**. In column 2, the maximum expected concentration based on this equation, with an emission rate according to the E1 criteria, is 0.093 mg/m^3. This is below the DNEL of 0.1 mg/m^3. When a higher emission rate for the flooring and ceiling materials is applied (e.g. 0.25 mg.m^{-2}.h^{-1}) a concentration of 0.13 mg/m^3 is calculated which shows a risk characterization ratio above 1. This means that, with the conditions chosen for this calculation, the E2 emission criteria does not lead to control of risks.

In column 3, the second scenario is presented which is calculated using the same input parameters, but with wood based panels classified as E1+ i.e. 0.080 mg/m^3 (0.064 ppm). The maximum expected concentration in that case is 0.079 mg/m^3.

Table 4. Calculated exposure scenario's based on E1 and E1+ emission categories

Salthammer (2010)	$C = (SER * L) / n$		
Classification	**E1**	**E1+**	**Units**
Ceiling / Flooring (Uncoated materials)			
SER (specific emission rate)	0.124	0.080	mg.m^{-2}.h^{-1}
L (loading)	0.08	0.08	m^2/m^3
n (ventilation rate)	0.5	0.5	per hour
C (concentration)	*0.020*	*0.013*	*mg/m^3*
Ceiling / Flooring (Coated materials)			
SER (specific emission rate)	0.0124	0.008	mg.m^{-2}.h^{-1}
L (loading)	0.72	0.72	m^2/m^3
n (ventilation rate)	0.5	0.5	per hour
C (concentration)	*0.018*	*0.012*	*mg/m^3*
Subtotal concentration (ceiling / floor)	**0.038**	**0.024**	mg/m^3
Wardrobe (Uncoated materials; particle board)			
SER (specific emission rate)	0.096	0.096	mg.m^{-2}.h^{-1}
L (loading)	0.06	0.06	m^2/m^3
n (ventilation rate)	0.5	0.5	per hour
C (concentration)	*0.012*	*0.012*	*mg/m^3*
Wardrobe (Coated materials; MDF)			
SER (specific emission rate)	0.04	0.04	mg.m^{-2}.h^{-1}
L (loading)	0.54	0.54	m^2/m^3
n (ventilation rate)	0.5	0.5	per hour
C (concentration)	*0.043*	*0.043*	*mg/m^3*
Subtotal concentration (wardrobe)	**0.055**	**0.055**	mg/m^3
Total concentration (ceiling/flooring + wardrobe)	**0.093**	**0.079**	mg/m^3

In the scenarios presented above, the values for the wardrobe are based on one specific source with emission rates that do not correspond to either E1 or E1+. Although we believe this represents a reasonable worst case, it complicates understanding of the influence of the emission rates and the loading rate on the formaldehyde concentration. To illustrate this, another set of estimates is made in which the equation described above is calculated for a situation where all emitting material, including furniture, has the same basic emission rate for uncoated material, but still assuming that 90% of the

loading rate relates to coated materials and that the reduction of emission by the coating is 90%. This is done for various emission rates and loading rates. These calculations are shown in

Figure 4.

Figure 4. Concentration of formaldehyde in relation to emission rates and loading rates
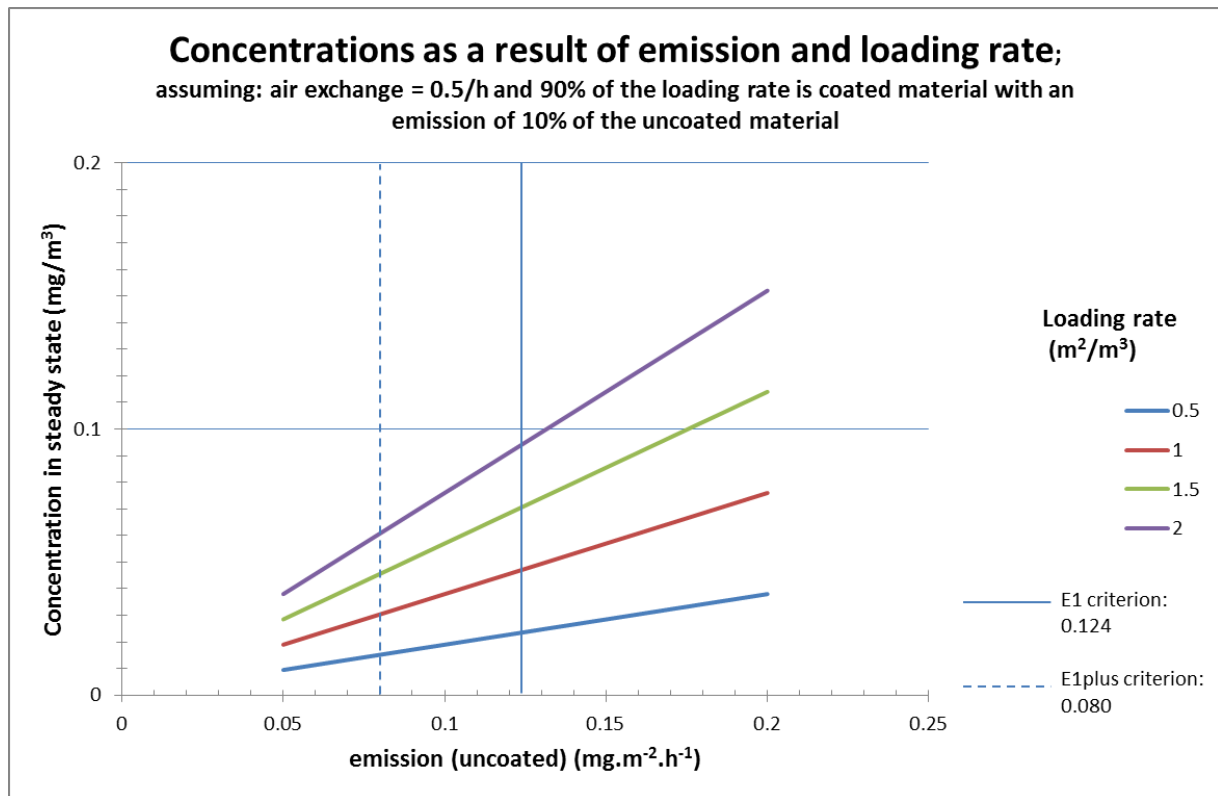
5. Discussion

There is a large number of studies providing information on real indoor air concentrations of formaldehyde. The majority of these studies is done with a method based on DNPH derivatisation, or with DNSH or acetyl acetone derivatisation methods. These methods are considered to be reliable, in contrast with the older chromotropic acid method (Hun *et al.*, 2010, Salthammer, Mentese and Marutzky, 2010). In general, the studies of indoor air concentrations do not precisely indicate the (potential) sources of formaldehyde. The loading rate of wood based panels and the exact type and quality of wood based panels is generally not mentioned. In some cases a differentiation is made between e.g. wood based flooring material and non-wood based flooring material (e.g. Schultz *et al.*, 2010). The total number of measurements in 13 recently performed studies was > 2500. Because of all the differences in methods and details of description, a formal statistical analysis of the total dataset is not possible. However, the estimated 'central tendency' value of the total dataset in (generally) older homes is approximately 0.025 mg/m^3. This is similar to values reported as 'averages' by Järnström (2007), Salthammer, Mentese and Marutzky (2010) and Sarigiannis *et al.* (2011). The approximate 'reasonable worst case value' from these studies was estimated at 0.085 mg/m^3 which is between the third and fourth highest maximum of the 13 studies. Only in one of these studies a maximum value above the reference value of 0.1 mg/m^3 is reported (Raw *et al.*, 2004). Some of the studies attempted to study the influence of potential determinants of concentrations. Whereas some studies did find an effect of e.g. season or new wooden furniture installed or low air change rates, even for the situations with the highest concentrations in these studies (except Raw *et al.*, 2004) all measured values were below 0.1 mg/m^3. New houses with new furniture may have higher formaldehyde concentrations for some time, but the relevant studies also show almost all values to be below the reference value. The 90th percentiles of measurements in new houses with wooden or wood based walls and flooring in an Austrian study are also clearly below the reference value.

The indoor air exposure measurements did not include situations in mobile homes. In the past high concentrations of formaldehyde have been found in mobile homes in the USA. However, the construction of the mobile homes in the USA is not comparable to normal mobile homes in Europe. Besides that, in the EU it is not very common to live in mobile homes. This is supported by the fact that no data have been found in European sources on concentrations of formaldehyde in mobile homes

Based on the indoor air concentration measurements it can be concluded that the reasonable worst case (*i.e.* a high value of the total distribution, but not the maximum; generally the 90th percentile value) indoor air concentrations both in older and in new houses, independent of the presence of more or less sources of formaldehyde, is below the reference value of 0.1 mg/m^3, e.g. at a level of 0.085 mg/m^3.

However, based on existing indoor air concentration data alone, it cannot be shown with sufficient certainty that with the use of present products, and specifically wood based panels, reasonable worst case concentrations will be below 0.1 mg/m^3. The issue here is that it cannot be shown that the loading rate in most studies is reasonable worst case. Therefore, also article emission data have been evaluated and some exposure scenarios have been constructed.

Concentrations of formaldehyde in chamber tests of wood and wood based panels show values below or close to the reference value with a maximum for uncoated urea formaldehyde based particleboard of around 0.11 mg/m^3. These values cannot be directly compared to values in real life, because the conditions in the measurement with the highest value were not typical with a temperature of 30 °C.

Emission rates for wood based products tested with the so-called gas analysis method are also not representative for real life (temperature 60 °C).

Emission rates tested with the small (or larger) chamber test methods tend to simulate real life situations better, with temperatures of around 23 °C and relative humidity of 45 or 50%. Both parameters are at the high end of values reported for the rooms in the indoor air concentration studies. The present voluntary industry emission criterion for wood based panels is E1, which is a steady state concentration of 0.12 mg/m^3 or a specific emission rate of 0.124 mg.m^{-2}.h^{-1}. Emission from uncoated urea formaldehyde based panels and perforated compressed wood have been found that were higher than this value, while other products (such as pure wood, uncoated phenol formaldehyde based panels and coated urea formaldehyde based panels) were clearly lower. Perforated wood based products have a much larger surface area (also not coated) than normal wood based panels and are therefore very worst case. However, these are not normal practice.

There is a paucity of publicly available data on emission or furniture made from wood based panels. The only relevant study found showed values below the E1 criterion (Yu and Kim, 2012).

Emission rates also (slowly) decrease over time, with a much higher absolute decrease for products with a higher initial emission. Long term small emissions of formaldehyde from articles take place due to chemical reactions like hydrolysis and diffusion.

Some paints also show emission rates higher than the E1 criteria when tested in a chamber test method under relevant conditions (Kelly, Smith and Satola, 1999), while emission from other paints is much lower (Kolarik *et al.*, 2011). Whether or not the formaldehyde emission is due to the use of formaldehyde based resins (only) or (also) due to the use of formaldehyde or formaldehyde donors as preservatives is not sure.

The emission rates of mineral wool, textiles and carpets and foams that were found in literature appear to be clearly below those of the wood based panels and because mineral wool and foams tend to be used behind walls (as insulation material) their emission into the rooms will be further reduced.

The combined information indicates that wood based panels, as construction material and/or in furniture, are the major source of formaldehyde from articles based on formaldehyde based resins in the indoor environment. Other sources are much less relevant, due to lower emissions and/or lower free surface areas.

To show what indoor air concentrations can be expected in a reasonable worst case situation, a few exposure scenarios have been constructed. In these exposure scenarios in a small room with a low air exchange rate flooring and ceiling with the maximum emission acceptable for the E1 criterion or the more stringent E1plus criterion was assumed as well as a large wardrobe with realistic emission values (relatively close to the E1 criterion) were accounted for. The calculations with ceiling and floor according to the E1 criterion indicate a concentration below the reference value and with the E1plus criterion the value will be even lower. In this scenario it is assumed that construction panes are not uncoated and that only part of them is uncoated. This is the general practice. The calculations with different emission rates and loading rates show that under normal indoor conditions and with wood based panels of which 90% of the surface is coated with a coating that reduces the emission by 90 percent even at a loading rates up to 2 m^2/m^3 the concentration of formaldehyde will stay below the reference value if the uncoated material adheres to the E1 criteria. However, the expert judgement that coating the wood based panel reduces exposure with 90 percent may need a further validation.

The combination of the large total dataset of indoor air concentrations of older and newer houses and the reasonable worst case exposure scenario with a high loading factor shows that it can be concluded that indoor air concentrations in a reasonable worst case situation will remain below the reference value of 0.1 mg/m^3 if all used wood based panels are largely coated and the uncoated panels adhere at least to the E1 emission criterion. However, if only wood based panels of E2 quality would be used, exceeding the reference value is well possible.

This report only describes the exposures of consumers. No estimation has been made of potential combined exposure for consumers that are also exposed to formaldehyde in occupational settings. Such an estimation cannot realistically be made by summing reasonable worst cases for workers and consumers, since the probability that a worker working in the reasonable worst case exposure situation is also living in a reasonable worst case exposure situation is low and summing of these two reasonable worst cases will overestimate the real reasonable worst case population risk of workers that are exposed to formaldehyde. To make an appropriate combined exposure estimation, a probabilistic approach should be used, based on the exposure distributions of worker groups and the exposure distribution in the indoor environment. Potentially, this should be modified for a possible correlation between working in an a work situation with exposure to formaldehyde and living in specific indoor situations.

6. Conclusions

1) The central tendency of formaldehyde indoor air concentration in Europe is about 0.025 mg/m^3 with almost all measured values well below the current REACH DNEL and an approximate reasonable worst case value of 0.085 mg/m^3.

2) In new build houses or due to renovations or redecoration the formaldehyde indoor air concentration may be increased. Literature indicates that even in these situations the indoor concentrations tend to be below the DNEL of 0.1 mg/m^3.

3) Emission rates of wood based products vary due to the different type of materials (plywood, particleboard, MDF), coated or uncoated and different test methods. Emission rates up to 2.7 mg.m^{-2}.h^{-1} have been described under laboratory settings with the gas analysis method that is intended to increase emission as much as possible. Emission rates with chamber test methods under conditions more relevant for emissions under normal conditions in houses and emission values measured in occupied buildings seem to be (much) lower, values up to 0.17 mg.m^{-2}.h^{-1} have been described in buildings, where the highest value is from perforated compressed wood material.

4) Other products like paints and insulation materials might also release formaldehyde. Under laboratory conditions emissions up to 0.65 mg.m^{-2}.h^{-1} have been described for paints in chamber testing, resulting in test chamber concentrations > 0.1 mg/m^3. The contribution of formaldehyde emissions from insulation materials like mineral wool is < 0.05 mg/m^3 especially since these materials are used behind the indoor walls which reduces the direct emission to the indoor environment.

5) A reasonable worst-case exposure scenario of a wardrobe in an European Reference room with both ceiling and floor made up of wood-based products, conforming to the E1 emission standard resulted in a maximum formaldehyde concentration of 0.093 mg/m^3. This is below the DNEL of 0.1 mg/m^3. If all uncoated material adheres to the E1 criteria and if 90% of the material is coated, leading to a reduction in emission of 90 percent, the reference level will not yet be exceeded, even at a loading rate of 2 m^2/m^3.

6) Based on the measured concentrations in real homes and the reasonable worst-case exposure scenario based on emission data and the E1 emission standard it can be concluded that the reasonable worst case exposure of the general population due to the use of articles made with formaldehyde based resins in Europe is below the DNEL.

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Annex I. List of abbreviations

REACH: Registration, Evaluation, Authorisation and restriction of CHemicals

CSR: Chemical Safety Report

DNEL: Derived No-Effect Level

EU: European Union

USA: United States of America

NIOSH: National Institute for Occupational Safety and Health (USA)

EPA: Environmental Protection Agency (USA)

DNPH: dinitrophenyl hydrazine

DNSH: dansylhydrazine

GM: Geometric Mean

GSD: Geometric Standard Deviation

Ppm: parts per million

RH: Relative Humidity

UF: Urea Formaldehyde

PF: Phenol Formaldehyde

PMDI: Polymeric Methylene Diisocyanate

TiO2:   Titanium Dioxide

VOC: Volatile Organic Compounds

MDF: Medium Density Fiberboard

SER: Specific Emission Rate

Annex II. Wardrobe used for Exposure Scenario

Besta – Storage combination with doors (1,80 x 1,92 x 0,40)
http://www.ikea.com/gb/en/catalog/products/S69903884/?query=699.038.84