Final Report

Literature survey on formaldehyde concentrations, formaldehyde emission sources and ventilation rates in the indoor environment of housings and public buildings in the United States of America

Fraunhofer WKI

Fraunhofer WKI

Department of Material Analysis and Indoor Chemistry

Bienroder Weg 54 E

38108 Braunschweig, Germany

Project Management:	Prof. Dr. rer. nat. habil. Tunga Salthammer
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Composite Panel Association

19465 Deerfield Avenue, Suite 306

Leesburg, Virginia 20176

United States

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Prof. Dr. T. Salthammer

(Head of Department
Material Analysis and Indoor Chemistry)

Dr. Erik Uhde

(Deputy Head of Department
Material Analysis and Indoor Chemistry)

Content

1 Introduction

Formaldehyde was described in the year 1855 by the Russian scientist Alexander Michailowitsch Butlerow. The versatility which makes it suitable for use in various industrial applications was soon discovered and the compound was one of the first to be indexed by the Chemical Abstracts Service (CAS). Between 1900 and 1930, formaldehyde-based resins became important adhesives for wood and wood composites. Today, formaldehyde is one of the most important chemical feedstock for numerous industrial processes. It is also used as a preservative, disinfectant and biocide. As far as the building products sector is concerned, its use as a component of thermosetting adhesives (urea-formaldehyde, melamine-urea-formaldehyde, phenol-formaldehyde, melamine-urea-phenol-formaldehyde) is of particular significance.

Indoor-related applications of formaldehyde in the past and present cover wood-based products (particleboard, oriented strand board (OSB), high-density fiberboard (HDF), medium-density fiberboard (MDF), plywood), cork products (flooring materials), insulation materials made of UF foam, mineral wool or glass wool, paper products, coating materials, paints and lacquers containing formaldehyde as preservative, textiles, cleaning and caring products, disinfectants and preservatives, photo-processing chemicals, cosmetics, etc.

Formaldehyde has been under discussion as an indoor air pollutant since Wittmann (1962) published his paper on the release of formaldehyde from particleboard during use. Adverse health effects from indoor exposure to formaldehyde, especially irritation of the eyes and upper airways, were first reported in the mid-1960s. Formaldehyde emissions from materials bonded with urea formaldehyde resin were soon identified as the cause of the complaints. Spengler and Sexton (1983) identified formaldehyde as one of the priority indoor air pollutants. In parallel, the first studies on the carcinogenicity of formaldehyde triggered more scientific work.

In 2004 the International Agency for Research on Cancer (IARC) classified formaldehyde as a human carcinogen (Group 1). The definition of a Group 1 carcinogen according to IARC is as follows: *“There is enough evidence to conclude that it can cause cancer in humans”.* The evaluation is based on information regarding the relationship between nasopharyngeal cancer and leukemia related to the exposure to formaldehyde (International Agency for Research on Cancer (IARC), 2006). The European Commission classified formaldehyde as a 1B carcinogen and mutagen 2 on June 5th, 2014 in the ordinance EU 605/2014. Category 1B states that the carcinogenic effect has been demonstrated in animal trials and is probable for humans.

Formaldehyde is ubiquitous in ambient and indoor air (Salthammer, 2013). The compound has always been a topic of environmental policy discussions as an air-polluting substance which primarily enters the body through respiration. In this report, a review of literature data on formaldehyde concentrations in air and of the major formaldehyde sources in the indoor environment is provided. This includes a comprehensive literature search of scientific databases like Web of Science, SCOPUS, PubMed, American Chemical Society, SpringerLink, ScienceDirect, WILEY, Taylor & Francis, Google Scholar and many more. Other references like reports and conference proceedings were also considered.

However, the estimation of human exposure to formaldehyde also requires comprehensive information on the living conditions, especially activities, indoor climate and air exchange rates. Moreover, infiltration from outdoor air, chemical reactions (usually known as indoor chemistry), possible sink effects and the influence of product aging must be considered. The presented results refer to the living behavior and indoor conditions in the United States of America. Other indoor formaldehyde concentrations, especially from Europe, are mentioned for purposes of comparison. Emission data from products and materials outside the US are considered if representative US data are not available or if the process is independent of the location (e.g. in case of combustion).[1]

References (Chapter 1)

International Agency for Research on Cancer (IARC), 2006. Formaldehyde, 2-Butoxyethanol and 1-tert-Butoxy-2-propanol. World Health Organization, Lyon, France.

Salthammer, T., 2013. Formaldehyde in the Ambient Atmosphere: From an indoor pollutant to an outdoor pollutant? Angewandte Chemie International Edition 52, 3320-3327.

Spengler, J.D., Sexton, K., 1983. Indoor air pollution: a public health perspective. Science 221, 9-17.

Wittmann, O., 1962. Die nachträgliche Formaldehydabspaltung bei Spanplatten. Holz als Roh- und Werkstoff 20, 221-224.

[1] A similar study was carried out for Europe on request of the European Chemicals Agency: Information requirements on formaldehyde given in the ECHA decision letter “DECISION ON SUBSTANCE EVALUATION PURSUANT TO ARTICLE 46(1) OF REGU-LATION (EC) NO 1907/2006, for formaldehyde, CAS No 50-00-0 (EC No 200-001-8)”. The report was submitted in October 2017.

2 Statistical background

2.1 Probability distributions

In our environment, many processes can be understood by application of stochastic approaches (Ott, 1995). The normal distribution, also known as Gaussian distribution, is an important probability model in statistics. The probability density function (see Equation 2.1) is symmetrical and bell-shaped (see Figure 2.1). A normal process results when a number of unrelated, continuous random variables are added together (Ott, 1995). Physical and chemical measurements can often adequately be explained with a normal distribution.

$$f(x) = \frac{1}{\sigma \cdot \sqrt{2\pi}} \cdot e^{-\frac{1}{2} \cdot \left(\frac{x-\mu}{\sigma}\right)^2} \quad (2.1)$$

In Equation (2.1) μ is the arithmetic mean value and σ is the standard deviation.

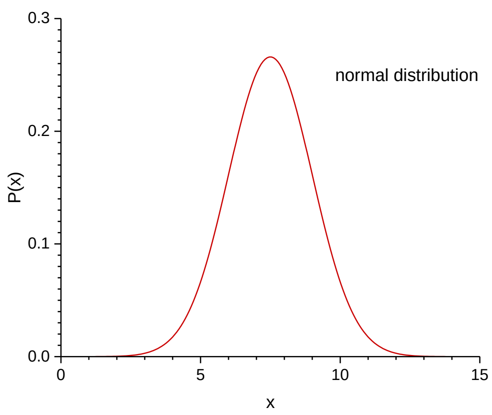

Figure 2.1: Example of a normally distributed probability function with μ = 7.5 and σ = 3.

However, most processes in the environment can be explained by the asymmetrical log-normal density Equation (2.2).

$$f(x) = \frac{1}{x \cdot \sigma \cdot \sqrt{2\pi}} \cdot e^{-\frac{1}{2} \cdot \left(\frac{\ln(x)-\mu}{\sigma}\right)^2} \quad (2.2)$$

A log-normal process is one in which the random variable of interest results from the product of many independent random variables multiplied together (Ott, 1995). For example, the size distribution of airborne particles usually follows a log-normal function. The normal and the log-normal distribution are interrelated by Equations (2.3) and (2.4).

$$GM\ (\mu_g) = e^{\mu} \quad (2.3)$$

$$\sigma_g = e^{\sigma} \quad (2.4)$$

GM (μ_g) is the geometric mean and σ_g is the geometric standard deviation.

On the basis of dilution theory Ott (1990) provided a theoretical model to demonstrate that concentrations of airborne pollutants follow a log-normal distribution. In other words: if formaldehyde concentrations are measured under identical sampling and analysis conditions in n randomly selected indoor environments the distribution of concentrations is log-normally distributed. The shape of the log-normal density function (2.2) is shown in Figure 2.2. On the logarithmic x-axis the log-normal curve becomes normally distributed.

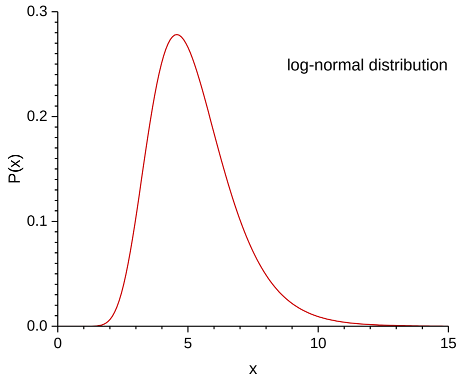

Figure 2.2: Example of a log normally distributed probability function with GM = 5.

2.2 Histogram and Box-Whisker plot

In many cases it is of advantage to display data as discrete values in histograms. An example is provided in Figure 2.3. Around 1000 data were measured in the interval $0 \le x \le 20$. These data were classified in intervals of $\Delta x = 1$ and displayed as a histogram. The analysis shows that the resulting distribution is asymmetrical and nearly log-normal.

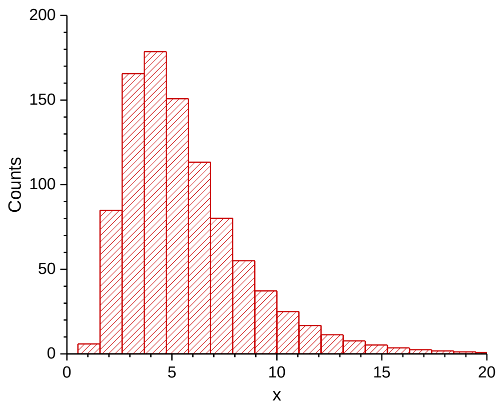

Figure 2.3: Example of a log-normally distributed histogram with about 1000 data $0 \le x \le 20$ and $\Delta x = 1$.

For normally distributed data, the common statistical parameters are the arithmetic mean value μ and the standard deviation σ. However, these parameters cannot be applied in case of a log-normal distribution, or generally speaking, if the distribution is skewed. Then a so-called non-parametric analysis using percentages (P-values) has to be applied, the result is often displayed as a Box-Whisker plot (see Figure 2.4) (Walpole et al., 2014).

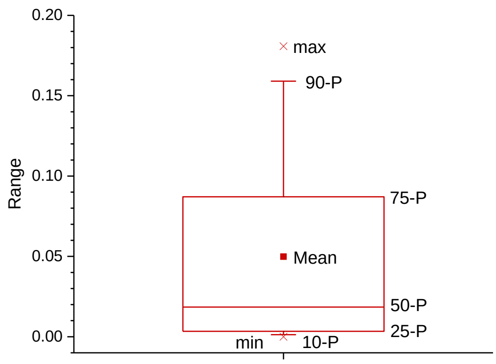

Figure 2.4: Example of a Box-Whisker plot.

In this figure, 50-P means that 50% of all data are below the 50-P value and 50% of the data are higher than the 50-P value, which is also known as the median. Usually, the lower and upper lines of the box are the 25-P and 75-P values (quartiles), respectively. The whiskers provide the 10-P and 90-P values. The arithmetic mean value is displayed as (■) and the min/max values as (x). Please note that for a log-normal distribution the arithmetic mean value is always higher than the median, which on the other hand is almost identical with the geometric mean.

In non-parametric tests the use of percentiles is common. 50-P (median) and 95-P are usually applied as the most important criteria. Reference values are often based on the 95-P to indicate that measured data, which exceed the 95-P, are conspicuous because they are significantly different from averages.

2.3 Monte-Carlo method

Environmental processes are often highly complex and influenced by manifold parameters, which are also dependent on the physical conditions. In many cases numerical approximations must be used. The Monte-Carlo method, which uses random sampling from probability density distributions, provides a powerful tool for the calculation of simulated experimental data from stochastic approaches (Bevington and Robinson, 2003).

The procedure is demonstrated in Figure 2.5 for a simple and fictitious example. The red bars represent a normal distribution of air exchange rates (AER). The blue bars represent a normal distribution of area specific formaldehyde emission rates (SER_A). The scientific question is as follows: what is the expected distribution of formaldehyde concentrations in the absence of sink effects and for a loading factor of L = 1 m²/m³? Firstly, random values are generated from the two distributions for AER and SER_A. From these two values a formaldehyde concentration is calculated using the equation $C_{HCHO} = SER_A/(AER \cdot L)$. This procedure is repeated until a statistically sufficient result is achieved. The green bars in Figure 2.5 represent the distribution of formaldehyde concentrations from a Monte-Carlo approach with 1000 runs.

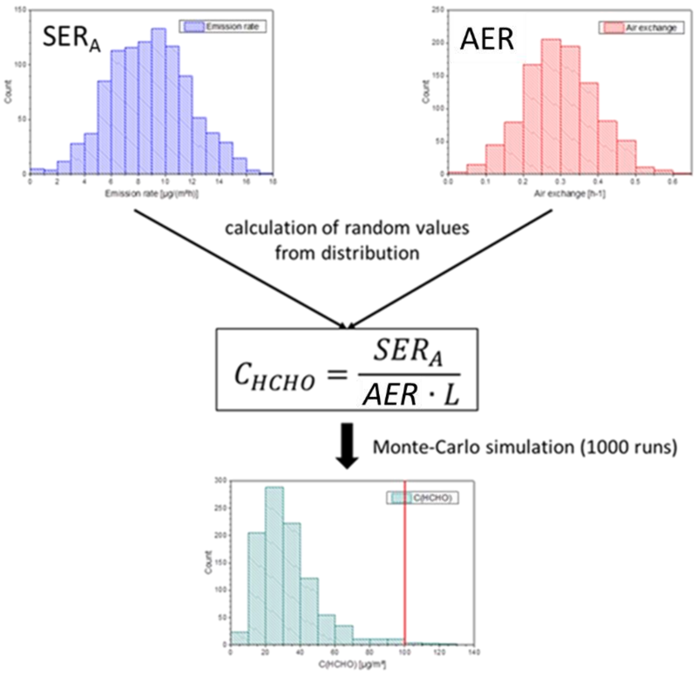

Figure 2.5: Monte-Carlo simulation of indoor formaldehyde concentrations (green) from air exchange rate (red) and emission rates (blue). L is the loading rate. Note: the example is fictitious and for demonstration purposes only.

The simulation of stochastic processes requires the generation of random numbers. This is usually achieved by application of pseudo random generators. In this work, scientific software OriginPro 2016G (OriginLab Corporation, Northhampton, USA) was applied. The random numbers were analyzed by appropriate statistical test as described by Morgan (1984) and the

OriginPro pseudo random generators were found to be applicable for the generation of uniformly, normally and log-normally distributed random numbers.

References (Chapter 2)

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Morgan, B.J.T., 1984. Elements of simulation. Chapman and Hall, London.

Ott, W.R., 1990. A physical explanation of the log normality of pollutant concentrations. Journal of the Air & Waste Management Association 40, 1378-1383.

Ott, W.R., 1995. Environmental Statistics and Data Analysis. Lewis Publishers, Boca Raton.

Walpole, R.E., Myers, R.H., Myers, S.L., Ye, K., 2014. Probability and statistics for engineers and scientists. Pearson Education Inc., Essex.

3 Indoor formaldehyde guidelines and reference limits

Publisher	World Health Organization
Value	**0.1 mg/m³ (30 minute average concentration)**
Type	Indoor Guideline
Update	2010
Comment	The short-term guideline will also prevent effects on lung function as well as long-term health effects, including nasopharyngeal cancer and myeloid leukaemia.
References	World Health Organization, 2010. WHO guidelines for indoor air quality: selected pollutants. WHO Regional Office for Europe, Copenhagen. Wolkoff, P., Nielsen, G.D., 2010. Non-cancer effects of formaldehyde and relevance for setting an indoor air guideline. Environment International 36, 788-799. Nielsen, G., Wolkoff, P., 2010. Cancer effects of formaldehyde: a proposal for an indoor air guideline value. Archives of Toxicology 84, 423-446. Nielsen, G.D., Larsen, S.T., Wolkoff, P., 2017. Re-evaluation of the WHO (2010) formaldehyde indoor air quality guideline for cancer risk assessment. Archives of Toxicology 91, 35-61.

Publisher	California‘s Office of Environmental Health Hazard Assessment (OEHHA)
Value	**55 µg/m³ (acute REL)** **9 µg/m³ (8 h REL)** **9 µg/m³ (Chronic REL)**
Type	Reference Exposure Limit (REL)
Update	2008
Comment	A concentration level at or below which no adverse health effects are anticipated for a specified exposure duration is termed the Reference Exposure Level (REL). RELs are based on the most sensitive, relevant, adverse health effect reported in the medical and toxicological literature. RELs are designed to protect the most sensitive individuals in the population by the inclusion of margins of safety. Since margins of safety are incorporated to address data gaps and uncertainties, exceeding the REL does not automatically indicate an adverse health impact. An acute REL is an exposure that is not likely to cause adverse health effects in a human population, including sensitive subgroups, exposed to that for the specified exposure duration on an intermittent basis. 8-hour RELs are developed for assessing potential non cancer health impacts for exposures to the general public that occur on a recurrent basis, but only

	during a portion of each day. 8-hour RELs are compared to air concentrations that represent an average (daily) 8-hour exposure. Chronic RELs are developed for assessing non cancer health impacts from long-term exposure. A chronic REL is a concentration level for inhalation exposure and in a dose at or below which no adverse health effects are anticipated following long-term exposure. Long-term exposure for these purposes has been defined by U.S. EPA as at least 12% of a lifetime, or about eight years for humans.
References	Office Of Environmental Health Hazard Assessment, 2015. Air Toxics Hot Spots Program. Risk Assessment Guidelines - Guidance Manual for Preparation of Health Risk Assessments. California Environmental Protection Agency, Sacramento California. https://oehha.ca.gov/air/general-info/oehha-acute-8-hour-and-chronic-reference-exposure-level-rel-summary (assessed: 27.10.2019)

Publisher	The German Committee on Indoor Guide Values (AIR)
Value	**0.1 mg/m³ (30 minute)**
Type	Indoor Guideline
Update	2016
Comment	Irritation of the human upper airways and cytotoxicity and carcinogenicity in animals studies following chronic exposure are the critical endpoints of inhaled formaldehyde. Regarding sensory irritation no valid LOAEC is available to derive a health hazard guide value (RW II) for formaldehyde in indoor air. Based on a NOAEC of 0.63 mg/m³ for sensory irritation in humans, a factor of 1 for time extrapolation and a factor of 5 for inter human variability the Committee derives a precautionary indoor air guide value (RW I) of 0.1 mg formaldehyde per cubic meter. The Committee recommends that this guide value should not be exceeded at any interval of half an hour during a day. For the assessment of the cancer risk of inhaled formaldehyde the Committee uses a non-linear approach due to the results of the animal studies showing an exponential increase of the risk curve: the additional theoretical cancer risk of a non-smoker following a continuous (80 years) inhalative exposure to 0.1 mg formaldehyde per cubic meter is assumed to be $3x10^{-7}$. In conclusion the indoor air guide value for formaldehyde is also protective against cancer risk of inhaled formaldehyde.
References	Ausschuss für Innenraumrichtwerte, 2016. Richtwert für Formaldehyd in der Innenraumluft. Bundesgesundheitsblatt 59, 1040-1044.

Publisher	French Agency for Food, Environmental and Occupational Health & Safety
Value	**100 µg/m³ (short term)**
Type	Indoor Air Quality Guideline
Update	2018
Comment	The updating of knowledge on the health effects of formaldehyde led ANSES to recommend a single short-term IAQG of 100 µg/m³ to protect the general population from acute and chronic effects. This value should be complied with for repeated and continuous short-term exposure over a day. ANSES insists on the need to develop suitable measurement methods for comparison with the single short-term IAQG of 100 µg/m³ to be complied with for repeated and continuous short-term exposure over a day. The current French regulations on the surveillance of indoor air quality in public-access buildings rely on regulatory IAQGs on the one hand and on a sampling strategy aiming to characterize long term exposure with samples taken over several days, repeated in two different periods of the year, on the other hand. These methods, especially the required sampling times, cannot be used to assess the variability of concentrations over time, in particular the existence of exposure peaks, and thus ensure compliance with the IAQG for formaldehyde set at 100 µg/m³ with a duration of application of one to four hours. Pending the possible definition of new surveillance methods in light of the proposal of a single short-term IAQG, a pragmatic option could be considered to interpret measurement results for concentrations obtained over several days with the aim of characterizing long-term exposure as currently recommended in the regulations. For this to happen, the authorities could apply an additional safety factor to the single short-term IAQG. This would enable a comparison with measurements obtained over several days by reducing the risk of the single IAQG of 100 µg/m³ being exceeded over short periods (concentration peaks).
References	OPINION of the French Agency for Food, Environmental and Occupational Health & Safety on the revision of ANSES's reference values for formaldehyde: occupational exposure limits (OELs), derived no-effect levels (DNELs) for professionals, toxicity reference values (TRVs) and indoor air quality guidelines (IAQGs). Maisons-Alfort, 2018. https://www.anses.fr/en/system/files/AIR2017SA0041EN.pdf (assessed: 25.11.2019)

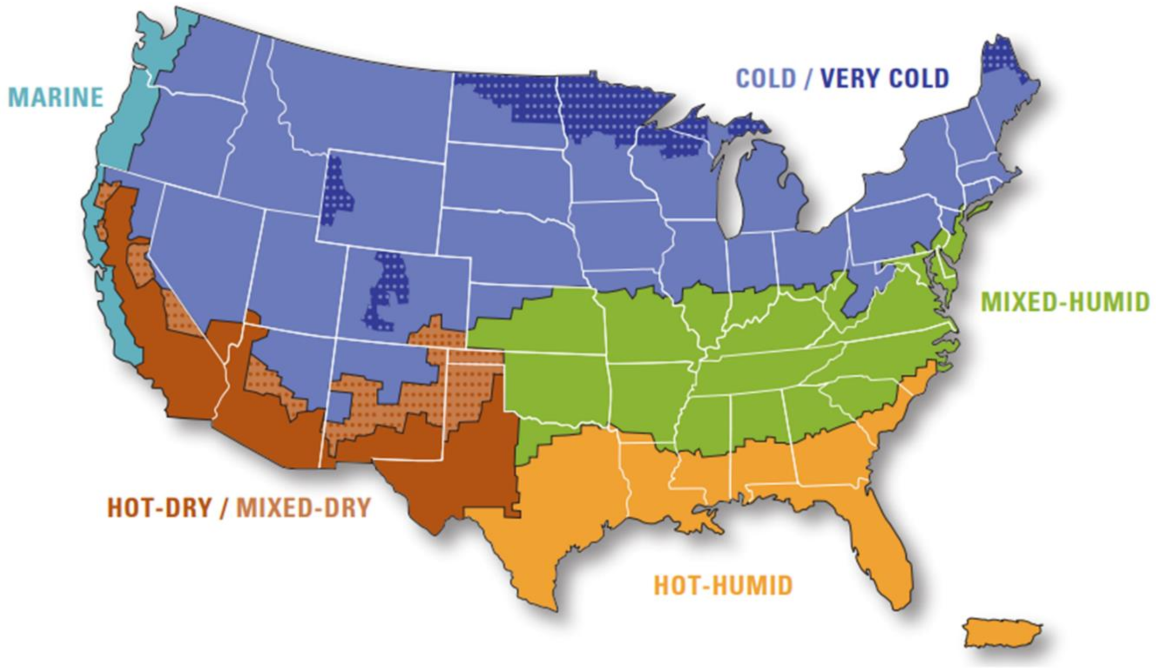
Publisher	Health Canada
Value	**123 µg/m³ (1-hour)** **50 µg/m³ (8-hour)**
Type	Residential Indoor Air Quality Guideline
Update	2006
Comment	A 1-hour exposure limit is established at 123 µg/m³ (100 ppb), which represents one fifth of the no observable adverse effects level and one tenth of the lowest observable adverse effects level found for eye irritation in the Kulle (1993) study. An eight-hour exposure limit is established at 50 µg/m³ (40 ppb), i.e., at the lower end of the exposure category associated with no significant increase of asthma hospitalization in the Rumchev, et al. (2002) study.
References	Health Canada, 2006. Residential indoor guideline - formaldehyde. Ottawa, Ontario, ISBN 0-662-42661-4. http://publications.gc.ca/collections/Collection/H128-1-06-432-1E.pdf (assessed: 28.10.2019). Kulle, T.J., 1993. Acute odor and irritation response in healthy nonsmokers with formaldehyde exposure. Toxicol. Ind. Health 5: 323-332. Rumchev, K.B., Spickett, J.T., Bulsara, M.K., Phillips, M.R., and Stick, S.M., 2002. Domestic exposure to formaldehyde significantly increases the risk of asthma in young children. Eur. Respir. J. 20: 403-406.

Publisher	Committee for Risk Assessment (RAC), European Chemicals Agency
Value	**0.05 mg/m³**
Type	Derived No Effect Level (DNEL)
Update	2019
Comment	DNEL is defined as the level of chemical exposure above which humans should not be exposed. RAC took note of the DNEL of 0.1 mg/m³ as proposed by the dossier submitter, based on an existing WHO guideline, derived from human sensory irritation data. RAC highlighted several limitations of the underlying data. RAC agreed on a weight of evidence approach considering human and animal data for the relevant precursor events deriving a chronic DNEL of 0.05 mg/m³ for the inhalation route based on a study with monkeys.
References	Minutes of the 49th Meeting of the Committee for Risk Assessment (RAC 49), RAC/M/49/2019. https://www.echa.europa.eu/documents/10162/22838445/RAC49%1f FinalMinutes_rev1.pdf/d27e5c54-9c86-ee7a-89a8-80672fab47d3 (assessed: 12.11.2019)

Section Summary: The classification of formaldehyde as a human carcinogen or suspected human carcinogen triggered intensive discussions on formaldehyde guidelines and regulatory levels. *The indoor air guideline of 0.1 mg/m³ as derived by the World Health Organization (WHO) can be considered as the most reliable value. Several studies showed that it is protective against both acute and chronic sensory irritation in the airways in the general population. Moreover, the formaldehyde WHO guideline value is also considered defendable for prevention of all types of cancer.*

4 Formaldehyde in ambient air

The US Department of Energy recognized eight climate regions in the United States. As shown in Figure 4.1, seven climate zones occur in the continental United States, the sub-artic climate zone appaears only in Alaska (Baechler et al., 2015). The mainland of the United States is between the 24th and the 49th latitude. Consequently, atmospheric reactions in the troposphere differ widely within the United States



<u>Figure 4.1:</u> Climate zones in the continental United States (the figure was taken from Baechler et al., 2015).

The tropospheric photochemical processes which lead to the formation of formaldehyde are well-documented (Luecken et al., 2012). The ozonolysis, e.g. following the mechanism described by Criegee (1975) and the reaction mechanisms of alkanes and alkenes with hydroxyl radicals and nitric oxide were described in detail by Pitts and Finlayson (1975) and Wagner and Zellner (1979).

The thermal decomposition of cellulose also forms a number of carbonyl compounds, including formaldehyde. The causes of this are mostly uncontrolled processes such as forest fires (Na and Cocker, 2008) or the controlled burning of wood in household wood-burning heating ovens (Hedberg et al., 2002). Baker et al. (2018) observed high formaldehyde concentrations during an extreme wildland fire episode in California. Shen and Gu (2009) postulate a mechanism by which furfural and formaldehyde are formed from degradation of the cellulose unit via levoglucosan.

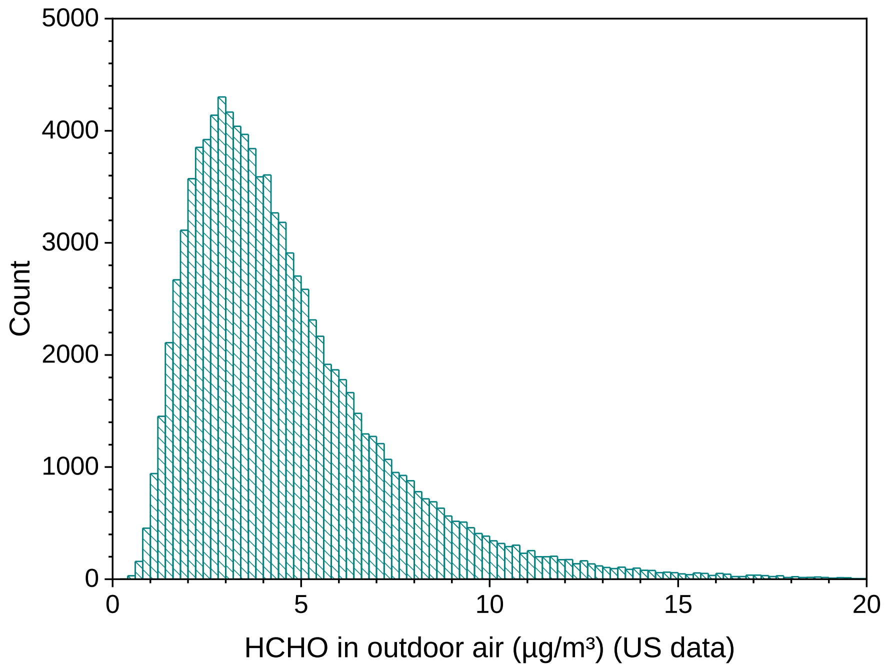
Formaldehyde has been identified as a relevant component in the exhaust of gasoline and diesel powered motor vehicles (Ban-Weiss et al., 2008; May et al., 2014). Intensive discussions are currently taking place concerning the sustainability of biofuels such as ethanol, higher alcohols, dimethyl esters and long-chain methyl esters. As these compounds already contain oxygen in their molecular framework, there are significant differences to the combustion chemistry of conventional hydrocarbons. Kohse-Höinghaus et al. (2010) reported on the reaction paths when combusting different biofuels. The authors conclude that such processes can generally be expected to produce carbonyl compounds, particularly formaldehyde. Leplat et al. (2011) come to similar conclusions following their research on the combustion chemistry of ethanol under varying conditions.

Table A1 in APPENDIX A shows a cross section of data which represents both the different areas of concentration and regional aspects in the United States. Data was taken both from the usual databases and previous research. Conventional atmospheric chemistry long considered formaldehyde as one of many target compounds in the complex reaction schemata of atmospheric components. It is also an important component of plant chemistry (Seco et al., 2008). Consequently, formaldehyde can be found as an atmospheric trace element in outlying areas and is thus to be considered as ubiquitous.

The formaldehyde concentrations in urban regions differ greatly. In northern and central Europe and in the United States, average values between 3 and 15 ppb are typical (Salthammer, 2013; 2019a; 2019b). The sometimes very high concentrations in Asian and South American megacities have different causes. The intensive solar irradiation combined with high concentrations of reactive organic compounds such as alkenes leads to photo smog causing a large proportion of the formaldehyde formation in metropolitan areas like Beijing, particularly in the summer months (Duan et al., 2008). Today, cities with high photochemical air pollution typically have an average formaldehyde concentration of between 20 ppb and 30 ppb with peaks of 40 ppb to 50 ppb. The highest formaldehyde values have been recorded in Rio de Janeiro for some years. Corrêa et al. (2010) measured a considerable increase in the formaldehyde concentration between 1998 and 2004, which they attributed to the increasing use of biofuels. Improved engine technologies then led to a gradual decline in the concentrations. Gaffney and Marley (2009) also closely investigated the increasing significance of formaldehyde as an outdoor air pollutant resulting from fuel emissions. Other sources also contribute to formaldehyde in ambient air. Gallego et al. (2016) reported increased formaldehyde concentrations around waste treatment plants in Barcelona.

Altshuller and McPherson (1963) measured extreme formaldehyde concentrations of 0.115 ppm (143 µg/m³) in the Los Angeles atmosphere in September 1961. Grosjean and Williams (1992) measured formaldehyde maximum concentrations of 26 ppb (32 µg/m³) and 15 ppb (19 µg/m³) at two Southern California smog receptor sites. Propper et al. (2015) point out that formaldehyde annual average concentrations declined by 22% in California between 1996 and 2012. However, their study does not consider extreme short-term events of LA-type summer smog (Haagen-Smit, 1952). Reactive oxidation chemistry is still an important formaldehyde source. In 2007 Choi et al. (2010) measured formaldehyde midday peaks between 15 ppb and 20 ppb over a forest canopy in California.

For this study it was assumed that formaldehyde outdoor concentrations range between 1 µg/m³ and 30 µg/m³ in urban areas and between 1 µg/m³ and 5 µg/m³ in other areas. This assumption includes short-time events like photo smog. It was also assumed that concentrations higher than 20 µg/m³ contribute with less than 1%. It was also taken into account that the distribution of atmospheric data is usually log-normal. The result of a Monte-Carlo simulation with a geometric mean of 3.98 µg/m³ and a geometric standard deviation of 1.75 µg/m³ is shown in Figure 4.2.



<u>Figure 4.2:</u> Monte-Carlo simulation of formaldehyde outdoor concentrations in the United States. The simulation is based on Table A1 (APPENDIX A). See Table 4.1 for statistical parameters.

Table 4.1: Statistical parameters of the Monte-Carlo simulation shown in Figure 4.2, representing formaldehyde outdoor concentrations in the United States (GM = geometric mean, σ_g = geometric standard deviation).

GM	σ_g	25-P	50-P	75-P	90-P	95-P	99-P
µg/m³							
3.98	1.75	2.73	3.98	5.80	8.16	9.98	14.64

References (Chapter 4)

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Section summary: *Today, the formaldehyde concentrations in outdoor air, particularly in polluted urban areas, sometimes already reach indoor levels. This is largely a result of chemical processes and the use of biofuels. In the medium term, this development might have consequences for the way buildings are ventilated and lead to a change in the way we evaluate human exposure (see Salthammer, 2013).*

5 Formaldehyde concentrations in indoor air under living conditions

For a comprehensive risk assessment the important question arises how and what extent dwellers are exposed to formaldehyde. Due to low permeation coefficient through skin the dermal pathway is not significant (Weschler and Nazaroff, 2014) and formaldehyde is not found in house dust. Therefore, inhalation is the major route of exposure. In their publication Salthammer et al. (2010) pointed out that *"…average exposure concentrations between 20 µg/m³ and 40 µg/m³ seem to be (more) realistic".* Their statement is based on a comprehensive literature review under special consideration of the German 2003 - 2006 Environmental Survey and several European studies from Finland, Sweden, France and Italy, which were carried out in the new Millennium.

In a follow-up study Salthammer (2013) stated: *"Taking into account further data (…), formaldehyde concentrations significantly under 40 ppb can be expected in normal living conditions in Central European and North American households"*. The author requires a change of paradigm because indoor formaldehyde concentrations are decreasing and outdoor formaldehyde concentrations are increasing. The differences in concentration levels are becoming smaller and in several cases an overlap can already be observed (see Figure 5.1).

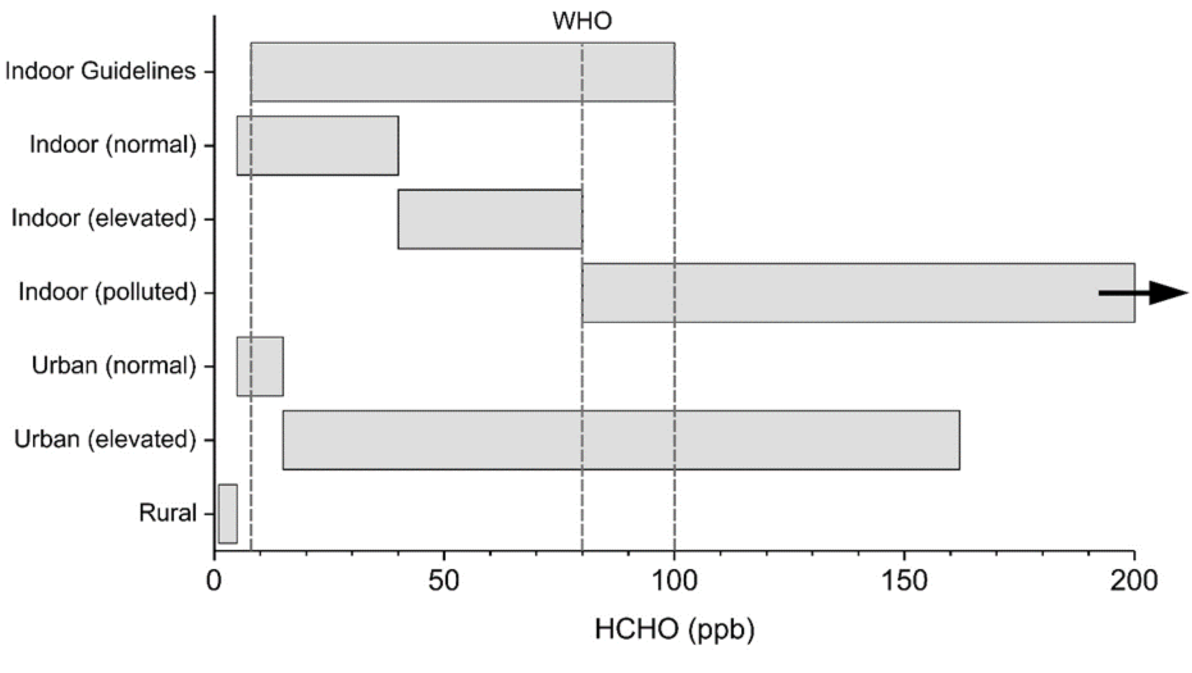

Figure 5.1: Range of formaldehyde concentrations in indoor and outdoor air. The range of current indoor guideline values between 8 ppb (10 µg/m³) and 100 ppb (124 µg/m³) is also provided. The figure was taken from Salthammer (2013), see this reference for more details.

Results from US studies, which present indoor formaldehyde concentrations under normal living conditions and might be considered as representative are provided in Table A2 (APPENDIX B).

The data in Table A.2 give a clear picture of indoor formaldehyde concentrations in the United States under normal living conditions and allow the derivation of a log-normally distributed probability function. Taking into consideration all available data, a log-normal distribution curve was calculated from a geometric mean of 23.0 µg/m³ and a geometric standard deviation of 3.0 µg/m³ by use of a Monte-Carlo simulation (see Figure 5.2 and Table 5.1).

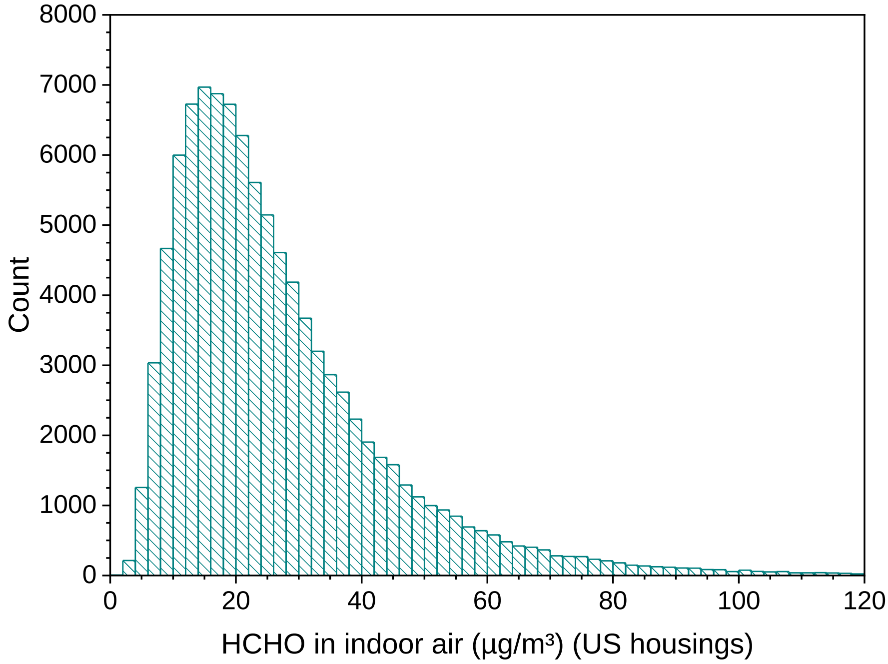

Figure 5.2: Monte-Carlo simulation of formaldehyde indoor concentrations in the United States under normal living conditions. The simulation is based on Table A2. See Table 5.1 for statistical parameters.

Table 5.1: Statistical parameters of the Monte-Carlo simulation shown in Figure 5.2, representing formaldehyde indoor concentrations under normal living conditions in the United States (GM = geometric mean, σ_g = geometric standard deviation).

GM	σ_g	25-P	50-P	75-P	90-P	95-P	99-P
µg/m³							
22.47	1.84	14.88	22.43	33.88	49.16	61.49	93.98

The situation is similar in Canada. On the basis of five studies, carried out between 1989 and 1995, Liteplo and Meek (2003) calculated a log-normal distribution of formaldehyde concentrations in Canadian homes with P-50 = 28.7 µg/m³, P-75 = 45.1 µg/m³, P-90 = 70.7 µg/m³, P-95 = 91.2 µg/m³ and P-97.5 = 113.8 µg/m³. Gilbert et al. (2005) measured a 50-P value of 29.6 µg/m³ (min = 5.5 µg/m³, max = 87.5 µg/m³) in residential indoor air (N = 59) in Prince Edward Island. Gilbert et al. (2008) measured the concentration of formaldehyde in 96 homes in Quebec City in dependence of the air exchange rate. The authors found a clear correlation between formaldehyde concentration and ventilation: P-50 = 30.6 µg/m³ at AER = 0 - <0.113 h^{-1} and P-50 = 22.6 µg/m³ at AER = >0.230 h^{-1}.

Several studies with relevance for United States are available, which are not considered in Figure 5.2 and Table 5.1. These studies review or re-evaluate already published data.

Logue et al. (2011) summarize the results of 77 published studies. As far as formaldehyde is concerned they distinguish between short-term concentrations during typical indoor activities and representative indoor air concentrations.

In a follow-up study Logue et al. (2012) estimated the chronic health impact of air pollutants in US residences. They assumed 69 µg/m³ as the formaldehyde population-average concentration. This value represents the calculated arithmetic mean from Logue et al. (2011). It was pointed out earlier that due to the asymmetric distribution of pollutant concentrations in atmospheric environments the arithmetic mean is not a representative parameter. In their publication Logue at al. (2011) also report a median (50-P) of 23 µg/m³ for formaldehyde.

Hun et al. (2010) re-evaluated data of the RIOPA study.

On the basis of previously published data Chan et al. (2016) calculate disability adjusted life years (DALYs) and come to the conclusion that chronic health effects are driven primarily by $PM_{2.5}$ exposure and secondarily by formaldehyde exposure.

Diaz and Siegel (2018) reviewed 50 studies and conclude that there is no evidence of higher exposure to HCHO in social housing.

The study by Noris et al. (2013) on indoor air quality after retrofits and the study by Hult et al. (2015) on ventilation and source control use indoor-outdoor data.

Zaatari et al. (2014) reviewed the ventilation and indoor air quality in retail stores. Three studies reported formaldehyde concentrations across a variety of store types with a maximum concentration of 26 ppb (32 µg/m³).

Frey et al. (2014) discuss the indoor air quality in senior apartment buildings. However the data set is the same as used in Frey et al. (2015) (see Table A2 in APPENDIX B).

In their study on indoor quality in mobile homes Murphy et al. (2013) use the same data as in Maddalena et al. (2008) (see Table A2 in APPENDIX B).

The study by Ben-David and Waring (2016) uses simulated data.

In the following publications the data were not representative for the indoor environment or the quality of the data was not sufficient: Maddalena et al. (2009), Iyiegbuniwe (2013), Xiong et al. (2015).

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Section summary: *Results from studies, which represent indoor formaldehyde concentrations under normal living conditions in US housings and which might be considered as representative, were evaluated. The available data give a clear picture of indoor formaldehyde concentrations in the United States under normal living conditions and allow the derivation of a log-normally distributed probability function with a geometric mean of GM = 22.5 µg/m³.*

6 Ventilation and air exchange

There is an ongoing debate about ventilation and air exchange rates in living spaces. It is often argued that the tightening of buildings significantly lowers the supply of fresh air. This statement is true if measurements are carried out under steady-state conditions with doors and windows closed. However, this scenario does not represent normal living conditions, where people are moving from one place to another and doors and windows are frequently opened and closed. Fanger (2001) has calculated ventilation requirements on the basis of human bio-effluents and concludes that for each individual in a room a fresh air supply of 10 l/s is needed to maintain a high air quality standard with less than 15% of persons being dissatisfied.

Carbon dioxide (CO_2) is also a useful indicator for adequate ventilation. The Committee for Indoor Guideline Values (AIR) of the German Federal Environment Agency evaluates CO_2 concentrations below 1000 ppm as “hygienically acceptable”, CO_2 concentrations between 1000 ppm and 2000 ppm as “hygienically noticeable” and concentrations above 2000 ppm as “hygienically unacceptable” (Ad hoc AG, 2008; Fromme et al., 2019). Internationally, the limit values for CO_2 indoors only differ slightly. Under steady-state conditions, the minimum air exchange rate to maintain a certain CO_2 concentration can be calculated from Equation (6.1).

$$AER = \frac{1000 \cdot N \cdot \dot{Q}_{CO2}}{(C_{CO2}(t) - C_{CO2(ambient)}) \cdot V} \quad (6.1)$$

N is the number of persons in the room, $\dot{Q}_{CO2}$ is the CO_2 exhalation rate per person in l/h, V is the room volume, $CO_2(t)$ is the concentration in indoor air and $CO_{2(ambient)}(t)$ is the concentration in ambient air.

Figure 6.1 has been taken from Schieweck et al. (2018) and provides a typical indoor scenario with four persons and an exhalation rate of 20 l/h and person. The CO_2 concentration in ambient air is assumed to be 400 ppm. The air exchange AER is plotted against the room volume. The green area represents indoor CO_2 concentrations <1000 ppm, the yellow area represents CO_2 concentrations >1000 ppm and <2000 ppm and the red area represents CO_2 concentrations >2000 ppm. Under these conditions a room with a volume of 100 m³ requires an air exchange rate of 0.5 h^{-1} to stay below 2000 ppm CO_2. For a CO_2 concentration <1000 ppm an air exchange of 1.3 h^{-1} is needed. In case of physical exercise the release of carbon dioxide is considerably higher (Persily and de Jonge, 2017), which also requires higher air exchange rates.

The use of an adequate, hygienically acceptable and realistic air exchange rate is frequently discussed. In their study on formaldehyde concentrations in US residences, Hult et al. (2015)

have established target air exchange rates between 0.2 h^{-1} and 0.8 h^{-1} and have also considered a reference air exchange rate of 0.35 h^{-1} as defined by ASHRAE.

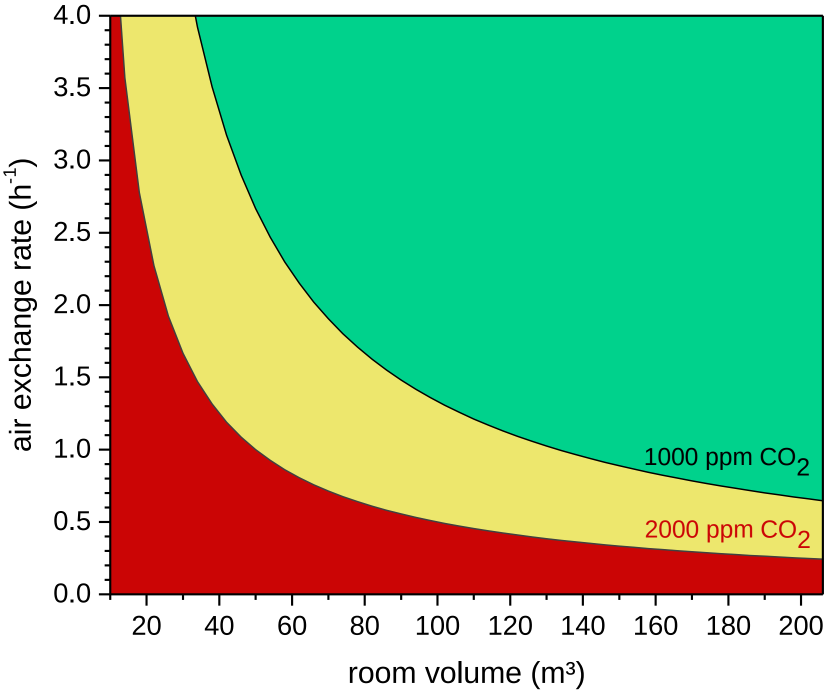

Figure 6.1: Air exchange rates (AER) in dependence of the room volume for N = 4, $\dot{Q}_{CO2}$ = 20 l/h and person, $CO_{2(ambient)}(t)$ = 400 ppm and two CO_2 indoor concentrations (1000 ppm and 2000 ppm). The figure is taken from Schieweck et al. (2018).

In their report on formaldehyde emissions from laminate flooring the Centers for Disease Control and Prevention (CDC) (2016) estimated an AER uniform distribution between 0.1 h^{-1} and 1.21 h^{-1} for the Monte-Carlo simulations of formaldehyde concentrations. The calculated median value is then 0.65 h^{-1}.

Other studies are summarized in Table A3 (APPENDIX C). These studies include different types of building (mechanically ventilated and non-mechanically ventilated). The calculated median vales (50-P) are between 0.17 h^{-1} and 1.43 h^{-1}. Willem et al. (2013) demonstrated that the measured air exchange rate depends on the ventilation setting. As shown by Bennett et al. (2011) the calculated air exchange value also depends on the measurement method (tracer gas vs. constant emitter). It is therefore difficult to estimate correct or realistic air exchange rate distributions in buildings. This was already pointed out by Persily (2016), who evaluated field measurements of ventilation rates. The author state that 10% of the studies did not report how the ventilation rates were determined and 75% did not describe the timescale over which measurements were made. Human activities and dynamics also influence the air exchange as shown by Lee et al. (2016) for the opening of doors. Reiß and Ehrhorn (2009) studied the air exchange rate in naturally ventilated buildings dependent on the opening of windows. As shown in Figure 6.2, the influence can be weak or strong, depending on the building conditions. However, the Figure shows that opening the window for 3 minutes per hour (0.05 h/h) results in air exchange rates between 0.35 h^{-1} and 0.6 h^{-1} (see also Salthammer, 2019).

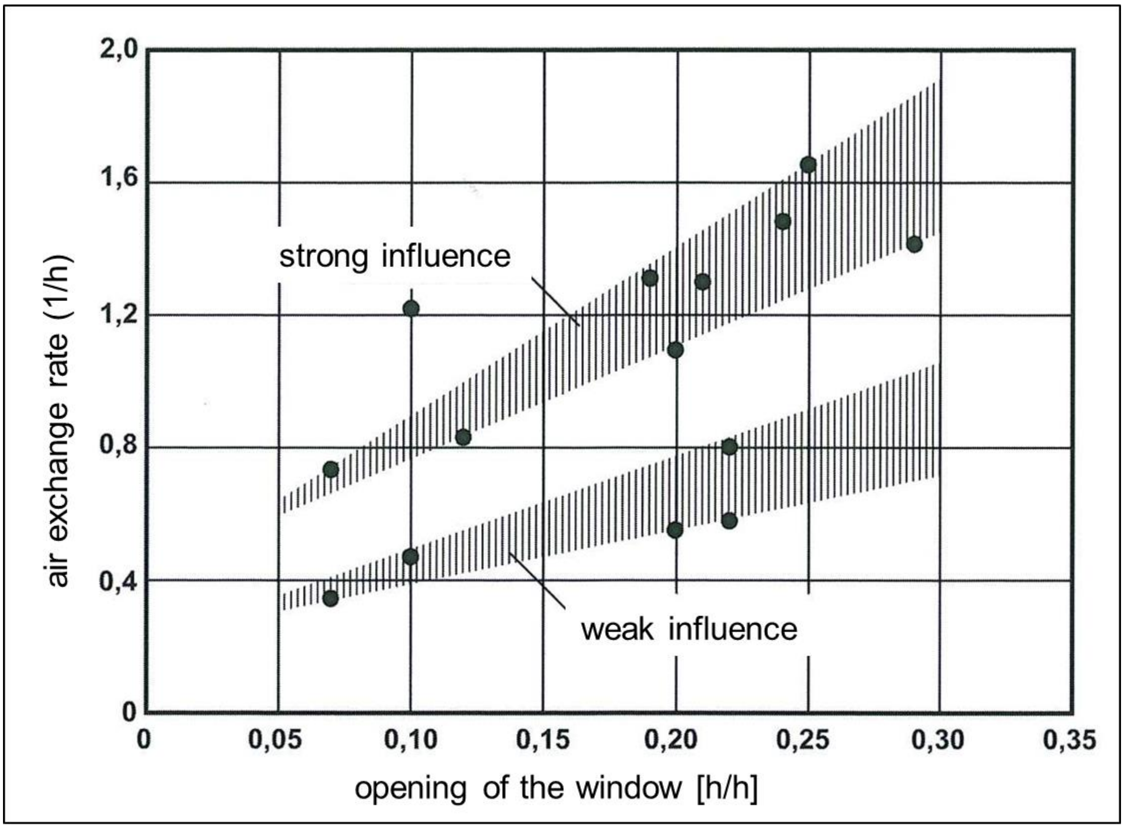

Figure 6.2: Influence of window opening on the air exchange rate in buildings (see Reiß and Ehrhorn, 2009).

However, when taking all these factors into account, an overall median of air exchange rates between 0.45 h^{-1} and 0.55 h^{-1} might be considered as realistic for U.S. housings and buildings. The data for the U.S. also show some similarity to the European data. On the basis of numerous studies Salthammer (2019) calculated a log-normal distribution of air exchange rates with a median of 0.52 h^{-1} for European housings (see Figure 6.3).

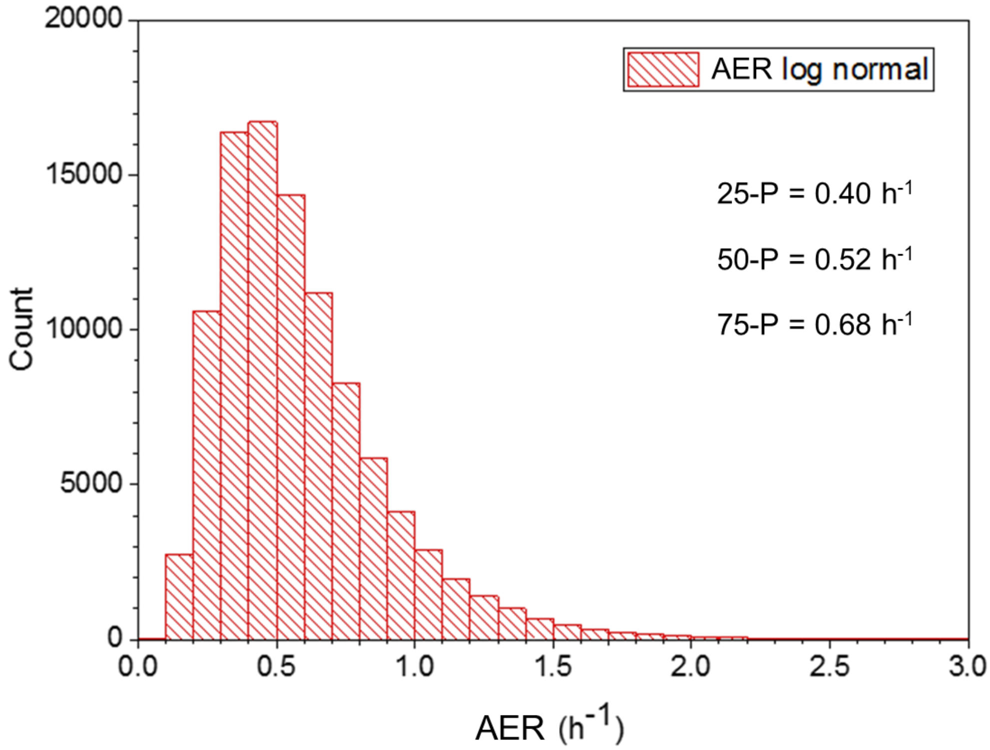

Figure 6.3: Monte-Carlo simulation of a log-normal distribution of air exchange rates for European housings. The figure has been taken from Salthammer (2019).

Many researchers found a strong dependence between air exchange rates and formaldehyde concentrations. Offermann et al. (2009) found a broad but linear relationship between log AER and the logarithmic indoor formaldehyde concentration in new homes (see Figure 6.4). Hult et al. (2015) measured formaldehyde concentrations in 9 U.S. residences for three different air exchange rates at each site (see Figure 6.5). The data in Figure 6.6 were taken from the study by Bradman et al. (2017), which was carried out in California early childhood education environments.

On the basis of existing data Rackes and Waring (2016) studied VOC and VVOC emission rates from building materials in dependency of air exchange rates. The authors found that the formaldehyde data could be described best by a dependent emissions model (DEM) and conclude that the air exchange rate influences the emission rate via the responded to the AER via the boundary layer between air and emission source. This result is in accordance with previous findings (Myers, 1984).

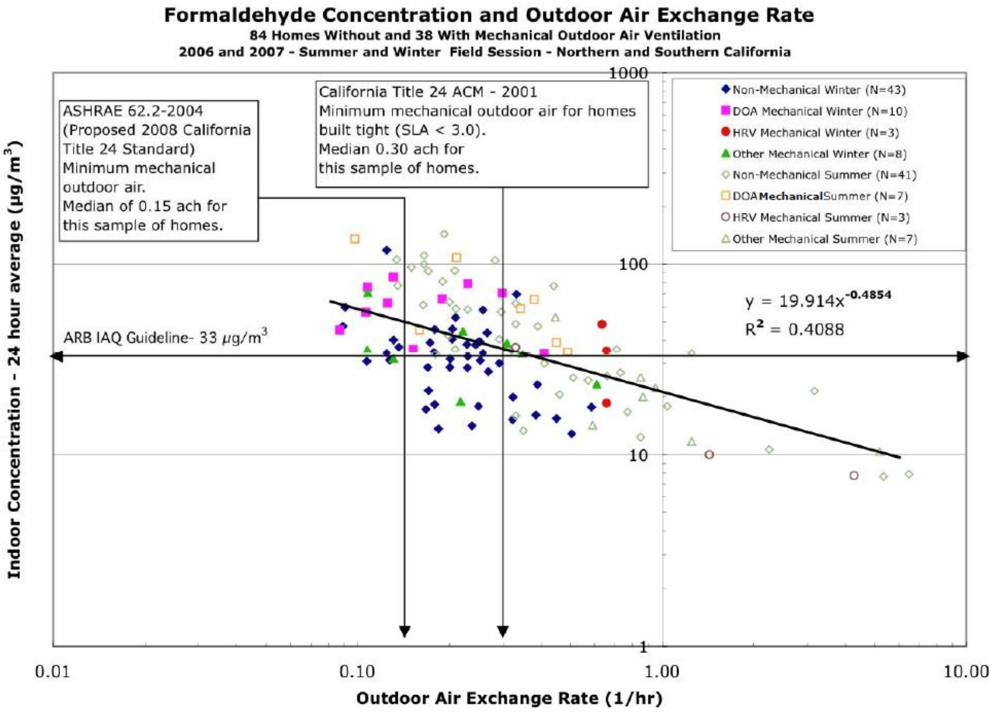

Figure 6.4: Indoor formaldehyde concentrations versus air exchange rates in California early childhood education environments. The figure has been taken from Offermann (2009).

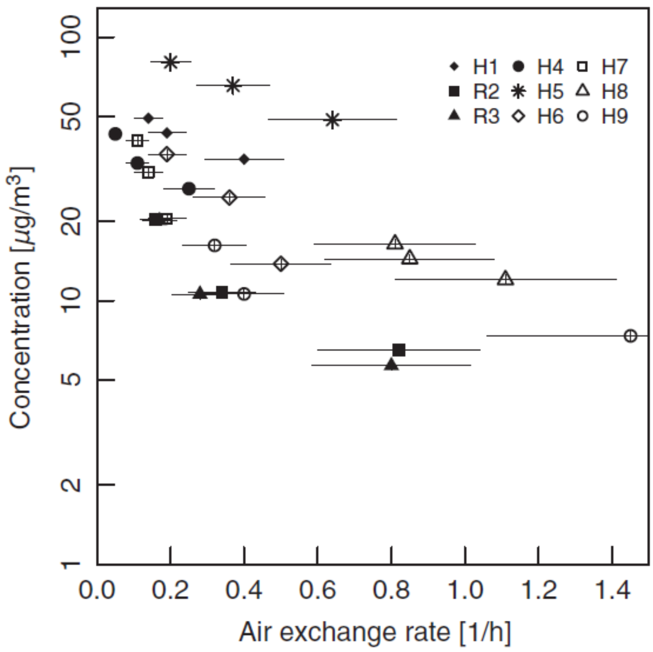

Figure 6.5: Indoor formaldehyde concentrations versus air exchange rates in 9 U.S. residences for three different air exchange rates at each site. The figure has been are taken from Hult et al. (2015).

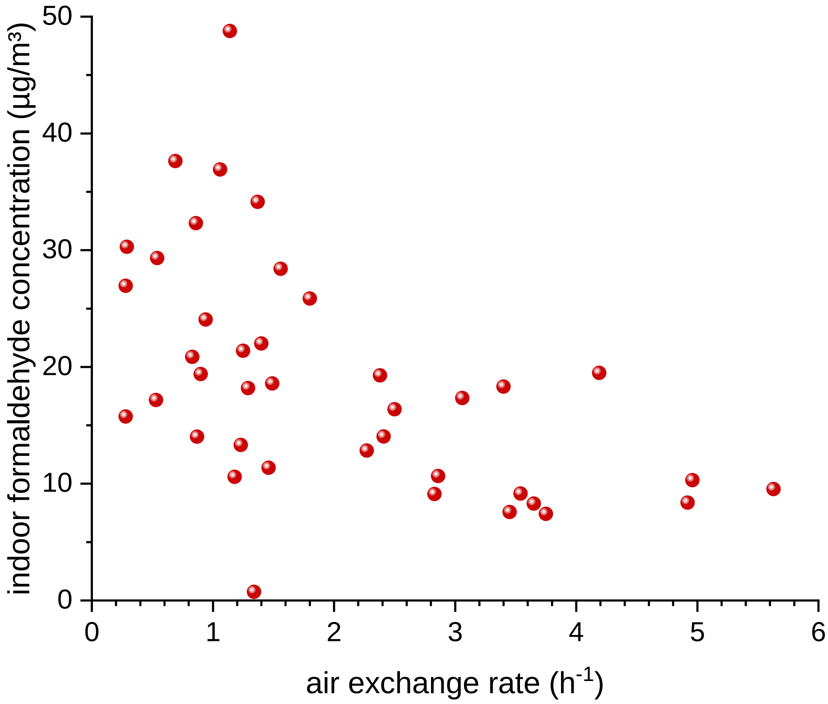

Figure 6.6: Indoor formaldehyde concentrations versus air exchange rates in California early childhood education environments. The data have been taken from Bradman et al. (2017).

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Section summary: *It is often argued that the tightening of buildings significantly lowers the supply of fresh air. This statement is only true if measurements are carried out in manually ventilated buildings under steady-state conditions with doors and windows closed. However, under normal living conditions, people are moving from one place to another and doors and windows are frequently opened and closed. Several studies showed that the assumption of a minimum average air exchange rate of 0.5 h^{-1}, under residential-typical conditions, is reasonable.*

7 Formaldehyde emission sources

In a recent publication, Salthammer (2019a) evaluated formaldehyde sources in the indoor environment. The corresponding data are published in Salthammer (2019b). From the available data, distribution functions (log-normal, normal, uniform) were calculated. The results are presented in Table 7.1. It must be pointed out, that the data represent typical distributions of emission rates. Worst case scenarios (see below) are not taken into account.

Table 7.1: Distribution functions (log-normal, normal, uniform) representing formaldehyde concentrations and emissions related to different products and processes in the indoor environment (from Salthammer (2019a), see also Salthammer (2019b)).

Category	Model	Parameters 1)
HCHO from indoor chemistry	log-normal	GM = 40 µg/h, σ_g = 1.65 µg/h
HCHO from burning candles 2)	log-normal	GM = 192.5 µg/h, σ_g = 1.42 µg/h
HCHO from burning incense sticks 2)	uniform	Min = 3 µg/m³, Max = 39 µg/m³
HCHO from burning mosquito coils 2)	uniform	Min = 0.54 mg/h, Max = 7.52 mg/h
HCHO from tobacco smoking	uniform	Min = 20 µg/m³, Max >1,000 µg/m³
HCHO from electronic cigarettes	uniform	Min = 1 µg/m³, Max = 135 µg/m³
HCHO from decorative fireplaces 2)	uniform	Min = 698 µg/h, Max = 10,637 µg/h
HCHO from wood-burning fireplaces 2)	uniform	Min = 5 µg/m³, Max = 48 µg/m³
HCHO from cooking	normal	µ = 700 µg/h, σ = 100 µg/h
HCHO from air cleaning devices 2)	uniform	Min = 2 µg/m³, Max = 25 µg/m³
HCHO from miscellaneous products 2)	uniform	Min = 1 µg/m³, Max = 5 µg/m³
HCHO from textile	log-normal	GM = 1.9 µg/(m² h), σ_g = 1.38 µg/(m² h)
HCHO from carpet	log-normal	GM = 3.9 µg/(m² h), σ_g = 1.65 µg/(m² h)
HCHO from surface coatings	log-normal	GM = 2.3 µg/(m² h), σ_g = 1.56 µg/(m² h)
HCHO from wallcoverings	log-normal	GM = 0.5 µg/(m² h), σ_g = 2.23 µg/(m² h)
HCHO from solid wood	normal	µ = 4 µg/(m² h), σ = 1 µg/(m² h)
HCHO from particleboard 3)	log-normal, normal	GM = 79 µg/(m² h), σ_g = 1.37 µg/(m² h)
HCHO from OSB 3)	log-normal, normal	GM = 39 µg/(m² h), σ_g = 1.96 µg/(m² h)
HCHO from MDF	n.a.	GM = 80 µg/(m² h)
HCHO from plywood	n.a.	GM = 48 µg/(m² h)
HCHO from laminate	log-normal	GM = 8.5 µg/(m² h), σ_g = 1.8 µg/(m² h)
HCHO from furniture	log-normal, normal	GM = 17.8 µg/(m² h), σ_g = 2.54 µg/(m² h)
HCHO from doors	log-normal	GM = 18.2 µg/(m² h), σ_g = 2.7 µg/(m² h)
HCHO from mineral wool	n.a.	GM = 31.0 µg/(m² h)

1) Conversion factor: 1 ppb = 1.24 µg/m³ (1013 mbar (101300 Pa), 23 °C (293 K), M(HCHO) = 30.03 g/mol)

2) One item

3) The data for particleboard and OSB represent the European market (Marutzky and Schripp, 2012).

From the available formaldehyde emission data distributions the area specific emission rates were calculated. Geometric mean values and, if available, geometric standard deviations were applied. Moreover, the emission rates were corrected for real-life conditions if necessary. During use, wood-based materials and mineral wool are usually covered with other materials, primer, paint or wallcoverings. As discussed by Salthammer (2019a), a correction factor of 0.25 (which means 75% reduction) should be applied to the wood-based products. A factor of 0.15 (which means 85% reduction) should be applied to mineral wool.

To demonstrate the covering (barrier) effect for particleboard, a set of experiments was performed by WKI in 2017. A particleboard (22 mm x 2070 mm x 4110 mm) being produced in the European Union (Perforator Value 5.4 mg/100 g atro, related to 6.5% humidity) was selected for the tests. Two shelves were cut from the particleboard, connected edge to edge, the back was then wrapped airtight into aluminum foil and a surface of 0.5 m^2 was left open.

In the first experiment the release of formaldehyde from this test specimen was studied in a 1 m^3 stainless steel chamber at T = 23 °C, RH (relative humidity) = 45%, AER = 0.5 h^{-1} and L = 0.5 m^2/m^3. After 116 hours the test specimen was removed from chamber, a water-based primer was applied to the open surface of the particleboard and dried outside the chamber for about 4 hours. Then the test was continued under the same conditions. Without primer, the chamber concentration was about 80 ppb, with a slightly decreasing trend (82 ppb after 6 h → 77 ppb after 116 h). With primer, the formaldehyde concentration in the chamber was significantly reduced. The measured values were between 27 ppb and 29 ppb (see Figure 7.1). It becomes clear that the primer is an effective diffusion barrier for formaldehyde. Under the chosen test conditions the formaldehyde concentration with primer is about 35% of the concentration without primer.

In a second series of experiments test specimen from the same particleboard were used. This time the freshly cut shelves were fixed on a wood frame (oak), the edges were sealed and different types of coatings were applied. The hollow space under the shelves was left empty and was not filled with insulation material. Six experiments were carried out in parallel in 1 m^3 stainless steel chambers at T = 23 °C, RH = 45%, AER = 0.5 h^{-1} and L = 0.5 m^2/m^3. In all cases a steady concentration was reached. In Test 2 with primer only, the formaldehyde concentration in the chamber was again approx. 30% of the concentration without primer. For a better comparison the results from Experiment 2 are presented in relative units (see Table 7.2). The results make clear that surface coatings cause a significant reduction of the formaldehyde emission rate from particleboard, which is between 70% and 98%. It is, however, difficult to estimate the reduction effect under real-life conditions. Under the assumption that most walls are coated with dispersion paint, plaster and wallpaper a reduction of 75% was assumed by Salthammer (2019a) for the calculation of indoor scenarios.

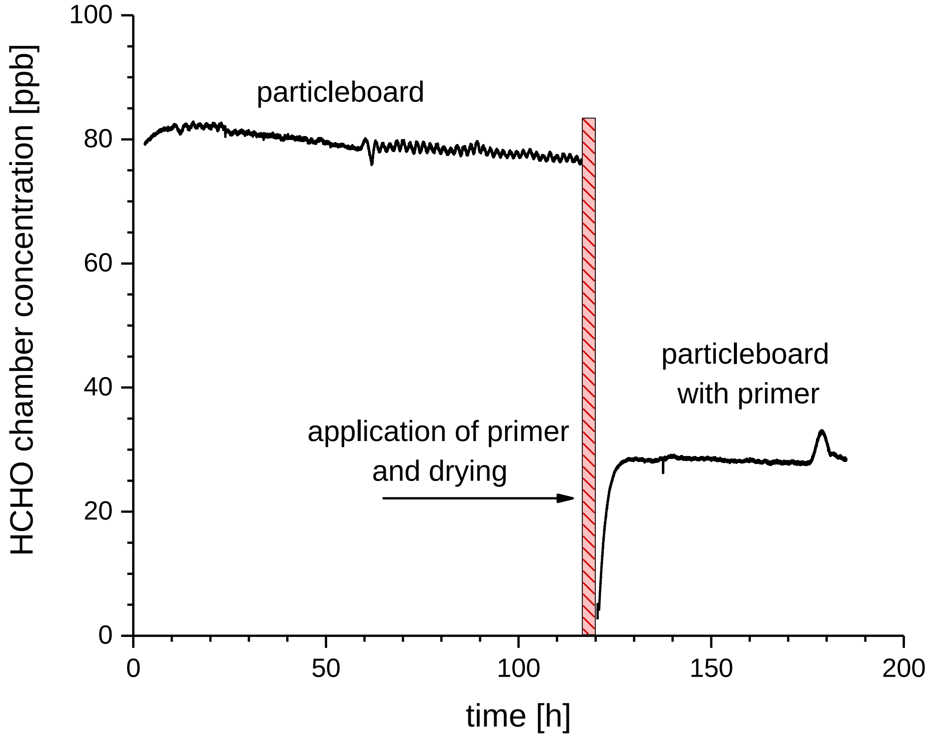

Figure 7.1: Experiment 1: formaldehyde concentration in a 1 m³ stainless steel chamber before and after application of primer (T = 23 °C, RH = 45 %, ACH = 0.5 h^{-1}, L = 0.5 m²/m³). The figure has been taken from Salthammer (2019a).

Table 7.2: Experiment 2: reduction of the area specific formaldehyde emission rate from particleboard by different types of covering for the described experimental scenario. The table has been taken from Salthammer (2019b).

Test	Covering	rel. SER_A [%]	reduction of rel. SER_A [%]
1	no covering	100	0
2	with primer	30	70
3	with primer and dispersion paint	24	76
4	with primer and plaster	22	78
5	with primer and wallpaper (fleece)	6	94
6	with primer and latex paint	2	98

In Figure 7.2 the blue squares (■) represent the geometric mean (GM) of the determined area specific emission rate (SER_A) distribution for wood-based materials and mineral wool on the basis of data from Table 7.1. The black dots (●) represent corrected values for a 75% reduction (wood-based materials) and 85% (mineral wool) of the formaldehyde emission from covering. The upper whiskers represent the emission rate of wood-based materials resulting from 70% reduction and the lower whiskers represent the emission rate resulting from 98% reduction.

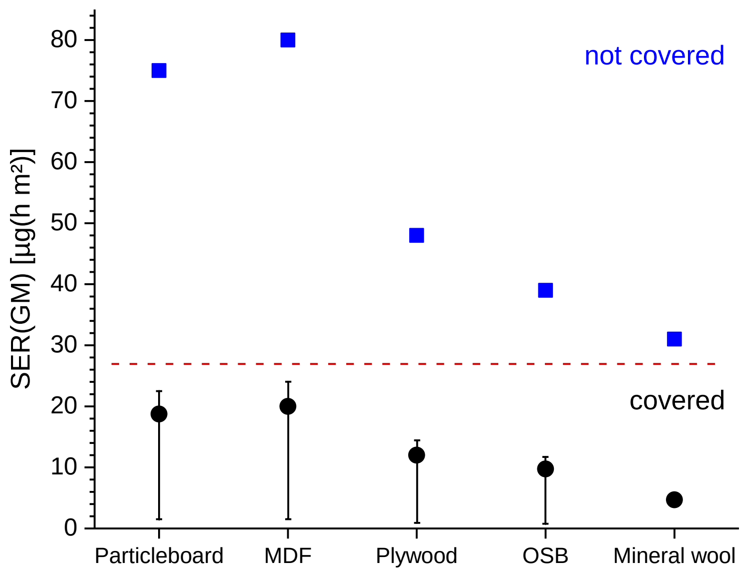

Figure 7.2: Effect of covering on the formaldehyde emission rate of wood-based materials and mineral wool. The data of uncovered materials are taken from Table 7.1.

Figure 7.3 provides a comparison of different building product groups. The data are taken from Table 7.1. The black squares (■) represent the geometric mean (GM) of the determined area specific emission rate (SER_A) distribution for a certain product group and the whiskers are the geometric standard deviations (σ_g). The blue squares represent the corrected values for wood-based materials and mineral wool (■).

For other product types (e.g. consumer products) area specific emission rates could not be derived. Therefore, these are not considered in Figure 7.3.

After correction, all area specific emission rates are found in a narrow range between 0.5 µg/(m² h) and 20 µg/(m² h). Please note that in Figure 7.3 the scale of the ordinate is linear. However, even under real-life conditions, the emission rates of wood-based products, laminate and furniture, all usually manufactured by use of formaldehyde containing resins, tend to be higher (8.5 µg/(m² h) to 20 µg/(m² h)) in comparison to textile, carpet, wallcoverings, surface coatings and solid wood (0.5 µg/(m² h) to 4 µg/(m² h)). An exception is OSB, which is often manufactured by use of formaldehyde-free resins. Consequently, the uncorrected and corrected geometric mean SER_A of OSB are lower in comparison to the other types of wood-based products. Mineral wool, usually produced by use of formaldehyde containing resins, also exhibits a low emission profile 4.7 µg/(m² h), which is due to the covering by other building materials (gypsum board, wood-based products, foil, primer, paint, wallcovering), which act as strong diffusion barriers.

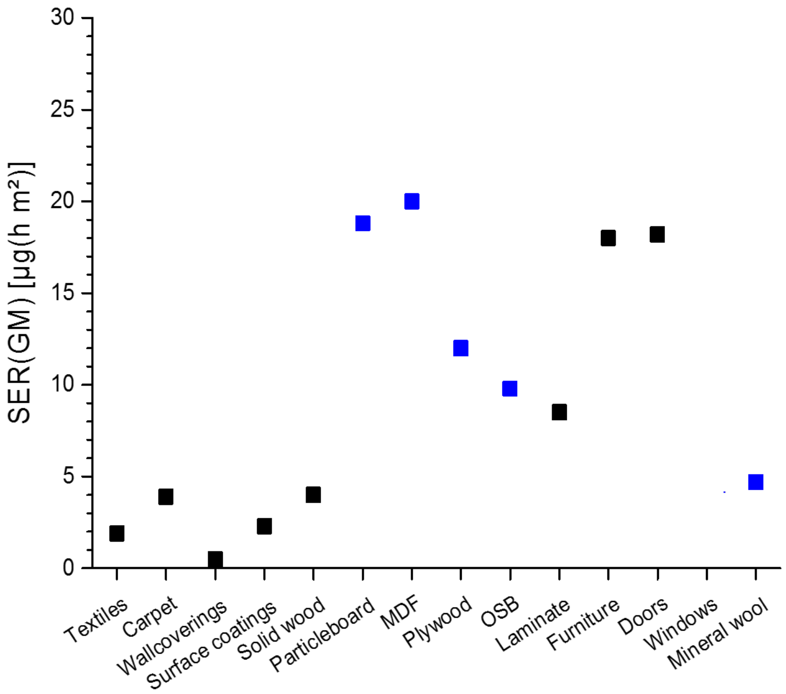

Figure 7.3: Comparison of area specific emission rates (geometric means) of indoor related materials and products (■). The blue squares (■) represent corrected values for covered wood-based materials and mineral wool. Please note that the data for wood-based materials are based on the study by Marutzky and Schripp (2012) and see text for further discussions.

In Figure 7.3 the data for wood-based materials are based on a German study from 2012, which summarizes results from a Research Project of DIBt Deutsches Institut für Bautechnik on raw particleboard, medium density fiberboard (MDF), oriented strand board (OSB) and plywood. According to the former regulations for emission testing, all samples were measured as produced in the factory. The measurements were carried out in accordance with DIN EN 717-1 (2005) at T = 23 °C, RH = 45%, AER = 1 h^{-1} and L = 1 m²/m³ (Marutzky and Schripp, 2012). The distribution is shown for particleboard in Figure 7.4 (red bars).

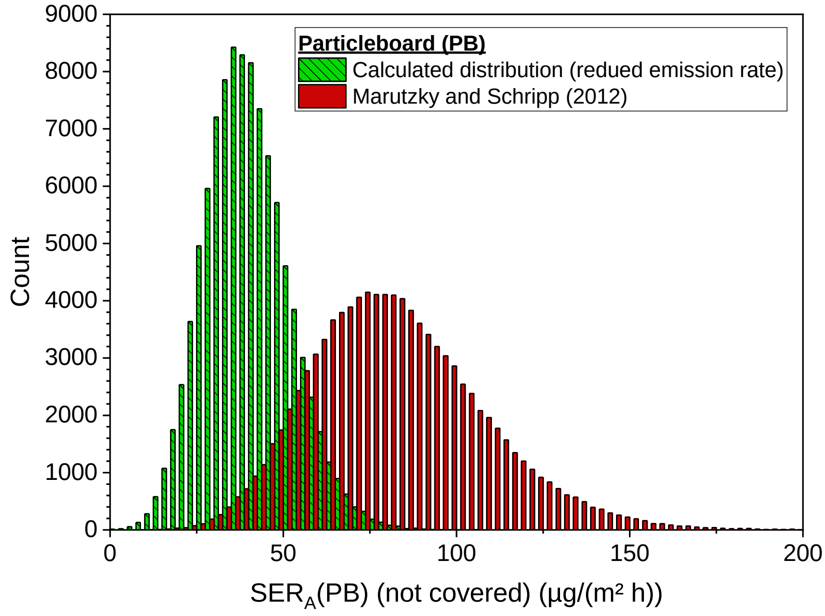

Figure 7.4: Histogram of particleboard formaldehyde emission rate distributions. Red: distribution as published by Marutzky and Schripp (2012) and used in Salthammer (2019a; b); Green: calculated distribution under the assumption of reduced emission rates (see text for details).

Since 2012, reduced emission can be assumed for wood-based materials, especially in the U.S., where strict regulations apply (see APPENDIX E). The green bars in Figure 7.4 are calculated from a Monte-Carlo approach and represent a distribution of formaldehyde emission rates from particleboard under the assumptions that the median of the formaldehyde emission rate distribution is approx. 40 µg/(m² h) and that approx. 15% of the samples exceed an emission rate of 50 µg/(m² h). This simulates a theoretical distribution, which can be expected from the revised German Chemicals Prohibition Ordinance for formaldehyde, which is 0.1 ppm < 2 x measured chamber concentration with regard to EN 717-1 (T = 23 °C, rel. humidity = 45%, AER = 1 h^{-1}, L = 1 m²/m³) or 0.1 ppm > measured chamber concentration according to EN 16516 (T = 23 °C, rel. humidity = 50%, AER = 0.5 h^{-1}, L = 1.8 m²/m³). The test conditions according to ASTM E1333 for particleboard are T = 25 °C, rel. humidity = 50%, AER = 0.5 h^{-1}, L = 0.43 m²/m³ (Hemmilä et al., 2019). Increased temperature (23 °C versus 25 °C and humidity (45% versus 50%) have an increasing effect on the release of formaldehyde from particleboard (Meyer et al., 2014). Roffael (2017) calculated that the CARB II formaldehyde limit value of 0.09 ppm (measured according to ASTM E1333 or ASTM D6007) corresponds approximately to 0.065 ppm according to DIN EN 717-1 (see APPENDIX D and E for more details on formaldehyde source/sink models and test regulations).

In order to compare different product groups, activities and processes directly, the geometric mean values (GM) of area and unit specific emission rates can be converted into Reference Room concentrations. The Reference Room is a European standard room with a volume of 30 m³, defined geometry and defined loading rates. Details are described in the European Standard EN 16516 (2017). An air exchange rate AER of 0.5 h^{-1} was used throughout. For the building products, loading rates L were taken from EN 16516 (2017). For furniture a loading rate of L = 1 m²/m³ was assumed. As far as the unit specific emission rates are concerned one single unit or item (e.g. one candle) was assumed. If it was not possible to derive a geometric mean or an emission rate, ranges are provided. The results of these calculations are shown in Figure 7.5. Please note that the scale of the ordinate is logarithmic

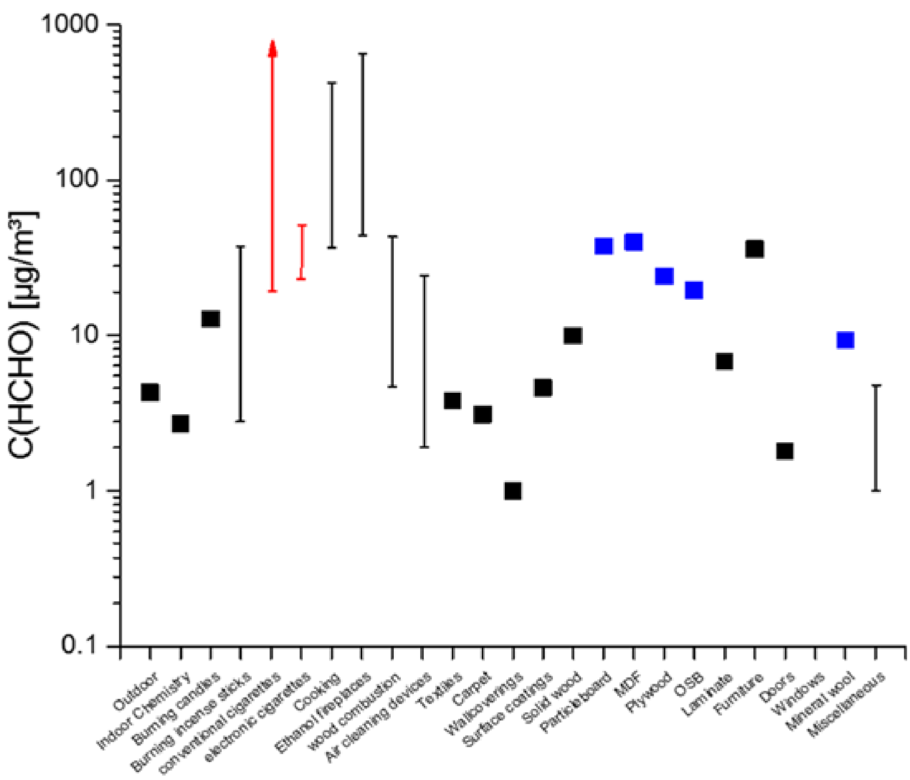

Figure 7.5: Comparison of Reference Room (30 m³) concentrations for a fixed air exchange rate (AER) of 0.5 h^{-1} for different products and materials (geometric means and ranges). For wood-based materials and mineral wool covered products are considered (■). See EN 16516 (2017) for the loading rates of building products. The loading rate for furniture is 1 m²/m³. The figure is taken from Salthammer (2019a). Please note that the data for wood-based materials are based on the study by Marutzky and Schripp (2012) and see text for further discussions.

It should, however, be pointed out that the data in Figure 7.5 do not necessarily represent indoor concentrations or even exposure scenarios, because neither living conditions nor sinks are considered. Moreover, the simple addition of Reference Room concentrations does not reflect real living scenarios. However, the comparison is highly useful for a ranking of emission sources.

In Figure 7.5 the blue squares (■) represent reference concentrations being calculated from the covered products. The red bars represent European Reference Room scenarios being calculated from conventional and electronic cigarettes.

It becomes clear that combustion processes, which are usually unregulated in the indoor environment, cause the highest concentrations by far. This is especially true for the ethanol fireplaces, which cover a range from 47 µg/m³ to 709 µg/m³. A single burning incense stick leads to concentrations between 3 µg/m³ and 39 µg/m³ and the geometric mean of a single burning candle is 12.9 µg/m³. Cooking is also a strong source of formaldehyde with concentration ranging between 37 µg/m³ and 417 µg/m³. It is estimated that wood combustion contributes with 5 µg/m³ to 48 µg/m³ in this scenario (Salthammer, 2109a, b).

On the basis of data provided by Marutzky and Schripp (2012) and the assumed covering (barrier) effect, wood-based materials and furniture range between 20 µg/m³ and 40 µg/m³. However, when taking Figure 7.4 into account it can be speculated that emission rates from covered wood-based materials are considerably lower today. Laminate floorings and covered insulation materials made from mineral wool are also at the lower site with a geometric mean of 6.8 µg/m³ and 9.3 µg/m³, respectively. With the exception of air cleaners, all other product groups and processes contribute with less than 5 µg/m³. Air cleaners work with different technologies and cause Reference Room concentrations under these conditions between 2 µg/m³ and 25 µg/m³.

It also must be considered that some of the sources are permanent, some are temporary and some are intermitting. Infiltration from outdoor air, building products and furniture certainly can be considered as permanent sources. Cooking, oven cleaning and most combustion processes belong to the group of temporary sources although in many homes wood combustion is now becoming a permanently applied technology for heating. In principle, indoor chemistry can be regarded as intermitting, as this process strongly depends on the presence of other indoor air pollutants like unsaturated hydrocarbons and the formation or infiltration of ozone. However, it is assumed that indoor chemistry permanently contributes with 2.7 µg/m³ (geometric mean) to formaldehyde indoor air pollution.

Air cleaning devices are also considered as intermitting sources. Stand-alone devices will be turned on and off by the dweller as desired. The application of inbuilt air cleaning devices, which are based on ozonization or corona discharge, in combination with mechanical ventilation is still uncommon in private living spaces. The activity of air-cleaning paint depends on sunlight or artificial light.

Finally, it should be mentioned that the estimated contribution of miscellaneous products (1 µg/m³ to 5 µg/m³) also refers to the application of a single item (e.g. one cleaning agent, one polish, etc…). Depending on the type, these sources can be classified as temporary or intermitting. Moreover, several items could be handled in parallel.

Only few representative data on emission sources are available for the United States.

The report by Bennett et al. (2011) deals with the impact of carpet and wood-based furniture on the indoor air quality in small and medium size commercial buildings. Interestingly, indoor formaldehyde concentrations were significantly higher with the presence of carpet but not sensitive to new carpet or new wood furniture.

Hult et al. (2015) investigated the impact of ventilation control and source control on formaldehyde exposure in US residences and distinguish between conventional materials and low-emitting materials. However, potential formaldehyde sources in the 9 selected residences were not studied. The authors conclude that both increased ventilation and the use of low-emitting building materials can significantly reduce residential formaldehyde exposures. This statement is certainly true as long as temporary sources are excluded from the discussion.

Chen et al. (2018) studied formaldehyde emissions from low- and high-emitting laminate flooring. The authors criticize that testing with no exposed seams and perimeter cut edges might allow a finished flooring product with a high-emitting core and high emissions after installation to meet low-emitting labelling criteria inappropriately.

The Centers for Disease Control and Prevention (CDC) (2016) and Pierce et al. (2016) studied the emission of formaldehyde from laminate flooring manufactured in China. Sheehan et al. (2018) calculated the potential exposure and cancer risk. The time-weighted-average (TWA) daily formaldehyde inhalation exposure within 899 investigated homes was estimated to be 17µg/day. Based on verified nonlinear cancer risk assessment models Sheehan et al. (2018) conclude that formaldehyde emissions from the installed Chinese-manufactured laminate flooring pose virtually no cancer risk to affected consumers.

Huangfu et al. (2019) studied the dependency of the room temperature on formaldehyde levels in four different types of housing using high time resolution monitoring. It was found that the formaldehyde sensitivity to temperature ranged from 3.0 to 4.5 ppb per °C. Specific sources were not identified but the authors discuss the possible impact of composite wood flooring and furnishings.

The effect of temperature and humidity on formaldehyde emissions from particleboard and plywood samples collected from temporary housing units was studied by Parthasarathy et al. (2011). It was found that increases of temperature and humidity contributed to an increase of formaldehyde emission factors. Maddalena et al. (2009) measured formaldehyde concentrations in four temporary housing units and determined the emission factors of potential formaldehyde sources.

The impact of cooking and cooking activities is described in the publications by Fortmann et al. (2006), Logue et al. (2014) and Militello-Hourigan and Miller (2018).

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Section summary: *A multitude of different permanent and temporary formaldehyde emission sources were identified. In addition to the typical building products, these also include chemical reactions occurring in indoor spaces, infiltrated outdoor air, combustion processes of all kinds, the operation of equipment such as air purifiers and emissions from human activities such as cooking and cleaning. It is also clear that a covering (barrier) effect should be considered for the release of wood-based materials under living conditions (see Salthammer, 2019a).*

8 Discussion

For this work, intensive literature research was carried out using scientific databases (Web of Science, SCOPUS, Google Scholar, etc.) and intelligent search algorithms. Furthermore, the bibliographies of the publications were scrutinized regarding additional relevant sources. Solely publications were evaluated which provide a representative image of formaldehyde concentrations (indoor and outdoor) and formaldehyde emission rates in accordance with the current state of technology. Many data were taken from the recent publications by Salthammer (2019a, b). No work was taken into account in which specific individual cases are reported upon. In general, publications published before 1990 were not included, unless the work therein is of fundamental interest. Geographically, the focus was placed upon the United States of America.

It was necessary to gather representative data in order to calculate realistic distributions of formaldehyde concentrations and emission rates. Unfortunately, only in few cases are emission data or emission related formaldehyde indoor data published in the necessary quantity and quality. Often these are not samples which were chosen by chance, but which are instead potential problem cases. Serving here as an example is the report from the Center for Disease Control and Prevention (CDC), which takes into account solely high-emission Chinese products which were marketed via a specific American company. The same is true for temporary housings and mobile homes. The publications by Maddalena et al. (2008) and Murphy et al. (2013) on formaldehyde in travel trailers, park models, and mobile homes refer to the hot and humid conditions of Louisiana and Mississippi. Consequently, the data in Table A2 can be used to calculate a distribution of formaldehyde concentrations over the different climatic regions in the United States (see Figure 5.2 and Table 5.1) but they are not sufficient to generally distinguish between formaldehyde concentrations in different types of indoor environments.

A frequently controversially discussed issue concerns the real air exchange in indoor spaces. In literature, extremely low values < $0.1\ h^{-1}$ are repeatedly communicated, which are interpreted as typical air exchange values. However, it is usually overlooked that these are values which are measured with closed windows and doors. These values serve standardized investigations under stationary conditions, but do not describe normal living conditions.

For the 30 m^3 European Reference Room discussed here, assuming one individual with a CO_2 emission of 20 l/h, an outside air concentration of 300 ppm and an air exchange of $0.5\ h^{-1}$, the CO_2 steady-state concentration is around 1600 ppm. In order to remain permanently at a concentration of under 2000 ppm under stationary conditions, a minimum air exchange of $0.4\ h^{-1}$ is necessary. In the case of two persons, with a CO_2 emission totaling 2 x 20 l/h, this necessary minimum air exchange increases to 0.8 h^{-1} (corresponding to 0.6 h^{-1} at 2 x 15 l/h). However, hygienically flawless conditions are not thereby achieved. In order to remain permanently in the range of 1000 ppm at a CO_2 emission of 2 x 20 l/h, a minimum air exchange of 1.8 h^{-1} would be necessary. Sustained air exchanges in the range of 0.1 h^{-1} are therefore implausible, as the inhabitants would already experience discomfort under the influence of the exhaled CO_2 and would undertake ventilation measures.

The literature data discussed in Chapter 6 appear to be realistic, but rather at the lower end of a ventilation rate recommended for residential hygiene reasons. For test house measurements in the USA, an air exchange rate of 0.35 h^{-1} is usually set. For investigations of laminates, the Center for Disease Control and Prevention (2016) assumed for Monte Carlo experiments an evenly distributed air exchange of between 0.1 h^{-1} and 1.21 h^{-1}, resulting in a median of 0.65 h^{-1}. Under the aforementioned aspects, the distribution determined from experimental air exchange data in Europe with a median of 0.52 h^{-1} (see Figure 7.4) can be accepted as reasonable and well-founded.

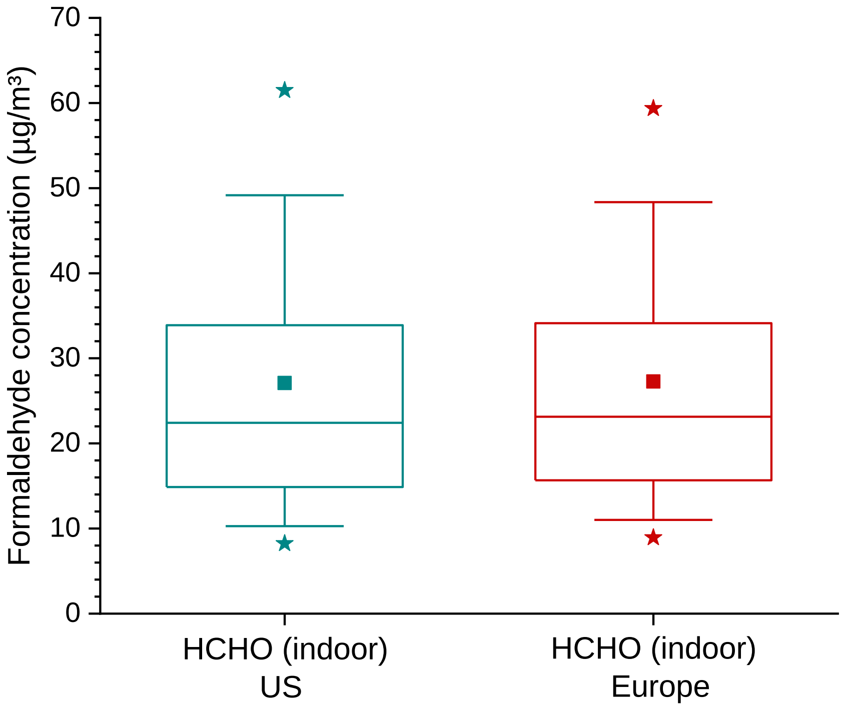

Figure 8.1: Comparison of indoor formaldehyde concentrations in the US and in Europe. The data for the US are taken from Table 5.1. The data for Europe are taken from Salthammer (2019a, b). The boxes represent 25-P, 50-P (median) and 75-P, the whiskers represent 10-P and 90-P, (■) is the arithmetic mean, (★) represent 5-P and 95-P, respectively.

Moreover, the question of residential-typical formaldehyde concentrations in the room air is a further point of permanent discussion. It is undisputed that particularly the use of formaldehyde-containing wood-based materials and other products has led in the past to high room air concentrations of formaldehyde (Gammage and Gupta, 1984; Roffael, 1993). On the other hand, there has been a significant decline in contamination of the indoor environment since the 1980s, which can be demonstrated by a comparison of the German environmental surveys from 1985/1986 and 2003 to 2006 (Salthammer et al., 2010).

A similar trend can be seen for the US when comparing studies from the 1980s (Stock and Mendez, 1985) and 1990s (Hodgson et al., 2000) with recent data. Today, under normal living conditions, the average formaldehyde concentrations in US housings lie within the range of 20 µg/m³ to 30 µg/m³.

It is also of interest to compare the distributions of indoor formaldehyde concentrations for the US and Europe. The box-whisker plots shown in Figure 8.1 demonstrate that there is almost no difference between the two distributions although different legislations, emission standards and formaldehyde indoor guidelines apply.

As far as formaldehyde indoor guidelines are concerned, the Risk Assessment Committee (RAC) of the European Chemicals Agency (ECHA) recently questioned the Derived No Effect Level (DNEL) as defined by the WHO. In 1989, the WHO established an indoor guideline value of 0.1 mg/m³, which was later re-evaluated and confirmed (World Health Organization, 2010). Several authors investigated the WHO assessment thoroughly and considered a formaldehyde guideline of 0.1 mg/m³ to be protective against both acute and chronic sensory irritation in the airways in the general population and also consider the guideline defendable for prevention of all types of cancer (Nielsen and Wolkoff, 2010; Wolkoff and Nielsen, 2010, Ausschuss für Innenraumrichtwerte, 2016). The comprehensive review by Golden (2011) provides a critical review of the air exposure limit for formaldehyde on the basis of the available literature. The author’s analysis data on sensory irritation, nasopharyngeal cancer and leukemia and comes to the conclusion that a formaldehyde indoor air limit of 0.1 ppm should protect even particularly susceptible individuals from both irritation effects and any potential cancer hazard.

In 2019, RAC questioned the validity of the WHO approach and agreed on a weight of evidence approach considering human and animal data for the relevant precursor events deriving a chronic DNEL of 0.05 mg/m³ for the inhalation route based on a study with monkeys (Rusch et al., 1983). RAC especially criticizes the small number of subjects in the study by Lang et al. (2008), which provides the basis for the WHO value. However, the Lang et al. (2008) study was evaluated and their validity was confirmed by several authors. Golden (2011) doubts the validity of regulatory guidance levels based on reports of nasal lesions in occupationally exposed workers and suggests that other co-exposures or levels of formaldehyde much higher than reported likely play contributory roles. Under consideration of original studies and critical reviews it is difficult to follow and understand the arguments of RAC.

In Chapter 7, the most important formaldehyde sources are discussed. A major problem for the future is the constant increase of formaldehyde in ambient air, as already pointed out in a publication by Salthammer (2013). In European metropolises, comparatively low concentrations are still being measured; the maximum concentration already attains, however, the 50-P value for indoor air. For comparison: for Beijing and Rio de Janeiro, formaldehyde concentrations in outdoor air of 62 ppb (77 µg/m³) and 113 ppb (140 µg/m³) have been published. Extreme formaldehyde concentrations in ambient air also occurred in California.

The data for wood-based materials were extracted from a report for the Deutsches Institut für Bautechnik (German Centre of Competence for Construction, DiBt) (Marutzky and Schripp, 2012). A comparable study has not yet been published. These data, with 30 to 48 samples respectively, were presumed to be representative of the emission behavior of wood-based panels on the European market. It is, however, unlikely that the data also represent the current situation in the United States, because severe restrictions apply for more than 10 years (see Figure 7.4 and detailed discussions in Chapter 7).

In accordance with the application of wood-based materials in indoor spaces, a shielding effect, determined through experimental investigations, can be presumed for these panels. A similar problem occurs during the determination of representative data for coated wood-based furniture. The results of regular investigations and examinations are not generally published. Case studies, which report exclusively on unusually high emissions, were not taken into consideration here for the reasons mentioned above

Furthermore, it is difficult to estimate the emission behavior of wood-based materials and furniture under real conditions over a longer period of time. Under the consistent conditions of a chamber test, the long-term emission behavior can be plausibly estimated. In contrast, the influence of temperature and humidity fluctuations can barely be calculated. In accordance with the modified Andersen equation (see APPENDIX E), the formaldehyde emission decreases by 23% when the temperature falls from 23 °C to 21 °C. Conversely, the emission increases by 14% when the relative humidity (RH) rises from 45% to 55%.

The assumptions of a reduction of 60% through aging and 25% through sink effects as assumed by Salthammer (2019a, b) for the calculation of European Reference Room concentrations would appear reasonable according to the available literature.

A largely underrated formaldehyde source can be summarized under the term "indoor chemistry". This essentially covers chemical reactions which are initiated through atmospheric species such as ozone and OH radicals (Morrison, 2010). Formaldehyde is one of the major intermediate and reaction products. The American Alfred P. Sloan Foundation has initiated its own funding program on this topic: https://sloan.org/programs/science/ chemistry-of-indoor-environments. According to Mendez et al. (2015), 2% to 11% of the formaldehyde concentration in indoor spaces can be explained through chemical reactions.

Strictly speaking, many air-purifying devices are also based on reactions which are described through "indoor chemistry". This applies in particular for techniques which work with ozone or photocatalysis. The mineralization of organic substances from the room air in water, carbon dioxide and, if appropriate, hydrogen halides is only in rare cases complete; generally, undesirable by-products are formed. These, however, are barely taken into account in investigations into the efficiency of air-purifying devices. The Clean Air Delivery Rate (CADR) (Waring et al., 2008) is generally stated, which takes into account solely the degradation of substances or particles but not, however, the possible formation of by-products.

The strongest emission sources by far are combustion processes, which are not included under "indoor chemistry". In particular, ethanol combustion is known for its high formaldehyde emissions (Sarathy et al., 2014; Schripp et al., 2014); candles and incense sticks are, however, also strong sources. The fact that, for example, non-ventilated ethanol ovens are not subject to any regulations whatsoever regarding the release of organic substances through ethanol combustion therefore presents an anachronism. The residential hygiene assessment is essentially based on the carbon dioxide value, whereby the maximum workplace concentration is used here for the evaluation instead of the indoor air guideline values. An assessment of the exposure to formaldehyde from combustion processes in indoor spaces is, however, difficult, as these are not permanent but temporary sources. If open combustion processes are active in indoor spaces, it can be expected that the WHO guideline, which is defined as a 30 min value, will be exceeded.

Furthermore, as already mentioned, temporary sources were not taken into account at all in this calculation scenario. These are often active for a short time, but usually lead to high peak concentrations. The emission rates for burning candles in Figure 7.5 refer to solely one source, but several candles are often ignited simultaneously. Ethanol ovens are only occasionally operated in households; however, comparable emissions of formaldehyde can also be expected during the operation of rechauds, which are used considerably more frequently. Taking the named aspects into account, it is surprising that ethanol ovens or air-purifying devices for use in indoor environments are not subject to stricter regulations or classified through appropriate environmental labels. In view of the discussed aspects, a further tightening of already existing regulations for building products would be barely effective. With such a measure, which simultaneously requires a high outlay, the average formaldehyde concentration could, at best, be reduced by a few percent; peak concentrations and therefore high exposures would, however, remain largely uninfluenced. This aspect is of particular importance in the case of formaldehyde, as it is a substance with a threshold effect.

Previous calculations illustrate that the Reference Room concept is only suitable for the comparison of the emission behavior of building products relative to one another (Salthammer, 2019a). It becomes clear that the simple addition or subtraction of emission rates does not in any way reflect real living scenarios. Consequently, formaldehyde concentrations determined for the European Reference Room are hardly suitable to evaluate realistic consumer exposure scenarios.

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<u>Section Summary:</u> *The contribution of permanent, intermitting and temporary emission sources to formaldehyde levels in the indoor environment was discussed. Temporary sources are often active for a short time, but they usually lead to high peak concentrations. Consequently, a further tightening of already existing regulations for building products would be barely effective. With such a measure the average formaldehyde concentration could, at best, be reduced by a few percent; peak concentrations and therefore high exposures would, however, remain largely uninfluenced. This aspect is of particular importance in the case of formaldehyde, as it is a substance with a threshold effect.*

9 Conclusion and Summary

A literature study was carried out with respect to emission and presence of formaldehyde indoors. Data concerning formaldehyde concentrations in indoor and outdoor air, as well as data on air exchange, were collected for the geographic region of the United States of America.

A multitude of different permanent and temporary formaldehyde emission sources were identified. In addition to the typical building products, these also include chemical reactions occurring in indoor spaces, infiltrated outdoor air, combustion processes, the operation of equipment such as air purifiers and emissions from human activities such as cooking and cleaning.

The various emission sources were compared with one another under standardized conditions. It was thereby necessary to bear in mind that, for example, the emission tests for raw wood-based materials and mineral wool do not take place under realistic conditions, as these products are not applied open (i.e. without coatings/coverings) in indoor areas. It could be demonstrated that coatings and coverings drastically reduce the release of formaldehyde into the room air. Overall, there was a distinction between permanent and temporary sources. It became clear here that peak concentrations are often caused by temporary sources whose release potential for formaldehyde is not subject to any regulations whatsoever.

In a previous study (Salthammer (2019) Building and Environment 150, 219-232) it became clear that the Reference Room concept in general greatly overestimates the formaldehyde concentrations in indoor areas when diverse sources are simply added together. When the aging and sink effects – which can be estimated from diverse works – were taken into account, an unrealistic picture also remained. The calculation results make clear that the Reference Room is neither suitable for the realistic comparison of the emission behavior of building products in an application-oriented manner, nor for a health related evaluation of indoor exposure to formaldehyde. Moreover, EN 16516 neglects many important factors like sinks, ageing and diffusion effects. However, as mentioned above, the Reference Room is very suitable for the comparison of the emission behavior of building products relative to one another.

The evaluation of current literature resulted in typical concentrations of formaldehyde within the range of 20 µg/m³ to 30 µg/m³ for US buildings under residential-typical conditions. The assumption of a minimum average air exchange of 0.5 h^{-1}, also under residential-typical conditions, is reasonable.

In view of the discussed aspects, as well as taking into account outdoor air conditions and diverse secondary sources, the potential problem of exposure to high formaldehyde concentrations in indoor areas can therefore not be solved through the further tightening of already existing regulations, in particular because peak concentrations and therefore high exposures would remain largely uninfluenced. This aspect is of particular importance in the case of formaldehyde, as it is a substance with a threshold effect. Therefore, in case of any concerns by regulators on formaldehyde exposure in indoor air, the most appropriate risk management option would be to address the peak concentrations originating from temporary sources.

APPENDIX A

Table A1: Formaldehyde concentrations in ambient air in the United States.

Type	n	GM	Min	Max	25-P	50-P	75-P	90-P	95-P	Reference
		µg/m³								
Los Angeles, Elisabeth, Houston (RIOPA)	353					6.42			9.95	(Liu et al., 2006)
FEMA temporary housing units	2		2.06	3.34						(Maddalena et al., 2009)
Small + Medium Size Commercial Buildings	40		0.15	8.31		2.59			6.65	(Bennett et al., 2011)
Modeling of regional sources			<0.1	12.4						(Luecken et al., 2012)
US retail stores (TX, PA) [1)]				4.2						(Nirlo et al., 2014)
Senior apartments before retrofit (AZ) [1)]						6.1				(Frey et al., 2015)
Senior apartments after retrofit (AZ) [1)]						7.5				(Frey et al., 2015)
Senior apartments 1 y after retrofit`(AZ) [1)]						4.7				(Frey et al., 2015)
High performance home (CA)	24	-	<LOD	6.5	1.1	2.0	3.0	-		(Less et al., 2015)
Green vs. non-green homes (OH) [2)]					5.0	6.6	7.4			(Coombs et al., 2016)
Healthy Homes Study (CA) [1)]	179	2.5						4.2	4.8	(Mullen et al., 2016)
Childhood education facilities (CA)	19	2.4	1.5	4.0	1.9	2.3	3.1	3.9		(Bradman et al., 2017)
Health efficient new gas homes (CA) [1)]	68					3.5				(Chan et al., 2019)

[1)] Converted from ppb to µg/m³ (1 ppb = 1.24 µg/m³)

[2)] Converted from ppm to µg/m³ (1 ppm = 1240 µg/m³)

APPENDIX B

<u>Table A2:</u> Formaldehyde indoor concentrations in US housings and facilities.

Type	n	GM	Min	Max	25-P	50-P	75-P	90-P	95-P	Reference
		µg/m³								
Los Angeles, Elisabeth, Houston (RIOPA)	353			136		20.1			32.5	(Liu et al., 2006)
Stores (non-residential) (MA)	138	19.6	1.58	90.6						(Loh et al., 2006)
FEMA temporary housing units (LA, MS) [1)]	519	95	4	732						(Maddalena et al., 2008)
New homes (CA)	105	-	4.8	47.0	-	36	-		-	(Offermann, 2009)
Medium-sized commercial buildings (CA)	40	16.4	1.41	102						(Wu et al., 2011)
Nail salons (UT) [2)]	14	19.8	10.8	39.7	15.8	19.8	26.0			(Alaves et al., 2013)
US retail stores (TX, PA) [1)]	14		5.7	83						(Nirlo et al., 2014)
Apartments (conventional) (MA)	41	9.4								(Colton et al., 2014)
Apartments (green) (MA)	18	12.1								(Colton et al., 2014)
Senior apartments before retrofit (AZ) [1)]	72					47				(Frey et al., 2015)
Senior apartments after retrofit (AZ) [1)]	54					53				(Frey et al., 2015)
Senior apartments 1 y after retrofit`(AZ) [1)]	55					32				(Frey et al., 2015)
US retail stores – grocery (CA)	7					9.4		15.8		(Chan et al., 2015)
US retail stores – furniture/hardware (CA)	7					26.4		54.4		(Chan et al., 2015)
US retail stores – apparel (CA)	5					15.7		27.8		(Chan et al., 2015)
Bedroom (high performance home, CA)	24	-	111.7	33.2	15.5	17.5	29.7		-	(Less et al., 2015)
Kitchen (high performance home, CA)	24	-	8.1	48.8	13.8	20.1	27.2		-	(Less et al., 2015)
Green vs. non-green homes (OH) [2)]	96				17.4	24.8	40.1			(Coombs et al., 2016)

Homes (pre-weatherization) (NC)	52					17				(Doll et al., 2016)
Homes (post-weatherization) (NC)	52					15				(Doll et al., 2016)
Portable classrooms [2)]	9		9	42		14				(Ribeiro et al., 2016)
Traditional classrooms [2)]	3		15	36		20				(Ribeiro et al., 2016)
Healthy Homes Study, bedroom (CA) [1)]	340	19						37	45	(Mullen et al., 2016)
Healthy Homes Study, kitchen (CA) [1)]	340	19						36	42	(Mullen et al., 2016)
Childhood education facilities (CA)	40	15.9	0.7	48.8	10.6	17.8	25.0	33.2		(Bradman et al., 2017)
Homes pre-weatherization (IL, IN) [1)]	71	35								(Francisco et al., 2017)
Homes post-weatherization (IL, IN) [1)]	71	29								(Francisco et al., 2017)
School buildings (OH, IL, IN, MI)	144			32		6.0				(Zhong et al., 2017)
Homes (pre-renovation) (MA)	10	16	4.4	27		17			26	(Dodson et al., 2017)
Homes (post-renovation) (MA)	10	10	1.5	28		11			26	(Dodson et al., 2017)
Homes pre-weatherization (USA) [1)]	514		<1	89		19				(Pigg et al., 2018)
School classrooms whole year (Midwest) [1)]	220		<10	40		<10				(Deng and Lau, 2019)
Nail salons (8 h average) (CO)	6		7.29	20.6						(Lamplugh et al., 2019)
Health efficient new gas homes (CA)	68					22.6			38.6	(Chan et al., 2019)

[1)] Converted from ppb to µg/m³ (1 ppb = 1.24 µg/m³)

[2)] Converted from ppm to µg/m³ (1 ppm = 1240 µg/m³)

APPENDIX C

Table A3: Air exchange rates (AER) in US housings and facilities.

Type	Ventilation/Comment	n	GM	Min	Max	50-P	Reference
			h^{-1}				
New homes (CA)	Outdoor AER (24 h average)	106	-	0.09	5.3	0.26	(Offermann, 2009)
Los Angeles (CA) (RIOPA)	(48 h average) see paper for details	182				0.87	(Yamamoto et al., 2010)
Houston (TX) (RIOPA)	(48 h average) see paper for details	164				0.47	(Yamamoto et al., 2010)
Elisabeth (NJ) (RIOPA)	(48 h average) see paper for details	163				0.88	(Yamamoto et al., 2010)
Commercial buildings (CA) 1)	Tracer gas method	40		0.3	9.07	1.04	(Bennett et al., 2011)
Commercial buildings (CA) 1)	Steady state method	40		0.12	6.26	0.73	(Bennett et al., 2011)
New homes, low ventilation setting (CA)	Constant emitters	9		0.05	0.81	0.17	(Willem et al., 2013)
New homes, med. ventilation setting (CA)	Constant emitters	9		0.11	0.85	0.31	(Willem et al., 2013)
New homes, high ventilation setting (CA)	Constant emitters	9		0.25	1.45	0.65	(Willem et al., 2013)
US retail stores – grocery (CA)	Tracer gas method	8		0.49	1.72		(Chan et al., 2015)
US retail stores – furniture/hardware (CA)	Tracer gas method	8		0.36	3.06		(Chan et al., 2015)
US retail stores – apparel (CA)	Tracer gas method	5		0.42	1.98		(Chan et al., 2015)
US residences (MI)	Constant emitters	170				0.35	(Du et al., 2015)
US residences, basement (MI)	Constant emitters	170				1.15	(Du et al., 2015)
High performance home (CA)	Natural (6 d average)	8	-	-	-	0.32	(Less et al., 2015)
High performance home (CA)	Mechanical (6 d average)	8	-	-	-	0.30	(Less et al., 2015)
Childhood education facilities (CA)	see paper for details	40	1.53	0.28	5.63	1.43	(Bradman et al., 2017)

1) Parallel measurements in the same room using different techniques.

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APPENDIX D

Formaldehyde emission and sink models

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APPENDIX E

Formaldehyde emission regulations

Standard	Chamber (m^3)	T (°C)	RH (%)	AER (h^{-1})	L (m^2/m^3)	Q/A ($m^3/(h\ m^2)$)	Material
DIN EN 717-1	0.225, 1, >12	23	45	1.0	1.0		Wood-based panels
EN 16516	> 0.02	23	50	0.5	1.8		Wood-based panels
ASTM E1333	≥ 22	25	50		0.95 0.43 0.26 0.13		Hardwood Plywood Wall Paneling Particleboard Flooring Materials Industrial Particleboard Panels Industrial Hardwood Plywood Panels Medium Density Fiberboard (MDF) Low Density Particleboard Door Core Grade
ASTM D6007	0.002 – 1	25	50			0.526 1.172 1.905 3.811	Hardwood Plywood Wall Paneling Particleboard Flooring Materials Industrial Particleboard Panels Industrial Hardwood Plywood Panels Medium Density Fiberboard (MDF) Particleboard Door Core

L = loading rate (m^2/m^3); Q = chamber air flow (m^3/h); A = sample surface (m^2)

Relationship between DIN EN 717-1 and ASTM E1333 according to Equation (A1), see also Meyer et al. (2014).

$$C = 0.0366 \cdot C_{st} \cdot (T - 13.15) \cdot (e^{0.0403 \cdot RH} + 2.073) \cdot \frac{1}{1 + 2.07 \cdot \frac{AER}{L}} \qquad \text{(A1)}$$

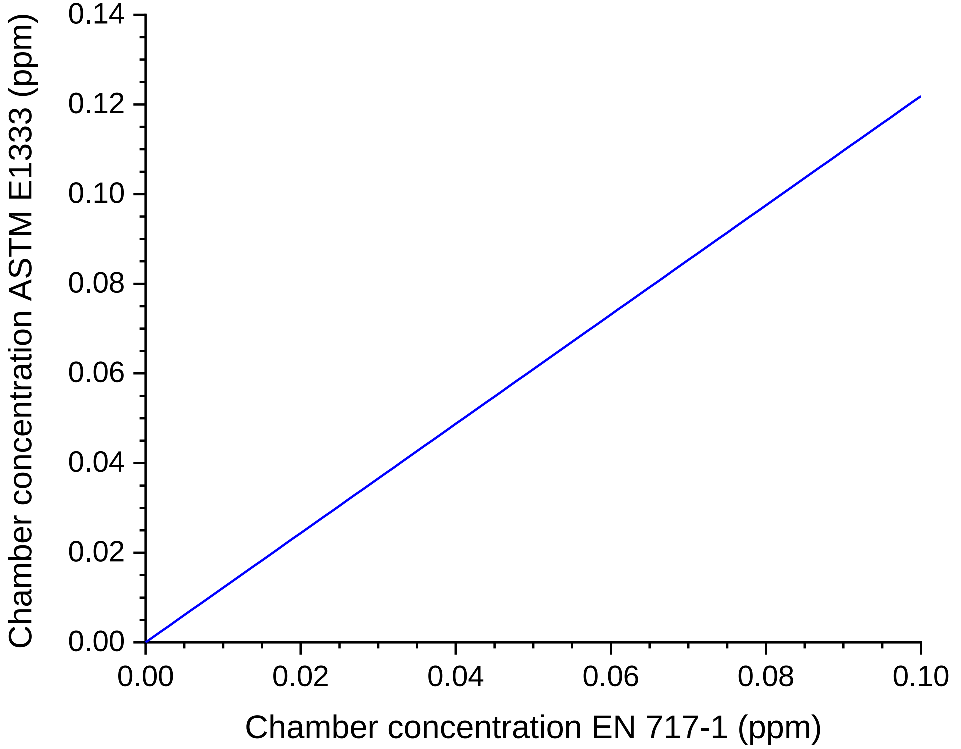
C_{st}: steady-state concentration in the DIN EN 717-1 test chamber (T = 23 °C, RH = 45%, AER = 1.0 h^{-1}, L = 1.0 m^2/m^3)

T: 25 °C (ASTM E1333);

RH: 50% (ASTM E1333);

AER: 0.5 h^{-1} 50% (ASTM E1333);

L: 0.43 m^2/m^3 (ASTM E1333).



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