CHARACTERIZATION AND ANALYSIS OF VERY VOLATILE ORGANIC COMPOUNDS AND ODORS FROM MEDIUM DENSITY FIBERBOARD COATED WITH DIFFERENT LACQUERS USING GAS CHROMATOGRAPHY COUPLED WITH MASS SPECTROMETRY AND OLFACTOMETRY

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Abstract. Volatile organic compounds (VOCs) from furniture and interior furnishing materials have been proven to pose adverse health effects. However, very VOCs (VVOCs) and odors were rarely taken into account. To bridge this gap, emissions of VVOCs and odors from medium density fiberboard (MDF) coated with different lacquers were characterized using gas chromatography coupled with mass spectrometry and olfactometry detection. The results demonstrated that the total VVOC at the 28th d ($TVVOC_{28}$) from the control sample was higher than that of the other three lacquered samples. Alcohol VVOCs were the most abundant chemicals from the control MDF, followed by ketones, esters, and ethers, accounting for more than 90%

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of the total concentration. Also, they were the major odor-contributing substances with higher odor intensities. Nitrocellulose (NC), polyurethane (PU), and water-based (WB) lacquer paintings had a suppressive effect on the emission of certain VVOCs but promotion of the others. After the lacquer paintings, alcohols and ethers were the major components, accounting for 82.3% ~88.0% of the total VVOC. In addition, odors were affected by these three lacquer paintings. Fruity was the dominant odor impression of MDF, NC, and PU decoration MDF, with an odor rating of 4.4, 7.1, and 4.9, respectively. A multiodor mixture was the major odor impression of WB decoration MDF, with an odor rating around 4.0. The odor may differ from one lacquer to another. Additionally, a newly added fishy-like odor arose in PU lacquer painting. From the data analysis of this study, PU and WB decoration MDF might be suitable for furniture or decorative materials due to their lower pollutants and odor emissions. Based on the comprehensive evaluation indices, the latter may be more preferred and was highly recommended for indoor applications.

Keywords: Medium density fiberboard (MDF), odor; lacquer painting, very volatile organic compounds (VVOCs), emission.

INTRODUCTION

With the rapid development of the economy and urbanization, various kinds of lacquered wood-based panels are widely used in the production of fashionable furniture for their beautiful patters, rich colors, and superior water resistance. People spend a substantial amount of time indoors daily (Klepeis et al 2001; Simon et al 2020) and the presence of volatile air pollutants is an increasingly widespread concern. It is a fact that volatile air pollutants from differently lacquer-covered wood-based panels have become one of the major contributors to indoor air pollution, which can directly affect people's mental emotion and production efficiency (Aatamila et al 2011; Wang et al 2022; Jiang et al 2018; Shao Y et al 2018) and even impair human physical health (Adamova et al 2020; Que et al 2013). By far, previous researches on indoor air pollutants have concentrated on volatile organic compounds (VOCs) that mainly included chemical components and emission levels (He et al 2012; Wang et al 2019b), testing and sampling methods (Kim et al 2010), emission mechanism models (He et al 2019; Wang et al 2021; Xiong et al 2019; Zhang et al 2021), environmental factors (Jiang et al 2017; Wang et al 2018), and health risk assessment (Capikova et al 2019; Tong et al 2019; Wang et al 2019a). These results not only contribute to a better understanding of the dynamic quality of indoor air and its relationship with the health of the residents, but also can be used to guide the choice of furniture materials. In addition, the existence of the unpleasant odors and sensory irritations can lead to complaints by a conspicuous number of inhabitants. Some odors might induce both physiological symptoms and reactions (irritation, dizziness, headache, nausea, hematopoietic, central nervous, and respiratory issues) and mental stress (Schiffman 1998). It is, therefore, worthwhile to monitor and optimize the volatile air pollutants emission level.

To date, little effort has been devoted to the emission of very VOCs (VVOCs) from wood-based panels. The German Committee on Health-related Evaluation Procedure for Volatile Organic Compounds from Building Products (AgBB) has proposed a retention range for VVOCs below C_6 (AgBB 2015), which should be given more attention because of their high volatility, strong toxicity, and carcinogenicity. In addition, the ISO 16000-6 standard designated substances that eluted before n-hexane on a nonpolar gas chromatography column as VVOCs. The VVOCs characteristics from solid wood with different lacquers were reported in another research (Wang et al 2020b). Esters and alcohols were determined to be the main VVOCs. Ethyl acetate from the ester VVOCs was the dominant odor substance that may originate from the solvents of UV coatings. More recently, Schieweck identified the presence of VVOCs in a study. The C_4 and C_5 alkanes were identified as the most abundant substances, and they were considered to be propellants from insulating materials. The findings further underlined that the proper selection of construction materials remained important to achieve an acceptable indoor air quality (Schieweck 2021).

The correlation between volatile components and odors (flavors from substances) can be achieved by using gas chromatography in combination with different detectors, including olfactometry (GC-O), mass spectrometry (GC-MS) and flame ionization (GC-FID) (Aith Barbara et al 2020). Gas chromatography–olfactometry harmoniously combines the separation capacity of GC with the sensitivity recognition of human olfactory and has proved to be a valuable and reliable tool for investigating the single-substance separation from complex compounds as well as for the detection and identification of the odorous compounds coming from a wide range of materials. So far, with its unique properties, this approach has found its way into a wide range of fields involving perfume (Ngassoum et al 2004) and food aromas (Zhu and Xiao 2018) and is becoming increasingly common in areas related to environment, medicine, and materials. Wang et al found more than 10 odor compounds from particleboards coated with water-based (WB) lacquer, and aromatics and alcohols were determined to be the main odor impression (Wang et al 2019b). Ghadiriasli et al demonstrated the composition of oak's odor through the extract dilution analysis (OEDA) and two-dimensional GC-MS/O. 97 odor substances were identified during the entire odor testing, consisting of terpenes, aldehydes, acids, and lactones, as well as a small portion of phenols (Ghadiriasli et al 2018). Dong et al examined the odor characteristics from PVC-overlaid medium density fiberboard (MDF) using GC-MS/O. A total of 23 odor compounds were detected and the dominant odor characteristics were determined to be aromatic, sour, and fresh scent, coming from toluene, ethylbenzene, phenanthrene, and dibutyl phthalate (Dong et al 2019). Liu et al identified 11 VOCs from wood-based panels that classified as hazardous air pollutants. Aldehydes had some unpleasant odors, with octanal being the main odor contributor to these aldehydes (Liu et al 2020). The work of Jiang et al investigated VOCs emissions from particleboard. A total of 44 VOCs were identified, consisting of alkanes, aromatic hydrocarbons, carbonyl compounds, alcohols, and esters. Aldehydes, particularly hexanal and pentanal, were listed as the main odorants (Jiang et al 2017).

To our knowledge, this is the first more comprehensive and detailed study of the characteristics of VVOCs and odors emitted from veneered MDF coated with different lacquers, typically used for furniture and decorative materials. The aim of this study was to better investigate VVOCs and odors using GC-MS/O, and to further broaden the detection scope of low-molecular-weight compounds, as well as to provide a clearer understanding of the hazardous substances from lacquer-covered wood-based panels. A 15-L environmental chamber with controlled conditions was used for gas sampling, which could realistically simulate indoor human living conditions. The total VVOC at the 28th d ($TVVOC_{28}$) and the chemical components were qualitatively and quantitatively analyzed, and the possible sources of VVOCs were simultaneously clarified. It will serve as a valuable guidance when choosing these lacquered MDF as furniture or decorative materials, and provide a reference for improving the painting process and developing the reduction measures.

MATERIALS AND METHODS

Materials

The MDF was obtained from a furniture manufacturer in Guangzhou, China, and had a formaldehyde (HCHO) emission level of E1 according to the standard EN 13896. The original dimensions were 1200 × 1200 × 18 mm, and eucalyptus was used as the main raw material for MDF production. The density and MC varied from 0.7 to 0.8 g/cm^3 and 8% to 12%, respectively. The hot-pressing temperature was 180-230°C, and the glues used for MDF production were urea–formaldehyde resin. The dimensions of the secondary machined specimen were 400 × 400 × 18 mm, and 0.25 mm thick *Fraxinus mandshurica Rupr* veneer was glued to the specimen surface on a hot press using a 6:4 mass ratio of urea–formaldehyde resin and polyvinyl acetate adhesive. The amount of glue to be used on one side veneer was 150 g/m^2. The hot-pressing temperature, time, and pressure for veneer lamination were 100°C, 3 min, and 1 MPa, respectively. The sample measuring 150 × 75 ×18 mm was prepared in a wooden factory. A self-adhesive aluminum foil was attached to the edges of samples to avoid leakage of gas. The samples were painted with nitrocellulose lacquer (NC lacquer), polyurethane lacquer (PU lacquer), and WB lacquer. The parameters for these three lacquer paintings were listed as follows: the NC lacquer, Bauhinia paints, transparent undercoat/white matte topcoat, main paint: diluent = 2:1, painted two layers of undercoat (150 g/m^2/session) and two layers of topcoat (150 g/m^2/session), at least 12 h among painting sessions; PU lacquer, Bauhinia paints, transparent undercoat/white topcoat, main paint: diluent: curing agent = 2:1:1, painted two layers of undercoat (150 g/m^2/session) and two layers of topcoat (150 g/m^2/session), at least 12 h among painting sessions; WB lacquer, Xinletian, transparent undercoat/twilight gray topcoat, main paint: diluent (distilled water) = 1:10, painted two layers of undercoat (150 g/m^2/session) and two layers of topcoat (150 g/m^2/session), at least 12 h among painting sessions. Four kinds of MDF were used as testing materials, namely, MDF, NC decoration MDF (NC-MDF), PU decoration MDF (PU-MDF), and WB decoration MDF (WB-MDF). Before the testing, these tested materials were stored indoors for 28 d at a temperature of 20-23°C and an RH of 50% ± 10%.

Methods

Sampling methods. According to GB/T 29899-2013, a 15-L environmental chamber was used for gas sampling, which was independently designed by Northeast Forestry University (Harbin, China). This chamber, consisting of glass materials and silicone hoses, did not release volatile contaminants. The schematic representation of a 15-L environmental chamber is shown in Fig 1. An RH of the chamber was regulated by allowing a portion of the airflow to bubble via distilled water in a glass bottle at a controlled temperature level. An automatic digital temperature and humidity sensor was installed in the air inlet of the chamber to continuously register temperature and humidity. Ultrapure nitrogen gas (99.999% purity, Harbin Liming Gas Co., Harbin, China) was introduced into the chamber as a carrier gas and exchanged with the external environment. The chamber was performed at a temperature of 23°C ± 0.5°C and RH of 50% ± 5% to be as close as possible to a typical indoor environment.

Before loading chamber with measuring sample, the chamber inner surfaces were cleaned until the quantity was acceptable. Blank sampling tube should have sufficiently low concentrations and not contain the target compound. Each measured sample was rapidly placed in the center iron holder of the chamber, with a total exposed area of 0.0225 m^2 and a loading rate of 1.5 m^2/m^3. Ultrapure N_2 was continuously introduced into the chamber at 250 mL/min. The electric fan mounted on the top of the chamber was powered on and held until the testing was completed.

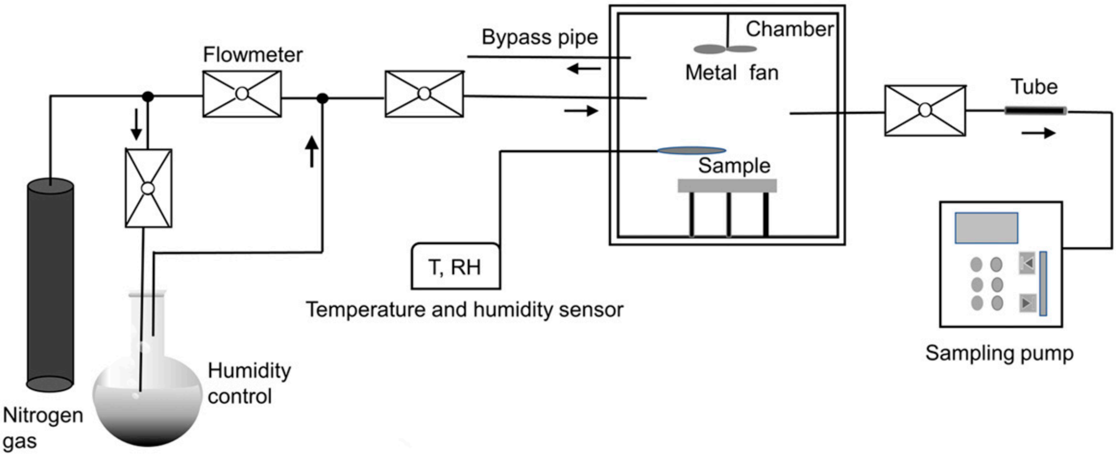

Figure 1. Schematic representation of a 15-L environmental chamber.

Thus, the gases in the chamber could be adequately mixed. The sample cycle time in the chamber was 3.5 h.

The stainless steel tube (produced by Markes International Inc., Llantrisant., UK) containing carbopack C, carbopack B, and carboxen 1000 was used for gas sampling. Before starting sampling, the tubes were pretreated at four temperature intervals (100°C, 200°C, 300°C, and 380°C) with a N_2 rate of 50 ~100 mL/min through a thermal analysis processor (TP-2040; Beijing Beifen Tianpu Instrument Technology Co., Beijing, China). The pretreated time was set to 15 min at each temperature point. The gas sampling flow rate was set at 250 mL/min, and the sampling time was 12 min, 3 L gas was collected into each tube using a miniature vacuum pump (ANJ6513; Chengdu Xinweicheng Technology Co., Chengdu, China). As soon as the sampling was completed, two ends of the tube must be tightly plugged with special brass caps supplied by the adsorption tube manufacturer. The collected gas samples were immediately desorbed for analysis.

Analytical method for GC-MS. The external standard method was used in this experiment. The gas mixture was analyzed using a thermal desorption instrument coupled with gas chromatography–mass spectrometry (TD-GC-MS) and quantified to Chinese National Standard GB/T 29899-2013. The automatic thermal desorption instrument was produced by Markes International. Gases were thermally transferred and refocused on a cryogenic capillary trap. Both cold trap desorption and thermal desorption were performed at a temperature of 300°C. The dry purge and tube desorption held for 5 min and 10 min, respectively. Ultrapure helium gas (99.999% purity) was used as the carrier gas for GC at 1 mL/min.

The DSQ II series quadrupole GC-MS was produced by Thermo Fisher Scientific, and a nonpolar GC column (DB-5MS, 30m length, 0.25 mm inner diameter, 0.25 μm film thickness, Agilent Technologies, State of California, USA) was employed for the gas separation. The temperature program for GC oven was initially started 40°C, and kept for 2 min, then increased to 50°C at 2°C/min and held 50°C for 4 min, and then increased to 150°C at 5°C/min and stayed 150°C for 2 min, and finally increased to 250°C at 10°C/min and maintained 250°C for 8 min. The desorbed gas components were identified based on the retention times and MS detector, and compared with National Institute of Standards and Technology (NIST) and Wiley libraries. The MS detector had a full scan mode and an electron impact energy at 70 eV. The mass-to-charge ratio varied from 40 to 450 amu. The ion source temperature was kept at 230°C. The relative percentage content of each VVOC component was obtained through the area normalization method. Repeated experiments were conducted in triplicate and the mean VVOC concentration was calculated from the duplicated measurements. The $TVVOC_{28}$ was obtained by the concentration summation of odor and nonodor compounds. The total odor intensity at the 28th d (TOI_{28}) was obtained from the summation of the intensities of all odorous substances.

Analytical method for GC-O. The sniffer 9100 olfactory port (Bruchbuhler, Switzerland) was applied in this sniffing experiment. The transmission line temperature was determined to be 150°C so as to prevent condensation of the analytes on the capillary walls. The gas mixture was separated by GC column after thermal desorption and part of it went to the MS for identification and the rest to the sniffer for sniffing operation. The GC effluents were split 1:1. The moist air supplied by a water bottle was continuously added to the glass conical port to reduce damage to nasal mucosa of the odor assessors due to dryness. The schematic representation of gas chromatography–mass spectrometry coupled with olfactory detector is shown in Fig 2. The time-intensity method was used in this experiment. Six-point odor intensity (from 0 to 5) was used to determine the odor grade according to Japanese Standards (Ministry of the Environment Law 1971). The correlation between odor intensity and characterization is shown in Table 1. Based on some certain screenings and training recommendations in accordance with International Organization for Standardization 2017 (ISO 12219-2017-7), five trained and experienced odor assessors

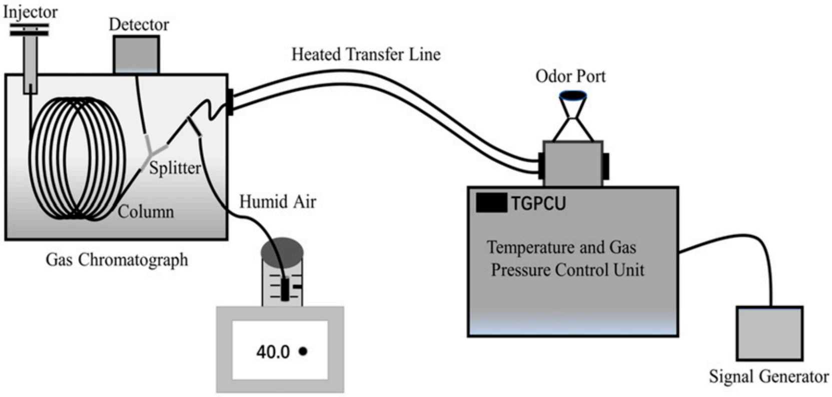

Figure 2. Schematic representation of gas chromatography–mass spectrometry coupled with olfactory detector.

(aged between 20 and 30 yr old, with no smoking history and no olfactory impairment or allergic rhinitis) were selected to form an odor assessment group to evaluate these odors. All assessors were already screened for sensitivity, motivation, ability to concentrate, and ability to remember and identify odor characteristics. Some activities, such as eating or drinking strongly irritating foods and chewing gum, were prohibited for 5 h prior to taking this GC-O testing. In addition, they were not allowed to use heavy cosmetics and perfumes, or strong deodorants during the day of the olfactory assessment. The sniffing results were simultaneously registered, including the retention time, odor type, and intensity grade of the odoriferous stimuli. At least two assessors smelled the same odor at the same retention time, which was used for the sniffing results. The final odor intensity was an average from the same sniffing results.

According to the National Standards Authority of Ireland in EN 13725-2003, the sniffing laboratory with a temperature of 21 ~23°C and a RH of 40%, was required a good ventilation and no other smell during the entire sniffing operation.

Risk evaluation method. Since some small changes in chemical structure may have a significant effect on biological activity, particularly if toxicity is mediated by binding to receptors, it is very, therefore, necessary to consider certain minimum criteria when using predictive methods for hazard assessment. The lowest concentration of interest (LCI) is an assessment level of pollutants, above which adverse effects are potentially to arise in the indoor environment (Lu et al 2020). Based on LCI guidelines (AgBB 2015), they are required to quantify using their individual calibration factors when the concentration of substances in the test chamber exceeds 5 μg/m^3. For each compound *i*, the R_i is equal to the ratio of C_i to LCI_i, where C_i is the chamber concentration of compound *i*. If R_i is <1, it indicates that there is no effect. When several compounds are detected at concentrations >5 μg/m^3, an additive effect is assumed, and then the risk index *R*-value (the sum of all R_i) will not exceed the value 1. However, because of insufficient studies and experimental data, the EU-LCI values for some chemicals are not directly obtained. In such case, if test data are available for a range of structurally closely related chemicals, it is possible to confidently extrapolate from data-rich compounds to data-poor ones. But even so, some EU-LCI data are still not available. According to health-related evaluation procedure criteria for VOC emissions from building products of AgBB, $TVVOC_{28}$, TOI_{28}, the risk index

Table 1. The correlation between odor intensity and odor characteristic.

Odor intensity	0	1	2	3	4	5
Characterization	None	Very weak	Weak	Moderate	Strong	Very strong

R-value, and the concentrations of nonassessed compounds were used as evaluation indicators. The VVOC components without LCI values are calculated and to avoid the risk of positive evaluation of materials that release substantial amounts of nonassessable substances. The detailed evaluation procedures were referred to Wang's previous study (Wang et al 2020a).

RESULTS AND DISCUSSION

VVOC Emission Characteristics and Source Analysis of MDF Coated with Different Lacquers

Figure 3 shows $TVVOC_{28}$ concentration and TOI_{28} of MDF coated with different lacquers. As shown in Fig 3, the control sample (MDF) had the highest maximum $TVVOC_{28}$ concentration of 426.78 μg/m^3, followed by NC-MDF (223.65 μg/m^3), WB-MDF (205.27 μg/m^3) while the PU-MDF was the lowest (196.05 μg/m^3). The maximum $TVVOC_{28}$ for the control sample were approximately 2-folds higher than the maximum $TVVOC_{28}$ values for the other three lacquered MDF. After the lacquer painting, $TVVOC_{28}$ concentration values displayed a downward trend, decreasing by 47.59%, 51.90%, and 54.06%, respectively. On the whole, $TVVOC_{28}$ concentration values of these three lacquer-covered MDF in the equilibrium stage tended to be at a stable level with little discrepancy. Certainly, as we expected, the three lacquer paintings provided an inhibiting effect on the $TVVOC_{28}$ emissions, this tendency was generally similar with earlier study (Wang et al 2020a). On the one side, when the control MDF was covered with a lacquer layer, part of the lacquer paintings could penetrate into the pore structure of the wood-based panel causing blockage, resulting in volatile pollutants not being easily evaporated and diffused, whereas on the other side, the cured painting film enclosed most of the contaminants within the panel. It was equivalent to adding a mass transfer barrier layer on the surface of MDF to increase the resistance of VVOCs diffusion, thereby reducing its release rate. However, it was found that the TOI_{28} was irregular when the control MDF was coated with different lacquers. Although the control had the largest $TVVOC_{28}$ concentration, the TOI_{28} was not the most powerful in these different kinds of MDF. It was obvious to see that the NC-MDF has the greatest TOI_{28} value, with an odor rating of 12.7, next were 10.9 for PU-MDF, 10.4 for MDF, and 10.0 for WB-MDF. After the lacquer treatments, the TOI_{28} values for these three lacquered MDF were slightly higher or comparable to that of the control MDF, implying that the odors of these lacquered MDF could not disperse sufficiently within a shorter time and required longer ventilation to dissipate. Generally speaking, the lacquer paintings could dramatically mitigate pollutant emission levels. Taking into account $TVVOC_{28}$ concentrations and TOI_{28} values with the gaseous pollutant level for the four kinds of MDF, the WB-MDF was probably the best, followed by the PU-MDF and the NC-MDF, and the control sample (MDF) may be the worst.

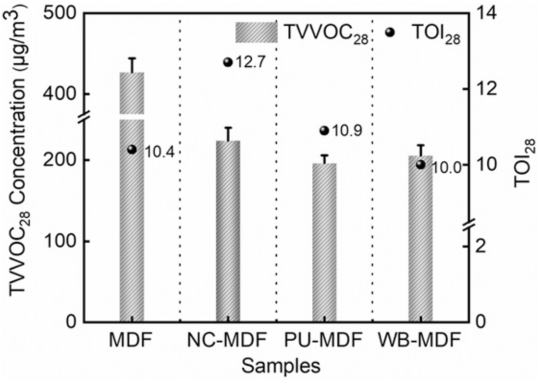

Figure 3. Total very volatile organic compounds at the 28th d ($TVVOC_{28}$) concentration and total odor intensity at the 28th d (TOI_{28}) of medium density fiberboard (MDF) coated with different lacquers.

Figure 4 shows the detailed VVOCs composition and their percentage content of MDF with different lacquers. As described in Fig 4, the major VVOCs detected from the control MDF were divided into the following six groups, namely alkanes, alcohols, aldehydes and ketones, esters, ethers, and others, of which concentrations of alkanes and other components were very small amounts, each

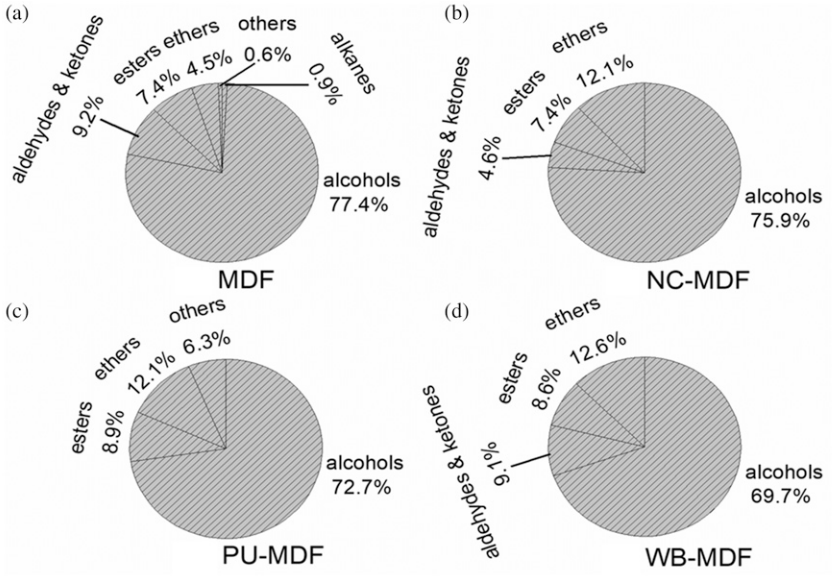

Figure 4. VVOC composition of MDF with different lacquers and their percentage content.

constituting $<1\%$ of the $TVVOC_{28}$ concentration. Alcohol VVOCs were the most abundant species for the control MDF, followed by aldehydes and ketones and esters, which together accounted for more than 90% of the total VVOC. Alcohol VVOCs made up $>75\%$ of the total emission, with ethanol being the most abundant component and 1-butanol as the second contributor, contributing 54.14% and 23.24% of the total VVOC emissions, respectively. Meanwhile, ketones, esters, and ethers VVOCs were the major chemical species detected in the control MDF. The concentrations of acetone and ethyl acetate were also the large fractions of the total emissions for the control MDF. The formation of alcohols and ketones VVOCs in the control MDF was more complicated and probably originated from the degradation of wood carbohydrates and lipids (Fagerson 1969). Also, these two groups also partially derived from adhesives and other additives. For example, ethanol and acetone were commonly used as solvents for adhesives and the latter was also applied as an important cleaning agent for residual adhesives on the edges after high temperature and pressure. The hydrolysis reactions of adhesives in panels could generate 1-butanol, which was consistent with the previous report (Karlsson et al 1989). In addition, ketones VVOCs were formed by chemical degradation of polyunsaturated fatty acids (Chatonnet and Dubourdieu 1998). Ethyl acetate from esters VVOCs was partially derived from the complex chemical reactions in the cellulose and hemicellulose during the hot pressing, the remainder may be generated from the adhesive solvent. The proportion of ethers VVOCs was only 4.5% of the total VVOC. There were also furan derivatives formed by repeated dehydration and rearrangement of carbohydrates during heating (Cullere et al 2013), and tetrahydrofuran of the ethers was produced by the thermal degradation of lignocellulose during hot pressing of the control MDF (Ning et al 2013). The concentrations of dichloromethane and 1,4-dioxane were very small, only 3.85 $\mu g/m^3$ and 2.75 $\mu g/m^3$ respectively (Table 2). These two substances could often be blended with other solvents to facilitate the dissolution of the resin used in the adhesive. Even in small amounts, attention should be paid to them because of their carcinogenicity. It was noted that the main compositions of VVOCs from these three lacquered MDF were roughly

Table 2. The odor characteristics of MDF coated with different lacquers.

Category	No	Compounds	LCI (μg/m^3)	RI	Formula	Odor	Concentration (μg/m^3), R*i* (C*i*/LCI*i*)			
							MDF	NC-MDF	PU-MDF	WB-MDF
Alkanes	1	dichloromethane	—	<600	CH_2Cl_2	—	3.85/—	x	x	x
Alcohols	2	ethanol	1860	<600	C_2H_6O	alcohol-like	231.08/0.1242	68.02/0.0365	62.91/0.0338	57.94/0.0311
	3	1-butanol	3000	647	$C_4H_{10}O$	alcohol-like, sweet	99.22/0.0330	x	x	x
	4	1,2-propanediol	2500	<600	$C_3H_8O_2$	sweet	x	101.78/0.0407	79.67/0.0318	85.16/0.0340
Esters	5	ethyl acetate	3620	<600	$C_4H_8O_2$	fruity	31.41/0.0086	8.75/0.0024	10.16/0.0028	9.49/0.0026
	6	2-methyl 2-propenoic acid-methyl ester	110	701	$C_5H_8O_2$	pungent	x	7.73/0.0702	7.37/0.0670	8.16/0.0741
Aldehydes and Ketones	7	acetaldehyde	1200	<600	C_2H_4O	pungent	x	x	x	18.70/0.0155
	8	3-methyl butyraldehyde	—	686	$C_5H_{10}O$	fruity, apple-like	x	5.56/—	x	x
	9	acetone	1200	<600	C_3H_6O	pungent	39.22/0.0326	x	x	x
	10	3-methyl-2(5H)-furanone	—	974	$C_5H_6O_2$	—	x	4.80/—	x	x
Ethers	11	tetrahydrofuran	1500	627	C_4H_8O	fruity, ether-like	19.25/0.0128	27.01/0.0180	23.51/0.0156	25.79/0.0171
Others	12	1,4-dioxane	73	700	$C_4H_8O_2$	—	2.75/0.0376	x	x	x
	13	1-aminobutane	—	780	$C_4H_{11}N$	—	x	x	3.61/—	x
	14	N, N-dimethylformamide	15	772	C_3H_7NO	fishy-like	x	x	8.82/0.5880	x

similar to that of the control MDF, but their emission levels varied considerably. Additionally, the emission results for these three lacquered MDF were quite similar and no obvious differences were found in the outlines of the components. Alcohol VVOCs were also the largest chemical group, which accounted for 69.7% ~75.9% of the total VVOC emissions. It could be clearly seen that the lacquer treatments had a remarkable barrier effect on the emitted alkane VVOCs, utterly blocking them and reducing them to zero. As far as alcohols VVOCs, 1-butanol was wholly inhibited and the concentration of ethanol decreased to a great extent after the lacquer treatment, but a relatively high level of 1,2-propanediol was found in these lacquer paintings. Similarly, esters and ethers VVOCs were relatively abundant components and were slightly affected when MDF was coated with different lacquers. In relation to ethers VVOCs, this value raised from 4.5% to 12.1% (NC-MDF), 12.0% (PU-MDF), and 12.6% (WB-MDF), a jump of 7.6%, 7.5%, and 8.1%, respectively. The alcohols, esters and ethers VVOCs may be partially derived from panels themselves and another large part was commonly from the solvents, cosolvents and auxiliary additives used in the production and application of these lacquered paintings. 1,2-propanediol was commonly added to coatings as a film-forming additive and 2-methyl 2-propenoic acid-methyl ester was traditionally used as a synthetic raw material for the lacquer coatings. In addition, aldehyde and ketone VVOCs were also correspondingly decreased, especially for PU-MDF, aldehyde and ketone VVOCs were completely confined internally, reducing the hazard posed by the emission of these low-molecular-weight carbonyl compounds. In general, these lacquer paintings could not only improve properties and esthetic characteristics, but also limited the emission levels of VVOCs. It was worth mentioning that the previously mentioned lacquers inhibited the emission of certain substances and provided complete barrier containment to dichloromethane, 1-butanol, acetone, and 1,4-dioxane but increased that of others. Because of the irritating and unpleasant odor of some of the newly added substances and even their high toxicity, the higher ventilation rates and additional placement time of these lacquered MDF needs to be added appropriately and to make sure of these added VVOCs were distributed as possible.

Characterization of Odor Compounds of MDF Coated with Different Lacquers

To further determine the odor compounds of MDF coated with different lacquers, GC-O olfactory technology was applied in this experiment. The odor compound was registered when any one of four specimens had an odor intensity >1. The detail odor characteristics of MDF coated with different lacquers is summarized in Table 2. As can be seen from this table, a total of 14 VVOCs were successfully detected from these samples. Figure 5 shows the odor–time intensity spectrum of MDF coated with different lacquers. As demonstrated in Fig 5, 10 odor compounds were simultaneously measured based on the olfactory analysis and they were mainly composed of alcohols (3 substances), esters (2 substances), aldehydes and ketones (3 substances), esters (1 substance), and others (1 substance). The vast majority of VVOC odorous substances from these lacquered MDF had a rather short retention time and mostly being detected within 10 min, concentrated between 3 and 8 min. Only 1 VVOC odorous substance was retained for longer than 10 min and that was N, N-dimethylformamide. The overall odor intensity of all odor compounds detected from these four samples was not too high, with the highest odor

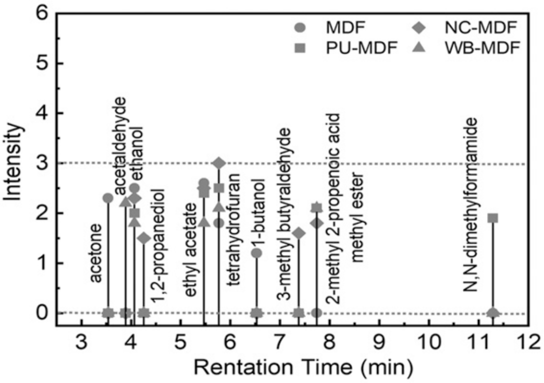

Figure 5. The odor-time intensity spectrum of MDF coated with different lacquers.

intensity being tetrahydrofuran from NC-MDF at 3.0. It was also found that the intensities of ethanol and ethyl acetate reduced to some extent after the lacquer treatments. The two odor compounds (acetone and 1-butanol) were not detected and they were well suppressed after treatment. The tetrahydrofuran was found in both the control MDF and the lacquered MDF, and its intensity was elevated after treatment. Another five odor compounds, namely, 1,2-propanediol; 2-methyl 2-propenoic acid-methyl ester; 3-methyl butyraldehyde; N, N-dimethylformamide; acetaldehyde, were not detected in the control group, and appeared after the lacquer paintings.

As evidenced in Table 2, acetone was reported as pungent odor, similar to the previous finding (O'Neil 2013). Acetaldehyde detected in this study had a pungent odor, similar to the pungent odor reported elsewhere (Hagemeyer 2014), whereas this substance was reported to have a fruity odor (Lewis 2007). Ethanol was perceived as having a pleasant and alcohol-like odor, a similar observation reported in the CAMEO chemicals hazardous materials database. In addition, a fragrant and vinous odor characteristic of ethanol was described by U.S. National Institute for Occupational Safety and Health (NISOH 2010). Similarly, our measured results found 1-butanol was considered to have an odor similar to that of alcohol-like, which agreed with the previous study (Verschueren 2001). In our present testing results, 1,2-propanediol was perceived as slightly sweet. Ethyl acetate was perceived to be a fruity odor, which was consistent with the description of NIOSH (2010), but Sax (1984) also believed this substance had a fragrant odor characteristic (Sax 1984). Our measuring results found that tetrahydrofuran had a fruity odor. Research showed that 3-methyl butyraldehyde had a fruity and apple-like odor characteristic. The 2-methyl 2-propenoic acid-methyl ester was detected with a pungent odor. The N, N-dimethylformamide showed a fishy-like odor, whereas it was also presented a faint and amine-like odor, as reported by NIOSH (2010). The odor compound characteristics are closely related not only to the concentration but also to the medium of the substance. The same substance may have a variety of odor characteristics when the concentration and the medium were changed. Certainly, as seen in Fig 5 and Table 2, ethanol had a considerable concentration but not the highest odor intensity. In contrast, the tetrahydrofuran had a lower concentration but the greatest odor intensity, indicating that the intensity of the same odor compound was influenced by the concentration and that there was no exact correlation between the intensities and their concentrations of different odor compounds.

When mixtures of different odor chemicals were combined together, there were four main ways that influenced the interaction and their relationships could be independence from one another, integration, synergism, or antagonism effect (Cain and Drexler 1974). With integration, the total intensity was the addition of the two odor intensities. With synergism, the total odor intensity was stronger than the addition of the two odor intensities. With antagonism, the total intensity was considered to be less than the addition of the two odor intensities. For an independent effect, the total odor intensity was mainly determined by the odor intensity of a certain odor compound. To better understand the differences before and after the lacquer paintings and to consider the complexity of the interactions of various odor compounds at the same time, the integration effect was used in this odor analysis based on the previous report (Schreiner et al 2017). According to the identification results, the odor characteristics from four samples were classified into the following six categories: alcohol-like, sweet, fruity, pungent, ether-like, and fishy-like. Figure 6 shows the odor radar profile spectra of MDF coated with different lacquers. As depicted in Fig 6(a), fruity was the major odor impression of the control MDF, with an odor rating of 4.4, followed by alcohol-like (3.7), both of which contributed the major determinants to the formation of the overall odor. In addition, pungent and ether-like odors had a relatively positive complement effect to the formation of the overall odor, with an odor rating around 2. Esters, ethers, and alcohols VVOCs were determined as the major odor sources for the control MDF, with the main odor-contributing substances

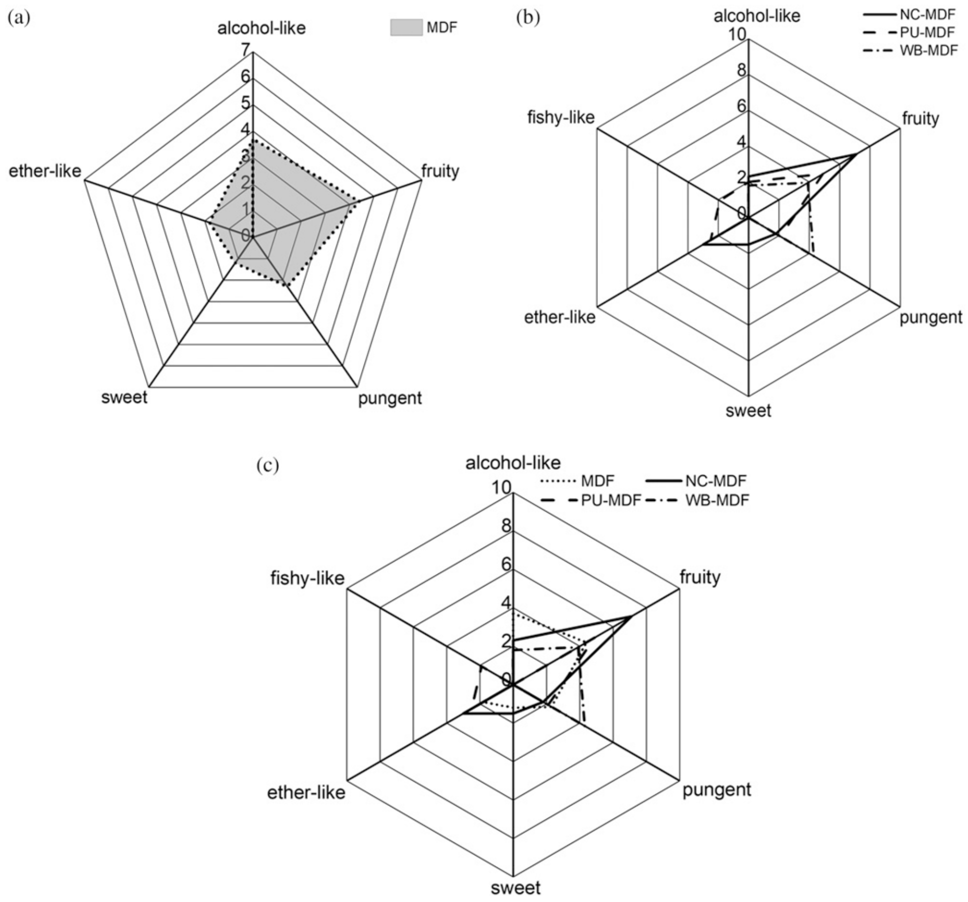

Figure 6. The odor radar profile spectra of MDF coated with different lacquers. (a) MDF; (b) three lacquered MDF; (c) MDF and three kinds of lacquered MDF.

being ethyl acetate, tetrahydrofuran, ethanol and 1-butanol. It could be seen in Fig 6(b) that the fruity was the dominant odor impression of the NC-MDF, with a higher odor rating of 7.1, playing a fundamental and decisive role in the formation of the overall odor, followed by ether-like (3.0), then alcohol-like and pungent, with an odor rating of about 2.0, having an auxiliary modification role to the formation of the overall odor. Esters, ethers, and aldehydes VVOCs were the major odor contributors and the main odor-contributing substances were ethyl acetate, tetrahydrofuran and 3-methyl butyraldehyde. And for the PU-MDF, fruity was also the major odor impression, with an odor rating of 4.9, acting as an active contributor to the formation of the overall odor. Next in order were ether-like (2.5), pungent (2.1), and alcohol-like (2.0), which were the fundamental modifiers to the composition of the overall odor. Esters and ethers VVOCs were the main odor contributors and the main odor-contributing substances were ethyl acetate and tetrahydrofuran. Conversely, the pungent was recognized as a predominant odor impression of the WB-MDF, with an odor rating of 4.3, followed by fruity (3.9) and ether-like (2.1). In general, the WB-MDF was endowed with a multitude of odor blends. Esters, ethers, and aldehydes VVOCs were the major odor providers and the main odor-contributing substances were ethyl acetate, 2-methyl 2-propenoic acid-methyl ester, tetrahydrofuran, and acetaldehyde.

Figure 6(c) shows a comparison of the odor radar before and after the lacquer paintings. As clearly seen from this figure, the odor radar profile after the lacquer painting was generally similar to that of the control group, but with clear differences. Fruity and a multiodor mixture remained the dominant odor impression in these three lacquered MDF. The major odor impression may be changed after the lacquer paintings, but they are basically pleasurable, meaning that most individuals would feel comfortable when they were in such an environment, with the exception of some allergic individuals. It was found that NC, PU, and WB lacquer paintings had a sealing effect on alcohol-like odor, especially for PU, where this odor characteristic completely disappeared after lacquering. And in the meanwhile, another newly added odor characteristic, fishy-like (1.9) occurred in the PU lacquer, which was attributed to the presence of N, N-dimethylformamide. This substance was used on the one hand as a solvent for the synthesis of PU, and on the other hand to dissolve low-solubility pigments. Research results showed that the fruity character of the NC and PU paintings increased in varying degrees, with an increase in intensity of 2.7 and 0.5, respectively. The intensities of more odor characteristics changed to a varying degree after the lacquer treatment. After the lacquer paintings, the odor intensity of pungent decreased slightly in the NC and PU paintings, whereas WB painting increased by 2.0. The sweet intensity remained weak, and was completely inhibited after the PU and WB lacquer paintings, which had little impact on the overall odor profile. The ether-like odor character slightly increased, and their intensities increased from 1.8 to 3.0, 2.5, and 2.1, increased by 1.2, 0.7, and 0.3, respectively, but which had little influence to the overall odor profile.

Risk Evaluation of MDF Coated with Different Lacquers

Figures 3 and 7 show all the risk assessment results of MDF coated with different lacquers.

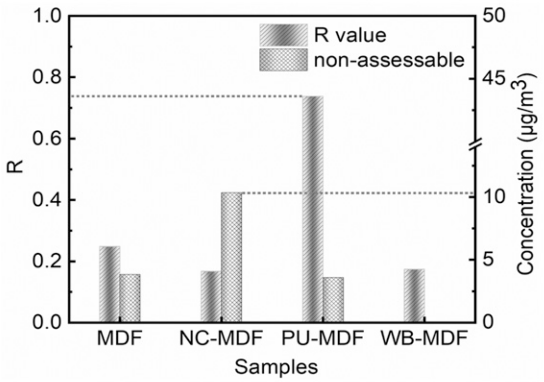

Figure 7. The risk assessment of MDF coated with different lacquers.

As seen in Fig 7, PU-MDF had the largest risk index *R*-value of 0.7390, followed by MDF (0.2488), WB-MDF (0.1744) while NC-MDF (0.1678) was the lowest. After the lacquer paintings, the *R*-value became weaker, with the exception of PU-MDF. The *R*-value of PU-MDF was approximately 3-folds higher than that of the control group, which was directly correlated with the lower EU-LCI value of N, N-dimethylformamide. The concentration of nonassessable substances of the NC-MDF was higher than that of the others, which should be given more attention. As previously mentioned, it could be concluded that the lacquer paintings did block the emission of VVOCs, but at the same time increased the odorous characteristics. Although the *R*-value and odor intensity of the control MDF were not very strong, the $TVVOC_{28}$ were the highest. In terms of VVOCs contaminant emissions and its evaluation levels, it was not recommended to use the MDF directly as furniture or other decorative materials for interior application whenever possible. Certainly, the MDF covered with various kinds of decorative materials or lacquer paintings is highly recommended as furniture materials or other decorative materials, which may be a great option for reducing pollutant emissions. If the MDF is truly needed, additional placement time can be added as appropriate. Even more importantly, manufacturers are expected to address the issue of pollutants and odor emissions at the source of wood-based panel production and they can seek greener, more environmentally friendly solvents to substitute the usage of alcohols, ketones, and ester solvents and use modified glues and low-odorous wood raw materials, thus, manufacturing environmentally friendly panels. Though the lacquer paintings reduced the pollutant emissions, the odors emitted may exceed those of the control group for a shorter limited time and thus also required more attention. Among these three lacquered MDF, NC-MDF had not only the largest $TVVOC_{28}$ concentration but also the maximum odor intensity value. As far as its environmental characteristics are concerned, it is not advocated for use as furniture or other decorative materials within a limited period of time after painting. It is, therefore, highly recommended to keep NC-MDF in a well-ventilated location for a considerable period of time before using it. If it is necessary to use NC-MDF indoors for a short time after lacquering, the high ventilation rates are one of the options for addressing indoor pollutant and odor problems. Although taking both energy consumption and air quality aspects sufficiently into account demands, an optimum air exchange rate might be confirmed, above which is economically viable. The PU-MDF and WB-MDF might be a better choice as furniture or other decorative materials due to their lower pollutants and odor emissions. When the risk index *R*-value and nonassessable compounds were simultaneously under consideration, the evaluation results may be changed. When emissions of materials were considered comprehensively, WB-MDF might be more preferable and was suggested for indoor applications based on the lower evaluation indices of $TVVOC_{28}$, TOI_{28}, *R*-value, and concentrations of nonassessment compounds. Certainly, VVOCs are only one type of volatile pollutants and do not represent the full information about these materials. When occupants choose these lacquer-covered materials for indoor applications, the emissions of VOCs, SVOCs, and carbonyl compounds should also be taken into account.

CONCLUSIONS

In this study, a 15-L environmental chamber, combined with multisorbent adsorption tubes, was used for gas sampling. Emissions of VVOCs and odors from MDF coated with different lacquers were investigated by gas chromatography coupled with mass spectrometry and olfactory detection. The results indicated that the lacquer paintings had an impact on VVOCs and odor emissions. The $TVVOC_{28}$ from the control MDF was higher than that of the other three lacquered samples. After painting, $TVVOC_{28}$ was decreased by 47.59%, 51.90%, and 54.06%, respectively. The most abundant VVOCs components from the control sample were alcohols, followed by ketones, esters, and ethers, accounting for more than 90% of the total. Also, they were the major odor-contributing substances with higher odor intensity values. After the lacquer paintings, alcohols and ethers VVOCs were the major chemicals, which accounted for 82.3% ~88.0% of the total VVOC. It was shown that these three lacquers had a suppressive effect on the emission of certain VVOCs but increased that of the others. In addition, odors were affected by these three lacquer paintings. The TOI_{28} for these three lacquered MDF were slightly higher or comparable to the control MDF. Fruity and a multiodor mixture were the dominant odor impression of these kinds of lacquered MDF. The three lacquers had a strong inhibition of the alcohol-like and sweet, but increased the ether-like. A newly added fishy-like arose in PU lacquer painting. The PU-MDF and WB-MDF might be a better choice as furniture or other decorative materials due to their lower pollutants and odor emissions. Based on the comprehensive evaluation indices of the $TVVOC_{28}$, TOI_{28}, *R*-value, and concentrations of nonassessment compounds, the WB-MDF may be more preferred and the WB paints is highly encouraged to use indoor to reduce indoor air pollution problems if possible.

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