Assessing Panelboard Volatile Organic Compound Emission Profiles through Renewables Use*

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Abstract

With an increasing prevalence of renewables used as binders in panelboard products, it is of interest to know how these renewables may impact manufacturing environments and product performance. In this study, soy and zein proteins together with a kraft lignin and condensed tannin extract have been evaluated for contributions to volatile organic compounds arising in press emissions from processing hardwood veneer plywood. Analysis revealed aldehydes tended to dominate press emissions, with both proteinaceous materials producing significantly higher acetaldehyde and hexaldehyde emissions relative to the wood veneer used. Other aldehydes, including formaldehyde, valeraldehyde, and propionaldehyde, were also produced but in relatively lower quantities. The use of polyphenolics led to lower formaldehyde content in press emissions with tannin, also reducing acetaldehyde and other aldehydes in press emissions. Chamber testing plywood panels found the composition of VOC emissions initially released from panels to be prominent compounds in press emissions produced during panel manufacture.

In panelboard manufacture, the wood furnish is typically heated below 200°C via rapid hot pressing, with this process potentially liberating a range of organic compounds either by volatilization or decomposition (Wang et al. 2003). These emissions may be released on pressing or retained within the wood matrix to potentially migrate from the panelboard with time. In manufacturing wood-based panelboards, the use of adhesive binders or other additives can complicate the volatile organic compounds (VOCs) liberated from this process; formaldehyde from urea formaldehyde resin is a well-known example of this. Formaldehyde is a VOC subject to extensive testing in panelboard production and postmanufacture emissions. Furthermore, with the increasing use of biobased materials for panelboard binders, it is of interest to know how the use of these renewables contributes VOCs to the manufacturing environments as well as within panel products. Both torrefaction and pyrolysis of biomass are examples of high-temperature processing, which can produce a range of gaseous VOCs together with chars and oils (Morgan and Kandiyoti 2014). At lower temperatures typical of panel processing, renewables can also degrade to liberate VOCs. Moreover, little is known of how renewables may contribute to panelboard emissions and whether this is complicated by their use in partial or fully substituted resin systems or differing application chemistries such as pH.

In this article, we describe an initial survey of various renewables and their impact on the VOC composition of

press and panelboard emissions as either individual components or in combination. This work draws on a greater study developing methodology for qualitative and quantitative assessments of VOCs arising from various renewables (Grigsby et al. 2019, Miyamoto et al. 2019). It provides a complementary assessment of those renewable binders that were presentation topics at the International Conference on Wood Adhesives. Agricultural processing by-products soy and corn gluten meals together with a technical grade (kraft) lignin and condensed tannin (poly-flavanoid) extract have been evaluated individually as binders and in combination together to manage or reduce overall VOC profiles in hardwood plywood. The plywood panel series was formed employing a press temperature of 170°C and an extended pressing time to ensure bondline cure and liberation of volatile compounds during manufacture. Gas emissions produced during panel hot pressing and consolidation were captured, segregated, and chemically analyzed using the methodology presented in Figure 1 (Grigsby et al. 2019). The emissions were qualitatively assessed to determine semivolatile (SVOC) to very volatile (VVOC) compounds and the VOC profiles produced. Chamber testing of panelboards was used to identify and quantify longer-term panel VOC emissions and compare these with emissions evolved on panel manufacture. Ultimately the outcomes from the greater study will be used to direct bio-based ingredient use in mixtures with other traditional or bio-based binders or aid adhesive design.

Experimental

Materials

Lauan (*Shorea* spp.) veneer (220 by 200 by 1.7 mm) was sourced from Niigata Gohan Shinko (Japan). Soy flour Soy 7B was sourced from Archer Daniel Midland (USA), and the lignin was Indulin AT, a technical kraft lignin sourced from Ingevity (USA). The corn gluten meal was Avon Gold sourced from NZ Starch Ltd. (New Zealand), and the tannin was a hot water–sodium sulfite extract of radiata pine (*Pinus radiata*) bark (Grigsby 2017). A commercial grade wheat flour was used to form glue mixes and was sourced from Oshika (Japan).

Glue mix formulation

Each renewable was thoroughly dispersed in water to form aqueous suspensions suitable for application to the veneer and plywood manufacture. No attempt was made to produce homogeneous, dissolved solutions of each ingredient or to make pH adjustments. For the bioadhesive combination, this resin (Grigsby et al. 2019) was combined with wheat flour (2%, wt/wt) and thoroughly mixed. Reference veneer-only panels were produced by applying water to the veneer using a water content calculated for the glue mixes (above) with the veneers and then pressed (as below).

Plywood manufacture

Each glue mix was applied to a single side of lauan veneer at a rate of 260 to 280 g/m^2 by a hand roller, and the veneers were assembled to form 3-ply panels. After an open assembly time of 5 minutes, the veneers were placed on platerns within a gas capture assembly apparatus (Miyamoto et al. 2010; Grigsby et al. 2019). Panels were then pressed at 170°C for a total of 600 seconds at 65 kg/cm^2 using a Kobayashi laboratory press. After pressing, panels were covered in foil, cooled to ambient temperature, and then stored in sealed bags prior to trimming and chamber testing.

As the hot-press closed, collection of press gas emissions was started with emissions drawn from the platern assembly at a rate of 2,000 mL/min (Fig. 1). The gases first passed through a condenser collection apparatus (0°C) before being collected in a Tedlar bag (GL Sciences, Japan). In total, 20 liters of press gas emissions was obtained per panel together with a liquid condensate.
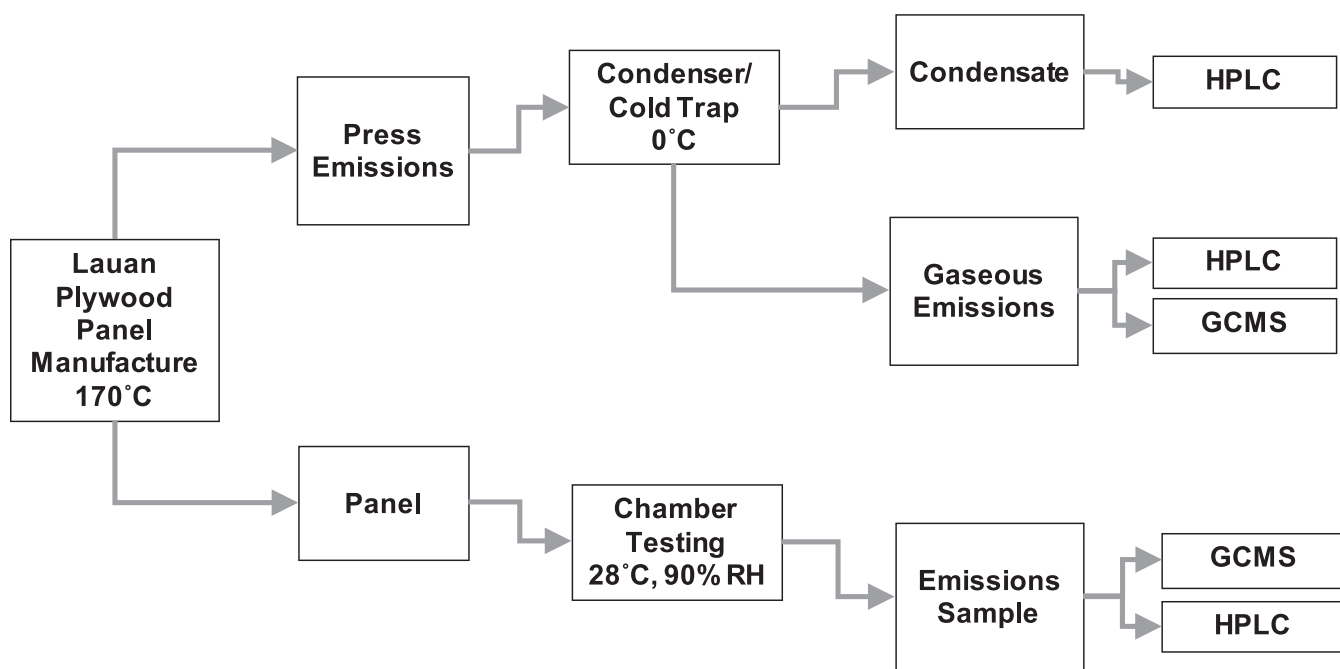

Figure 1.—Methodology for panel manufacture, press emissions capture, and segregation and chemical analysis of segregated fractions. HPLC = high-pressure liquid chromatography; GCMS = gas chromatography–mass spectrometry.

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Condensate analysis–HPLC analysis

Using a published procedure (Miyamoto et al. 2010), each liquid condensate was recovered and washed from the condenser trap assembly before being made to a total volume of 100 mL with distilled water. A subsample of condensate solution (0.5 mL) was combined with acetonitrile and passed through an InertSep™ miniAero 2,4-dinitrophenylhydrazine (DNPH) cartridge (GL Sciences, Japan) and then further washed through with acetonitrile (2 × 1 to 2 mL). This treated solution was then made up to 5 mL with acetonitrile for high-pressure liquid chromatography (HPLC) analysis.

A Shimadzu LC-10Avp System instrument was used for HPLC analysis. An octadecyl-silica (ODS) column (Inertsil® ODS-3 5 μm, 4.6 mmφ by 250 mm; GL Science, Japan) was used running a mixed acetonitrile-water gradient system at 40°C with an ultraviolet detector at 360 nm wavelength (Grigsby et al. 2019).

GC-MS analysis

Following a published methodology, a 1-liter subsample press emission gas sample was passed through a DNPH cartridge to absorb aldehydes (Miyamoto et al. 2010). This reacted DNPH cartridge was then processed using HPLC preparation and analysis methodology above. Volatile compounds were absorbed to Tenax cartridges (Tenax®TA, Perkin Elmer) by passing a second gas subsample (500 mL) through the Tenax cartridge. Gas chromatography–mass spectrometry (GC-MS) analysis used a Shimadzu GC2010 instrument fitted with a Perkin Elmer Turbomatrix automated thermal desorber and Shimadzu QP2010 quadrapole detector system. Cold trap desorption was from −5°C to 270°C over 42 minutes. Gas chromatography was performed using a J&W DB-624 column with a three-step temperature ramp from 40°C (4 min at 7°C/min, then to ~100°C at 10°C/min, followed by ~240°C [10 min]). Chromatogram analysis and interpretation used Shimadzu LabSolutions and GCMSolution 4.11 SU2 software.

Chamber testing of panelboard emissions

Small chamber testing was undertaken on selected samples according to JIS A 1901 (Japanese Standards Association 2015) at 28°C and 50 percent relative humidity. For each panel set, panelboards were trimmed to 165 by 165 mm and two panels were taped together to give two exposed surfaces (147 by 147 mm) with a total exposure area of 0.0432 m^2. These panels were then mounted inside the chamber, and the chamber was sealed. After 24 hours (and again at 8 days), the chamber emissions were sampled by drawing 0.5 liter of gas separately through both Tenax and DNPH cartridges. The DNPH and Tenax cartridges were each processed as described in earlier text in preparation for HPLC and GC-MS analyses, respectively.

Results

Press emissions

As outlined in Figure 1, pressing lauan veneer panels with each renewable produced press emissions that were segregated into condensate and gaseous fractions. These emissions were dominated by water loss (condensate) with press emissions also consisting of both SVOCs and VVOCs. Aldehydes, ketones, and terpenes were identified as the primary components of press emissions from each panel set (see the Supplementary Materials available at https://doi.org/10.13073/FPJ-D-18-00014.s1). This was not unexpected because processing lauan produces moisture below 150°C, with acetaldehyde, formaldehyde, and acetone predominantly liberated above this temperature as the wood undergoes the onset of thermal degradation (Chen et al. 2011). However, analysis of gaseous fractions revealed differences in the composition of VOCs in press emissions across the panel series compared with using lauan veneer only.

For the gaseous fractions of press emissions, the VOC components were further segregated into hydrazine-reacted and Tenax-absorbed fractions (Fig. 1). For the wood-only panel, HPLC of the hydrazine-reacted fraction revealed acetaldehyde (ca. 5,000 μg/m^3) and acetone (ca. 5,000 μg/m^3) in greater quantity together with hexaldehyde (ca. 2,000 μg/m^3; Fig. 2). Panels bonded with soy flour were dominated by acetaldehyde (>17,000 μg/m^3), with both acetone (ca. 6,000 μg/m^3) and hexaldehyde (ca. 4,000 μg/m^3) also found in relatively higher proportions than the veneer-only panel. Propionaldehyde and butanone were found in similar proportions (ca. 300 μg/m^3) for the soy flour and wood-only panels. Formaldehyde, a key compound in panelboard emissions, was found to be higher in soy panels (1,470 μg/m^3) compared with pressing veneer (460 μg/m^3). GC-MS analysis of the Tenax-absorbed gaseous fraction identified terpenes as a significant contributor to VOCs, with these found to be variable (120 to 1,000 μg/m^3) across all panels. This was attributed to inherent variations in veneer thickness, density, and extractables distribution across the lauan substrate used (Grigsby et al. 2019). The soy flour panels were also distinguished by 2-methylpropionaldehyde (ca. 100 μg/m^3) and hexane (ca. 600 μg/m^3) contributing to VOCs in press emissions. This latter compound likely arises as a residue from soybean oil extraction (Lusas and Riaz 1995) and provides some context to the quantities of other compounds found in press emissions. Collectively, the total VOC compounds in the Tenax fraction was relatively higher with soy flour panels (2,700 μg/m^3) compared with press emissions from the lauan veneer alone (<1,200 μg/m^3; Fig. 2).

Corn gluten meal (CGM) was also used to bond panels and provided a contrast in both amino acid composition and carbohydrate content compared with soy flour. The gaseous fraction of press emissions of CGM panels was dominated by acetaldehyde (ca. 13,500 μg/m^3) and hexaldehyde (>16,000 μg/m^3; Fig. 2). In these panels, hexaldehyde was significantly higher with use of soy flour (ca. 4,500 μg/m^3), whereas the acetaldehyde content was relatively lower than found with soy flour (>17,000 μg/m^3). Analysis also revealed propionaldehyde (800 to 1,200 μg/m^3) and valeraldehyde (1,800 to 2,200 μg/m^3) were significantly higher than with either lauan or soy flour. While variable, acetone (8,000 μg/m^3) and formaldehyde (160 μg/m^3) were comparable to using veneer only. For the Tenax-absorbed compounds, GC-MS revealed the gaseous fraction to also contain hexaldehyde (ca. 1,800 μg/m^3), with 2-methylpropanal (ca. 200 μg/m^3) and nonanal (<50 μg/m^3) as other aldehydes identified in this gaseous fraction. CGM does not additionally contribute to terpenes (350 to 500 μg/m^3) and, overall, had a total VOC content of 9,000 μg/m^3 in the Tenax fraction, which was the highest of all renewables evaluated in this study.
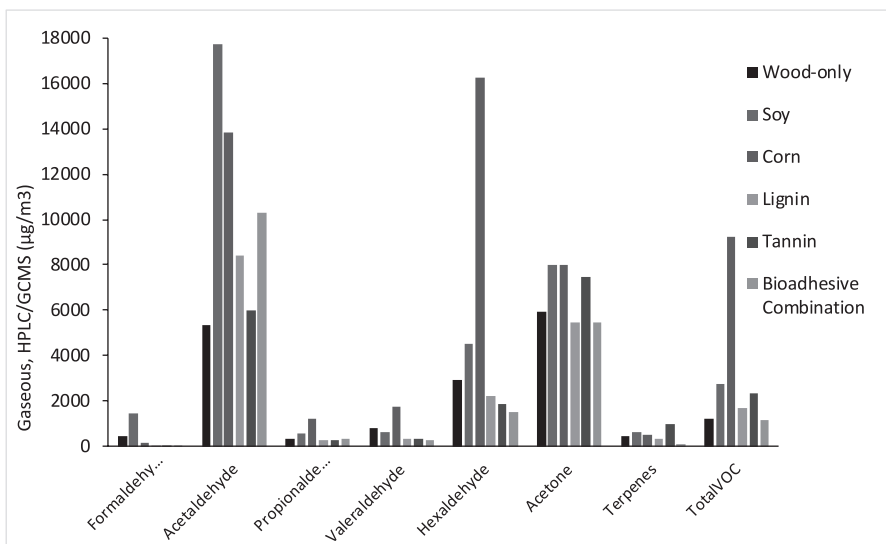
Figure 2.—Results of high-pressure liquid chromatography and gas chromatography–mass spectrometry analyses of the gaseous press emissions capture on panel manufacture.

Kraft lignin and condensed tannin were evaluated for their respective contributions to press emissions to provide a contrast to the proteinaceous materials. Both polyphenolics were found to contribute to differences in VOC profiles. Analysis of the lignin panel press emissions revealed a low formaldehyde content (<30 µg/m^3) in the gaseous fraction, which was substantially lower than with soy flour, CGM, or wood only (>450 µg/m^3). This may reflect the inherent chemical reactivity of lignin toward formaldehyde (Pizzi 2006). While the acetaldehyde content (ca. 8,200 µg/m^3) of the lignin panel press emissions was revealed to be higher than lauan, this was significantly lower than with either soy flour or CGM use. For the other aldehydes, HPLC revealed propionaldehyde and valeraldehyde were lower than wood, with hexaldehyde at a relatively similar content as wood only, with all significantly lower with lignin use than found using either protein source. Other aldehydes, such as nonanal, 2-methylpropanal, and tolualdehyde, isolated from pressing with soy flour or CGM were not identified in the top 20 VOCs isolated from lignin panels. The acetone content was comparable to lauan with butanone also isolated in lower quantity. In the Tenax fraction of the lignin panel press emissions, acetone and hexaldehyde were comparable to lauan. Acetic acid and the kraft process residual by-product dimethyl disulfide were other compounds also identified in the press emissions of these lignin panels.

Wood panels prepared with condensed tannin revealed significant differences in VOC profiles of gaseous press emissions. Analysis revealed tannins also contributed to substantially lower formaldehyde emissions (<25 µg/m^3) compared with wood only (ca. 500 µg/m^3). The acetaldehyde content (6,000 µg/m^3) was also the lowest of the renewables used, as was valeraldehyde (300 µg/m^3) and hexaldehyde (1,850 µg/m^3), but these values did not substantially differ from wood only. The terpenes and acetone contents of tannin panel press emissions were comparable to other renewables. The tannin panel press emissions, like lignin, were also distinguished by a greater acetic acid content (>200 µg/m^3), but the significance of this is unknown because the relative contributions of acetic acid from the other renewable panel sets were likely masked by the higher amounts of other VOCs in their press emissions.

For panels bonded with a bioadhesive composition formulated from soy flour, lignin, and tannin, analysis of press emissions also revealed contrasting VOC profiles compared with proteinaceous material use or wood-only panels (Miyamoto et al. 2019). The hydrazine-reacted gaseous fraction found acetaldehyde in higher amounts ($>10,000$ µg/m^3) than the wood-only panel (ca. 5,500 µg/m^3), whereas acetone (ca. 5,500 µg/m^3) and hexaldehyde (ca. 1,500 µg/m^3) were found in relatively similar proportions in press emissions. This contrasted the press emissions using soy flour for which acetaldehyde was in substantially greater quantity (8,000 µg/m^3) with both acetone ($>6,000$ µg/m^3) and hexaldehyde (ca. 4,000 µg/m^3), which were also found in relatively higher proportions. Formaldehyde (20 µg/m^3) and valeraldehyde (250 µg/m^3) were also found to be proportionately lower in the bioadhesive panel press emissions than in the control panel. Propionaldehyde and 2-methylpropionaldehyde (ca. 100 µg/m^3) were found in similar proportions as the wood-only and soy panel press emissions. Hexane was also identified (<200 µg/m^3) in bioadhesive panel press emissions and can be attributed to the soy component in this formulation. Dimethyl disulfide (ca. 100 µg/m^3) and acetic acid (<200 µg/m^3) were also found in bioadhesive panel press emissions and derived from the lignin component in this formulation. Overall, the total VOC compounds in the bioadhesive press emissions (1,100 µg/m^3) were comparable to lauan veneer ($<1,200$ µg/m^3) panels, with the total VOCs higher in soy flour panel press emissions ($>2,400$ µg/m^3).

Discussion

For the renewables used to produce panels with lauan veneer, it was evident that use of proteinaceous materials contrasted the VOC profiles produced by the polyphenolics. Oxidative degradation products such as aldehydes were prominent in the VOC profiles with both soy flour and CGM and found to contribute significantly to higher acetaldehyde

and hexaldehyde emissions. Acetaldehyde ($>$14,000 $\mu g/m^3$) was significantly higher for both these renewables compared with lauan (ca. 5,000 $\mu g/m^3$), which itself is known for acetaldehyde emissions (Li et al. 2004). Hexaldehyde featured in both the HPLC and GC-MS analyses for soy flour and CGM press emissions, but it was CGM that was revealed as a significant contributor of this longer chain aldehyde. For formaldehyde, it was evident that soy flour contributed to emissions of this compound, which were in relatively higher amounts compared with wood only. In the case of other aldehydes, CGM promoted emissions of propionaldehyde and valeraldehyde, whereas soy flour contributed only relatively smaller increases in propionaldehyde. Soy flour and CGM also led to emissions of methyl propionaldehyde. With acetone and, to a lesser extent, butanone, there were no significant increases of these compounds with use of any one renewable component. This suggests the lauan veneer as the primary source of acetone in press emissions.

In contrast to the proteinaceous materials, lignin and particularly tannin were determined to contribute to significantly lower formaldehyde and acetaldehyde emissions on pressing. Tannin promoted the lowest press emissions of formaldehyde (12 $\mu g/m^3$). For acetaldehyde emissions it was evident both lignin and tannin contributed to lower emissions of this compound compared with either soy or corn use. The lowest acetaldehyde emissions were with tannin (6,000 $\mu g/m^3$) while use of lignin contributed ca. 8,500 $\mu g/m^3$. This result can be attributed to the reactivity of lignin and particularly tannin toward both formaldehyde and acetaldehyde, consistent with the inherent chemistries of these renewables (Garcia et al. 2016). Moreover, the presence of lignin and tannin in the bioadhesive resin also conferred significantly lower formaldehyde emissions ($<$20 $\mu g/m^3$) than lauan veneer. However, in the case of acetaldehyde, press emissions from the bioadhesive panel were relatively higher (ca. 10,000 $\mu g/m^3$) and attributed to using soy flour in this resin formulation.

Panel emissions

Panel emissions postmanufacture were evaluated to compare proportions of VOCs generated on panel manufacture with those liberated from panels over time. Initial panel emission VOC releases were sampled over an 8-day period for the wood-only, soy, and bioadhesive panels (Tables 1 and 2). Chamber testing air samples at 24 hours and 8 days were segregated into DNPH and Tenax-absorbed fractions as above. This revealed generally that those compounds obtained in high proportions in press emissions were prominent in panel VOC emission profiles. Analysis found five individual components released at a rate of $>$6 $\mu g/m^2/h$ from the lauan veneer panel, together with a range of terpenes after 24 hours (Table 1). Formaldehyde (27 $\mu g/m^2/h$), acetaldehyde (195 $\mu g/m^2/h$), and acetone (51 $\mu g/m^2/h$) were predominantly released from this panel. The release of these compounds was significantly higher than the terpenes fraction (32 $\mu g/m^2/h$) with acetic acid (8 $\mu g/m^2/h$) and 1-butanol (12 $\mu g/m^2/h$) also identified as significant VOC emissions. For the soy flour panel, chamber testing revealed a formaldehyde emission rate of 20 $\mu g/m^2/h$, with acetaldehyde (175 $\mu g/m^2/h$) being comparable to the lauan panel. The soy flour panel had lower emissions of acetone ($<$40 $\mu g/m^2/h$) and terpenes (10 $\mu g/m^2/h$) than wood only. In the case of the bioadhesive panel, this showed a

Table 1.—Summary of emission rates from panels after 24-hour chamber testing (including significant volatile organic compounds [VOCs] $>$0.5 g/m²/h).

	Compound emission rate ($\mu g/m^2/h$)		
	Veneer only	Bioadhesive	Soy flour
Formaldehyde	27.4	13.3	19.9
Acetaldehyde	195.1	140.7	176.6
Acetone	51.0	35.5	35.6
Acetic acid	7.7	9.6	6.5
Butanol	11.8	4.3	3.4
Butanone	—	—	—
Total terpenes	32.3	5.4	10.1
Total VOCs	53.2	19.3	20.6

significant reduction in both formaldehyde ($<$14 $\mu g/m^2/h$) and acetaldehyde (ca. 140 $\mu g/m^2/h$) emission rates. Other VOC components were also lower, including acetone ($<$40 $\mu g/m^2/h$) and terpenes ($<$10 $\mu g/m^2/h$). Only the acetic acid emission rate remained unchanged compared with the wood-only panels. Valeraldehyde and hexaldehyde were not identified in the top five panel emissions of any panel sets, despite being significant contributors to VOC profiles in press emissions.

Air sampling at 8 days revealed lower rates of aldehyde emissions (Table 2). Formaldehyde (16 $\mu g/m^2/h$) and acetaldehyde (38 $\mu g/m^2/h$) emissions were significantly lower for the lauan panel. A formaldehyde emission of ca. 15 $\mu g/m^2/h$ was found for the soy panel, which was higher than the bioadhesive panel ($<$10 $\mu g/m^2/h$). However, the soy flour panel retained a significantly higher acetaldehyde emission rate (90 $\mu g/m^2/h$). Across the panel sets, the acetone (ca. 55 $\mu g/m^2/h$) and acetic acid (ca. 10 $\mu g/m^2/h$) emissions rates remained relatively unchanged at 8 days compared with 24 hours. Butanone (ca. 5 $\mu g/m^2/h$) emerged as a significant VOC emission at 8 days, with an associated loss of butanol ($<$0.5 $\mu g/m^2/h$). Overall, while panel emissions have been undertaken over a relatively short period, testing has revealed prominent aldehyde and ketone compounds found in press emissions (Fig. 2) were also present in panel VOC emission profiles (Tables 1 and 2). Acetaldehyde together with acetone were major components of press emissions and observed as primary components of panel VOC emissions. The relative volatility of formaldehyde and terpenes also contributed to their presence in chamber testing.

Table 2.—Summary of emission rates from panels after 8-day chamber testing (including significant volatile organic compounds [VOCs] $>$0.5 g/m²/h).

	Compound emission rate ($\mu g/m^2/h$)		
	Veneer only	Bioadhesive	Soy flour
Formaldehyde	15.8	6.1	14.9
Acetaldehyde	38.1	52.8	88.9
Acetone	55.2	40.9	45.0
Acetic acid	10.1	8.5	7.6
Butanol	—	—	—
Butanone	5.6	7.9	5.1
Total terpenes	8.9	1.7	2.6
Total VOCs	19.0	10.2	10.3

Findings

Overall, analysis of press emissions has revealed the composition of VOCs evolved during panel manufacturing using common renewables used in panelboard adhesives and resins. In processing lauan panels at 170°C, the renewables have variously produced thermal degradation products that contributed VOCs to both press and panel emissions. Both soy flour and CGM variously contributed to formaldehyde, acetaldehyde, and hexaldehyde in VOC profiles of press emissions. These proteinaceous materials also produced valeraldehyde and propionaldehyde. In contrast, using polyphenolic materials led to reduced contributions of formaldehyde in press emissions. While using lignin led to lower formaldehyde, tannin significantly reduced both formaldehyde and acetaldehyde in press emissions as well as contributing to reductions of other higher aldehydes. In initially evaluating panel emissions over 1 week, chamber testing revealed those prominent VOCs in press emissions such as acetaldehyde and acetone also manifest in panel emissions.

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