論　文（Original article）

Emission of volatile organic compounds during drying of veneer: Red meranti (*Shorea* sect. *Rubroshorea*), larch (*Larix* sp.), and sugi (*Cryptomeria japonica* D. Don)

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Abstract

Estimation of the amount of volatile organic compounds (VOCs) emitted from the wood-based material industry has become an urgent matter following the 2004 amendment of the Air Pollution Control Law to prevent air pollution from suspended particulate matter (SPM) and photochemical oxidants by controlling VOC emission from factories. At the same time, aldehyde emission from wood products has raised concerns over its effect on human health. In this study, emissions of VOCs and aldehydes during veneer drying were estimated. Wood species commonly used for veneer, red meranti (*Shorea* sect. *Rubroshorea*), larch (*Larix* sp.), and sugi (*Cryptomeria japonica* D. Don), were dried at 140–180ºC, and the discharged air was analyzed by high-performance liquid chromatography (HPLC) and gas chromatography / mass spectroscopy (GC/MS). The emissions of VOCs and aldehydes increased with increasing temperature for all three species. Emissions from sugi wood were higher than from the other species, likely because of the longer drying time due to its high moisture content. The dominant compounds in the discharged air were terpenes. Moreover, discharged amounts of VOCs and aldehydes from the industrial drying process were estimated using the experimental data obtained in this study. These results can be of help in a voluntary approach by the wood-product industry to establish effective emission control of VOCs and aldehydes.

Key words : Veneer, Drying, Volatile organic compounds (VOCs), Aldehydes

1. Introduction

Estimation of the amount of VOCs emitted from the wood-based material industry has become an urgent matter following the 2004 amendment of the Air Pollution Control Law to control the emission of VOCs from factories (Ministry of the Environment, 2004a). When a mixture of VOCs and nitrogen oxides (NOx) is irradiated by ultraviolet rays, photochemical oxidants are generated, and VOCs from factories generate suspended particulate matter (SPM) (Ministry of the Environment, 2004b). The amendment adopts the concept of "best policy mix", an appropriate combination of legal control and voluntary approach by business entities to establish effective emission control of VOCs. As for the wood products industry, facilities including those involving the coating process with high emission of VOCs are subjected to legal control, and the rest require a voluntary approach. However, the range of legal control is to be reexamined if the voluntary approach does not give satisfying results, and those factories presently excluded from legal control are likely to come under control. Meanwhile, complaints about the odors from wood products facilities have been made by neighboring residents. At the same time, aldehydes released from wood and wood products have raised concerns over its effects on human health, and the Ministry of Health, Labour and Welfare proposed a set of guidelines (Ministry of Health, Labour and Welfare, 2002; Tohmura et al., 2005). Under these circumstances, it is important to clarify the components, particularly VOCs and aldehydes, of discharged air from wood products facilities in Japan.

The Air Pollution Control Law stipulates that the concentration of VOCs must be measured using a flame ionization detector (FID) or non-dispersive infrared (NDIR) analyzer, and be described in parts per million carbon (ppmC). This equipment makes it possible to easily determine whether or not the concentration of VOCs exceeds the limit value of the regulation; however, the kind and amount of compounds in discharged air cannot be evaluated. A number of foreign research projects have used this equipment to measure VOCs released during the wood drying process; however, each emitted compound is not always analyzed (Ishikawa, 2007; Milota, 2000).

Even in the few cases where each compound is measured, the measurement systems differ due to the variation in equipment and conditions of the drying processes (Bengtsson & Sanati, 2004; Cronn et al., 1983; Fritz et al., 2004; Granström, 2003; McDonald et al., 2002; Otwell et al., 2000).

In this study, therefore, VOCs and aldehydes released during the veneer drying process were analyzed by high-performance liquid chromatography (HPLC) and gas chromatography / mass spectroscopy (GC/MS). Using this data, the amount of VOCs and aldehydes discharged during drying of 1 m^3 of veneer was calculated in order to estimate emissions from industrial wood-drying facilities. These results can contribute to a voluntary approach by the wood products industry to establish effective emission control of VOCs and aldehydes.

2. Materials and methods

Red meranti (*Shorea* sect. *Rubroshorea*), larch (*Larix* sp.), and sugi (*Cryptomeria japonica* D. Don) veneer measuring 130 × 130 × 3 (thick) mm was used.

2.1 Measurement of VOCs and aldehydes

Fig. 1 shows a schematic representation of measuring the discharged air during drying. Seven pieces of the specimens were heated in an oven (inner size: 30×30×34 cm, DKN302, Yamato Scientific Co. Ltd., Tokyo) at 140–180℃ until the moisture content was reduced to below 10%. The drying temperature and final moisture content were determined based on the conditions of industrial veneer drying. Experimental conditions and moisture content of the specimens before and after drying are listed in Table 1. During the drying process, the discharged air was passed through a heat-resistant tube and a drain trap, and was collected in a sampling bag (10 L, GL Science Co. Ltd., Tokyo) fixed in a sampling case equipped with a pump (MP-Sigma 30, Shibata Scientific Technology, Tokyo). Hereafter, the trapped water and the gas collected in a sampling bag are described as "discharged moisture" and "discharged gas", respectively. The discharged air was sampled from the beginning to the end of the drying, and the air flow rate of Pump A was determined so that the amount of discharged gas did not exceed the capacity of the sampling bag. After drying, the discharged gas was sampled to a 2, 4-dinitrophenylhydrazine (DNPH) cartridge (LpDNPH S10L, Supelco Inc., Tokyo) and a Tenax-TA tube (25090-U, Supelco Inc., Tokyo) by Pump B, and the
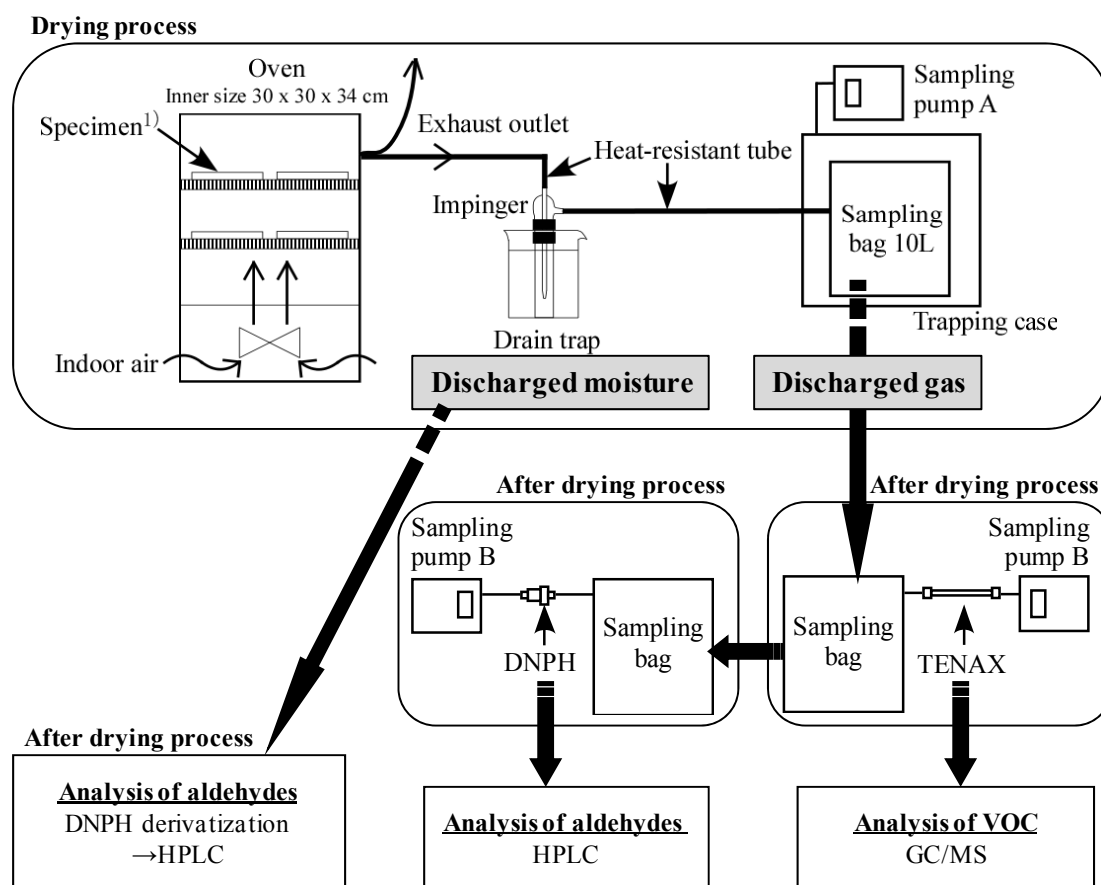

Fig. 1. Schematic representation of the experiments.
[1] Seven pieces of the specimens were put in the oven at a time.

Table 1. Experimental conditions and moisture content of specimens before and after drying.

Specimen	Drying temperature (°C)	Drying time (min)	Flow rate (pump A) (L/min)	Collected air volume (L)[1]	Flow rate (pump B) (L/min)	Sampled air volume (L)		Moisture content (%)[2]	
						DNPH	TENAX	Before drying	After drying
Sugi	140	43	0.20	8.60	0.10	2.50	2.50	109	6
	160	35		7.00				115	6
	180	27		5.40				105	2
Larch	140	14	0.45	6.30	0.10	2.50	2.50	38	7
	160	13		5.85				38	5
	180	12		5.40				38	2
Meranti	140	22	0.45	9.90	0.10	2.50	2.50	66	6
	160	19		8.55				67	5
	180	15		6.75				61	0
Sugi[3]	160	11 (0-11)	0.45	4.95	0.10	2.00	2.00	92	1
		11 (14-25)		4.95					
		11 (28-39)		4.95					

[1] Collected air volume = Drying time × Flow rate (pump A)
[2] n: 3-5
[3] Discharged air was collected three times during drying: stage 1 (0-11 min), stage 2 (14-25 min), stage 3 (28-39 min).

discharged moisture was DNPH derivatized. Moreover, part of the discharged gas was analyzed using an FID (VOC Monitoring System VMS-F, Shimadzu Co. Ltd., Kyoto) in order to compare the concentration values obtained by GC/MS and the regulation values of the Air Pollution Control Law.

Prior to each test, the oven interior was wiped with distilled water and kept at 200°C for 30 min to remove volatile compounds. A blank air sample was taken after the oven settled at 140–180°C. The blank value was omitted from the VOC and aldehyde calculation.

The collected volatiles were removed from the Tenax tube by heating the trap with an automatic thermal desorption system (ATD400, PerkinElmer Japan Co., Ltd., Kanagawa) at 280°C for 15 min. The compounds were cryofocused in a cold trap (air monitoring trap) at 0°C. By heating the cold trap, volatiles were transferred to a HP-5MS capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness, Agilent Technologies Japan, Ltd., Tokyo) and analyzed by GC/MS (GC type 6890, MSD 5973, Hewlett-Packard Japan, Ltd., Tokyo). The temperature program began at 40°C (hold 15 min), thereafter rising to 180°C at 4°C/min (hold 15 min), and to 280°C at 5°C/min (hold 15 min). All mass numbers between 15 and 550 m/z were recorded (SCAN technique). Individual compounds were identified by comparison of their mass spectra with the NIST (National Institute of Standard and Technology) Library, published values and NMR spectra as detailed in a previous report (Yatagai et al., 1985). The total volatile organic compound emission was first calculated by combining the peak areas of all identified compounds, after which the relative proportion of individual compounds from the total emission was calculated. Standard toluene

reagent (Kanto Chemical Co., Inc., Tokyo) was used, and the amounts of VOCs were indicated as toluene equivalents. The DNPH derivatives in a DNPH cartridge were desorbed and diluted up to 5 mL using HPLC-grade acetonitrile. The solution was injected into the HPLC (LC-10 ADvp, Shimadzu Co. Ltd., Kyoto). The analysis conditions are listed in Table 2. Standard reagent (CARB 1004 DNPH Mix 2, Supelco Inc., Tokyo) was used for the quantitative determination. The air concentration of VOCs and aldehydes was calculated by dividing the obtained mass values by sampled air volume.

Three to five tests were performed for one condition.

2.2 Discharge amount during drying of 1 m³ of wood

In order to estimate emissions from industrial wood-drying facilities, the amount of VOCs and aldehydes discharged during drying of 1 m³ of veneer was calculated. First, the air exchange rate of the oven used in the experiment was measured based on JIS A 1406 (Method for Measuring Amount of Room Ventilation (Carbon Dioxide Method)) (Japanese Industrial Standard, 1974). The oven was filled with CO_2 gas, and a digital CO_2 gas monitor (Fuso Rikaseihin Co., Ltd., Tokyo) was used to measure the

Table 2. Analysis conditions of HPLC.

Apparatus	SHIMADZU LC-10 Avp System
Column	two STR-ODSI (150 mm × 4.6 mm ϕ) (Shinwa Chemical Industries, Ltd.)
Mobile phase	Acetonitrile/Distilled water
Gradient	40:60 (0-5 min), 100:0 (5-30 min), 100:0 (30-35 min), 40:60 (35-36 min), 40:60 (36-50 min)
Flow rate	1.0 mL/min
Injection volume	20 μL
Oven temp.	40 °C
Detector	UV 360 nm

concentration. The air exchange rate was calculated using the measured values according to JIS A 1406. Then, the discharged amount from 1 m³ of wood was calculated using the following equation:

Discharged amount from 1 m³ of wood (g) = Air concentration (g/m³) × Air exchange rate (m³/h) × Drying time (h) / Total volume of specimens (m³) …… (1)

where air concentration is the value obtained for VOCs and aldehydes in the experiments described in 2.1, and total volume of specimens is the sum of specimen volumes placed in the oven. Formaldehyde was detected both in discharged gas and discharged moisture; therefore, the discharged amount was calculated using Eq. (2):

Discharged amount of formaldehyde from 1 m³ of wood (g) = {Air concentration (g/L) + Concentration in discharged moisture collected during sampling of 1 L of discharged gas (g/L)} × 1000 × Air exchange rate (m³/h) × Drying time (h) / Total volume of specimens (m³) …… (2)

2.3 Ether extraction

The amount of ether extractives included in specimens before and after drying was measured in order to estimate the amount of compounds included in the specimens before and after drying. First, half the sugi, larch, and meranti specimens were dried at 160°C for 35, 13, and 19 min as well as under the conditions of the experiment described in 2.1. Undried and dried samples were cut into pieces, and ground in a mill to pass through a 2.0-mm screen. Ground samples in 5-g units were Soxhlet-extracted with diethyl ether at 40°C for 5 h. The solvents were evaporated in vacuo at 40°C. Other 1-g ground samples were oven-dried at 105°C until their weight stabilized, after which the moisture content was calculated. From these results, the rate of ether extractives relative to the oven-dried weight of the wood was calculated. Compounds in the extractives were then analyzed using GC/MS. The analysis conditions were the same as described above.

3. Results and discussion

VOCs, formaldehyde, and acetaldehyde were detected from the discharged gas, and formaldehyde was detected in the discharged moisture.

Formaldehyde concentration in discharged moisture and discharged gas is shown in Table 3 and Fig. 2, respectively. In order to determine the ratio of formaldehyde collected
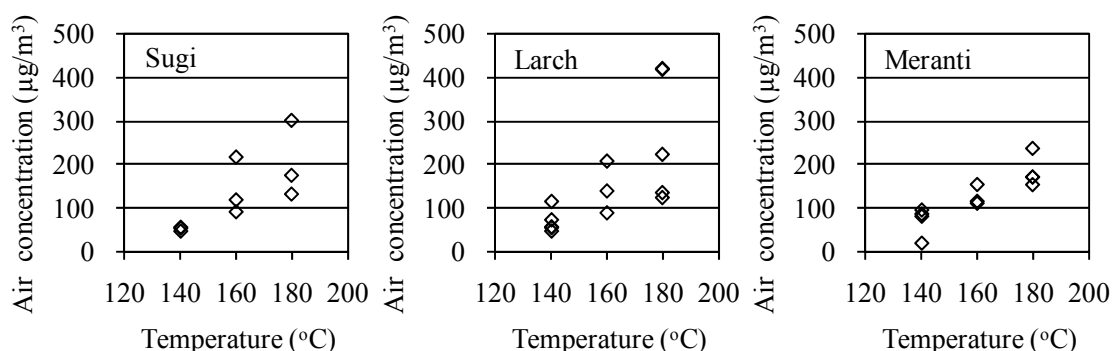
Fig. 2. Formaldehyde concentration in discharged gas.

Table 3.   Formaldehyde concentration in discharged moisture.

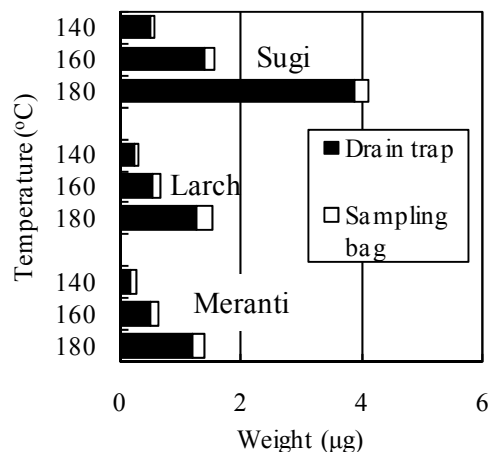| Specimen	Temperature (°C)	Formaldehyde concentration (µg/ml)
Sugi	140	5.2
	160	10.9
	180	21.0
Larch	140	3.1
	160	4.1
	180	7.8
Meranti	140	2.6
	160	3.7
	180	7.0

Fig. 3. Comparison of formaldehyde amount in drain trap and sampling bag during sampling 1L of discharged gas.

Emission of volatile organic compounds during drying of veneer: Red meranti (*Shorea* sect. *Rubroshorea*),
larch (*Larix* sp.), and sugi (*Cryptomeria japonica* D. Don)

119

in the trapped water to the total discharged amount, the mass of formaldehyde collected in discharged moisture and discharged gas during sampling of 1 L of discharged air was calculated from the data in Table 3 and Fig. 2, and compared in Fig. 3. The data clearly shows that most of the discharged formaldehyde was collected in the drain trap. This result indicates that condensing and collecting discharged moisture or using a water scrubber at wood processing plants efficiently removes formaldehyde.

Fig. 4 and Fig. 5 show the air concentration of acetaldehyde and VOCs in discharged gas, both of which increased with increasing drying temperature. Fig. 6 shows the major compounds emitted during drying. Sesquiterpenes were mainly released during sugi drying and monoterpenes and sesquiterpenes were detected during larch and meranti drying. Tables 4 and 5 list the results of ether extraction. Many compounds detected from the drying process were found in ether extractives, and they are reported to be included in untreated sugi, larch, and meranti wood (Arihara et al., 2004; Bisset et al., 1971; Gornostaeva et al., 1982a, b; Kolesnikova et al, 1972; Nagahama and Tazaki, 1993; Nagahama et al., 1995; Nagahama et al., 1996; Nagahama et al., 1998; Nagahama et al., 2000; Ruuskanen et al., 2007). It has also been reported that sugi generally includes ferruginol, but the amount depends on the individual tree and it is not susceptible to wood drying (Kano et al., 2004). In this study, a high amount of ferruginol was found in the ether extractives of sugi wood; however, the amount in the discharged gas was lower than that of the other compounds,

and no significant relationship with drying temperature was found. These results indicate that most ferruginol included in untreated sugi wood is not likely to be discharged during drying.

Table 6 shows the VOC concentration measured by GC/MS and FID. In literature concerning the Air Pollution Control Law, the concentration is often measured by FID. However, the values of air concentration ($\mu g/m^3$) obtained by GC/MS and parts per million carbon (ppmC) measured by FID differ significantly as indicated in Table 6. Therefore, care must be taken not to confuse these two units.

Next, the discharged amount from 1 m^3 of wood was calculated by assigning air concentration and concentration in discharged moisture to Eq. (1) or (2). The results are listed in Table 7. The air exchange rate of the oven, calculated based on JIS, was 0.48 m^3/h. The discharged amount of VOCs and aldehydes increased with increasing temperature for all three species. Sugi showed the highest discharge amount, followed by larch, with the lowest being meranti. One reason for the high discharged amount for sugi is considered to be the longer drying time due to its higher initial moisture content compared to that of the other species. Therefore, the relationship between discharged amount and drying time was investigated (Fig. 7). The discharged amount of formaldehyde, acetaldehyde, and VOCs increased with increasing drying time. The discharged amount increased even after the moisture content was decreased to nearly 0%. It has been reported that the
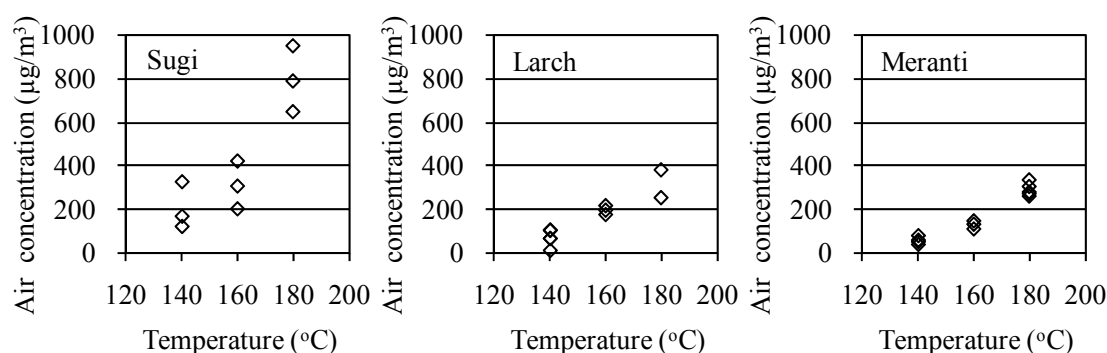

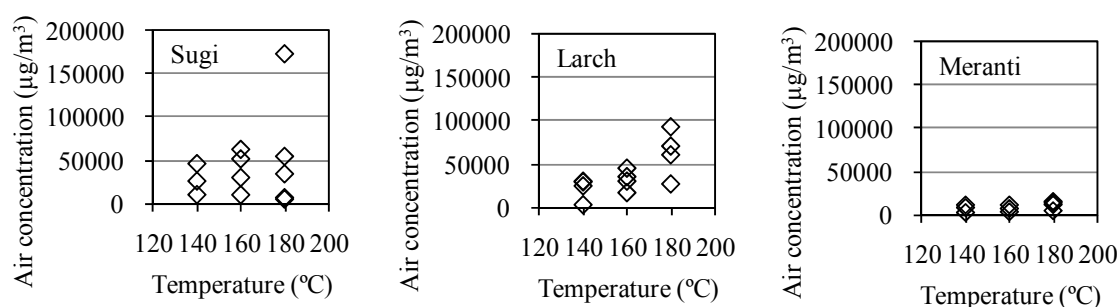Fig. 4. Acetaldehyde concentration in discharged gas.

Fig. 5. VOCs concentration in discharged gas.

discharged amount of VOCs and formaldehyde increases with decreasing moisture content (Granström, 2003; Otwell et al., 2000). These results indicate that the discharged amount of VOCs and aldehydes could be decreased by preventing extra drying time.

Assuming that the difference between the amount of ether extractives of undried and dried wood is the maximum amount of VOCs discharged during drying, the maximum amount of VOCs discharged during drying 1 m³ of wood can be calculated to be 254−2204 g as shown in the rightmost column in Table 4. During actual industrial drying, only a part of the maximum amount would be

Table 4. Ether extractives of undried and dried wood.

| Specimen | Ether extractives (%)[1] | | | | | | Oven-dried weight of 1m³ wood (g) [3] | Ether extractives (g/m³ wood) [4] |
	Undried	Average	Dried[2]	Average	Undried - Dried	Average		Undried - Dried
Sugi heartwood	2.47		1.13		1.34			
	1.31	1.75	0.91	1.08	0.40	0.67	330,000	2204
	1.45		1.19		0.27			
Sugi sapwood	0.30		0.14		0.16			
	0.63	0.54	0.14	0.17	0.49	0.37	320,000	1176
	0.68		0.24		0.45			
Larch	1.08		1.03		0.05			
	0.99	1.02	0.91	0.97	0.08	0.05	500,000	254
	1.00		0.98		0.02			
Meranti	0.96		0.75		0.21			
	0.97	0.95	0.88	0.79	0.09	0.16	410,000	662
	0.91		0.73		0.19			

[1] Relative to oven-dried weight
[2] Drying temperature: 160ºC
[3] Calculated from the oven-dry specific gravity of sugi, larch, and meranti
(Fushitani et al., 1985; Terasawa & Tsutsumoto, 1992; Sato & Sumi, 1971).
[4] Ether extractives (g/m³ wood) = Ether extractives (undried - dried) (%) / 100 × Oven-dried weight of 1 m³ wood (g)

Table 5. Results of GC/MS analysis of ether extractives. (%)

| Compounds | Sugi heartwood | | Sugi sapwood | | Larch | | Meranti | |
	Undried	Dried	Undried	Dried	Undried	Dried	Undried	Dried
Ferruginol	16.0	26.4	7.8	31.7				
15-Phyllocladene	13.6	12.1	3.0	6.0				
δ -cadinene	3.2	2.5	5.1	1.2	4.4	1.0	0.6	2.6
Calamenene	1.5	1.4					0.4	0.6
Cubebene	1.4	1.2			1.2			
8,11,13-Abietatriene	1.2	0.7						
α -Gurjunene	1.1							
γ -Muurolene	1.0							
Cadalene	0.4							
β -Cubebene	0.2							0.5
α -Cubebene	1.7			0.8				0.2
3-Copaene		1.4	13.8					
Acetyl ferruginol		0.9						
1(10)-Aristolene		0.8						
Guaiazulene		0.3						
γ -Eudesmol			6.6					
3,11-Eudesmadiene			4.0					
Cryptopinone				3.1				
Dehydroabietal					11.5	21.0		
M⁺272					9.6	8.4		
γ -Cadinene							1.3	1.5
β -Elemene							0.9	2.7
β -Caryophyllene							0.6	
3-Copaene							0.5	1.1
α -Amorphene							0.5	2.3
Ledene								2.1
Tricosane								0.4
α -Muurolene								0.3
Total	41.2	47.6	40.2	42.9	26.6	30.4	4.7	14.2

Fig. 6. Major compounds released during drying.

Fig. 7. Relationships between drying time, moisture content,
and emissions from 1m³ of veneer. Specimen: sugi;
temperature: 160℃.
[1] Cumulative amount.
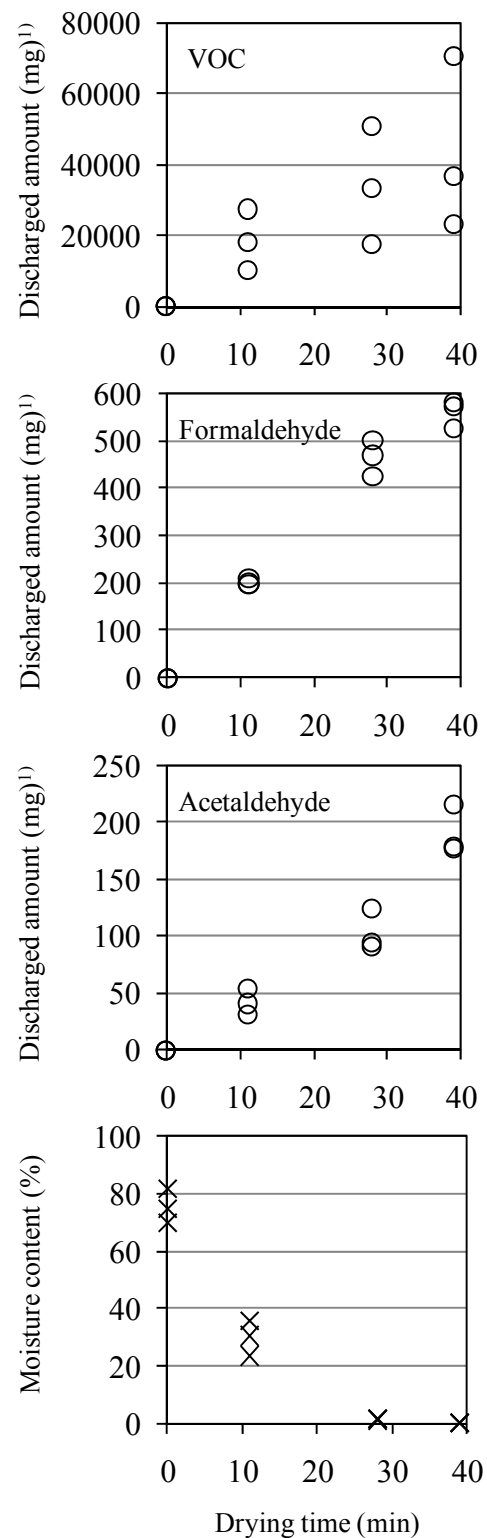
Table 6.   Comparison of VOC concentrations
measured with FID and GC/MS.

Temperature (℃)	FID (ppmC)	GC/MS (µg/m³)
140	10	1,682
160	16	12,634
180	21	22,458

Specimen: larch

emitted. As shown in Table 7, the amount of discharged VOCs during drying of 1 m³ of sugi, larch, and meranti was estimated to be 0.5−35 g. The amount of discharged VOCs during drying of other species (pine, fir, hemlock, etc.) at 49−160ºC has been reported to be 20−4000 g/m³ of wood (Ingram et al., 2000; McDonald et al., 2002; McDonald et al., 2004; Milota, 2003; Milota, 2006; Milota & Mosher, 2006; Shmulsky, 2000a; Shmulsky, 2000b; Shmulsky, 2000c). Our results are somewhat lower but almost the same level as these published values. The discharged amount of formaldehyde and acetaldehyde during drying of radiata pine at 100ºC dry-bulb temperature (DBT) with 70ºC wet-bulb temperature (WBT) has been reported to be 1.1 and 8.7 g/m³ of wood, respectively (McDonald et al., 2002). Our results were lower than these values (Table 7), possibly because of the shorter drying time in our experiments.

Moreover, the discharged amount during industrial drying was calculated from data reported by Kawarada et al. (2008) using Eq. (1), and compared to our results (Table 8). The discharged amounts of VOCs in our results were lower than that in the industrial drying. The reasons for the difference are considered to be the individual varieties of sugi, irregularity of air concentration and contamination in the industrial drying duct, and the difference in species (a species of Southeast Asia was dried together with sugi during the industrial drying).

4. Conclusions

Red meranti, larch and sugi veneer was dried under conditions generally used in industrial drying processes (140−180°C), and discharged VOCs and aldehydes were evaluated. The following results were obtained:

1. VOCs, formaldehyde, and acetaldehyde were emitted from all three species during drying. The amounts increased with increasing drying temperature and time.

2. Most of the discharged formaldehyde was removed by means of a drain trap.

3. Main VOCs emitted during drying were terpenes.

4. Discharged amount from industrial drying was estimated using the results obtained in this study.

These results indicate that emissions of VOCs and aldehydes can be decreased by controlling drying time and temperature. Terpenes, however, are reported to have several beneficial functions, and it is desirable to investigate their efficient utilization. Formaldehyde, on the other hand, is a carcinogen and should be removed. It is also indicated from the results obtained in this study that formaldehyde can be efficiently removed from discharged air by water scrubbing.

Table 7.   Emissions of VOC and aldehydes during drying.

Specimen	Temperature (ºC)	Time (min)	Emissions from 1m³ green wood (g)			Ovendried weight of 1m³ wood (g)[1]	Discharged VOC (%)[2]
			VOC	Acetaldehyde	Formaldehyde		
Sugi	140	43	13.1	0.20	0.54	330,000	0.0040
	160	35	26.5	0.25	1.23	330,000	0.0080
	180	27	34.8	0.48	2.86	330,000	0.0105
Larch	140	14	0.5	0.02	0.10	500,000	0.0001
	160	13	3.7	0.06	0.20	500,000	0.0007
	180	12	6.1	0.08	0.42	500,000	0.0012
Meranti	140	22	0.5	0.03	0.13	410,000	0.0001
	160	19	0.5	0.05	0.27	410,000	0.0001
	180	15	1.4	0.10	0.47	410,000	0.0003

[1] Calculated from the oven-dry specific gravity of sugi, larch, and meranti
   (Fushitani et al., 1985; Terasawa & Tsutsumoto, 1992; Sato & Sumi, 1971).
[2] Discharged VOC (%) = Emissions from 1m³ green wood (g) / Ovendried weight of 1m³ wood (g) × 100

Table 8.   Comparison of the data in this study with an industrial drying process (Kawarada et al., 2008).

	Our results (Sugi, 180ºC)	Kawarada et al. (Sugi and other species, 180ºC)
Air concentration of VOC (μ g/m³)	57,197	62,600
Ventilation rate (m³/h)	0.48	16,100
Dried veneer volume (m³/h)	7.9 x 10⁻⁴*	5.4
VOC released from 1m³ veneer (g)	35	187

* Calculated from the dried veneer volume, 3.55×10^{-4}m³, during 27 minutes in our experiment.

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レッドメランチ、ラーチ、スギの単板乾燥工程で排出される
揮発性有機化合物

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要 旨

　大気汚染防止法の改正により工場からの揮発性有機化合物（VOC）の排出規制が始まり、木質建材製造工程における VOC 排出の実態解明が急務となっている。また、アルデヒド類についても木材からの放散が懸念されている。本研究では、合板に使用される代表的樹種、レッドメランチ、ラーチ、スギの生単板を工場での乾燥条件に近い温度（140 ～ 180℃）で乾燥し、この間に排出される VOC とアルデヒド類について、ガスクロマトグラフ質量分析（GC/MS）および高速液体クロマトグラフィー（HPLC）により定性定量分析を行った。いずれの樹種についても、VOC、ホルムアルデヒド、アセトアルデヒドは、乾燥温度が高くなるほど排出量が多くなる傾向が認められた。これらの排出量は、同じ温度ではスギが最も多く、次いでラーチ、レッドメランチの順であった。スギは他の樹種よりも生材含水率が高く乾燥時間が長いことが、排出量の多い原因の１つであると考えられた。また、排出される主な VOC はテルペン類であることが明らかになった。さらに、実験値をもとに工場での排出量を推定することができた。これらの結果は、工場における VOC とアルデヒド類の排出実態把握および排出低減化の自主的取り組みに役立つものである。

キーワード：単板、乾燥、揮発性有機化合物（VOC）、アルデヒド