

Effects of time, temperature, and humidity on acetaldehyde emission from wood-based materials

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Received: 26 December 2013 / Accepted: 5 March 2014 / Published online: 9 April 2014
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Abstract Acetaldehyde emissions from particleboard, fiberboard, and plywood were studied using the small chamber method and high-performance liquid chromatography analysis. The chamber tests were conducted for 1 month to examine the time dependence of acetaldehyde emission. Effects of temperature and relative humidity were also examined. Acetaldehyde emissions from these wood-based materials decreased rapidly and their behavior could be described by an exponential function or by the sum of two exponential functions. This result suggests that in an adequately ventilated atmosphere, the acetaldehyde emission factor decreases quickly following the board's production. Under fixed absolute humidity conditions, the initial acetaldehyde emission factor was larger under higher temperature conditions, but tended to show almost the same value after 14 days. This suggests that higher temperatures promote higher initial concentrations and a faster decline of acetaldehyde. A semi-empirical linear equation was obtained for the early stage relationship between the emission factor and temperature. Under fixed temperature conditions, higher relative humidity caused a larger acetaldehyde emission factor throughout the testing

period, and it did not result in a significantly faster decline in emissions. The relationship between acetaldehyde emission and relative humidity can be described using an exponential function.

Keywords Acetaldehyde · Long-term emission · Temperature · Relative humidity · Wood-based materials

Introduction

Acetaldehyde is one of the common volatile organic compounds (VOCs) derived from wood [1] and wood-based materials [2, 3]. It is also commonly detected in both indoor and outdoor environments, although its concentration in indoor environments is larger than in the surrounding environment [4–6]. Acetaldehyde is classified by the International Agency for Research on Cancer, World Health Organization [7], as a group 2B carcinogenic substance, meaning that it is possibly carcinogenic to humans and thus, in Japan, it has indoor air guideline value of $48 \mu\text{g}/\text{m}^3$ [8].

A Japanese governmental survey, conducted from 2002 to 2005 [9], revealed that about 10 % of newly built dwelling house exhibited excess values of acetaldehyde concentration. Park and Ikeda [10] surveyed 1417 housings, including 311 newly built homes, and reported that the median of acetaldehyde concentration was $101 \mu\text{g}/\text{m}^3$. Sax et al. [5] reported that indoor acetaldehyde concentration increased in winter. This is contrary to formaldehyde concentration, which is the most common pollutant emitted from wood-based materials, and suggests that the behavior of acetaldehyde concentration differs from that of the surrounding environment. Higher concentrations indicate the existence of sources of acetaldehyde emission in dwellings.

Parts of this study were presented at the 29th Annual Meeting of the Wood Technological Association of Japan, Okayama, October 2011, the 62nd Annual Meeting of the Japan Wood Research Society, Sapporo, March 2012, and BIOCOMP 2012, Shizuoka, October 2012.

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Wood, and wood-based materials and their ingredients, i.e., adhesives, are often presented as major sources of acetaldehyde in dwellings [11, 12]. Their contribution to indoor acetaldehyde concentration is estimated to be larger than that from smoking or drinking alcohol indoors [13]. Yagi et al. [14] reported a large acetaldehyde emission factor of 535 $\mu\text{g}/\text{m}^2 \text{ h}$ from laminated lumber using the small chamber method. Tohmura et al. [15, 16] revealed that the addition of ethanol to adhesives causes significant emissions of acetaldehyde from glue-laminated timber, and that acetaldehyde was generated by an enzymatic process of alcohol dehydrogenase contained within wood tissue. Although most cases of acetaldehyde emission from wood-based materials are relatively small [3] and unlikely engaged with ethanol addition to adhesives, acetaldehyde emissions from wood-based materials can still affect indoor air quality [11]. The behavior of acetaldehyde emissions from wood-based materials in residential environment remains unclear. Knowledge of the emission behaviors of building materials and the influencing factors would be helpful for estimating and reducing indoor air pollution. Specially, panel-shaped wood-based materials are usually used for interior finishing materials in dwellings and have larger emission area in indoor spaces. They could have larger influence in indoor air quality than beam-shaped materials such as glued laminated timber. However, the properties of acetaldehyde emission from panel-shaped wood-based materials have not been understood well enough. Therefore, in this study, the effects of time, temperature, and humidity on acetaldehyde emission from wood-based materials such as particleboards, fiber boards and plywood were examined using the small chamber method.

Materials and methods

Materials

Table 1 shows a list of specimens. The particleboard (three layered) and fiberboard specimens were commercial products provided directly from manufacturers. The specimens were sealed in plastic bags and transported to the laboratory. Plywood specimens were fabricated from logs in a laboratory. Adhesives for the plywood specimens were also commercial products sourced directly from manufacturers and were prepared according to the instructions provided. Karamatsu (Japanese larch, *Larix leptolepis* Gordon) and sugi (Japanese cedar, *Cryptomeria japonica* D. Don) specimens were intended to represent structural plywood. Todomatsu (Sakhalin fir, *Abies sachalinensis* FR. Schmidt) specimens were intended to represent interior plywood. The specimens were cut into 160 \times 160 mm test pieces. The

Table 1 List of specimens

Specimen	Adhesive	Thickness (mm)	Plies	Density (g/cm^3)
Particleboard M ^a	MUF	12	–	0.72
Particleboard U ^a	UF	12	–	0.69
MDF M ^a	MUF	9	–	0.72
MDF U ^a	UF	9	–	0.72
Insulation board ^a	Not used	10	–	0.26
Hardboard ^a	Not used	5	–	1.00
Karamatsu plywood	PF	9	3	0.46
Sugi plywood	PF	9	3	0.35
Todomatsu plywood 1	MUF	9	3	0.43
Todomatsu plywood 2	Latex-based non-formaldehyde	9	3	0.43

MUF melamine–urea–formaldehyde, UF urea–formaldehyde, PF phenol–formaldehyde

^a Commercial product

rear sides and edges of the test pieces were sealed with VOC-free aluminum tape leaving a 148 \times 148 mm emission surface. All specimens were examined for time dependence. For temperature and relative humidity dependence, manufacturer-provided boards were examined.

Small chamber method

Small chamber testing was conducted according to JIS A 1901 [17]. Twenty-liter stainless steel chambers with clean air supply equipment were used with an air exchange rate of 0.5 h^{-1} . A pair of test pieces was placed into the chamber to obtain a loading factor of 2.2 m^2/m^3 . For the test of time dependence, the chambers were kept at 28 $^\circ\text{C}$ and relative humidity of 50 % for 4 weeks. Air sampling was performed at 1, 3, 7, 14, and 28 days after placing the test pieces in the chambers. For the test of temperature dependence, the chambers were set at 23, 28, and 33 $^\circ\text{C}$. Absolute humidity was maintained at 13.6 g/m^3 to achieve an equal water vapor inflow to the standard test condition (28 $^\circ\text{C}$, 50 % R.H.). Air sampling was performed after 1, 3, 7, and 14 days. For the test of relative humidity dependence, the chambers were kept at relative humidity conditions of 20, 50, and 80 % at 28 $^\circ\text{C}$. Air sampling was performed after 1, 3, 7, and 14 days. The chamber air was collected with a 2,4-dinitrophenylhydrazine (DNPH) cartridge (GL-Pak mini AERO DNPH, GL sciences Inc., Tokyo, Japan) using an air sampling pump (SP208-1000 Dual, GL Sciences). The sampling volume was 10 L at a sampling rate of 167 mL/min.

Determination of acetaldehyde

High-performance liquid chromatography (HPLC) analysis was performed to determine the concentration of acetaldehyde. The DNPH cartridge was diluted with HPLC-grade acetonitrile in a 5-mL volumetric flask. Then, the solution was injected into an HPLC system (L-7000 Series, Hitachi Ltd., Tokyo, Japan) with an octadecylsilyl column (Intersil ODS-3 5 μm , 4.6×150 mm, GL sciences). The injection volume was 20 μL . The HPLC column temperature was maintained at 40 $^{\circ}\text{C}$. The flow of the solvent (acetonitrile/distilled water, 55/45) was maintained at 1.0 mL/min. The wavelength of the UV detector was set at 360 nm. Acetaldehyde determination was calibrated using an acetaldehyde-2,4-DNPH standard (SUPELCO 442434, SIGMA-ALDRICH, MO, USA). The limit of determination was 4 $\mu\text{g}/\text{m}^3$. The emission factor of acetaldehyde was calculated as follows:

$$R = C \times \frac{n}{L} \quad (1)$$

where R is the emission factor ($\mu\text{g}/\text{m}^2 \text{ h}$), C is the chamber air concentration ($\mu\text{g}/\text{m}^3$), n is the ventilation rate (h^{-1}), and L is the loading factor (m^2/m^3).

Results and discussion

Time dependence

Figure 1 shows the relationship between acetaldehyde emission factor and elapsed time. Acetaldehyde emission decreased relatively faster during the first week. The acetaldehyde emission factor was 4–33 $\mu\text{g}/\text{m}^2 \text{ h}$ on day 1. On day 14, no specimen showed an emission factor greater than 10 $\mu\text{g}/\text{m}^2 \text{ h}$. Four weeks later, the emission factor had decreased by 40 to 90 % from day 1, and the highest value was 6 $\mu\text{g}/\text{m}^2 \text{ h}$ for insulation board. The decrease in emission factor became slower as time elapsed. A least squares fitting method was performed with an exponential model [18] as follows:

$$R(t) = R_0 e^{-kt} \quad (2)$$

where $R(t)$ is the emission factor at time t (d), R_0 is the initial emission factor, and k is the decrease coefficient (h^{-1}). A two-phase model describing the sum of two exponential functions [19] was also examined, as shown below:

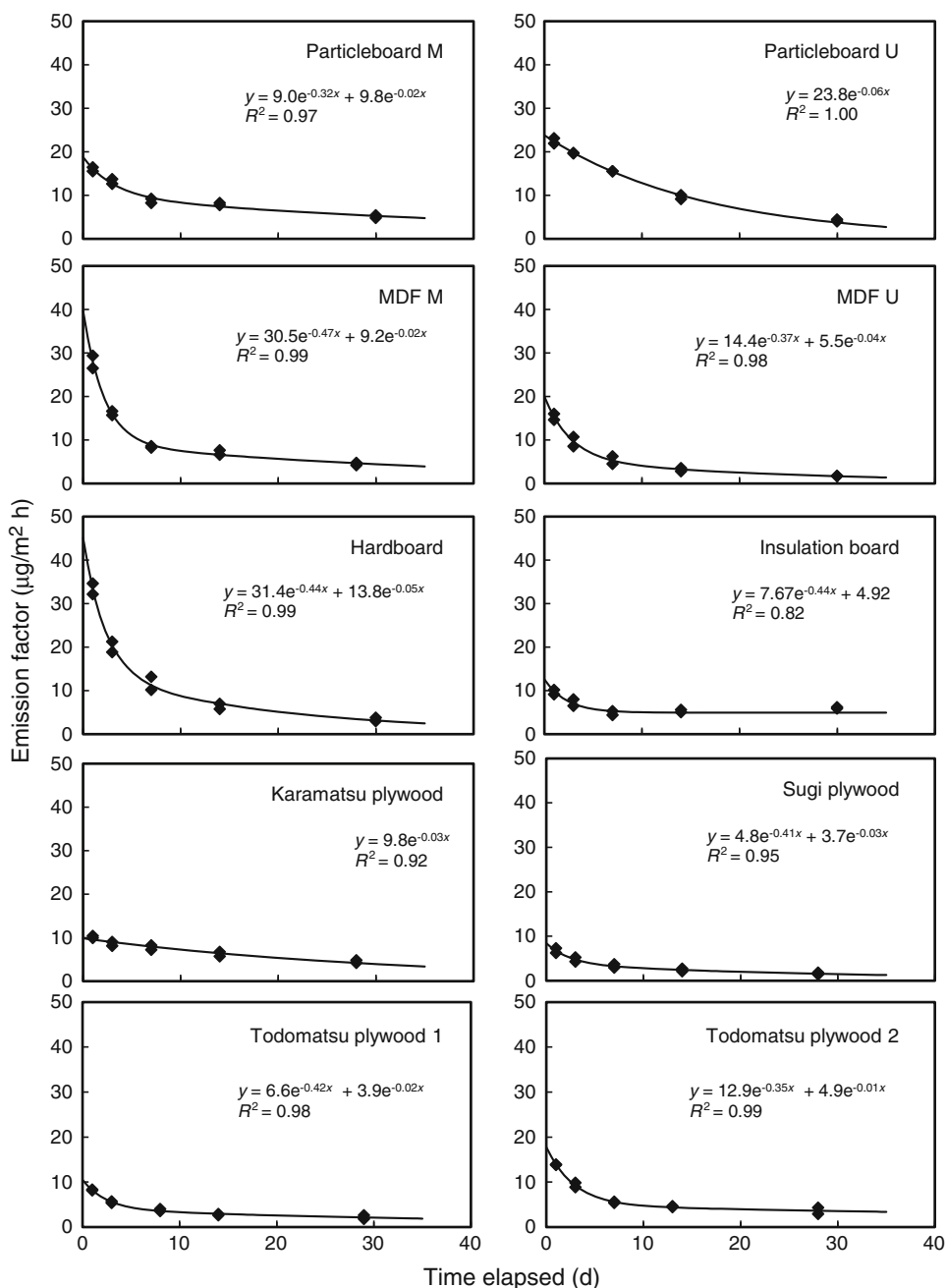
$$R(t) = R_1 + R_2 = R_{01} e^{-k_1 t} + R_{02} e^{-k_2 t} \quad (3)$$

where R_1 is the emission factor for phase 1, R_2 is the emission factor for phase 2, R_{01} and R_{02} are the initial emission factors, and k_1 and k_2 are the coefficients of

decrease for phases 1 and 2, respectively. In the earlier stage of emission (phase 1), R_1 with a larger value of k_1 is dominant. As time elapses, R_1 declines rapidly and R_2 with an apparently smaller value of k_2 becomes dominant (phase 2). This model has been developed to describe emissions from ‘wet’ materials such as wood stain. R_1 and R_2 are presumed to express an evaporation process in the coating surface and a diffusion process in the coating film, respectively [19]. This model can be applied to solid ‘dry’ materials such as polyvinyl chloride flooring [20] and pressed wood products [21], for which R_1 is considered a desorption process in the materials’ surfaces, and R_2 is considered a diffusion process in the materials [20]. The calculation was performed using GNU Octave [22] version 3.2.4 with the optim package version 1.0.17. Particleboard M, MDF M, MDF U, hardboard, sugi plywood, todomatsu plywood 1, and todomatsu plywood 2 were all expressed by Eq. 3; however, the second term showed a constant value ($k_2 < 0.01$) because of an equivalent emission factor observed on days 7 and 14. Particleboard U and karamatsu plywood were expressed by Eq. 2.

These results suggest that in an adequately ventilated atmosphere, the acetaldehyde emission factor will decrease rapidly following the board’s production. It is inferred from the value of parameter k compared with k_1 and k_2 in Eq. 3 that the emissions from particleboard U and karamatsu plywood were assumed dominated by internal diffusion. This result suggests faster acetaldehyde depletion from the surface and the observed emissions were of phase 2, but the reason for this remains unclear. Previous studies summarizing long-term VOC emissions from building materials [23, 24] have expressed emissions from wood-based materials as empirical power law models; however, their parameters do not bear any physical meanings [25]. Figure 2 shows a comparison between Eq. 3 and the power law model of Particleboard M. The power law model tends to overestimate the acetaldehyde emission factor over longer periods. Park and Ikeda [26] suggested that the concentrations of indoor air pollutants of newly built houses are influenced by the aging of emission sources. The emission decrease observed here suggests a rapid decline of indoor concentrations of acetaldehyde in newly built dwellings. Ohira [1] examined a series of solid hardwood and softwood samples with the 20-L small chamber method under the same conditions as those in this experiment, and reported that maximum value of acetaldehyde emission rate on day 28 was 16.8 $\mu\text{g}/\text{m}^2 \text{ h}$. However, the specimens observed in this study showed smaller values on day 28, suggesting that adhesives used in the production of wood-based materials are not major sources of acetaldehyde emissions. Acetaldehyde emissions from the wood-based materials examined in this study declined

Fig. 1 Relationships between acetaldehyde emission factor and elapsed time



rapidly in a properly ventilated environment. Results suggest that the wood-based materials did not exhibit particularly significant influence on indoor acetaldehyde concentration compared with solid woods.

Temperature dependence

Figure 3 shows the relationship between elapsed time and the acetaldehyde emission factor for various temperatures. In an equivalent absolute humidity atmosphere, the emission factor shows a greater value at higher temperature on day 1, but tends to indicate almost

equivalent values by day 7. The emission factor of the earlier period is affected largely by the initial emission factors R_{01} and R_{02} . The relationship between the initial emission factors and absolute temperature is as follows [20]:

$$\log R_{01} \propto \frac{1}{T} \tag{4}$$

$$\log R_{02} \propto \frac{1}{T} \tag{5}$$

where T is the absolute temperature (K). For the earlier emission stage, Eq. 4 can be translated as follows:

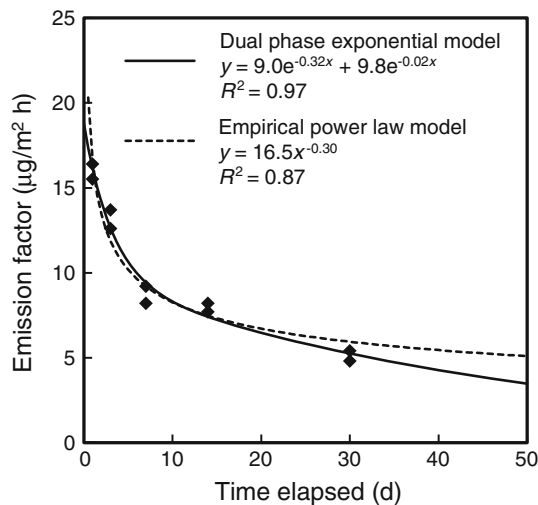


Fig. 2 Comparison between two acetaldehyde emission models (particleboard M): dual phase exponential model and empirical power law model

$$\log R(T) \approx \frac{A}{T} + B \tag{6}$$

where $R(T)$ is the emission factor at absolute temperature T , and A and B are constants. The emission factors of days 1 and 3 are normalized by dividing by values at 28 °C. Figure 4 shows the relationship between $1/T$ and the normalized emission factors on days 1 and 3. Equation 6 and the coefficients determined from the experimental results for days 1 and 3 provide the following empirical formula:

$$R_s(T) = e^{\left(\frac{-3638}{T} + 12\right)} \tag{7}$$

where $R_s(T)$ is the normalized emission factor at absolute temperature T . A first order approximation of Eq. 7 at 301.15 K (28 °C) gives:

$$R(\theta) = R_{028}[1.0 + 0.04(\theta - 28)] \tag{8}$$

where $R(\theta)$ is the emission factor at the temperature of θ (°C) and R_{028} is the emission factor at 28 °C. Equation 8 describes the relationship between the temperature and emission factors before day 3.

Ichihara et al. [27] measured the emissions from plywood of formaldehyde, another important indoor air pollutant, under several temperature conditions. They reported that the plywood specimens retained larger emission factors under higher temperatures over a period of 100 days, compared with the emission profile of a formaldehyde-immersed calcium silicate plate. They also suggested that this emission behavior was due to the hydrolysis of adhesives. Conversely, higher temperatures produced larger acetaldehyde emissions in the early period and smaller emissions in the later period because of faster decline. This emission behavior is apparently explained by the faster

reduction of acetaldehyde content from the surface due to large phase 1 emissions caused by higher temperatures, and relatively smaller phase 2 emissions controlled by inner diffusion. Accordingly, a bake-out process which gives materials higher temperature with ventilation is assumed applicable in reducing acetaldehyde emissions by new materials. Equation 8 can be useful in estimating the acetaldehyde concentration in a bake-out process.

Relative humidity dependence

Figure 5 shows the relationship between elapsed time and the acetaldehyde emission factor under conditions of various relative humidity at a temperature of 28 °C. Under the fixed temperature conditions, higher relative humidity produces a greater acetaldehyde emission factor throughout the test period. Acetaldehyde emission factors do not show a significantly faster emission decline, contrary to the temperature dependence. Particleboard M and U display distinctive emission behaviors in days 1–3 with relative humidity of 80 % compared with other conditions. MDF U also shows a distinctive emission profile under conditions of 80 % relative humidity. The emission factors for day 14 are normalized by dividing with values for relative humidity of 50 %. The relationship between acetaldehyde emission and relative humidity on day 14 can be described by an exponential function (Fig. 6). The formula determined from experimental results is as follows:

$$R(h) = R_{h50} \times 1.02^{(h-50)} \tag{9}$$

where h is the relative humidity (%), $R(h)$ is the emission factor at h % relative humidity, and R_{h50} is the emission factor at 50 % relative humidity. This equation is assumed applicable to phase 2 emissions.

Miyamoto et al. [28] measured particleboard samples for 21 days under conditions of relative humidity of 20, 50, and 80 %, and reported that higher relative humidity produced larger emission factors throughout the test period, which is in accordance with our results. Using a vinyl chloride film, Tamura et al. [29] demonstrated that relative humidity affects both the film moisture content and organic gas emission, and they suggested that the diffusion coefficient is affected by moisture content. Kagi et al. [30] measured internal diffusion coefficients of several VOCs of a vinyl chloride film under different humidity conditions and revealed that higher relative humidity causes a larger diffusion coefficient. They also indicated that the emission factor of phase 2 is affected by the moisture content of the materials, and suggested that relative humidity also affects the mass transfer coefficient of hydrophilic substances on the surface. On the assumption that the results obtained for vinyl chloride film can be applied to wood-based materials and

Fig. 3 Relationships between acetaldehyde emission factor and elapsed time for various temperature conditions with absolute humidity of 13.6 g/m³

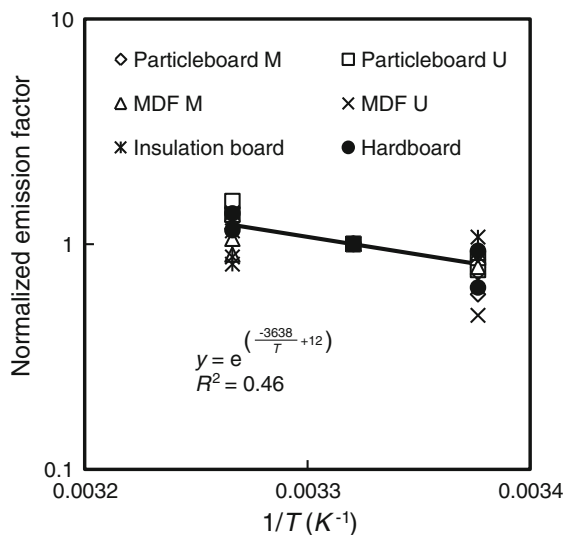
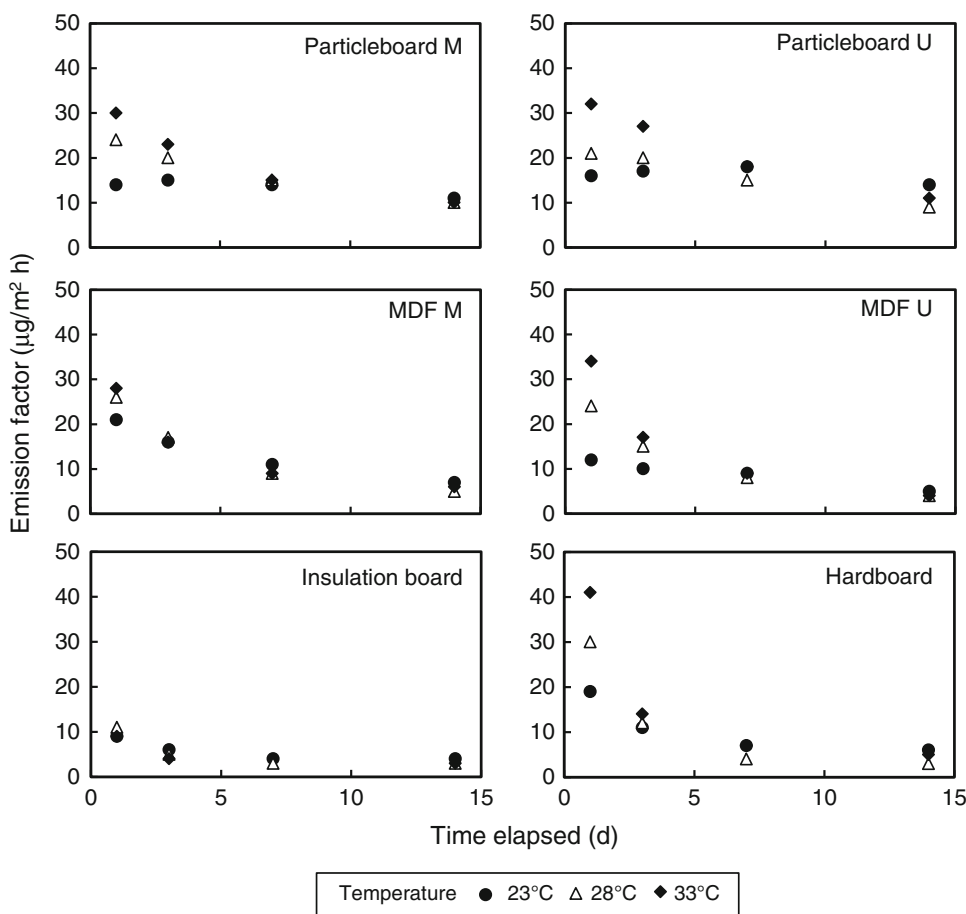


Fig. 4 Relationship between 1/T and normalized acetaldehyde emission factor for days 1 and 3 with relative humidity of 50 %

acetaldehyde, the emissions under conditions of varying relative humidity are explained by variations in water content of the materials. Specifically, the distinctive

emission behaviors shown for conditions of 80 % relative humidity could be explained by emission increase emerged by water absorption. The continuous emission of particleboard M and the increasing emission of particleboard U, observed in the earlier phase, could be associated with an increase of water content on the surface. The continuous emissions observed in the later phase for insulation board and hardboard appear associated with water absorption to the inner layers of the material. The distinctive behavior of MDF U could be associated with water absorption throughout the period. The larger emission factors observed during the later period under conditions of 80 % relative humidity suggest that diffusion coefficients of acetaldehyde are more sensitive to relative humidity or water content than temperature. However, the mechanism underlying these phenomena remains unclear. Higher relative humidity produces larger formaldehyde emissions from plywood and a significantly slower decline; after 100 days, emissions are still half the level of the initial emission factors [31], because of the hydrolysis of the adhesives. Conversely, conditions of higher relative humidity also demonstrated higher acetaldehyde emission in the initial stage, but a simple

Fig. 5 Relationships between acetaldehyde emission factor and elapsed time under various conditions of relative humidity for temperature of 28 °C

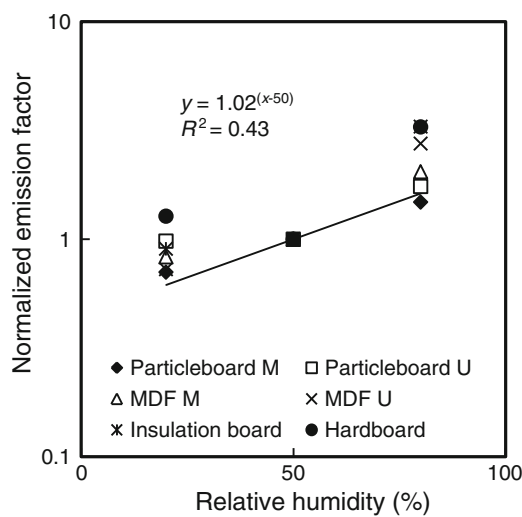
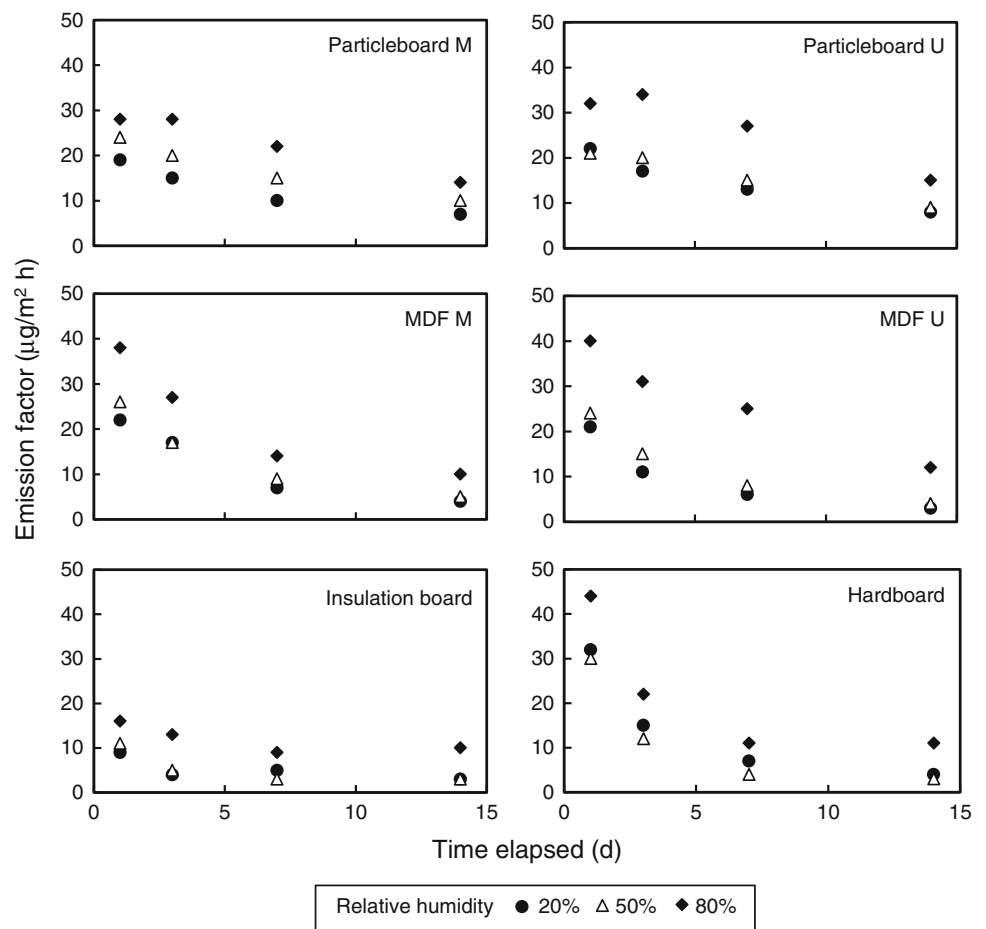


Fig. 6 Relationship between relative humidity and normalized acetaldehyde emission factor for day 14 at temperature of 28 °C

decrease in the later stage. Acetaldehyde emissions of phase 2 under conditions of higher relative humidity are supposed to decline as the content reduces.

Conclusions

Acetaldehyde emissions from several wood-based materials were investigated using the small chamber method. Acetaldehyde emission factors for wood-based products diminish rapidly in a properly ventilated environment. After 2 weeks, the acetaldehyde emissions from the materials in this study were no greater than 10 µg/m² h. The relationship between the elapsed time and emission factor can be described by the sum of two exponential functions or by a single exponential function. Higher temperatures increase the initial acetaldehyde emissions and accelerate the decrease in later emissions. A semi-empirical formula for the relationship between temperature and the initial acetaldehyde emission factor was obtained from the results. However, conditions of higher relative humidity gave greater values of emission factor. An empirical formula for the relationship between relative humidity and acetaldehyde emission factor was obtained from the results. In the later stage of emissions, the dependence of the acetaldehyde emissions on temperature is relatively smaller than the dependence on relative humidity. The effect of material moisture content on acetaldehyde emission should be examined in future studies.

Acknowledgments The authors would like to thank the Japan Fiberboard and Particleboard Manufacturers Association for providing the specimens. This study was supported by the Forestry and Forest Products Research Institute research grant #201002.

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