VOLATILE ORGANIC COMPOUND EMISSIONS ARISING FROM THE HOT-PRESSING OF MIXED-HARDWOOD PARTICLEBOARD

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ABSTRACT

This study investigated the effects of adhesive type and press variables on the volatile organic compound (VOC) emissions arising from hot-pressing mixed-hardwood particleboard. Three adhesive types, urea-formaldehyde resin (UF), phenol-formaldehyde resin (PF), and polymeric methylene diisocyanate resin (pMDI), were evaluated in this study. A 2^{5-1} fractional factorial design was used to evaluate the primary effect of five press variables (press temperature, press time, mat resin content, mat moisture content, and board density) and their interactions. A total of 27 chemical compounds were identified and quantified in the VOC emissions using four analytical techniques. Formaldehyde, methanol, acetic acid, and HMw VOCs with hexanal being the predominant chemical were the major compounds comprising the VOC emissions. The results revealed that formaldehyde and methanol emissions from UF particleboard, as well as the methanol emissions from PF particleboard were the most abundant components of the VOC emissions, and they contributedabout 92 and 72 percent of the total identified VOCs, respectively. Lower levels of formaldehyde and acetic acid were released during the hot-pressing of PF particleboard. Acetic acid and HMw VOC emissions were the most abundant components of the VOC emissions arising from pMDI-bonded particleboard. pMDI significantly reduced the methanol emissions from the mixed-hardwood particleboard. The most significant press variables controlling VOC emissions were press time, mat resin content, press temperature, and interactions among these three variables. These press variables had different effects on the individually identified compounds based on the adhesive type used. In general, formaldehyde emissions arising from hardwood particleboard hot-pressing were significantly lower than those from softwood particleboard. However, formaldehyde emissions from UF-bonded hardwood particleboard were significantly higher than from the softwood UF-bonded particleboard.

Today, there are 48 particleboard mills in the United States with a total annual capacity of 9,606,000 m^3 (Anonymous 2000), and the demand for particleboard is projected to continue. With implementation of the 1990 Clean Air Act, volatile organic compound (VOC) emissions and hazardous air pollutants (HAPs) arising during the hot-pressing of particleboard are a major concern of particleboard manufacturers. The measurement of VOC emissions from hot-pressing has been the subject of recent research (Wang and Gardner 1999; NCASI 1999, 1996; Wolcott et al. 1996; Broline et al. 1995; Carlson et al. 1995).

The VOC press emissions in wood composite manufacturing arise from both the wood particles and adhesive binder, and are dependent on numerous factors, such as wood species, adhesive type, and pressing conditions. The adhesives and the wood itself contain volatile compounds, which may be emitted

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during manufacturing. Additionally, thermal degradation of the wood may contribute to VOC emissions during manufacturing.

Lower levels of VOC emissions are expected from hardwood manufacturing processes compared to VOCs from softwood manufacturing processes (NCASI 1996, 1989) due to the large amount of volatile extractives in softwoods. Researchers evaluating emissions of nine different wood species found that the primary emissions are: terpenes from softwoods and acetic acid from hardwoods (Risholm-Sundman et al. 1998). Carlson et al. (1995) attributed the emissions of acetic acid, formic acid, methanol, and furfural during hot-pressing of particleboard to xylan decomposition as the thermally unstable hemicellulose is broken down. Previous research on emissions from southern pine press operations found that the wood extractives a -pinene and b-pinene are the predominant VOC emissions (Wang 1999, Carlson et al. 1995, Ingram et al. 1994), although degradation and reaction products of the wood and extractives, e.g. hexanal, pentanal, and pentanol, were also present.

Adhesive type may have a significant effect upon emissions during panel manufacture both because of the chemicals present in the adhesive and the interaction of the adhesive with the wood. The primary adhesives used in wood panel manufacture are urea-formaldehyde (UF), phenol-formaldehyde (PF), and polymeric methylene diisocyanate (pMDI). During hot-pressing of wood panels bonded with UF or PF adhesives, free-formaldehyde and methanol are released because both UF and PF resins contain small amounts of methanol and free-formaldehyde (Pizzi 1983). Using average test data from four UF-bonded particleboard mills, researchers (NCASI 1999) have extrapolated that an annual production of 265,000 m^3 (150 million $ft.^2$, 3/4 in. basis) releases approximately 60 tons/yr. of total HAPs, 40 tons/yr. of methanol, and 20 tons/yr. of formaldehyde emissions from particleboard hot-pressing. pMDI is a non free-formaldehyde containing adhesive. It forms chemical bonds by reacting with active hydrogen atoms and water on the wood surfaces and cures very fast. pMDI has become a replacement for conventional formaldehyde adhesives within wood-based panel manufacturing industries since the late 1980s. However, there is a possibility that some lower molecular weight pMDI may be released during hot-pressing, and there are concerns about worker health when using pMDI. Information about other VOC emissions from pMDI panel production is limited.

VOC press emissions are significantly affected by pressing conditions. Wolcott et al. (1996) studied the pressing variables affecting formaldehyde and methanol emissions from UF-bonded particleboard with a mixture of Douglas-fir and southern pine furnish. They found that formaldehyde emissions increased with increasing press time, platen temperature, moisture content, resin level, and formaldehyde-to-urea (F/U) mole ratio. Methanol emissions increased with increasing press time and moisture content (MC). Broline et al. (1995) employed a two-level half-fractional factorial design to determine the effects of process variables on formaldehyde, methanol, and ammonia press emissions from UF-bonded mixed-pine particleboard production. Five press variables (MC, resin loading, resin mole ratio, press time, and panel density) were investigated. They found that MC, resin loading, resin mole ratio, and press time were significant factors impacting VOC emissions. Carlson et al. (1995) investigated the effect of press variables on VOC emissions from laboratory PF-bonded aspen oriented strandboard (OSB). They reported that formaldehyde emissions increased with increasing press temperature, mat MC, resin treatment, resin free formaldehyde, and pressing time.

Above all, wood species, adhesives, and press conditions are significant factors affecting VOC emissions from particleboard hot-pressing. However, many of the previous studies focused on the VOC press emissions from softwood furnish. There is a lack of information on the type and quantity of VOC press emissions from hardwood furnish, and very little work has been done in evaluating the effects of adhesives and pressing variables on VOC press emissions.

The objectives of this research focused on:

1) Identification and quantification of hardwood VOC press emissions;

2) Evaluation of VOC emissions from hardwood-adhesive and hardwood furnish subjected to thermal treatment;

3) Evaluation of the effect of adhesive type on hardwood VOC press emissions;

4) Evaluation of the effect of five major press variables and their interactions on hardwood VOC press emissions.

MATERIALS AND METHODS

MATERIALS

Hardwood particles used in this study were obtained from Rodman Industry, Marinette, Wisconsin. They were composed of 60 to 70 percent northern hardwoods, 30 to 40 percent aspen, and a small amount of softwood. The MC of the particles was about 12 percent. The adhesives selected were UF resin (product no. 9-2011) from Southeastern Adhesive Company, PF resin (product no. NRC139b) from Neste Resin Corporation, and pMDI resin (product no. G541) from Bayer Corporation.

The chemicals used in this study for the collection and analysis of VOCs were methylene chloride, formaldehyde, methanol, formic acid, acetic acid, propionic acid, butyric acid, hexanal, a -pinene, b-pinene, octanal, D-limonene, borneol, naphthalene, hexadecane, heptadecane, and octadecane. Methylene chloride was used for extracting the VOCs from the hot-press process stream, and performing liquid-liquid extractions with the resulting VOC-water solutions. The other listed chemicals were used as standards in the various chemical analyses to be described in subsequent experimental sections.

PANEL MANUFACTURE

The resin was applied to the particle furnish in a drum blender with a spinning disk atomizer at 15,000 rpm. The target size of the particleboard was 305 by 305 by 19 mm (12 by 12 by 3/4 in.) All mats were hand formed in a 305- by 305-mm (12- by 12-in.) deckle box, and were pressed in the hot-press maintained at a temperature of ± 1°C around the target setpoint.

VOC EMISSION COLLECTION

During hot-pressing, VOC emissions were collected using a collection system comprised of a caul plate, two 250-mL scrubbers, and a vacuum pump. The two 250-mL scrubbers were chilled in a cooler bath to approximately 5°C. The first scrubber contained 100 mL fresh distilled water, and the second scrubber contained 150 mL methylene chloride. Using a vacuum oil pump, VOC emissions were pulled through the caul plate and absorbed in the two scrubbers. The collection of VOC emissions began when the caul plate was closed. The recovery efficiency of the collecting system was greater than 90 percent, based on previous experiments (O'Neill 2000).

After the hot-pressing cycle was completed and the press was opened, a 20-mL water solution was taken from the first scrubber for determination of low molecular weight (LMw) compounds in the VOC emissions. The remaining water solution was combined with methylene chloride solution from the second scrubber, and was extracted in a separatory funnel. The combined solution was extracted twice with 30 mL methylene chloride. The methylene chloride extraction solution was used for the determination of high molecular weight (HMw) compounds in the VOC emissions.

VOC EMISSION ANALYSIS

Four analytical techniques **(Fig. 1)** were utilized to identify and quantify the chemical compounds contained in the VOCemissions.

Gas chromatography/mass spectrometry (GUMS). — GC/MS was used to analyze the HMw VOC emissions in the methylene chloride solution. The GC/MS analysis was performed using a Hewlett-Packard GC-MS system consisting of an HP 6890 gas chromatograph and an HP quadrapole mass selective detector. A 30-m HP-5 column with 0.25-mm ID, and 5 percent crosslinked phenymethylsiloxane was used to separate the chemical compounds. The GC injector temperature was set at 270°C. The carrier used was helium with a flow rate of 0.7 mL/min. The GC oven temperature was started at 40°C and held for 4 minutes. Then the temperature was programmed to 280°C at 10°C per minute and held for 8 minutes. The ionizer voltage of the MS detector was set at 70 ev, and the temperature was set at 200°C. The scan range of molecular weights was 40 to 500 amu.

The total HMw VOC emissions were calculated by combining the estimated amount of the 50 largest peaks, which accounted for more than 97 percent of total VOCs detected in the GC/MS total

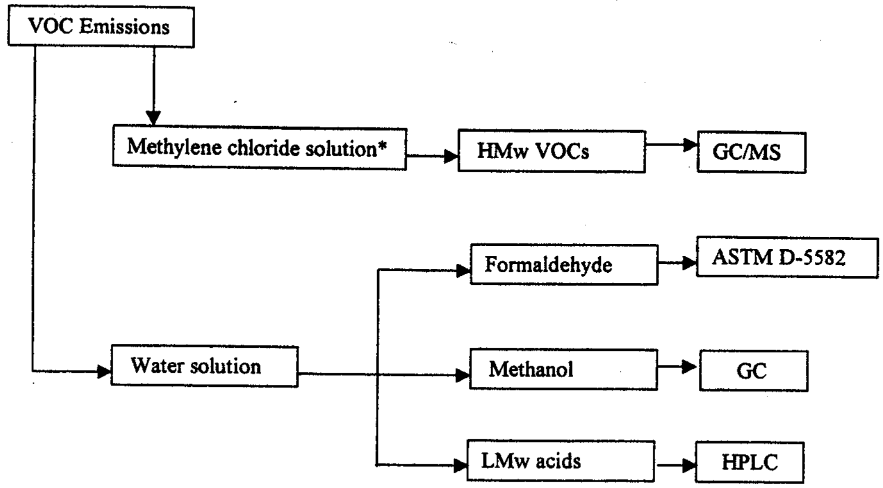

Figure 1. — Analytical techniques for measuring VOC emissions including extract from the water scrubber.

TABLE 1. — Press variables and their levels during hot-pressing ofhardwood particleboard.

Treatment levels and adhesives	Press temp.	Press time	Resin content	Mat MC	Board density
	(°C)	(min.)	---------- (%) ----------		(g/cm^3)
Low level					
UF	165	3	4	12	0.71
PF	165	4	3	13	0.71
pMDI	165	3	2	12	0.71
Center point					
UF	182	5	6	13	0.11
PF	182	6	5	14	0.11
pMDI	182	5	3.5	13	0.71
FH1[a]	182	5	0	13	0.11
FH2[a]	182	6	0	14	0.11
High level					
UF	199	7	8	14	0.83
PF	199	8	7	15	0.83
pMDI	199	7	5	14	0.83

[a] Press conditions of neat hardwood.

ion chromatograph. The quantity of HMw VOCs was calculated using external standards. Ten chemicals (hexanal, octanal, a -pinene, b-pinene, limonene, borneol, naphthalene, hexadecane, heptadecane, and octadecane) were selected as the external standards in this study. Each standard compound was selected based on molecular formulas, functional groups, and a range of diverse chemical structure. More details on the quantification methodology for the VOC emissions can be found in the work of O'Neill(2000).

Gas chromatography/flame ionization detector (GC/FID). — A GC/FID technique was used to analyze the water solution samples. The analysis was performed using a Hewlett-Packard 6850 gas chromatograph and an FID. The column used was an HP-WAX capillary column (15 m by 320 µm by 0.5 µm) bonded with polyethylene glycol. The injector temperature was set at 200°C

TABLE 2. — The main VOCs arising during the hot-pressing of mixed-hardwood particleboard.

Compounds	Boiling points	Analysis method	Retention time	External standard
	(°C)		(min.)	
Formaldehyde	−19.5	ASTM D-5582		Formaldehyde
Methanol	64.6	GC	−1.46	Methanol
Formic acid	100.7	HPLC	−3.51	Formic acid
Acetic acid	117.9	HPLC	−3.95	Acetic acid
Hexanal	131	GC/MS	−4.05	Hexanal
Propionic acid	140.7	HPLC	−4.89	Propionic acid
a -Pinene	156.2	GC/MS	−7.13	Alpha-Pinene
Butyric acid	163.5	HPLC	−6.55	Butyric acid
b-Pinene	166	GC/MS	~8.04	Beta-Pinene
2-Pentyl furan	64 to 66 at 23 mm	GC/MS	~8.39	Hexanal
Octanal	171	GC/MS	~8.65	Octanal
3-Carene	N/A[a]	GC/MS	~8.72	Limonene
Limonene	178	GC/MS	~9.08	Limonene
(E)-2-octenal	N/A	GC/MS	~9.68	Octanal
Nonanal	191	GC/MS	~10.49	Octanal
Borneol	209	GC/MS	~11.54	Borneol
2-Methylnaphthalene	241	GC/MS	~13.58	Naphthalene
Tetradecane	253.7	GC/MS	~14.76	Hexadecane
Longifolene	150 to 151 at 36 mm	GC/MS	~14.99	Hexadecane
Hexadecane	287	GC/MS	~17.24	Hexadecane
Heptadecane	301.8	GC/MS	~18.37	Heptadecane
Octadecane	316.1	GC/MS	~19.45	Octadecane
Eicosane	342.7	GC/MS	~21.45	Octadecane
Heneicosane	356.5	GC/MS	~22.39	Octadecane
Docosane	368.6	GC/MS	~23.29	Octadecane
Tricosane	380.2	GC/MS	~24.13	Octadecane
Tetracosane	391.3	GC/MS	~24.96	Octadecane

[a] N/A = not available,

with an injection volume of 1.0 µL. The carrier used was helium with a flow rate of 4.7 mL/min. The GC oven temperature was initially held at 50°C for 2 minutes. Then the temperature was programmed to 200°C at 25°C per minute and held for 2 minutes. The FID was set at 300°C, an H_2 feed rate of 30 mL/min, an air feed rate of 400 mL/min., and a helium makeup feed rate of 25 mL/min. Methanol was identified and quantified with external standards in the GCanalysis.

High performance liquid chromatography (HPLC) analysis. —The analysis of LMw acids in the water solutions was carried out using an HPLC with a UV-VIS spectrophotometer (Hitachi L-4500 diode array). An Adsorbosphere C18 5µ HPLC column (150 mm by 4.6 mm) was used, and 0.1 mM H_3PO_4 was selected as the mobile phase with a flow rate of 0.5 mL/min. The absorbance at 210 nm was measured to quantify the LMw acids. Four chemicals (formic acid, acetic acid, propionic acid, and butyric acid) were selected as external standards in the HPLC analysis.

Chromotropic acid method (ASTM 0-5582). — An ultraviolet visible spectrophotometer (Hewlett Packard 8452A diode array) was used to determine the amount of formaldehyde in the water solution, according to ASTM D-5582.

EXPERIMENTAL DESIGN AND STATISTICAL ANALYSIS

A two-level half-fractional factorial design (2^{5-1}) was used to evaluate the major effects of five press variables. One center point with triplicates was included in the design to check linearity, experimental error, and to provide hot-pressing variables approximating industrial production (Montgomery 1991). The five press variables included press temperature, press time, mat resin content, mat moisture content, and board density. **Table 1** shows the press variables and their levels (low, center, and high) for three adhesives, and press variables at center point for neat hardwood furnish. There are a total of 19 experimental runs for each adhesive. The run number was randomly chosen during the experiment. Statistical analysis of the experimental results was carried out for all classified compounds from the VOC emissions. In the analysis of variance (ANOVA) of the data, the variable with a probability value less than 0.05 was considered statistically significant. Based on the assumption of no interactions between variables, the relative effect of each variable was calculated to easily compare effects on VOC emissions among the press variables and adhesives.

$$Coef = \frac{\Delta \bar{Y}i}{\sum_{i=1}^{5} |\Delta \bar{Y}i}$$

where *Coef* = the relative effect of press variable *i*; DY = the mean effect of variable *i*. The Fisher LSD multiple comparison test was used for the comparison of VOC emissions from three hardwood-adhesive furnishes and neat hardwoodfurnish.

RESULTS AND DISCUSSION

VOC EMISSIONS CLASSIFICATION

Twenty-seven chemical compounds were identified in the VOC emissions arising from the hot-pressing of mixed-hardwood particleboard **(Table 2).** However, the compounds emitted were analyzed by several different techniques. Classifications of compounds in the discussions that follow are based on the chemical species and the technique used to identify and quantify them.

HMW VOCs. —The HMw VOCs are compounds that were identified in the methylene chloride trap and extracts. These compounds were identified by GC-MS. Twenty-two chemical compounds were included in this classification, with the main compounds being hexanal, pinenes, 2-pentyl-furan, 2-octenal, nonanal, 2-methyl-naphthalene, and longer chain alkanes (C_{14} - C_{24}). Under the press conditions of this study, hexanal emissions accounted for 20 to 51 percent of HMw VOCs, and it was the predominant single chemical. Because of the limitation of the GC/MS analytical technique, the compounds that elute earlier than the methylene chloride do not appear in the GC/MS to-

TABLE 3. — Summary of experimental results of VOC emissions from hardwood-adhesive furnish and neat hardwood.[a]

VOC compounds	Amount of VOC emissions				
	UF	PF	pMDI	FH1	FH2
	(mg/kg OD board)				
Formaldehyde					
Average	396.6	8.3	0.3	1.0	1.2
SD[b]	17.5	1.3	0.3	0.5	0.4
Methanol					
Average	328.2	216.2	5.1	12.3	9.7
SD	61.8	50.0	2.1	0.9	0.9
Acetic acid					
Average	31.7	19.8	26.2	29.5	36.9
SD	0.5	3.2	2.5	1.5	3.1
LMw acids (C1-C4)					
Average	32.5	29.4	27.6	32.0	41.9
SD	0.4	2.2	3.3	0.8	3.5
Hexanal					
Average	11.5	7.6	12.2	30.1	28.7
SD	1.5	2.9	7.0	1.6	7.6
HMw VOCs					
Average	34.4	29.8	33.6	66.2	69.7
SD	1.5	8.6	10.9	3.3	21.8
Total VOCs[c]	791.8	283.7	66.7	111.4	122.4

[a] Refer to the press conditions in Table 1.
[b] SD = standard deviation.
[c] Total identified VOCs including formaldehyde, methanol, LMw acids (C1-C4), and HMw VOCs.

TABLE 4. — Multiple comparisons of VOC emissions between UF particleboard and PF particleboard, pMDI particleboard, FH1, and FH2.

	PF	pMDI	FH1	FH2
Formaldehyde	***[a]	***	***	***
Methanol	***	***	***	***
Acetic acid	***	***	NS	***
LMw acids	NS[a]	***	NS	***
Hexanal	NS	NS	***	***
HMw VOCs	NS	NS	***	***

[a] *** = comparison significant at 0.05 level; NS = not significant.

talion chromatograph and were not included as HMw VOCs.

Formaldehyde. —Formaldehyde was identified through chromatropic acid analysis of the aqueous impinger solutions.

Methanol. —Methanol was identified through GC-FID analysis of the aqueous impinger solutions. Although other compounds were identified in the GC-FID analysis, only methanol was reported quantitatively.

LMw Acids. —Low molecular weight organic acids (C1 to C4) were identified and quantified using the HPLC analysis. Acetic acid, a primary compound in the LMw acids, accounted for about 33 to 99 percent of the LMw acids occurring at the press conditions examined in this study.

The components of VOC emissions

In determining the components of VOC emissions, there are two main points of interest: the type of significant VOC emissions for each adhesive used, and the effects of each adhesive on the identified VOCs in the wood-adhesive furnish. To determine the effect of the adhesive type on press emissions, panels were pressed at the center point conditions shown in **Table 1.** Panels were pressed with each of the adhesive types (UF, PF, and pMDI) and with wood particles to which no resin had been added (FH1 and FH2). The FH1 wood-only panels and the UF- and pMDI-bonded panels had similar press conditions (press temperature: 182°C, press time: 5 min., Mat MC: 13%, board density: 0.77 g/cm3 (48 lb./ft.^{3}), and the FH2 wood-only panels were pressed under conditions similar to the PF-bonded panels (press temperature: 182°C, press time: 6 min., Mat MC: 14%; board density: 0.77 g/cm^3 (48 pcf).

Table 3 summarizes the VOC emissions from hardwood panels bonded with UF, PF, and pMDI, and the hardwood panels pressed with no added adhesive (FH1 and FH2). The results of multiple comparisons among them are summarized in **Tables 4** to **6.**

Effect of UF resin. — For the UF-bonded hardwood panels, formaldehyde and methanol were the largest components of VOC emissions during pressing, accounting for about 92 percent of the total identified VOCs. Comparison with the emissions from the FH1 panel with no added adhesive shows that more than 95 percent of these two emissions came from the UF adhesive. Formaldehyde emissions arise from three sources: 1) excess free-formaldehyde in the resin; 2) thermal decomposition of the wood; and 3) hydrolysis of the UF resin during the heating under moist conditions in the panel (Marutzky and Margosian 1994). Methanol emissions may be accounted for by the methanol present in the formaldehyde used to make the resin (Carlson et al. 1995) and by the thermal decomposition of wood to make methanol (commonly known as wood alcohol). Therefore, these much higher levels of formaldehyde and methanol emissions from the UF particleboard, relative to panels with no resin, are expected.

Acetic acid emissions during pressing of the UF-bonded panel did not differ significantly from the emissions from the hardwood panel with no resin (Table 4). This is because acetic acid is formed from the hydrolysis of acetyl groups contained in the hemicellulose of the hardwood particles (Carlson et al. 1995). Xylan, the major hemicellulose component in hardwood, is highly acetylated at C-2 or C-3 (or both). On the average, there are 7 acetyl groups for every 10 backbone units (Sjostrom 1981).

Thus, a relatively high emission of acetic acid from hardwoods was expected (Sollidayet al. 1999, Risholm-Sundman et al. 1998).

HMw VOC emissions were the largest type of VOCs originating in the hardwood particles, and they were significantly reduced when the UF resin was added **(Table 4).** Hexanal and some other components in the HMw VOCs, for example pentyl-furans, arise from the decomposition products of the hardwood furnish (Baumann et al. 1999, Fengel and Wegener 1984). These components are released at a later time in the press run because they have higher boiling points. The reduction in HMw VOC emissions when UF resin is added may be due to the UF creating a barrier on the wood particles and restricting the emission of these higher boiling compounds.

Effect of PF resin. — For the PF hardwood-adhesive furnish, methanol was the largest component of VOC hot-pressing emissions, accounting for about 76 percent of the total identified VOCs. Comparison with the panels pressed with no resin (FH2) shows that 95 percent of the methanol emissions can be attributed to PF resin, with the remaining being present in the wood. Formaldehyde emissions comprised only about 3 percent of the total VOCs from the panel, with about 85 percent of the emissions attributed to the PF resin. Methanol and formaldehyde were emitted from both the wood and the resin with the major source being the resin due to the free formaldehyde and methanol in the resin. The lower levels of formaldehyde could possibly be attributed to a lower level of free formaldehyde (< 0.1%) and an additive in the PF resin (Wang 1999). The additive contained an amine functional group, which could react with formaldehyde to reduce the emissions during the hot-pressing process.

The acetic acid emissions from the PF panels, however, were significantly lower than those from panels pressed with no adhesive. This may be attributed to the higher pH value (12.2 to 12.7) of the PF resins used in this study., The basic components contained in the PF probably react with acetic acid, and reduced the acetic acid emissions. HMw VOCs emissions from the PF particleboard pressing were also significantly lower than those from the neat hardwood furnish because the pathways of the emissions with higher boiling points might be blocked after the PF resin cures. The presence of the PF resin decreased the rate of acetic acid and HMw VOCs release.

TABLE 5. —Multiple comparisons of VOC emissions between PF particleboard and UF particleboard, pMDI particleboard, FH1, and FH2.

	UF	pMDI	FH1	FH2
Formaldehyde	***[a]	***	***	***
Methanol	***	***	***	***
Acetic acid	***	***	***	***
LMw acids	NS[a]	NS	NS	***
Hexanal	NS	NS	***	***
HMw VOCs	NS	NS	***	***

[a] *** = comparison significant at 0.05 level; NS = not significant.

TABLE 6. — Multiple comparisons of VOC emissions between pMDI particleboard and UF particleboard, PF particleboard, FH1, and FH2.

	UF	PF	FH1	FH2
Formaldehyde	***[a]	***	NS	***
Methanol	***	***	***	***
Acetic acid	***	***	NS	***
LMw acids	***	NS	***	***
Hexanal	NS"	NS	***	***
HMw VOCs	NS	NS	***	***

[a] *** =comparison significant at 0.05 level; NS = not significant.

Effect of pMDI. —For hardwood panels bonded with the pMDI, acetic acid and HMw VOCs were the largest components of VOC emissions. They account for about 45 and 50 percent of the total identified VOC emissions, respectively. Methanol and free-formaldehyde are not present in pMDI resin, so the only contribution to these emissions was from the wood particles. Methanol and HMw VOCs were significantly decreased in the pMDI-bonded panels relative to the panels with no adhesive applied (FH1). Moisture in the panel acts as a carrier for some VOC emissions (Wang 1999, Wolcott et al. 1996), but pMDI reacts rapidly with water in the furnish panel (Wendler et al. 1995). This could cause the reduction of the VOC emissions, especially formaldehyde and methanol emissions. The fast curing of pMDI may be the second factor, blocking pathways of VOC emissions during the later stages of hot-pressing process. The third possible reason is that pMDI reacts with hydroxyl groups. Because methanol contains a hydroxyl group, the reaction between pMDI and methanol could occur during hot-pressing. Further studies could address why pMDI affects methanol, but not formaldehyde emissions.

VOC EMISSIONS AS A FUNCTION OF ADHESIVE TYPE

The results of the VOC emissions from the hot-pressing of mixed-hardwood particleboard are summarized in **Tables 7** and **8.**

Formaldehyde. — Formaldehyde emissions from UF-bonded panels were the largest component of VOC emissions among the three adhesive types (UF, PF, and pMDI). The emissions ranged from 52.4 to 640 mg/kg (oven-dry weight [OD]), and were significantly higher than those from panels bonded with either PF or pMDI resin **(Tables 4** and **7).** Formaldehyde emissions for panels bonded with PF and pMDI resin ranged from 2.7 to 10.6 mg/kg (OD), and from 0.02 to 0.85 mg/kg (OD), respectively. The pMDI panels had significantly lower formaldehyde emissions compared to both the UF and PF panels, due to the lack of formaldehyde in pMDI resin.

Methanol. — Methanol emissions were the second largest components of VOC emissions from panels bonded with either UF or PF resins. Panels bonded with pMDI had significantly lower methanol emissions. The metha-

TABLE 7. — Summary of experimental results of VOC emissions in 2^{5-1} experimental design.

Run no.	Press conditions[a]					Formaldehyde			Methanol			Acetic acid		
	Press temp.	Press time	Resin content	Mat MC	Board density	UF	PF	pMDI	UF	PF	pMDI	UF	PF	pMDI
						(mg/kg OD board)								
1	-	-	-	-	+	52	2.7	0.02	5	65	0.8	0.2	0.4	0.8
2	+	-	-	-	-	174	5.6	0.07	164	152	4.8	15.0	9.4	12.0
3	-	+	-	-	-	326	10.5	0.15	210	229	10.4	33.6	17.5	30.2
4	+	+	-	-	+	397	5.6	0.51	248	330	15.5	83.8	30.5	50.8
5	-	-	+	-	-	141	6.5	0.09	24	338	1.7	0.8	1.7	4.9
6	+	-	+	-	+	250	6.4	0.09	227	329	3.1	12.0	15.4	12.1
7	-	+	+	-	+	417	14.5	0.20	436	548	7.9	38.0	16.5	29.1
8	+	+	+	-	-	565	12.1	0.90	233	222	9.6	64.4	61.6	48.0
9	-	-	-	+	-	121	5.1	0.09	17	136	2.6	2.0	3.0	2.8
10	+	-	-	+	+	225	4.1	0.16	119	154	5.6	17.4	8.5	13.8
11	-	+	-	+	+	388	8.3	0.16	115	142	13.1	53.9	18.9	39.3
12	+	+	-	+	-	604	6.8	0.34	203	112	9.7	78.1	75.3	37.3
13	-	-	+	+	+	144	6.1	0.03	22	254	0.9	1.3	2.5	2.5
14	+	-	+	+	-	285	6.0	0.10	303	296	2.1	18.9	13.3	10.9
15	-	+	+	+	-	525	12.1	035	277	370	8.2	40.5	15.9	26.2
16	+	+	+	+	+	640	10.6	0.85	312	176	5.4	107.5	73.8	44.4
17	0	0	0	0	0	385	9.9	0.31	390	213	2.7	32.0	16.3	23.6
18	0	0	0	0	0	389	7.6	0.23	329	168	6.6	32.0	22.4	28.5
19	0	0	0	0	0	417	7.5	0.33	266	268	6.0	31.1	20.8	26.4

[a] Refer to Table 1, the symbols +, -, and 0 represent high level, low level, and center points, respectively. For example, in run 5, the symbols mean press temperature = 165°C, press time = 3 minutes, mat resin content = 8%, mat MC = 12%, and board density = 0.71 g/cm³.

TABLE 8. — Summary of experimental results of VOC emissions in 2^{5-1} experimental design (continued).

Run no.	Press conditions[a]					LMw acids (C1 to C4)			Hexanal			Hmw VOCs		
	Press temp.	Press time	Resin content	Mat MC	Board density	UF	PF	pMDI	UF	PF	pMDI	UF	PF	pMDI
						(mg/kg OD board)								
1	-	-	-	-	+	0.8	1.5	0.9	1.2	6.4	6.1	4.5	21.0	15.5
2	+	-	-	-	-	15.2	16.1	13.4	26.8	10.6	31.7	52.8	30.0	69.0
3	-	+	-	-	-	34.1	22.7	31.4	29.7	12.8	28.6	62.5	51.1	67.0
4	+	+	-	-	+	86.7	57.1	57.7	27.8	7.2	29.7	73.3	25.3	81.8
5	-	-	+	-	-	1.0	2.9	5.1	1.3	20.9	9.8	4.4	48.8	21.1
6	+	-	+	-	+	12.3	28.4	12.8	3.5	21.9	17.0	11.4	58.5	36.9
7	-	+	+	-	+	38.3	29.5	29.4	9.3	29.7	22.6	38.8	87.6	54.4
8	+	+	+	-	-	85.5	119.1	55.7	9.4	25.9	15.5	42.2	67.5	43.8
9	-	-	-	+	-	2.0	6.6	2.9	7.6	7.4	12.6	16.3	20.5	26.2
10	+	-	-	+	+	47.6	11.0	14.1	12.7	3.2	19.7	25.9	10.9	47.0
11	-	+	-	+	+	54.2	24.2	40.7	27.7	11.0	29.7	65.8	47.5	73.9
12	+	+	-	+	-	99.6	107.2	42.6	25.6	8.2	14.0	68.6	37.3	34.6
13	-	-	+	+	+	1.4	7.2	2.8	1.7	20.0	12.6	7.3	43.4	26.6
14	+	-	+	+	-	19.5	22.0	11.2	6.7	23.2	13.4	19.1	54.6	30.2
15	-	+	+	+	-	41.6	29.1	27.6	10.1	27.0	19.1	37.0	70.3	43.2
16	+	+	+	+	+	109.6	131.9	52.2	8.1	2.7	11.3	38.8	11.0	36.1
17	0	0	0	0	0	32.7	27.4	24.0	12.5	5.9	12.7	36.1	25.0	29.7
18	0	0	0	0	0	32.7	31.7	30.5	12.1	10.9	18.9	33.3	39.7	46.0
19	0	0	0	0	0	32.1	29.1	28.3	9.7	6.1	5.0	33.8	24.8	25.3

[a] Refer to Table I, the symbols +, -, and 0 represent high level, low level, and center points, respectively. For exam le, in run 5, the symbols mean press temperature = 165°C. press time = 3 minutes, mat resin content = 8%, mat MC = 12%. and board density = 0.71 g/cm³.

TABLE 9. - Effects ofpress parameters and their interactions on VOC emissions from UF-, PF-, and pMDI-bonded particleboard. [a]

	Press parameters and their interactions							
VOC emissions	Temp.	Time	Resin	MC	Density	Temp. * time	Time * resin	Time * MC
	(°C)	(min.)	-------- (%) --------		(g/cm^3)			
Formaldehyde (ASTM D 5582)								
UF	***[b]	***	***	***				***
PF		***	***			***	***	
pMDI	***	***	***			***		***
Methanol								
UF	***	***	***			***		
PF			***					
pMDI		***	***					
Acetic acid								
UF	***	***				***		
PF	***	***				***		
pMDI	***	***				***		
LMw acids (C1 to C4)								
UF	***	***		***		***		
PF	***	***				***		
pMDI	***	***				***		
Hexanal								
UF		***	***					
PF			***					
pMDI		***	***			***	***	
HMw VOCs								
UF	***	***	***					
PF			***					
pMDI		***	***			***		

[a] Temp. = press temperature; time = press time; resin = resin content; MC = mat MC; and density = board density.
[b] *** = 95 percent significant.

nol emissions from panels bonded with UF resin ranged from 5.4 to 436 mg/kg (OD), and with PF resin ranged from 64.7 to 547.7 mg/kg (OD). Methanol is present in formaldehyde solutions used in UF and PF adhesive manufacture, thus accounting for the significantly higher methanol emissions from panels pressed with these two resins. Under similar hot-pressing conditions, the UF-bonded panels emitted significantly higher levels of methanol than the PF panels. Interestingly, multiple comparisons of the data given in **Table 6** indicate that application of pMDI resin resulted in decreased emissions from the hardwood furnish.

Acetic acid. **—** Acetic acid emissions ranged from 0.2 to 107.5 mg/kg (OD) for UF-bonded panels, 0.4 to 73.8 mg/kg (OD) for PF-bonded panels, and 0.8 to 50.8 mg/kg (OD) for pMDI-bonded panels **(Table 7)** under the press conditions of this study. The results from the multiple comparison indicated that the acetic acid emissions from the PF panels were significantly lower than those from either the UF or pMDI panels **(Table 5).** This reduction in acetic acid emissions for panels bonded with PF resin is due to the higher pH value of PF resins (12.2 to 12.7), which would have reacted with the acetic acid. The significant difference in acetic acid emissions between UF-bonded panels and pMDI-bonded panels, under the same press conditions, is not readily explained because the acetic acid emissions are attributed to the hardwood furnish, not the resin.

HMw VOCs. — HMw VOC emissions from three types ofpanels (UF, PF, and pMDI) were emissions from the wood rather than the adhesives, and ranged from 4.4 to 73.3 mg/kg (OD), 11 to 87.6 mg/kg (OD), and 15.5 to 81.8 mg/kg (OD), respectively. As shown in **Tables 4, 5, 6,** and **8,** under similar press conditions (center points), there was no significant difference in HMw VOC emissions among these three panels. All three adhesives could possibly decrease the emissions of HMw VOCs by blocking the emission pathways after resin curing.

VOC EMISSIONS AS A FUNCTION OF PRESS VARIABLES

Formaldehyde. — **Table 9** and **Figures 2** and **3** summarize the ANOVA results of the effect of five press variables and their interaction on formaldehyde emissions. Formaldehyde emissions were significantly increased by increasing press time and resin content for panels bonded with all three adhesive types. Significant increases in formaldehyde emissions with increasing temperature were observed for the UF- and pMDI-bonded panels, but not the PF-bonded panels. Longer press times, higher resin contents, and higher press temperatures (except for the PF-bonded panels) can cause higher levels of formaldehyde emissions from both adhesive and hardwood furnish. **Figure 2** shows that increasing press time alone accounted for approximately 50 percent of the total effects, while MC had a small significant

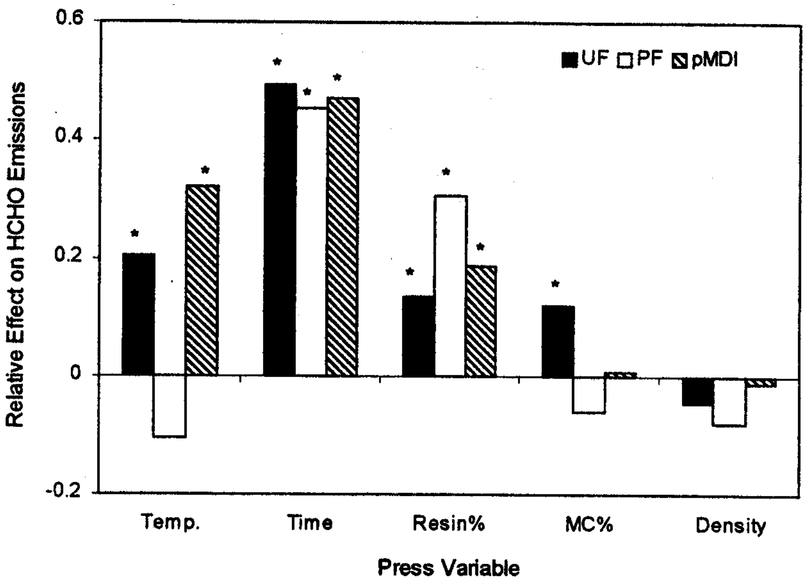

Figure 2. — Relative effects of press variables on formaldehyde emissions during hot-pressing of panels bonded with UF, PF, and pMDI (relative effect: single effect neglecting interactions; I = 95% significant).

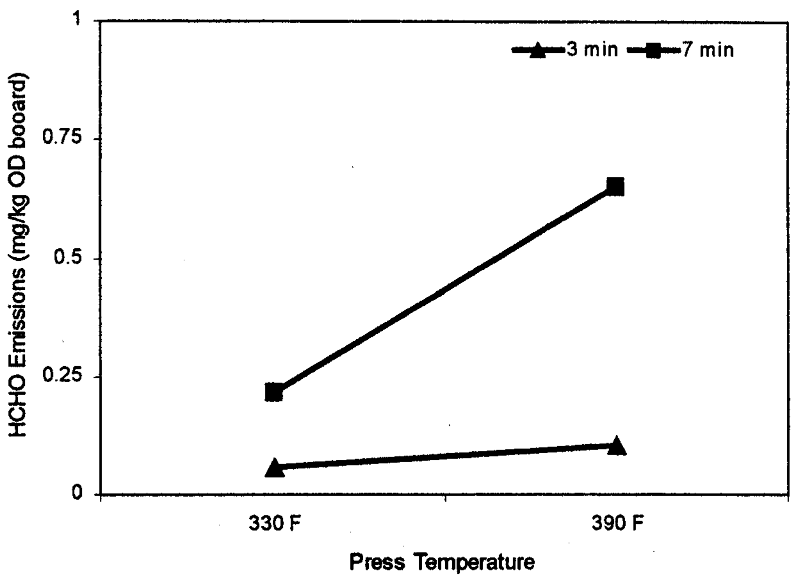

Figure 3. — Temperature-time interaction effect on formaldehyde emissions during hot-pressing of pMDI-bonded panels.

effect and panel density showed no effect. There were significant interactions of press temperature-press time and press time-resin content on the formaldehyde emissions from pMDI-bonded panels **(Fig. 3),** and a significant interaction of press time-MC on the formaldehyde emissions from UF-bonded panels.

Methanol. — **Table 9** and **Figures 4** and **5** summarize the ANOVA results of the effect of five press variables and their interaction on methanol emissions. Emissions of methanol during pressing of UF-bonded panels were significantly increased by increasing press temperature, press time and resin content. Significant increases in methanol emissions were observed from PF-bonded panels by increasing resin content and from pMDI-bonded panels by increasing press time or decreasing resin content. The negative dependence of methanol emissions on resin content for pMDI-bonded panels can be attributed to the reaction of pMDI with methanol and with the water, which acts as a carrier for the methanol emissions (Ingram et al. 1994). The combined effects of press time and resin content contributed about 60 percent of the total effects on the methanol emissions for all three adhesive types **(Fig. 4).** Because methanol emissions came from both adhesive (excluding pMDI adhesive) and hardwood furnish, it is reasonable that the methanol emissions increased with increasing press time, resin content (excluding the pMDI adhesive), and press temperature (excluding the PF adhesive). There was one significant interaction term of press temperature-press time found for the methanol emissions from UF-bonded panels **(Fig. 5).**

LMw acids. — **Table 9** and **Figure 6** summarize the ANOVA results of the effect of five press variables on emissions of organic acids. Acetic acid emissions significantly increased at increasing press temperature and press time for all three adhesive types, and the relative effects of these two press variables accounted for about 80 percent of the total effects **(Fig. 6).** A possible explanation for this result can be attributed to acetyl groups contained in the hemicellulose of the hardwood particles hydrolyzing into acetic acid during hot-pressing. Because the speed of hydrolysis reaction increases as the reaction temperature is raised, acetic acid emissions increased with increasing press temperature. Under the same pressing temperature, acetic acid emissions increased with increasing press time. Emissions of LMw acids increased with increasing mat MC for panels pressed with UF resin. Resin content did not have a significant effect on the acetic acid emissions, since acetic acid was attributed entirely to the hardwood particles. A significant interaction term of press temperature-press time was found for the acetic acid emissions from the UF- and PF-bonded panels.

HMw VOCs. — **Table 9** and **Figures 7** and **8** summarize the ANOVA results of the effect of five press variables and their interaction on HMw VOC emissions. The HMw VOC emissions were significantly reduced with decreasing press time and increasing resin content for both the UF- and pMDI-bonded panels. However, as shown in **Figure 7,** resin content had a significantly positive effect on the HMw VOC emissions from the PF-bonded panels, although all three adhesives decreased the emissions of HMw VOCs from hardwood particles at the level of center points. These results are further illustrated in **Figure 8:** HMw VOCs emissions rapidly decreased with increasing resin content for the UF-bonded panels. The emissions from the PF-bonded panels slightly decreased with increasing resin content up to 6 percent resin content, then sharply increased as resin content was raised. For the pMDI-bonded panels, HMw VOCs sharply decreased with increasing resin content below 4.5 percent resin content, then increased slightly as resin content increased. These results indicated that the three adhesives had different effects on the HMw VOCs emissions among the different range of resin contents. As shown in **Table 8,** the HMw VOC emissions at higher levels of resin content (runs 5, 6, 7, 8, 13, 14, 15, and 16) were higher than those at lower levels of resin content (runs 1, 2, 3, 4, 9, 10, 11, and 12) for the PF-bonded panels, except for run 16. This is because 20 to 51 percent of HMw VOC emissions were hexanal emissions that were higher at higher levels of resin content. Press temperature had a significant effect on the HMw VOC emissions only from the UF-bonded panels. There was a significant interaction of press temperature-press time on the HMw VOC emissions from the pMDI-bonded panels.

It was surprising that greater effects of MC on emissions were not observed. However, this lack of dependence upon MC concurs with previous research by Carlson et al. (1995). Carlson, while measuring emissions from pressing of PF-bonded oriented strandboard, found that if the mat MC was above 8 percent, changes in mat MC appeared to have less of an effect on the formaldehyde emissions. Because the MC levels in this study ranged from 10 to 14 percent for UF and pMDI panels, and 11 to 15 percent for PF panels, it is possible that there was sufficient moisture in the panels, even at the low point, that inhibiting of the carrier effects of the moisture were not observed.

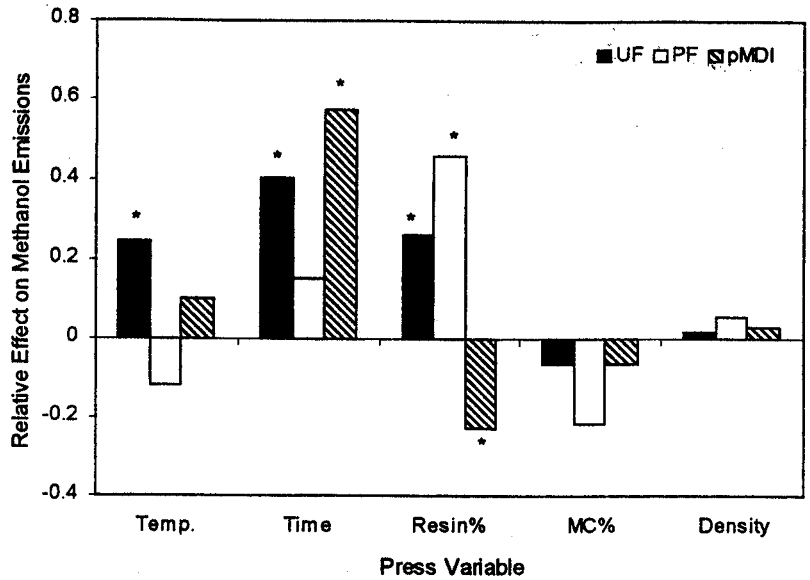

Figure 4. — Relative effects of press variables on methanol emissions during hot-pressing of panels bonded with UF, PF, and pMDI (relative effect: single effect neglecting interactions; * = 95% significant).

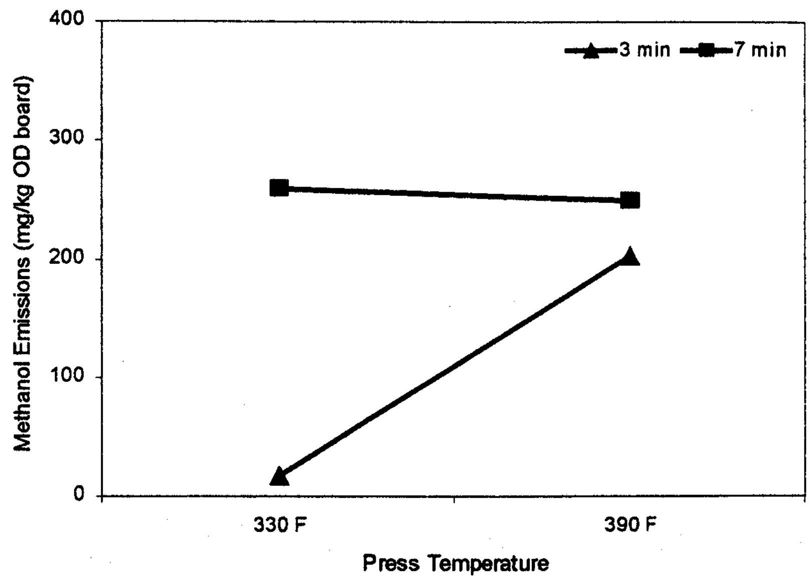

Figure 5. —Temperature-time interaction effect on methanol emissions during hot-pressing of UF-bonded panels.

COMPARISON OF FORMALDEHYDE EMISSIONS BETWEEN HARDWOOD AND SOFTWOOD

Using published data of VOC press emissions from southern pine particleboard (Wang 1999), the formaldehyde emissions from hardwood and softwood particleboard were compared **(Table 10).** The results show that formaldehyde emissions from hardwood particleboard bonded with the UF resin were much higher than those from softwood particleboard bonded with the UF resin. This might be attributed to resin batch differences between the pine and mixed-hardwood studies or the acetic acid from the hardwood furnish contributing to UF resin hydrolysis, and subsequently, larger emissions of formaldehyde. However, the formaldehyde emissions from hardwood particleboard were significantly lower than those from softwood particleboard for the other adhesive typesexamined.

CONCLUSIONS

The VOC emissions arising from the hot-pressing of mixed-hardwood particleboard with three adhesive types were identified and quantified using four analytical techniques. A two-level half-fractional factorial experimental design (2^{5-1}) was utilized to determine the primary effect of five press variables and the interactions between any two press variables on the VOC emissions. Twenty-seven compounds were identified in the hardwood VOC emissions, and the primary components in the VOC emissions were formaldehyde, methanol, acetic acid, and HMw VOCs in which hexanal was the predominant single chemical. Among the three adhesive types, formaldehyde and methanol emissions from the UF particleboard were the most abundant components of the VOC emissions, accounting for about 92 percent of the total identified VOCs in the UF particleboard. Methanol emissions from the PF particleboard were the second largest component of the VOC emissions, contributing about 75 percent of the total identified VOCs in the PF particleboard. More than 95 percent of these emissions came from the adhesives used. Lower levels of formaldehyde and acetic acid emissions were found in the PF particleboard. Compared with UF and PF, pMDI had much lower levels of formaldehyde emissions, and also significantly reduced the methanol emissions from the hardwood particles. Acetic acid and HMw VOC emissions were the largest components of VOC emissions from pMDI particleboard. Press time, resin content, press temperature, and interactions among these three variables are the most significant press variables controlling VOC emissions arising during the hot-pressing of hardwood particleboard. Most of the identified VOCs were significantly increased with increasing press time, mat resin content, and press temperature.

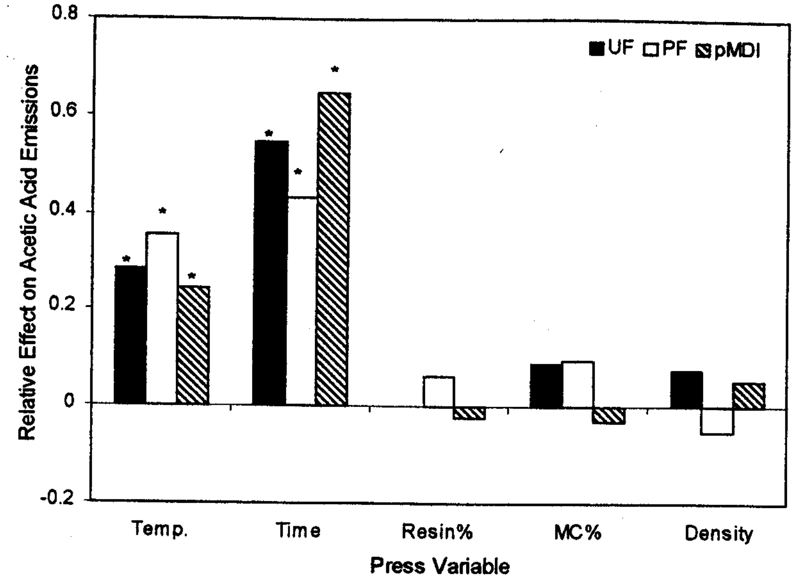

Figure 6. — Relative effects of press variables on acetic acid emissions during hot-pressing of panels bonded with UF, PF, and pMDI (relative effect: single effect neglecting interactions; * = 95% significant).

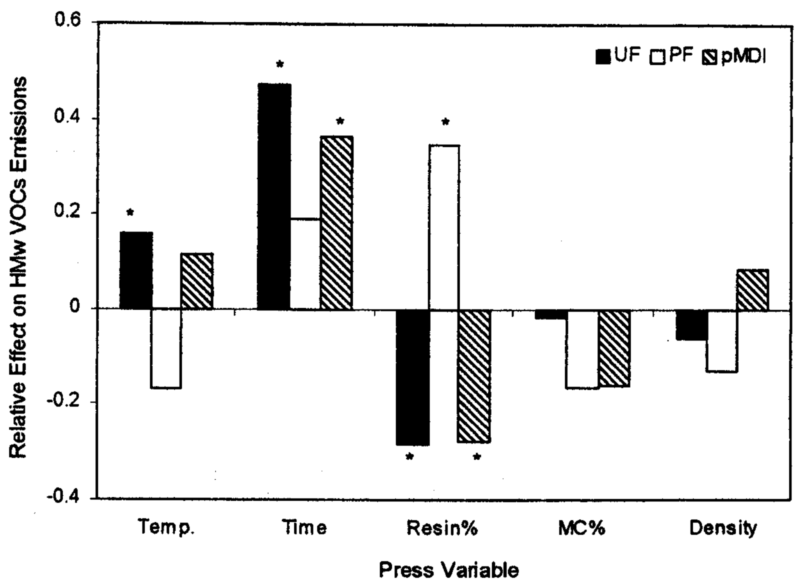

Figure 7. — Relative effects of press variables on HMw VOC emissions during hot-pressing of panels bonded with UF, PF, and pMDI (relative effect: single effect neglecting interactions; * = 95% significant).

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TABLE 10. — *Comparison of formaldehyde press emissions between hardwood (HW) and softwood (SW).* [a]

Press conditions								
Resin type	UF	UF	PF	PF	pMDI	pMDI	FH	FH
Wood type	HW[b]	SW[b]	HW	SW	HW	SW	HW	SW
Press time (min.)	5	6	6	8	5	6	5	6
Resin content (%)	6	7	5	6	3.5	4.5	0	0
Mat MC (%)	13	12	14	13	13	12	13	12
Formaldehyde (mg/kg OD board)	385	244	10	11	0.3	35	1.4	42
	389	257	8	13	0.2	32	0.4	41
	417	249	7	12	0.3	32	1.1	40
Average	397	250	8	12	0.3	33	1.0	41

[a] All panels were pressed at 182°C with a panel density of 0.77 g/cm ^{3}.
[b] Results from two different studies.

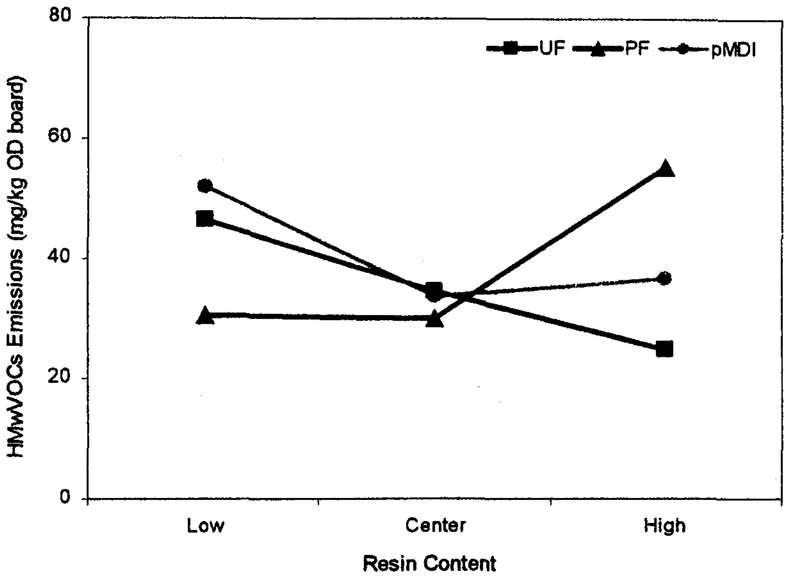

Figure 8. — HMw VOC emissions during hot-pressing as a function of resin content for panels bonded with UF, PF, and pMDI.

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