THE QUALITY OF INDOOR AIR IN WOODEN BASED BUILDINGS AND THE FACTORS WITH IMPACT OF THEM

PETR ČECH, DANIELA TESAŘOVÁ, JANA HADAČOVÁ, EVA JEŘÁBKOVÁ
MENDEL UNIVERSITY IN BRNO, FACULTY OF FORESTRY AND WOOD TECHNOLOGY
DEPARTMENT OF FURNITURE, DESIGN AND HABITATION
BRNO, CZECH REPUBLIC

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ABSTRACT

The primary aim of the paper is to characterize the emissions of volatile organic compounds (VOC) in the indoor air of wood-based buildings in the Krkonoše Mts. region in relation to season, age of the building, air moisture content and engineering composition of building materials.

It aims to identify the impact of important factors on the amount and content of VOC emitted into indoor air. The experimental part focuses on the assessment of influence of applied wood-based materials in the wood-based buildings on the quantity and quality of VOC emissions in indoor air. The second aim is to determine the impact of age of a building and the time of VOC measuring on the amount and content of emitted VOC, particularly on the amount of emitted terpenes and aldehydes. VOC emissions were analysed by gas chromatograph Agilent GC 6890 N with a mass spectrometer with cryofocusation, thermal desorption and library of spectra NIS 05.

KEYWORDS: Wooden based buildings, VOCs, TVOC, quality of indoor air, gas chromatography.

INTRODUCTION

Although numerous studies have investigated the levels of indoor air pollutant and emission measurements in laboratories, research on systematic in-field studies, linking the VOC concentrations to their indoor sources is rather limited (Jarnstrom et al. 2007). Indoor air quality is extremely variable and depends on activities of the people (Morawska et al. 2003, Edwards et al. 2006, Eklund et al. 2008, Buonanno et al. 2009, 2012), home furnishings (Yrieix et al. 2010), building materials (Missia et al. 2010) and season (Schlink 2004). Current research is involved with the constantly rising amount of sources, the complexity of mixtures, and the role of outdoor
air (Carslaw et al. 2009). The diversity of compounds, their variable toxicity and the Addressed peer group complicate the determination of guidelines for concentrations of volatile organic compounds (VOCs) in indoor environments.

The formation of VOCs in indoor environments is difficult to understand and to reconstruct (e.g. in experiments). On the one hand, compounds originate exclusively from indoor sources (a point of origin of gases or other materials, which appears constantly in a similar way) and, on the other hand, they are formed by mixtures of indoor and outdoor pollutants. In most cases, indoor VOC concentrations are significantly higher than outdoor levels (Batterman et al. 2007). This is influenced by type and age of building materials (Missia et al. 2010) and personal activities, e.g. renovation processes, that cause elevated levels. Increased levels occur directly after renovations and then normalize to lower concentrations (Jia et al. 2008a, Herbarth and Matysik 2010). Seasonal variations cause higher indoor levels to accumulate due to abated ventilation in winter (Dodson et al. 2008, Matysik et al. 2010). Furthermore, local conditions, such as industry or busy roads, create emission sources that differentiate the pollution amount of homes in industrial, urban, and nonurban regions (Jia et al. 2008b). This high variety of possible sources in indoor and ambient air poses a big challenge for scientists to assign different compounds to their point of origin.

Building materials are major contributors to indoor emission sources of volatile organic compounds (VOCs). Some of VOCs are of particular concern due to their potential health impact on human (Marchland et al. 2006), e.g. formaldehyde and benzene are some of the most studied pollutants since they are classified in Group 1 of human carcinogens by the International Agency for Research on Cancer (IARC, 2004; Liu et al. 2013, Jiang et al. 2013, Plaisance et al. 2014). In an INDEX project the existing knowledge worldwide has been assessed in terms of type and levels of chemicals in indoor air, as well as, the available toxicological informatic (Kotzias et al. 2004). It was concluded that VOCs such as benzene, formaldehyde, acetaldehyde, toluene and xylenes have to be considered as priority pollutants with respect to their health effects (Kotzias et al. 2004). Many studies have shown significant VOC sources indoors. For example, the main sources of aldehydes and BTEX indoors may include building materials (BM), such as hardwood, plywood, laminate floorings, adhesives, paints and varnishes (Marchland et al. 2006, Zhang et al. 2007, Lin et al. 2009, Corsi and Lin 2009, Weschler and Shields 1996, An et al. 2011, Böhm et al. 2012; Shinohara et al. 2009), adhesives and decoration materials. In addition to these primary emissions, numerous past researches also indicated that ozone reactions with BMs result in secondary emissions of aliphatic aldehydes, secondary organic aerosols and other products that are more important (Kim et al. 2010, Morrison et al. 1998, Weschler 2006, Poppendieck et al. 2007, Hoang et al. 2009, Cros et al. 2012, Gall et al. 2013, Lin and Chen 2014, Huang et al. 2011, Yu et al. 2011).

Weschler et al. (1992) have demonstrated that with the presence of ozone (60-100 mg.m⁻³) in freshly carpeted stainless-steel chamber, gas-phase concentrations of VOCs including styrene, 4-VCH and 4-PCH significantly decreased while the concentrations of aldehydes (C₁-C₁₀) increased. In addition, the total concentrations of VOCs also increased markedly. Wang and Morrison (Wang and Morrison 2006) observed substantial secondary emissions which include C₁ (Formaldehyde), C₂ (Acetaldehyde) and C₅-C₁₀ (Pentanal-Decanal) aldehydes from walls, carpet, floors and countertops in the presence of elevated ozone concentrations. Huang et al. (2012) performed small-scale environmental chamber experiments on a painted hardwood panel and found that the formaldehyde concentration within the chamber is found to increase by 215.8 % given an ozone concentration of 200 ppb and a reaction time of 3.0 h.
As people pay increasing attention to the impact of emissions from BMs, the market for environmentally friendly green building materials is gradually growing. Although Green Building Material (GBM) is intended to have low toxicity and minimal chemical emission, measurement of primary emissions alone may not be sufficient since secondary emissions due to ozone reactions may affect the perceived air quality in the long run (Kundsen et al. 2003). Kagi et al. (2009) demonstrated that natural wood material with low formaldehyde emission after being exposed to ambient ozone produced secondary pollutants, including formaldehyde, acetaldehyde, cyclohexanone and benzaldehyde.

For many of these chemicals, the risk on human health and comfort is almost unknown and difficult to be predicted because of the lack of toxicological data. In the frame of the INDEX project (Kotzias et al. 2004) the existing knowledge worldwide has been assessed in terms of type and levels of chemicals in indoor air, as well as, the available toxicological information. It was concluded that VOCs such as benzene, formaldehyde, acetaldehyde, toluene and xylenes have to be considered as priority pollutants with respect to their health effects. On the other hand, chemicals such as limonene and α-pinene require further research with regard to human exposure or dose response and effects.

Reported indoor concentration of individual VOCs are generally below 50 μg.m⁻³, with most below 5 μg.m⁻³ (Wolkoff et al. 2006). Formaldehyde indoor concentrations were varied between 38 and 310 μg.m⁻³ (Schleibinger et al. 2001). Formaldehyde mean values could reach 134±93 μg.m⁻³ and 86±58 μg.m⁻³ in new and old buildings, respectively (Park and Ikeda 2006). Additionally, typical European indoor exposure concentrations for xylenes, acetaldehyde and terpenes varied from 2–37, 10–18 and 6–83 μg.m⁻³, respectively (Kotzias et al. 2009).

Combined indoor/outdoor air quality measurements have shown that there exist significant VOC sources indoors. For example the aldehyde concentrations are usually 2-10 times higher than outdoors (Marchland et al. 2006). It has been pointed that in renovated or completely new buildings, the VOCs concentration levels are often several orders of magnitude higher (Kim et al. 2006). The main sources of aldehydes in homes include building materials, hardwood, plywood, laminate floorings, adhesives, paints and varnishes and in some cases they are products of ozone- initiated reactions (Marchland et al. 2006, Weschler et al. 1992). For example, interior coatings can increase indoor air pollution due to VOC emissions (Kwok et al. 2003). Some of the major VOCs emitted from an oil-based varnish were ethylbenzene, m, p-Xylene, o-Xylene and formaldehyde (McCrillis et al. 1999).

Formaldehyde is known to be released by press wood products used in home building construction such as MDFs, and paneling and products made by urea- formaldehyde resins (Kelly et al. 1999). The contribution of building materials and furnishing to indoor air pollution has been demonstrated by a study of VOC emissions in newly built, unoccupied houses at BRE (Yu and Crump 1999). The sampling of VOCs in indoor air has shown that the contribution of building material emissions was significant during the first six months.

Although, numerous studies have investigated the levels of indoor air pollutants and emission measurements in laboratories, research on systematic in- field studies, linking the VOC concentrations to their indoor sources is rather limited (Jarnstrom et al. 2007). The primary aim of the present work is to characterize building materials as indoor VOC emission sources by conducting indoor concentration and emission measurements at houses and public buildings including schools, across Europe. The measurements cover mainly carbonyls, BTEX and terpenes with emphasis to the abovementioned priority compounds.
MATERIAL AND METHODS

Characteristics of the assessed buildings
The quantitative and qualitative structure of VOC emissions in indoor air was measured in four wood-based buildings in the foothills of the Krkonoše Mts., two of which are situated on a ridge at 1000 – 1030 m above sea level and the other two in a valley at 476 – 527 m. The monitored detached buildings differ from one another in terms of their age, the material composition of their walls and their use.

Family house in Horní Maršov
The family house in Horní Maršov was built in 2006 at an altitude of 527 m, close to a forest and with low traffic load. The windows are south-west facing. The house is heated by a solid-fuel stove.

Technical specifications
The load-bearing part of the house consists of double timber frame – external and internal. The timber frame consists of spruce beams of 150 x 150 mm. The horizontal load is carried by rails, struts, thresholds and lintels which reinforce the entire construction. The load-bearing part is assembled using the original tenon and mortise jointing strengthened by wooden pegs. The external timber frame is covered by 30 mm wooden cladding which has an aesthetic function as well. The cladding is separated from the external structural frame by a 30 x 50 mm wooden lath grate and a diffusion foil.

An office in Mladé Buky
A ground floor building in the centre of Mladé Buky was built in 2000 at an altitude of 476 m with a medium traffic load. The entire area of walls is covered by tongue and groove panels. The room has a double-hung euro window with double glazing. The office is heated by an electric storage heater.

Technical specifications
The wood-based building was built as a framed construction in the 625 mm module. The load-bearing part consists of 120 x 120 mm spruce beams. Its exterior is covered with 18 mm OSB cladding. On the interior, the load-bearing structure is covered by vapour-tight foil JUTAFOL fitted with 30 x 50 mm wooden profiles creating a stud wall. Vertical interior surfaces are covered by 15 mm SECCA tongue and groove panelling treated with a water-based varnish.

Log cabin in Horní Lysečiny
A traditional wood-based building typical for the Krkonoše Mts. region, this log cabin was built in 1780 at an altitude of 1000 m. Its floor is made of solid wood boards joined by butt joints. The entire room is heated by a stove. The surroundings are subject to minimum traffic load.

Technical specifications
The wall construction is made from square-cut beams placed horizontally on top of each other. The wall beams are joined lengthwise with butt joints, the gaps filled with clay and covered in lime coating. The beams of two perpendicular walls are joined by dovetail joints.
Log house Velké Tippeltovy Boudy

This detached log house dating from the 17th century was built at an altitude of 1030 m on a forest clearing. It is heated by a tiled stove. The floor is made of solid wood boards joined by butt joints.

Technical specifications

The wall construction of log cabin is made from square-cut beams placed horizontally on top of each other and joined by dovetail joints in corners. The interior walls are treated with linseed oil.

Methods of air sampling in the monitored wood-based buildings

The concentration of volatile organic compounds in indoor air is determined using the adsorption method. The air samples are drawn by a pump through a Tenax TA thermal desorption tube at a constant speed and a defined flow rate.

- the air samplers were placed in the centre of the rooms, a minimum of 1 m from walls and at the height of 1 – 1.5 m above the floor (breathing zone).

Concurrently with the indoor air quality measurements, control samples were taken outside the buildings (not less than 1 m and no further than 100 m from the building), ideally at the floor height of the building corresponding to the monitored room (Methodological instructions for the measurements and determination of chemical, physical and biological indicators of indoor environment quality according to Regulation No. 6/2003 Col.).

Sampling procedure

In the first step there is necessary to airing of the monitored room. The airing of the room was following by placing the pump with the sorbent tube in the assessed room and in external environment. Next step of the measuring is launching the VOC emissions sampling, taking the time duration of single sampling of indoor air was 3 hours and outdoor was 1 hour. Upon reaching this limit, the pump was stopped and the steel sampling tube was exchanged. The VOC concentration in the indoor and outdoor air was determined using sampling pump Gilian LFS-113 DC. The air flow rate in the interior was 12 l/h and 6 l/h in the exterior. The sampling of VOC was done using steel tubes filled with Tenax TA (100 mg Tenax TA per tube). The VOC emission structure was determined along with the amount of individual emitted compounds trapped in the steel sampling tube were analysed by using a gas chromatograph with a mass spectrometer and a thermal desorption. The conducted analyses provide qualitative and quantitative data on the concentrations of selected VOC and the total volatile organic compounds (TVOC) in µg.m⁻³.

RESULTS

Family house in Horní Maršov

Fig. 1 shows the influence of relative moisture content on the quality of indoor air, especially of Pentanal emissions in indoor air of the wooden base building (Horní Maršov) in dependence on a season. The amount of concentration of Pentanal is growing in winter time compared with the other seasons. The highest concentration of Pentanal was found at conditions 24 % of relative humidity in winter time, conversely the lowest concentration of the compounds was measured at conditions 21 % of relative humidity in spring.
On the Fig. 2 we can see the influence of the relative moisture content from living room on an amount of emissions of Hexanal in family house in Horní Maršov. The highest concentration of Hexanal was measured at conditions 24 % of relative moisture content (39.3 µg.m⁻³) in winter. The bigger amount of Hexanal was found at conditions 40 until 47 % of relative humidity in autumn.

![Fig. 1: Pentanal from living room in family house in Horní Maršov (left).](image1)

![Fig. 2: Hexanal from living room in family house in Horní Maršov (right).](image2)

Fig. 3 presents data of TVOC (Total Volatile Organic Compounds) from living room in family house in Horní Maršov. Parameter of TVOC describing total content of Volatile Organic Compounds emitted into indoor air from the living room in wooden base building. The amount of TVOC is growing in winter and in autumn in compared with the emissions in spring season. The highest amount of TVOC was measured at conditions 24 % relative moisture content (148 µg.m⁻³) in winter.

![Fig. 3: TVOC from living room in family house in Horní Maršov.](image3)

**An office in Mladé Buky**

On the Fig. 4 there is presenting the influence of relative moisture content on a quality of indoor air, especially of emissions of Pentanal, from the office in wooden base building (Mladé Buky) in dependence on a season. The highest concentration was measured at conditions 31 % of relative moisture content (4.6 µg.m⁻³) in an autumn, followed by conditions 20 a 30 % of relative moisture content (4.1 and 4.2 µg.m⁻³) in spring season. The lower concentrations of Pentanal were found in winter time (0.1 until 0.2 µg.m⁻³).

![Fig. 4: Pentanal from living room in office in Mladé Buky.](image4)

Fig. 5 shows the influence of relative moisture content on concentration of Hexanal from the office in wooden base building in Mladé Buky. The highest concentration of Hexanal was measured at conditions 31 % of relative moisture content (31.8 µg.m⁻³) in an autumn, followed by values of concentrations, there were detected in spring season at conditions 20 a 30 % of relative moisture content (23.8 and 23.4 µg.m⁻³).
On the Fig. 6 we can see the influence of relative humidity on an amount of parameter TVOC emitted by indoor air from office in wooden base building in Mladé Buky. The highest values of TVOC were measured at conditions about 20 % in winter, followed by values of concentrations TVOC, there were detected in the autumn. The lower concentrations of parameter TVOC were found during spring season.

**Log cabin in Horní Lyséčiny.**

On the Fig. 7 there is presenting the influence of relative humidity from the room on concentration of Pentanal in the wooden base building in Horní Lyséčiny. The highest concentration of Pentanal was measured at conditions 42 % of relative moisture content in spring season (2.0 µg.m⁻³), other values of concentration were found in very small amount (0.1 until 0.3 µg.m⁻³) in other seasons.

Fig. 8 shows the influence of relative moisture content from the room on concentration of Hexanal in the wooden base building in Horní Lyséčiny. The highest concentration of Hexanal was measured at conditions 42 % of relative moisture content in spring season (8.9 µg.m⁻³). Other measured values of concentration of Hexanal were found in small amount (0.3 until 1.2 µg.m⁻³) in other seasons.
Fig. 7: Pentanal from the room in log cabin in Horní Lysečiny (left).

Fig. 8: Hexanal from the room in log cabin in Horní Lysečiny (right).

Fig. 9 presents data of parameter TVOC, emitted by indoor air from the room in the log cabin in Horní Lysečiny. The highest values of TVOC were measured on the conditions 21, 32 and 46 % of relative moisture content in winter. Other values of parameter TVOC were found in relatively lower concentrations (from 12 till 43 µg.m\(^{-3}\)) in other seasons.

Log house Velké Tippeltovy Boudy.

Fig. 10 shows the influence of relative moisture content on a quality of indoor air, especially of emissions of Pentanal in indoor air of wooden base building (Velké Tippeltovy Boudy) in dependence on the season. The amount of Pentanal concentration is growing in spring and winter time). The highest concentration of Pentanal was found at conditions 54 % of relative moisture content in winter time (3.7 µg.m\(^{-3}\)), followed by values of concentration of these compounds there were measured in spring.

On the Fig. 11 there is presenting the influence of relative humidity in log house on concentrations of Hexanal in wooden base building. The highest concentration of Hexanal was found at conditions 34 % of relative moisture content in spring season and followed by value, there was measured at condition 54 % of relative moisture content in winter time. The lower values of concentration of Hexanal were found in autumn.
DISCUSSION

Based on the results listed in this paper we can assess the indoor air quality in individual wood-based buildings.

Family house in Horní Maršov

Results of VOC emission measurements are shown in Figs. 1 - 2 which show the impact of relative air humidity on the amount of emitted volatile organic compounds, particularly aldehydes – namely Pentanal and Hexanal, over the period of three seasons of the year. The presented results reveal a dependence of the emitted Pentanal and Hexanal concentrations on relative air moisture content in winter and spring. The concentration of emitted compounds decreases with decreasing moisture content. The highest concentration was measured in winter, which is related to the onset of heating season. The study (Schieweck and Bock 2015) has also described the same behavior for indoor without the describing the amount of Pentanal and Hexanal emissions.

The measured data reveal a minimum impact of relative air moisture content on the...
concentration of BTEX. Concentration of the said VOC was very low, which means that their amount was below the level of quantification (LOQ).

Owing to the fact that the assessed buildings were wood-based, the monitored substances included terpenoid compounds as accompanying characteristic wood product emissions from the perspective of wood-based building interior emission load. However, only the concentration of emitted 3-D-Limonen was dependent on relative air humidity in all the three monitored seasons. As an exception, the concentration of emitted Limonen increased during the last measurement in winter. This increase can be accounted for the use of an air freshener and detergents.

The values of the so-called total volatile organic compounds (TVOC) are among the key monitored parameters. TVOC represent the total amount of VOC emitted in the indoor air of the assessed wood-based building. Fig. 3 demonstrates the impact of relative air moisture content on the total amount of emitted VOC in relation to the season. Based on the presented results, dependence of TVOC concentration on relative air humidity cannot be determined. The highest TVOC value, 192 µg.m⁻³, which borders on the recommended maximum limit of 200 µg.m⁻³, was measured in winter.

An office in Mladé Buky

The highest Pentanal and Hexanal concentrations in the indoor air of this building were measured in autumn. The obtained data did not reveal any dependence of the said concentrations on relative air moisture content.

Based on the conducted measurements of indoor air quality of this wood-based building it can be stated that concentrations of the monitored aromatic VOC compounds (Benzene, Toluene, Ethylbenzene and Xylenes) are dependent on relative air moisture content, as was demonstrated in measurements conducted in winter and spring. With decreasing relative moisture content the concentrations of emitted compounds decrease as well. Only the measurement conducted in autumn revealed a sharp rise in concentrations of the said compounds. This phenomenon might have been strongly affected by heating in the monitored building (onset of the heating season).

The obtained measurement data also reveal a dependence of monitored terpenes, or terpenoid compounds, on relative air moisture content, particularly in measurements conducted in winter and spring. A similar trend as that in aromatic compounds can thus be observed, upon which concentrations of emitted compounds decrease with decreasing relative air moisture content. Autumn is an exception again, as concentrations of the monitored substances increase sharply. This rise may be accounted for by the onset of the heating season. In another study, (Bari et al. 2015) there was identified the fragranced consumer products as the source of terpenes.

The total amount of emitted VOC in the indoor air of the monitored wood-based building, or the TVOC parameter, is dependent on relative air moisture content particularly in winter, which is illustrated by the fact the decreasing relative moisture content is accompanied by decreasing TVOC concentrations.

Log cabin in Horní Lysečiny

Based on the conducted measurements of indoor air quality of this wood-based building it can be stated that dependence between relative air moisture content and concentrations of the monitored aldehydes (Pentanal and Hexanal) was not established. In both cases the concentrations of monitored substances rise sharply in spring.

In the monitored representatives of aromatic compounds, dependence of concentrations on relative air moisture content was established in autumn and winter measurements. It follows that with decreased relative air humidity the concentrations of monitored compounds decrease as
well. In some monitored compounds (Benzene) a sharp increase in concentrations was observed in spring measurements, which may have been due to use of the building. As for the monitored terpenoid compounds, dependence of their concentrations on relative air moisture content was not established. The similar observation was published in other studies (Park and Ikeda 2006).

On the other hand, total volatile organic compounds (TVOC) show a clear dependence on relative air moisture content in three seasons. Accordingly, decreasing relative air moisture content is accompanied with decreasing TVOC concentration. The highest TVOC value was measured in winter.

Log house in Velké Tippeltovy Boudy

Based on the conducted measurements of indoor air quality of this wood-based building it can be stated that dependence of concentrations of the monitored aldehyde emissions (Pentanal and Hexanal) on relative air moisture content in winter and spring was not established. On the other hand, the dependence was established in autumn measurements.

Based on the measured data it can also be observed that dependence of concentrations of the monitored aromatic compound emissions (Benzene, Toluene, Ethylbenzene and Xylenes) on relative air moisture content was established. This phenomenon is apparent particularly in measurements conducted in autumn and spring. Measurements conducted in winter did not prove such dependence.

With respect to total emission load, or the TVOC parameter, dependence of TVOC concentration on relative air moisture content was not established in the results obtained, particularly in autumn and winter measurements. However, it needs to be stressed that the TVOC values measured in winter range from 215 to 274 µg.m⁻³, i.e. they exceed the recommended limit of 200 µg.m⁻³. (The TVOC value is given including measurement uncertainty).

It can also be observed that in spring measurements dependence of TVOC concentration on relative air moisture content was established. However, the measured high TVOC concentrations need to be taken into account. In spring the TVOC values ranged from 394 to 481 µg.m⁻³, which exceeds the recommended limit twice. Heating of the building by a tile stove can probably be accounted for this fact, but the same results of the great TVOC amount 300 µg.m⁻³ in the indoor air of apartment was published in the other papers (Schlink et al. 2004).

CONCLUSIONS

The primary aim of this paper was to determine the impact of relative moisture content of indoor air in wood-based buildings on the quality of their internal environment. Based on the obtained measurement results it can be stated that concentrations of VOC emissions in the indoor air of wood-based buildings in the Krkonoše Mts. region are influenced not only by air moisture content and the given season but by other factors as well, including the age of the building, material composition of walls, furnishings and type of heating.

Based on the results obtained in the experimental part of this paper we can conclude the following:

1. The highest concentrations of monitored VOC were measured in the log house Velké Tippeltovy Boudy. This is probably due to the fact that a tile stove is situated directly in the monitored room. Heating releases waste gasses and consequently increases temperature in the monitored room, which results in releasing of a higher amount of VOC (or higher VOC concentrations). The conducted measurements did not validate the commonly published
assertion that the higher the age of a building, the lower concentrations of emitted harmful substances. The highest values of the TVOC parameter were measured in a building which is over 200 years old.

2. Results of the experimental part reveal that relative air moisture content affects the VOC concentrations as well as the TVOC values. The dependence of VOC emissions on relative air moisture content was established in almost all the selected representatives of volatile organic compounds.

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Petr Čech, Daniela Tesařová*, Jana Hadačová, Eva Jeřábková
Mendel University in Brno
Faculty of Forestry and Wood Technology
Department of Furniture, Design and Habitation
Zemědělská 1
613 00 Brno
Czech Republic
Corresponding author: tesar@mendelu.cz