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Formaldehyde emission monitoring from a variety of solid wood, plywood, blockboard and flooring products manufactured for building and furnishing materials

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ABSTRACT

The measurements of formaldehyde emission (FE) from solid wood, plywood, flooring and blockboard used for building and furnishing materials were obtained using the European small-scale chamber (EN 717-1) and gas analysis (EN 717-2) methods to identify the major sources of formaldehyde among construction and wood products in the Czech Republic. The differences in the FE values reported for various wood products were a function of their structural differences. These results showed that the wood species, plywood type and thickness significantly affected the FE measured by EN 717-2 ($P < 0.001$). The FE values from solid wood ranged between 0.0068 and 0.0036 ppm and 0.084–0.014 mg/m^2 h. The initial FE ranged from 0.006 mg/m^3 for engineered flooring with polyvinyl acetate (PVAc) to 0.048 mg/m^3 for painted birch blockboard. Furthermore, the FE dropped noticeably by the end of the measuring period, ranging between 0.006 mg/m^3 for engineered flooring with PVAc and 0.037 mg/m^3 for painted beech blockboard. Additionally, the initial FE was higher for the painted blockboard (0.035–0.048 mg/m^3) than for the uncoated boards (0.022–0.032 mg/m^3). In the first week after manufacturing, the FE was high, but the decrease in FE was noticeable at the two-week measurement for all of the materials, especially for the painted blockboards.

1. Introduction

The adhesives used to produce plywood contain formaldehyde, which presents a serious problem in mobile homes because they are typically constructed from plywood. Additionally, formaldehyde is a potential human carcinogen and, because of its high danger level, is classified differently than most other pollutants. It is normally present at low levels, usually less than 0.03 ppm, in both indoor and outdoor air, while levels above 0.1 ppm can cause acute health problems [1–4].

The International Agency for Research on Cancer (IARC), a division of the World Health Organization, has recently established that formaldehyde is undetectable by smell at concentrations of less than 0.1 ppm. At concentrations between 0.1 ppm and 0.5 ppm, formaldehyde is detectable by smell, with some sensitive individuals experiencing slight irritation to the eyes, nose and throat. At levels from 0.5 to 1.0 ppm, formaldehyde produces irritation of the eyes, nose and throat in most people, while at concentrations above 1.0 ppm, exposure to formaldehyde produces extreme discomfort [5].

Indoor air quality is currently determined by measuring and monitoring the formaldehyde emissions (FEs) from building materials, including plywood, solid wood, wall coverings, coating products and flooring panels. These materials are used widely for the construction, decoration and furnishing of homes, offices, schools, as well as other non-industrial work places, many of which have high FE due to the use of such things as solvent-borne paints and adhesives.

The widespread use of urea-formaldehyde (UF) adhesives with products used for interiors and their presence within the home exacerbates this problem [6]. Phenol-formaldehyde (PF) resin is used to manufacture plywood for exterior applications because of its excellent water resistance. PF tends to be more chemically stable and less susceptible to hydrolysis than UF or melamine-urea formaldehyde resin (MUF) and is considered waterproof, while UF is not [7,8]. In both the US and Europe, products bonded with PF adhesives are classified as non-emitting and are exempt from FE regulations.

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Plywood used for interior applications (PLY) is suitable for making furniture, joinery production and interior furnishings. This product is also used as an exterior product (PLYs) that is suitable for moist and wet use, carrying and non-carrying construction elements. Moreover, among the different types of wood flooring, the solid wood and engineered floorings, which consist of PLY with a thin veneer, are bonded to the face of PLY using either UF or MUF resins as hot press adhesives. Additionally, PVAc has been added as a replacement for melamine-based resin systems to reduce the FEs from the adhesives used between plywood and fancy veneers [9,10].

Blockboard is a softwood strip-core joinery board with a veneer facing made from a central core of 25-mm wide strips with vertically arranged growth rings that are individually interlocked. Blockboard can be used to make shelves, doors, panelling and partitions, and the surface can be painted on both sides to equalise the surface tension. Blockboard is not covered by the Airborne Toxic Control Measure (ATCM) because it is not included in the definition of hardwood plywood (HWPW). The ATCM approved by the California Air Resources Board (CARB) in April 2007 applies to panel manufacturers, third party certifiers (TPC), distributors, importers, fabricators, and retailers of HWPW, particleboard, MDF, and finished goods containing those products that are sold in or delivered to California [2,3]. However, the FE limits from blockboard as well as other wood-based panels can be calculated from the European emission regulations from the standards (E1 $\leq$ 0.1 ppm or 0.124 mg/m^3) or (E1 $\leq$ 3.5 mg/m^2 h) as measured by the chamber (EN 717-1) and gas analysis (EN 717-2) methods, respectively. In 2008, CARB passed a regulation applying to any interior composite wood products sold in California. This law requires a low FE and a TPC with much tighter quality controls than have typically been employed in the industry [2,3].

Although regulations have tightened, questions arise about the FE from the wood itself [11,12]. In many studies, the FE from wood was insignificant compared to the FE emitted from the UF moiety of traditional composite wood panels. However, within the new CARB limits, the wood-derived FE was a more significant part of the total emitted formaldehyde concentration [13].

The FE of wood increases at elevated temperatures and prolonged heating times [14]. Additionally, the FE is elevated, even in the absence of resin in the wood [15]. Certain reports have mentioned that the FE is produced from the wood during hot-pressing of composite panels, and it is generally accepted that the FE from wood is an insignificant contributor to the total measurable level of FE in a composite wood product [13].

However, there are many factors influencing the FE from wood products and flooring materials, including the board thickness, resin type, floor heating, added substrates in the resin, raw materials and manufacturing techniques of the panels, the drying and hot-pressing techniques, the surface finishing materials and the moisture content [9,10,16–21]. The FE values of poplar and spruce plywood panels decrease with increasing veneer moisture content for UF and MUF glue types [22]. During hot pressing, acetyl groups are released from wood, forming acetic acid, which is considered to be a formaldehyde scavenger. Beech wood contains more acetic acid than pine [23].

A variety of test methods have been developed for determining the initial emittable formaldehyde concentrations from wood-based products. Each method has its own special set of board conditioning and test conditions that possess both positive and negative attributes. The large chamber tests, such as the European large chamber (EN 717-1), are expensive, time-consuming and need a large quantity of the sample. Consequently, it is impractical to use this technique for quality assurance in commercial production. Other methods, such as gas analysis (EN 717-2), using a perforator (EN 120) or a desiccator (JIS A 1460), are widely used in quality production control for factories and save time compared with the referenced methods (chambers), which need 7–28 days. Risholm-Sundman et al. [24] compared the results from many standards and reported that the correlation between the desiccator JIS A 1460 and the chamber and perforator methods were not convincing. However, the correlations among the test methods were convincing when using the same type of board with the same thickness [17]. Newer and simpler methods can rapidly and simultaneously measure the FE while saving time and have been validated using the characteristic parameters determined in the closed chamber experiment [25–27].

The objectives of this study were (1) to measure and monitor the emission of formaldehyde from six solid woods (beech, spruce, pine, oak, birch and poplar) using the environmental small-scale chamber and gas analysis methods; (2) to investigate the effect of the choice of wood species as well as different manufacturing techniques (UF or PF resin, board thickness) on the FE; (3) to determine the FE from the manufactured flooring and blockboard made using some combination from the previous solid wood, veneers and plywood; and (4) to show the correlation between the values of FE measured with the environmental small-scale chamber and gas analysis methods.

2. Experimental

2.1. Wood materials

The forest land area in the Czech Republic reported in Table 1 [28] consists mainly of Norway spruce (*Picea abies* L.), Scots pine (*Pinus sylvesrtis* L.), European beech (*Fagus sylvatica* L.), birch (*Betula pendula* Roth), European oak (*Quercus robur* L.) and hybrid poplar (*Populus nigra* × *P. maximowiczii*). These wood types are extensively used to produce wood products that are widely used for manufacturing furniture. Thus, monitoring and measuring the FE from these products is very important to control the human health hazards due to formaldehyde.Twelve samples from each of the previous freshly felled wood species were used to measure the FE (6 samples for EN 717-1 and 6 samples for EN 717-2). The wood species were stored separately under controlled conditions at a temperature of 20 °C and 65% relative humidity (RH) until testing. After a defined air-drying period, the formaldehyde measurements were carried out immediately after sampling. The pine strips (20–25 mm wide) were manufactured for further use.

2.2. Plywood manufacturing

Plywood panels were classified into two groups:

Table 1
Species composition in ha and % of the total timberland area.[a]

Species	Area of forest stands (ha)	Area of forest stands (%)
Norway spruce	1,352,820	52.16
Fir	25,274	0.97
Pine	437,466	16.86
Larch	100,853	3.89
Other conifers	6212	0.24
Conifers	1,922,625	74.12
Oak	176,397	6.8
Beech	187,027	7.21
Birch	72,895	2.81
Other broadleaves	207,408	8.01
Broadleaves	643,728	24.83
Total without unstocked areas	2,566,353	98.95

[a] Data from the report on the state of forests and forestry in the Czech Republic [28].

Table 2
Properties and composition of urea-formaldehyde adhesive resin used for manufacturing of plywood panels for interior uses.

Parameter	UF
Solid resin content (%)	66 ± 2
Viscosity (mPa s at 20 °C)	400–650
pH at 20 °C	7.5–8.7
Density	cca 1300 kg/m^3
Hardener (NH_4Cl)[a]	15% water solution
Wetting agent	Carboxymethyl cellulose (CMC)—active substance 63% by weight
Retarder of chemical reaction	Hexamethylenetetramine ($C_6H_{12}N_4$)—10% to the hardener solution
Technical flour	20–23 kg/120 kg UF
F/U molar ratio	<1.0 (<1.0:1)
Free formaldehyde	< 0.2%

[a] Percent based on solid content of UF resin.

1. Softwood plywood (SWPW), manufactured from spruce and pine veneers and bonded with PF and UF resins.
2. Hardwood plywood (HWPW), manufactured from beech, birch and poplar and bonded with PF and UF resins. The oak wood was used to produce fancy or decorative veneers for manufacturing the engineered flooring panels.

HWPW was used mostly in protected applications using the traditional UF as an adhesive resin, while the plywoods in this study were manufactured using either UF or PF.

Fresh veneer sheets (500 mm × 500 mm × 1.2–4.0 mm) were milled from logs with 40 cm diameters from spruce, pine, beech, birch and poplar. After manufacturing, the veneers were air-dried and conditioned in a climate chamber to 5–7% moisture content (MC). Plywood panels (PLY and PLYs) with 8 mm and 22 mm thicknesses were manufactured from veneers with different MCs. Cold pre-pressing (specific pressure 1.6–1.8 MPa) for all the thicknesses of softwood (spruce and pine) and hardwood (beech, poplar and birch) was used.

2.3. Adhesives system

The adhesive mixture was spread on the single surface of veneers using a glueing machine. UF adhesive was used to produce PLY panels, and the technical characteristics of this adhesive are presented in Table 2. Boards (2500 mm × 1250 mm × 8–22 mm) were produced from the wood species under investigation using a pressing machine-HBR 6/15 (2600 mm × 1700 mm) (Královopolská strojírna, Brno, Czech Republic). The pressing conditions and the properties of the veneers are given in Table 3. The PF adhesive resin was used to manufacture the PLYs panels. The properties and compositions of the PF resin are presented in Table 4. Boards (2440 mm × 1220 mm × 8–22 mm) were produced using a Siempelkamp pressing machine (2600 mm × 1350 mm) (1971, Germany). The pressing conditions are presented in Table 5. At least 6 panels were manufactured for each thickness and wood species.

Table 3
The pressing conditions for the plywood panels bonded with urea-formaldehyde adhesive resin.

Parameter	Value
Specific pressure	0.9–1.5 MPa[a]
Pressing temperature	100–110 °C
Pressing time	60 s/1 mm[b]
Veneer moisture	Face: 5–9%, core 5–12%
Veneer thickness	1.2–4.0 mm
Adhesive spread	180–250 g/m^2 [c]

[a] 0.9–1.4 MPa for softwood plywood, 1.5 MPa for hardwood plywood.
[b] Basic curing time for UF adhesive is 3 min and for the heating of 1 mm veneer thickness, it is 1 min. The heating of aluminium base, which is inserted to the press with stacked set, adds 1 min.
[c] It depends on several variable parameters—e.g., nominal veneer thickness and number of layers. With the increasing thickness of veneers the adhesive spread also increases. It also affects the veneer quality and type of plywood.

Table 4
Properties and composition of phenol-formaldehyde adhesive resin used for plywood.

Parameter	PF
Solid resin content (%)	46.5–49.5
Viscosity (mPa s at 20 °C)	250–1050
pH at 20 °C	min. 11.5
Density	1210–1250 kg/m^3
Foaming agent[a]	Oxyethylene castor oil and mixture of non-ionic tensides
Free formaldehyde	max 0.1%

[a] PF resin + foaming agent (PF resin/foaming agent—100:1) with density (PF resin + foaming agent): 750–850 kg/m^3.

2.4. Flooring and blockboard panels

Flooring and blockboard panels were produced from solid wood, veneers, pine strips and plywood in combinations to create more useable, high quality products at a good price with a low FE and good resistance to water, abrasion, and UV. We used the same source of wood furnish to eliminate the possibility that the high yield of FE is due to the raw material.

The FE measurements for the flooring and blockboard panels bonded either with PF or MUF resins or replaced with PVAc were performed by the EN 717-1 method.

The types of flooring and blockboard were produced as follows:

1. Solid wood flooring (3-layer floor) was manufactured using an upper layer of 3.8 mm solid beech wood (veneer), a middle layer of 8 mm spruce wood as a core, and a bottom layer of 2 mm spruce veneer. A UV-curable layer (0.3 mm) made from a natural oil was used as a surface finishing, and PF was used as the resin.
2. Engineered flooring was produced as follows: 3.8-mm-thick fancy oak veneer was glued to an 8-mm-thick spruce plywood sheet using MUF resin and was pressed at approximately 190 °C. A UV-curable layer was coated on the fancy veneer. The manufacturing steps were divided into three steps: spruce plywood only (veneers were bonded with PF resin), fancy veneer (oak) bonded on plywood using MUF resin [16] and the UV-curable layer coated on fancy veneer with plywood. Additionally, the same engineered flooring was manufactured using PVAc instead of MUF. Engineered flooring originally manufactured with UF resin during the pressing process is the cheapest option. However, the highest levels of formaldehyde off-gassing occurred in these materials. The industry changed

Table 5
The pressing conditions of plywood bonded with phenol-formaldehyde adhesive resin.

Parameter	Value
Specific pressure	0.9–1.5 MPa[a]
Pressing temperature	118–124 °C
Pressing time	60 s/1 mm[b]
Veneer moisture	5 ± 2%
Veneer thickness	<3.2 mm
Adhesive spread	150–200 g/m^2 [c]

For a and c, see Table 3.
[b] Basic curing time for PF adhesive is 3 min and for the heating of 1 mm veneer thickness is 1 min.

its standards to overcome this issue by introducing PF resin, as well as No Added Formaldehyde resins (NAF).

3. Blockboard was made from 20 to 25-mm-wide core pine strips. The strips were placed edge to edge and were sandwiched between hardwood veneers (birch, beech and poplar) on both sides, which were then glued with MUF under high pressure. Subsequently, the blockboards were classified into uncoated and painted boards. An oil-based paint was applied to the surface of blockboards using a high volume low pressure (HVLP) sprayer. Examples of some of the wood products used in this study are presented in Fig. 1.

Fig. 1. Examples of some of the wood products used in this study. Samples (from left to right, up to down): 3-layer solid wood flooring (upper layer: 0.3 mm UV-curable layer, 3.8 mm oak veneer; core: 8 mm spruce wood; bottom layer: 2 mm spruce veneer); engineered flooring: 0.3 mm UV-curable layer, 3.8 mm fancy oak veneer and 8 mm plywood; 3-layer solid wood flooring: bottom side.

2.5. Estimation of formaldehyde emission

The FE measurements were conducted in the experimental laboratory of the Timber Research and Development Institute (Prague, Czech Republic). Six replicates from each of the solid woods, floorings and blockboards were used to measure the FE by the small-scale chamber test (0.225 m^3) according to the EN 717-1 standard. Additionally, six separate replicates from each solid wood were measured by the gas analysis method (EN 717-2). The plywood panels bonded with UF and PF were conditioned for 4 weeks at 20 °C and 65% RH before measuring the FE with EN 717-2.

2.5.1. Chamber method (EN 717-1)

Briefly, in the quality assurance or referenced method (EN 717-1) (Fig. 2A), two test pieces (0.2 m × 0.28 m × board thickness) with a total area of 0.225 m^2 were used for measuring the FE. The samples (solid wood, flooring and blockboard) were not

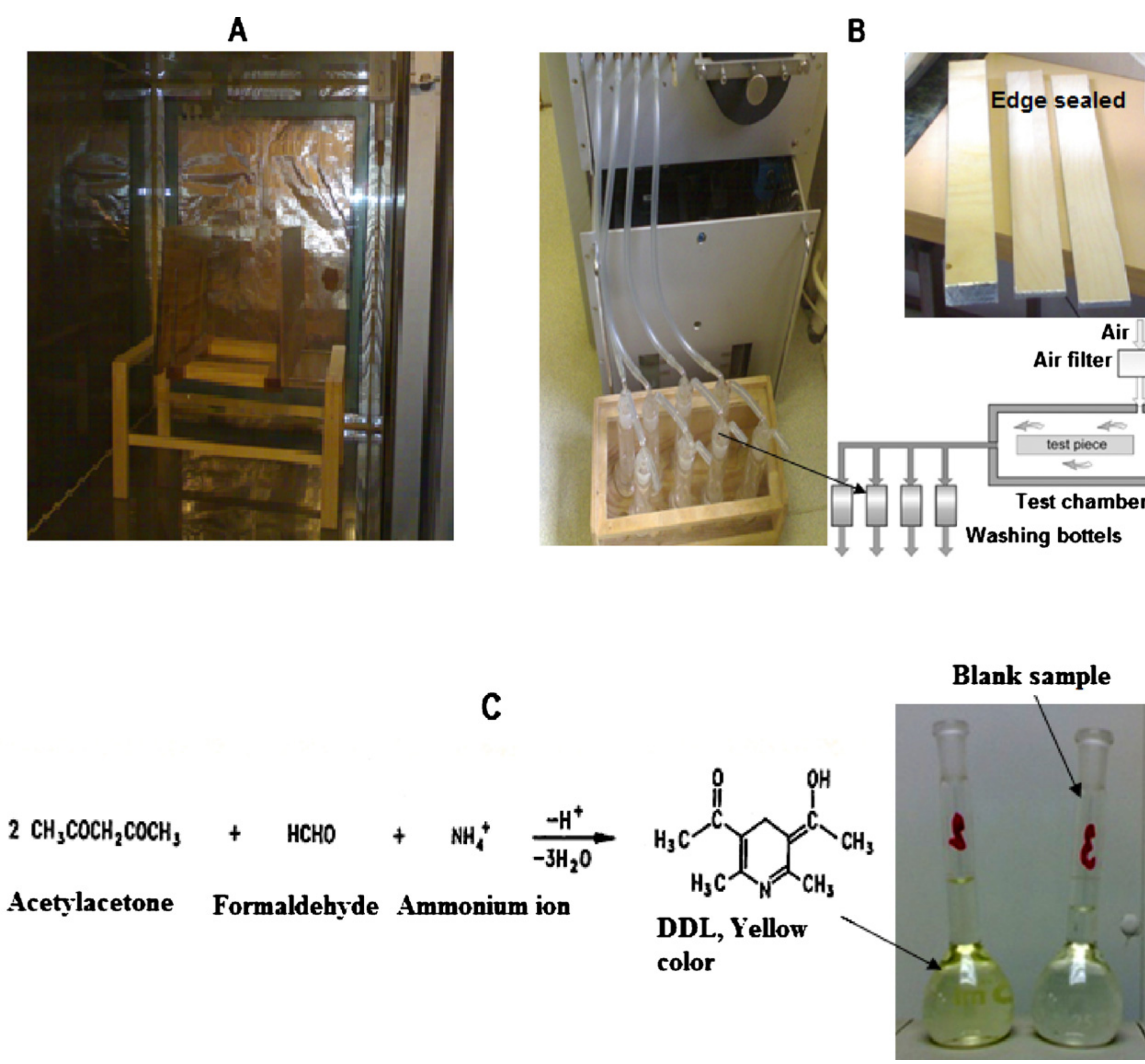

Fig. 2. The small-scale chamber (A), gas analysis (B) and the reaction scheme of the acetylacetone methods (C).

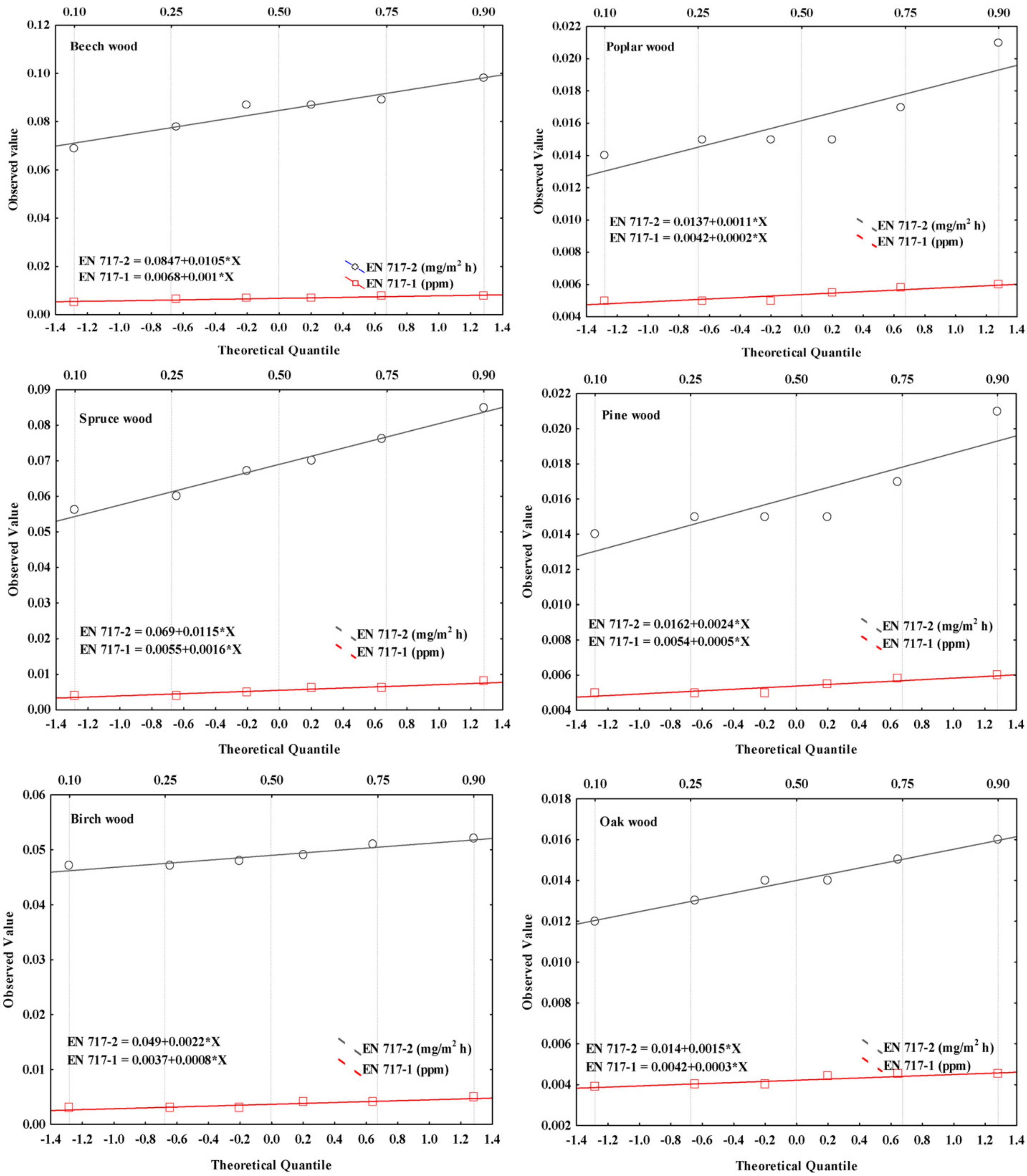

Fig. 3. Normal Q–Q plot distribution of formaldehyde emission from different types of solid wood measured by EN 717-1 and EN 717-2.

conditioned before the test. The loading factor was 1 m^2/m^3, which means the edges were partly sealed (1.5 m open edge/m^2). The edges were sealed with aluminium foil to obtain a constant ratio of the length (U) of the open (unsealed) edges to the surface area (A), so that $U/A = 1.5\ m/m^2$. The temperature and RH were $23 \pm 0.5\ ^\circ C$ and $45 \pm 3\%$, respectively. The formaldehyde emitted from the test pieces mixes with the air in the chamber, and a specified volume of air is drawn from the chamber twice a day. Samples are periodically measured until the formaldehyde concentration in the chamber has reached a steady-state. The values after 2–4 weeks of testing are given as the steady-state emissions values. The E1-emission class ≤0.1 ppm (0.124 mg/m^3) and E2 >0.1 ppm were used as a standard limits measured by EN 717-1 method.

2.5.2. Gas analysis method (EN 717-2)

The gas analysis method is used for factory production control. A test piece of 400 mm × 50 mm × board thickness is placed in a 4 L chamber with controlled temperature (60 ± 0.5 °C), relative humidity (RH ≤ 3%), airflow (60 ± 3 L/h) and pressure. Air is continuously passed through the chamber at 1 L/min over the test piece whose edge is sealed with self-adhesive aluminium tape before testing (Fig. 2B). The determination was made in duplicate using two different pieces and the actual formaldehyde value was the average of two pieces after 4 h. The E1-emission class ≤3.5 mg/m^2 h and E2 >3.5 ≤ 8 mg/m^2 h were used as standard limits measured by EN 717-2 method.

The emitted formaldehyde was absorbed in water from both methods and was determined photometrically by the acetylacetone (AcAc) method. The determination is based on the Hantzsch reaction [29] in which aqueous formaldehyde reacts with ammonium ions from ammonium acetate and AcAc to yield 3,5-diacetyl-1,4-dihydrolutidine (DDL), which has a characteristic yellow colour (Fig. 2C). The reaction is highly specific to formaldehyde. The formaldehyde concentration was determined by reading the colour of the DDL at 412 nm using a spectrophotometer.

2.6. Statistical analysis

The test for the normality of the formaldehyde values (mg/m^3 and mg/m^2 h) from solid wood was performed using a normal Quantile–Quantile plot (Q–Q plot). The FE values measured by EN 717-1 and EN 717-2 for solid wood and by EN 717-2 for plywood were statistically analysed using the General Linear Models (GLM) procedure in SAS version 8.2 [30] in a completely randomised design to test the differences among factors and levels. The comparison among the least square means (LS Means) with 95% confidence intervals (95% CI) was performed, using a least significant difference (LSD) at a 0.05 level of probability. Linear correlations were applied to EN 717-1 versus EN 717-2.

3. Results and discussion

3.1. Formaldehyde emissions from solid woods

The Q–Q plot was used to compare a sample of data on the vertical axis to a statistical population on the horizontal axis. Plots were measured based on their agreement with a fitted distribution of the observed data and were sometimes used to fit a distribution to the data. The Q–Q plots shown in Fig. 3 indicate that the distribution of the FE values measured by EN 717-1 had a higher homogeneity than those measured by EN 717-2. Fig. 3 shows that the points of the FE values measured by EN 717-2 from pine wood had a strongly nonlinear pattern, suggesting that the data were not normally distributed [$X \sim N(0,1)$], and the offset between the line and the FE values suggests that the mean of the data was not 0. Conversely, the linearity of the FE values from beech, spruce, poplar, oak and birch suggests that the data were normally distributed, regardless of whether they were measured by EN 717-1 or EN 717-2.

The rate at which the individual wood species' FE differed was associated with their steady state concentrations or emission rates (Table 6). The values ranged between 0.0068 and 0.0036 ppm, as measured by EN 717-1, after a test period of 15–21 days, while they varied between 0.084 and 0.014 mg/m^2 h, as measured by EN 717-2. Beech wood showed the highest FE, at 0.0068 ppm and 0.084 mg/m^2 h, as measured by EN 717-1 and EN 717-2, respectively, followed by spruce wood (0.0055 ppm) and pine wood (0.0053 ppm). Birch wood had the lowest amount (0.0036 ppm), as measured by EN 717-1, while poplar and oak woods (0.014 mg/m^2 h) had the lowest values when measured by EN 717-2.

Table 6
Formaldehyde emission values measured with EN 717-1 (ppm) and EN 717-2 (mg/m^2 h) from six solid woods.

Wood species	MC% after drying	Density (g/cm^3)	Formaldehyde emission values[a]	
			ppm[b]	mg/m^2 h
Beech	9	0.76	0.0068 ± 0.001[a]	0.084 ± 0.009[a]
Poplar	8	0.33	0.0042 ± 0.0002[c]	0.014 ± 0.001[d]
Birch	7	0.69	0.0036 ± 0.0008[c]	0.049 ± 0.002[c]
Oak	8	0.72	0.0042 ± 0.0003[c]	0.014 ± 0.001[d]
Pine	8	0.41	0.0053 ± 0.0004[b]	0.016 ± 0.002[d]
Spruce	9	0.43	0.0055 ± 0.001b	0.069 ± 0.011[b]
$LSD_{0.05}$			0.001	0.0072
P value			<0.0001	<0.0001
CV%			17.11	14.92
R^2			0.65	0.96

[a] Values are the mean ± SD; means with the same letter within the same column are not significantly different, according to $LSD_{0.05}$.
[b] At 23 °C and 1013 hPa, the following relationship exists for formaldehyde measured by EN 717-1: 1 ppm = 1.24 mg/m^3 or 1 mg/m^3 = 0.81 ppm.

Formaldehyde is emitted from wood under very high heat and is not expected to be a significant source of formaldehyde in composite wood products during service. Furthermore, when the wood samples from the six species were air-dried (25–30 °C), formaldehyde was formed with only relatively slight differences in the values between the wood species (Table 6).

Wood itself generates a significant amount of formaldehyde when exposed to certain conditions common to the composite panel manufacturing process, which is caused by the thermal degradation of polysaccharides in the wood [11,14]. However, this phenomenon does not explain FEs from wood that has never been heated [31]. Formaldehyde has been shown to be impermanent and rapidly decreases to levels below those set by the EN 717-1 and EN 717-2 standards [13]. Moreover, pyrolysis of spruce and pine wood at 450 °C generated formaldehyde, which was attributed to the breakdown of the polysaccharide fraction of the wood [32]. During hot pressing, acetyl groups released from the wood form acetic acid, which is considered to be a formaldehyde scavenger. Beech wood contains more acetic acid than does pine [23].

The correlations between the EN 717-1 and 717-2 measurements for FE for the six wood species are presented in Fig. 4. The correlations between the emittable formaldehyde values were varied, with an R^2 ranging between 0.54 and 0.94. Furthermore, the linear regression in the relationships between the values of the European small-scale chamber and gas analysis exhibited very good positive correlations with R^2 values of 0.94 (spruce), 0.93 (beech) and 0.87 (birch). In addition, a linear regression for the formaldehyde values produced R^2 values of 0.74 (oak) and 0.73 (pine), which are considered as moderate relationships. By contrast, the value of R^2 for poplar (0.53) was not convincing.

3.2. Formaldehyde emissions from plywood panels

Table 7 presents the evaluation of the differences in the FE values based on board thickness and wood species used in plywood panels as measured by the gas analysis method. The comparison between means is presented graphically in Fig. 5 to evaluate the level of significance.

In Table 7, the samples with a thickness of 22 mm had higher FEs than those with 8 mm thicknesses for all of the studied types of plywood. Additionally, among the 22-mm samples, the spruce

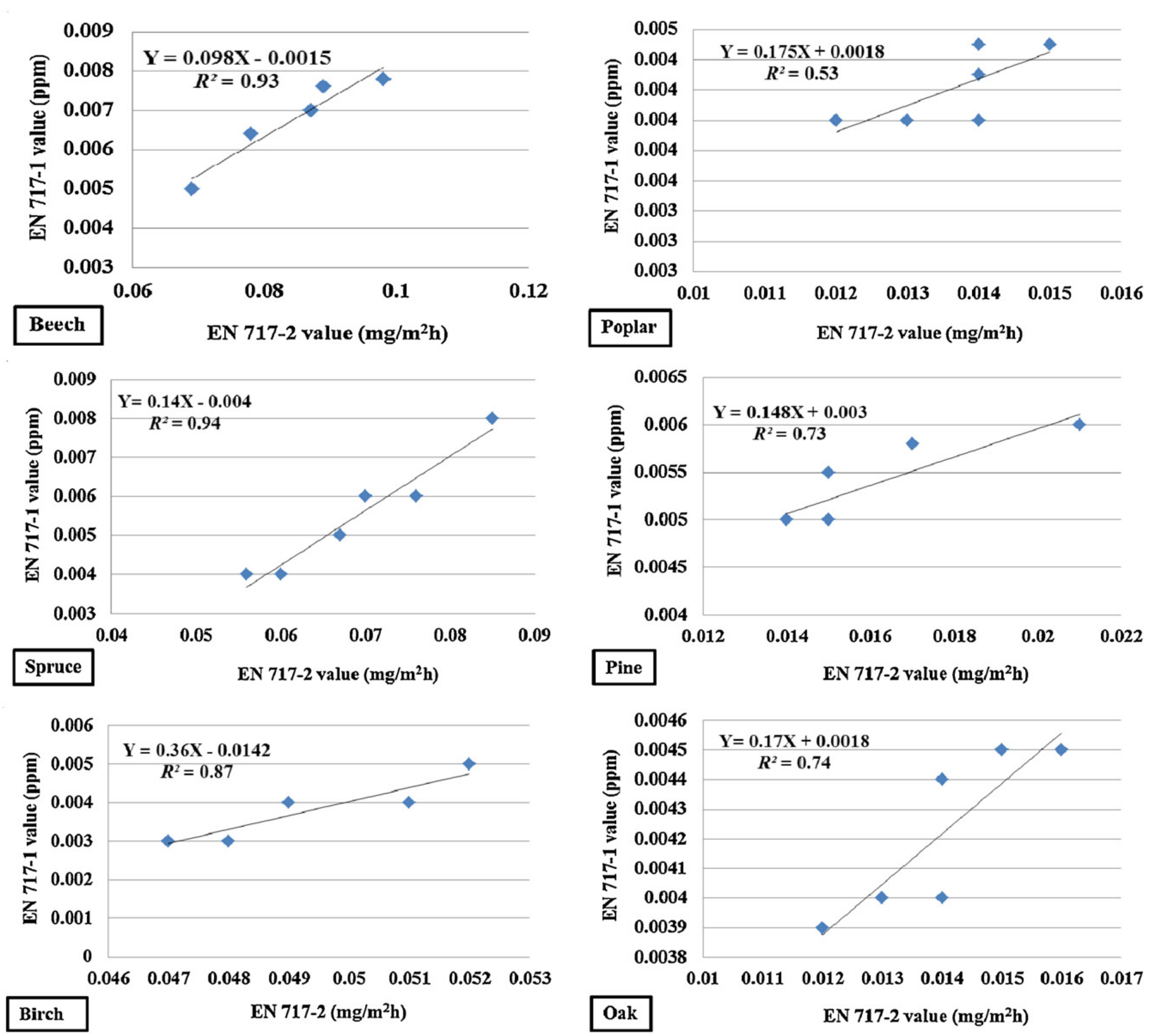

Fig. 4. Correlations between the formaldehyde values measured by EN 717-1 and EN 717-2 methods for different wood species.

PLY (2.65 mg/m^2 h) and beech PLY (2.61 mg/m^2 h) had the highest amount of FE, followed by birch PLY (2.21 mg/m^2 h) and pine PLY (1.79 mg/m^2 h). Furthermore, birch PLY (1.66 mg/m^2 h) and spruce PLY (1.58 mg/m^2 h) had the highest FEs for the 8-mm thick boards, followed by poplar PLY (1.25 mg/m^2 h) and beech PLY (1.24 mg/m^2 h).

In general, PLYs had lower ($P < 0.001$) amounts of FE for both board thicknesses (8 mm and 22 mm) than those from PLY. In particular, the UF resin had the highest FE rate because it contains a large amount of incompletely cured UF resin, which results in free formaldehyde after the hydrolysis of the cured UF resin. All of the panels were classified as E1-grade emission ($\leq$3.5 mg/m^2 h) and were considered as acceptable products for indoor applications.

The FE values of the PLYs ranged from 0.13 to 0.72 mg/m^2 h for the 8-mm-thick samples and from 0.36 to 0.85 mg/m^2 h for the 22-mm-thick samples. The FE of the boards was close to that of solid wood; thus, such low free formaldehyde values may be emitted from the wood itself because the C—C bonding in the PF resin was very stable against hydrolytic attack [7]. Moreover, this low susceptibility to hydrolysis is one of the reasons why PF resins are considered waterproof, while UF is not. PF resins are generally the resin chosen for manufacturing exterior grade structural panels. At such low levels of free FE, the boards are considered to be formaldehyde free.

The effect of individual parameters (wood species, board type and thickness) and the interaction between two parameters on the FE of the manufactured plywood panels was measured by EN 717-2 (Figs. 6 and 7). The FE was significantly affected ($P < 0.001$) by all of the parameters used for manufacturing.

The density and anatomy of the wood from which the veneers were made should be taken into account when evaluating the emissions results. For example, the formaldehyde present in the resin interacts in a distinct way with each wood species; thus, the FE values were affected by the anatomy of the respective wood species [17]. The plywood panels produced from poplar veneer [low specific gravity (*SG*, 0.33)] with a simple anatomy produce lower FEs values. Additionally, the amount of adhesive used was related to *SG*. An increase in *SG* (beech and birch plywood) causes more adhesive to be used to make the boards and consequently releases more formaldehyde. Furthermore, the free formaldehyde content of the beech plywood was higher than the values of the poplar plywood, which agrees with the previous study [33]. However, the highest

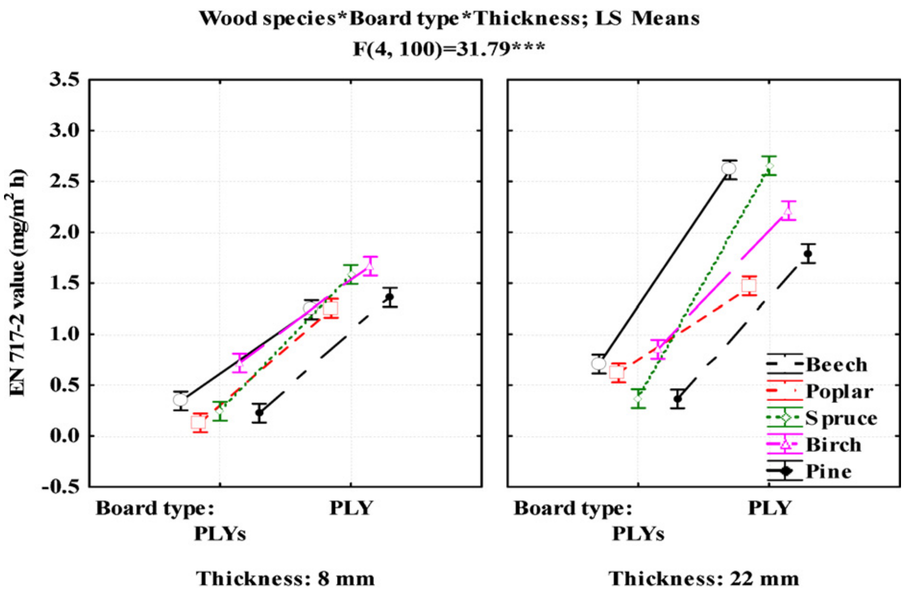

Fig. 5. Effective hypothesis decomposition of the interaction among wood species, board type and thickness and their effects on the formaldehyde emission measured by EN 717-2 method. Vertical bars denote 0.95 CI.

amount of formaldehyde released during hot pressing was related to the wood's structure (beech wood consists of bigger vessel surfaces). Furthermore, the FE of the beech plywood panels was higher and could be related to the higher density of the beech wood.

The major sources of indoor pollutants are the compressed wood products, including plywood and their adhesive resins. Thus, the FE from interior plywood can last for years with little decay. However, the manufacturers can now modify the production processes and use alternative adhesives to reduce the FE. Moreover, as shown in our study, the choice of veneer material for making plywood panels is very important in predicting the rate of FE when these panels are used in interior applications. Additionally, many countries have regulations in place that limit the maximum amount of FE from wood products. In the Czech Republic, the FE is mostly tested by the gas analysis and perforator methods from the manufactured wood-based panels, which do not necessarily correlate with the actual emission rates found by the referenced chamber method. From the previous results, all of the emission levels obtained from the manufactured plywood panels were below the E1-emission limit, even when measured by the EN 717-2 method.

The referenced method (EN 717-1) was operated under average indoor environmental conditions (temperature of 23 ± 0.5 °C and 45 ± 3% RH). Thus, the results of the FE from the samples were normally equal to the predicted FE values from building and furniture materials. At 29 °C, the measurements did not emit detectable formaldehyde levels, while at a temperature of 50 °C, a high initial emission of formaldehyde was found, which decreased with time [34]. Additionally, previous studies observed that the initial emittable formaldehyde concentration ($C_{m,0}$) from the dry building materials was increased significantly, by approximately 507%, when the temperature was elevated from 25.2 to 50.6 °C [35]. Furthermore, Zhang et al. [36] showed that the temperature has a significant effect on both the partition coefficient (K) and the diffusion coefficient (D) of formaldehyde emissions from some building materials, as measured by the C-history method [25,26], which was used to measure the D and K of formaldehyde in dry building materials at a range of temperatures (18, 30, 40 and 50 °C). A formula can be derived by relating the partition coefficient and its related factors. This formula can predict the theoretical partition coefficient and can provide insight for fitting experimental data, which agrees well with the experimental results. Thus, when the temperature was increased, K decreased, while D increased.

The referenced method needs to be operated between 7 and 28 days, which is time-consuming and requires expensive equipment. By contrast, the second method (EN 717-2) is operated at high temperatures (60 ± 0.5 °C) and low RH (≤3%), which does not approximate the actual indoor environment. These results mean that most of the formaldehyde in the building material (overestimation of FE) is not emitted at room temperature. This method is time-saving (approximately 4 h) compared to the 7–28 days needed for EN 717-1 and has gained wide acceptance for assessing the FE from wood-based panels and is a CARB-approved quality

Table 7
The initial emittable formaldehyde content from different types of plywood measured by EN 717-2 method (mg/m² h).

Wood species	Plywood type	Thickness (mm)	Formaldehyde value (mg/m² h)		
			Mean ± SD	−95% CI	+95% CI
Beech	PLYs	8	0.35 ± 0.06^{j}	0.25	0.44
		22	0.71 ± 0.11^{i}	0.61	0.80
	PLY	8	1.24 ± 0.04^{g}	1.14	1.33
		22	2.61 ± 0.21^{a}	2.52	2.70
Poplar	PLYs	8	0.13 ± 0.01^{k}	0.03	0.22
		22	0.62 ± 0.17^{i}	0.52	0.71
	PLY	8	1.25 ± 0.02^{g}	1.16	1.34
		22	$1.47 \pm 0.19^{e,f}$	1.38	1.56
Spruce	PLYs	8	$0.24 \pm 0.02^{j,k}$	0.15	0.33
		22	0.37 ± 0.06^{j}	0.27	0.46
	PLY	8	$1.58 \pm 0.24^{d,e}$	1.49	1.67
		22	2.65 ± 0.17^{a}	2.56	2.74
Birch	PLYs	8	0.72 ± 0.07^{i}	0.62	0.81
		22	0.85 ± 0.03^{h}	0.76	0.94
	PLY	8	$1.66 \pm 0.04^{c,d}$	1.57	1.75
		22	2.21 ± 0.04^{b}	2.12	2.30
Pine	PLYs	8	$0.22 \pm 0.01^{j,k}$	0.13	0.31
		22	0.36 ± 0.02^{j}	0.27	0.45
	PLY	8	$1.36 \pm 0.05^{f,g}$	1.27	1.45
		22	1.79 ± 0.15^{c}	1.69	1.88

Different letters represent significant differences between the averages of the values.
Means with the same letter are not significantly different at a 0.05 level of probability, according to the $LSD_{0.05}$ test.

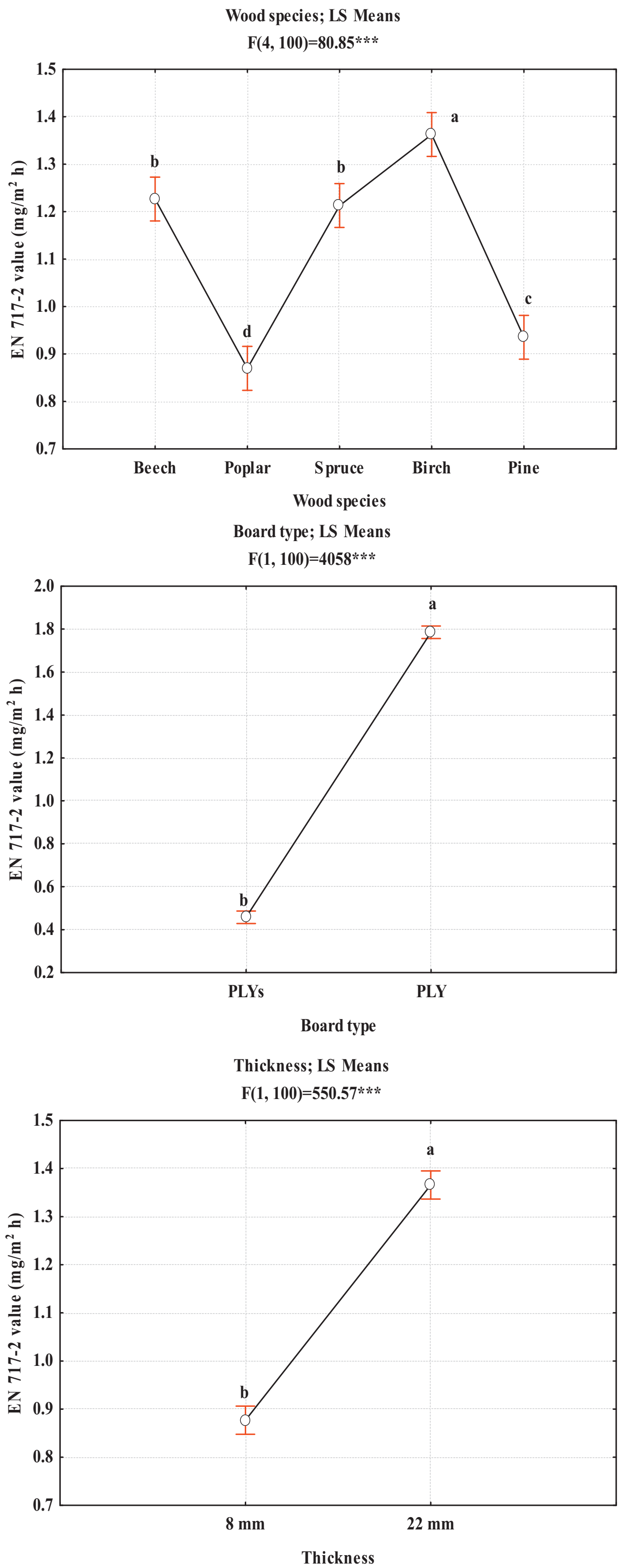

Fig. 6. Effect of individual parameters from the manufactured plywood panels on the formaldehyde emission measured by EN 717-2. Vertical bars denote 0.95 CI. Means with the same letter are not significantly different at 0.05 level of probability according to $LSD_{0.05}$ test.

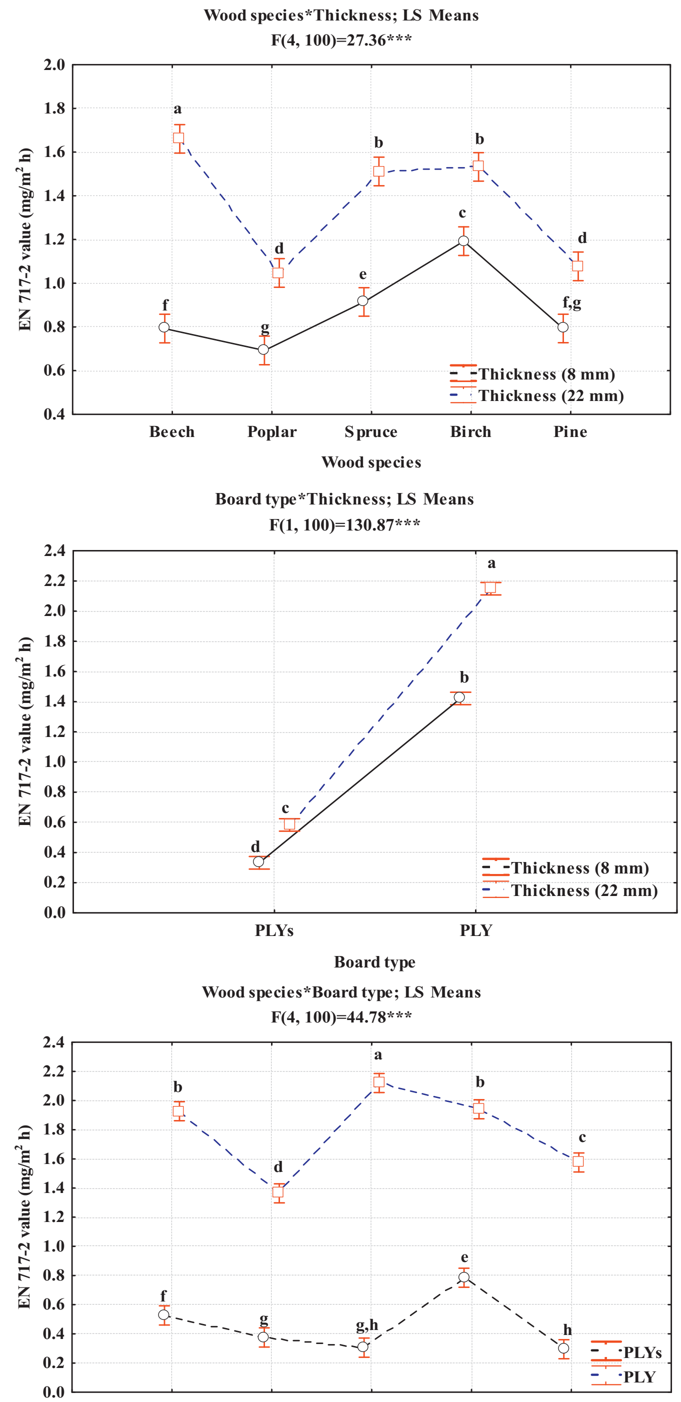

Fig. 7. Effect of the interaction between each two parameters from the manufactured plywood panels on the formaldehyde emission measured by EN 717-2. Vertical bars denote 0.95 CI. Means with the same letter are not significantly different at 0.05 level of probability according to $LSD_{0.05}$ test.

control test method [37]. Additionally, each sample emits enough formaldehyde to be classified as an E1 grade, even when measured by secondary methods (EN 717-2).

The total formaldehyde concentration measured by the gas analysis method cannot be emitted at standard room conditions (temperature and RH) and thus cannot be taken as an accurate index for the pollution level for tested building materials, as measured by the referenced room chamber method (EN 717-1). Thus, the products should be evaluated by intra-laboratory

Table 8
The initial and steady-state formaldehyde concentrations from different types of flooring and blockboard measured by EN 717-1 method (mg/m^3).

Material	Initial formaldehyde concentration	Steady state formaldehyde concentration[a]
Solid wood flooring with PF	0.015	0.013
Engineered flooring with MUF	0.02	0.018
Engineered flooring with PVAc	0.006	0.006
Birch blockboard		
Uncoated	0.023	0.020
Painted	0.048	0.03
Poplar blockboard		
Uncoated	0.022	0.015
Painted	0.035	0.025
Beech blockboard		
Uncoated	0.032	0.023
Painted	0.045	0.037

[a] The steady state concentrations express the concentration obtained at the end of the measuring period (ranging between 10 and 21 days).

and inter-laboratory comparisons to overcome the problems with the emission levels of different products in different regions or countries because there have been many studies focused on the variations in the FE measurements between laboratories. The most significant variations were due to heterogeneities in the chamber conditions, such as volume, materials, sampling air, and specific differences in test conditions [38–40].

The previous statements may apply to many other regions because building material testing has been initiated in Europe and North America. Furthermore, products manufactured using recently developed low formaldehyde emitting adhesives (MUF, UF and PF) have shown significantly lower emissions.

3.3. *Formaldehyde emission from flooring and blockboard*

The FE values from different types of flooring and blockboards are presented in Table 8 as both initial and steady-state formaldehyde concentrations (mg/m^3). The different types of flooring and blockboards measured by EN 717-1 showed the average emission to be much lower than the E1-emission class. The initial formaldehyde concentrations from the investigated materials ranged between 0.006 mg/m^3 for the engineered flooring with PVAc and 0.048 mg/m^3 for the painted birch blockboard. At the end of measuring period, the concentrations decreased and ranged from 0.006 mg/m^3 for the engineered flooring with PVAc to 0.037 mg/m^3 for the painted beech blockboard.

The measurements started one week after the materials were manufactured and continued for three weeks. The initial concentrations of FE were higher for the painted blockboard (0.035–0.048 mg/m^3) than for the uncoated blockboard (0.022–0.032 mg/m^3). Additionally, the FE in the first week after manufacturing was higher for all materials but decreased noticeably after two weeks, with the values for the painted blockboard drastically decreasing.

The engineered flooring bonded with PVAc had a steady state formaldehyde concentration similar to the initial one (0.006 mg/m^3). Previous studies showed that the FE level in engineered flooring decreased with increased amounts of PVAc [9,41]. Conversely, the painted blockboard emitted more formaldehyde than the uncoated boards [17,42]. However, when a UV-curable coating was applied to make flooring and fancy veneers with the plywood for engineered flooring, the FE decreased and returned to the same emission level as that of plywood [17,41]. Furthermore, the previous studies concluded that the FEs from flooring materials were much lower than those of furniture materials under heating at 20, 37 and 50 °C, which is significant because the furniture materials mostly exist at room temperature [19,43,44].

Thus, the resulting low FEs values may occur because (1) the measurements were started after one week from the production, (2) the resin used to manufacture the solid wood flooring was PF and (3) the plywood types used to produce the engineered flooring and all types of blockboards were bonded with PF resin, as described in our study. Thus, the released formaldehyde could only come from the wood species themselves, and the low amount of free formaldehyde came from the MUF resin. However, the current European regulations are forcing wood-based products to have zero emissions or to meet the E0 emission class ($\leq$0.07 ppm or 0.086 mg/m^3); the results reported here are roughly equivalent to SE0 ($\approx$0.04 ppm or 0.049 mg/m^3). Additionally, at this extremely low level of emission, approximately 50% of the measured formaldehyde is released naturally by the wood itself. Moreover, products manufactured using the low emission formaldehyde resin have shown significantly lower emissions.

The levels of FEs in Europe are super (SE0), E0, E1 and E2 emission classes. Boards of Class E1 can be used without causing an indoor air concentration of formaldehyde $\leq$0.1 ppm ($\leq$0.124 mg/m^3) as measured by the referenced method (EN 717-1) or $\leq$3.5 mg/m^2 h by factory production control (EN 717-2) method. In accordance with the European Standard EN 13986 [45], the formaldehyde release from wood-based panels used in internal applications will be classified as either Class E1 or Class E2. Class E2 with EN 717-1 is >0.1 ppm (>0.124 mg/m^3) and >3.5 $\leq$ 8.0 mg/m^2 h by EN 717-2. E0 is an updated class of E1 with much more stringent standards that requires FEs to be equal to or less than 0.07 ppm. Therefore, composite wood products, such as laminate flooring or engineered hardwood flooring, that meet the E0 standards would be considered a safer, greener choice than those that only meet the E1 standards. That said, the E1 standards are equivalent to those set forth by the U.S. Department of Labor Occupational Safety & Health Administration (OSHA) and are often used in engineered hardwood and laminate floors that are deemed by manufacturers as "eco-friendly" [46].

However, the Japanese *F*** class (1.5–2.1 mg/L) is more equivalent to the European E1-class, while the *F****/E0* (0.5–0.7 mg/L $\approx$ 0.07 ppm) and *F*****/SE0* (0.3–0.4 mg/L $\approx$ 0.04 ppm) emission limits were much lower than those of the E1 class. Thus, the emission from *F***** boards is close to the emission of solid untreated wood (i.e., between 0.008 and 0.01 ppm for spruce wood flakes) [47]. The resulting Phase 1 (P 1) CARB emissions regulations for plywood, started in 2010, of 0.08 ppm are roughly comparable to *F****/E0*, and the Phase 2 (P 2) emissions regulations, started in 2012, of 0.05 ppm are roughly equivalent to *F*****/SE0*. Both the CARB (P) and the Japanese Emission Standards (JIS/JAS *F*****) are more stringent; therefore, materials that meet these stricter regulations are preferred over those that only meet E1 or E0 standards.

A comprehensive and user-friendly emissions database was constructed, containing up-to-date emissions data for building materials because of the prioritisation of building materials as indoor pollution sources (BUMA emission data base) [48]. The BUMA prioritisation scheme provides a simple screening tool to rapidly classify building materials with respect to exposure levels of toxic emitted substances in the indoor environment. The BUMA emission database recommends that the specific emission rate from floorings be 25 μg/m^2 h (0.25 mg/m^2 h) with a range of 0–125 μg/m^2 h and that the steady-state emissions range from 0.006 mg/m^3 (engineered flooring-PVAc) to 0.018 mg/m^3 (engineered flooring-MUF) in the present study. The specific emission rate found by the BUMA Database for a wood-based panel was 144 μg/m^2 h (0.144 mg/m^2 h) with a range of 0–1580 μg/m^2 h. Additionally, the lowest formaldehyde concentrations were predicted to occur where the ceiling panel was covered with water-based paint after 3 and 7 day measurements. However, our study found that the coated blockboard with oil-based paint had the highest concentration of formaldehyde compared to the uncoated boards (Table 8).

Based on these data, we suggest that the materials produced in this study reduce the health hazards of FE and could be used as furniture or building materials under all of the manufacturing parameters with large-scale production. The real value of indoor formaldehyde emissions could be even higher because our measurements did not include emissions from textiles, carpets, cleaning products, etc. Additionally, decorative surface finishes can also either increase or decrease surface emissions, depending on the nature of the finish and the substrate. In fact, using low-emission softwood products for building materials maximises the use of natural forest resources in the Czech Republic.

In this article, the manufacturing process, standards and criteria for selecting materials to reduce formaldehyde emissions were created and discussed. The products were partially manufactured on a large scale to assess their commercial viability, which can be included in the quality assurance procedures in building design and construction processes.

4. Conclusion

The data obtained in this study resulted in some interesting observations. The wood veneer species apparently affected the final FE from the produced plywood when manufactured with the same base wood species. A similar efficiency was observed with the solid wood, wood veneer and plywood applied to produce flooring and blockboard panels. Thus, plywood behaves similarly to its solid wood source with respect to its FE values as well as in the production of flooring and blockboard.

The wood species, plywood type and thickness all significantly affected the FE measured by EN 717-2 ($P < 0.001$). The initial formaldehyde concentrations from the investigated materials ranged from 0.006 mg/m^3 (engineered flooring with PVAc) to 0.048 mg/m^3 (painted birch blockboard) and decreased by the end of the measuring period. Formaldehyde emissions in the first week after manufacturing were higher but showed pronounced decreases after two weeks of measurements for all of the materials, with dramatic decreases in the painted blockboard's emissions.

The results provide an in-depth understanding of the variables that affect formaldehyde emissions from wood products during the manufacturing process, which allow formaldehyde emissions to be controlled. The concentrations of formaldehyde in these products were below the mandated limits in the Czech Republic and EU.

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