

Emissions of hazardous air pollutants from lumber drying

Mike Milota*
Paul Mosher

Abstract

NCASI Method 105 was used during lumber drying to measure emissions of methanol, phenol, formaldehyde, acetaldehyde, propionaldehyde, and acrolein from red alder (*Alnus rubra*), ponderosa pine (*Pinus ponderosa*), white wood (a mix of western pines, fir, and spruce), Douglas-fir (*Pseudotsuga menzeisii*), western hemlock (*Tsuga heterophylla*) and white spruce (*Picea glauca*). Methanol and acetaldehyde were emitted in the greatest quantities. Results indicate a strong dependence on temperature for methanol and formaldehyde while the other compounds do not show a consistent trend. At conventional temperature, the acetaldehyde was often emitted in a greater amount than the methanol. At the higher temperature the reverse was true. The information should be helpful to mills drying these species for making a decision about whether they are a major source for HAPs as defined by the Environmental Protection Agency. The results should also be helpful in planning future testing.

The emission of organic compounds is of great importance to the forest products industry due to current and pending federal maximum achievable control technology (MACT) rules related to the hazardous air pollutants (HAPs) emitted from dryers, presses, and boilers. Hazardous air pollutants are a subset of the total organic material or volatile organic compounds (VOCs) emitted during processing. For purposes of the MACT rules, methanol, phenol, formaldehyde, acetaldehyde, propionaldehyde, and acrolein are considered surrogates for all HAPs from wood dryers and presses. These HAPs were measured for some western species in the work presented.

A facility with the potential to emit greater than 10 t/y (tons per year) of any one HAP or 25 t/y of combined HAPs from the site is considered a major source for HAPs and must comply with the MACT rules. This means adding end-of-pipe control equipment on certain dryers, presses and boilers, continuous monitoring, and certain reporting requirements. Lumber dry kilns were excluded from the requirement for controls; however, this exclusion is in review due to court rulings in June of 2007.

Facilities had the opportunity to demonstrate that the concentration of HAPs crossing the fence line did not pose a health risk to neighbors in lieu of control equipment (known as the low-risk option). Qualifying for this required knowing how much HAPs are emitted and modeling the dispersion of the HAPs into the environment. Lumber dry kilns were particularly problematic in qualifying for this option because of small amounts of acrolein and the lack of a single discharge

point. This option was also affected by recent court rulings and will probably not be available in the future.

Three methods are generally accepted by regulatory agencies for HAP testing from wood processing equipment. All can be found in the National Council for Air and Stream Improvement's Methods Manual (NCASI 2007). Methanol, formaldehyde, and phenol are measured using NCASI Method CI/WP-98.01 by bubbling a gas sample through water in chilled impingers and absorbing the HAPs into the water phase. The other HAPs are too volatile or unstable for Method 98.01 to work well. This is compensated for in NCASI Method 99.02 by sampling the gas leaving the impingers using an evacuated Summa canister. This method, however, is expensive, much more complex, and can give variable results. NCASI Method ISS/FP-A105.01 was recently developed as a modification to Method 98.01 to eliminate the need for canisters by derivatizing the aldehydes to more stable and less volatile aldehyde oximes which remain in the water phase.

NCASI (2002), one in a series of technical bulletins on the emissions from many types of wood processing equipment,

The authors are, respectively, Professor and Research Assistant, Wood Science and Engineering, Oregon State University, Corvallis, Oregon (Mike.Milota@OregonState.edu, Paul.Mosher@OregonState.edu). The authors appreciate support from AMEC, Hampton Affiliates, National Council for Air and Stream Improvement, Rosboro Lumber, West Fraser, and Weyerhaeuser. This paper was received for publication in August 2007. Article No. 10394.

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Table 1. — Past studies of MACT HAP emissions.

Source	Species	Temperature (°F (°C))	MC		Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
			Initial	Final					
NCASI 2002	Loblolly pine ¹	235 (112.7)	NA	22	0.240	0.018	0.044	0.002	0.006
NCASI 2002	Loblolly pine ²	235 (112.7)	NA	12 to 15	0.200	0.013	--	--	--
Milota 2005	White spruce ¹	221 (105)	32.4	15	0.021	0.0014	0.018	NA	0.0006
McDonald 2002	Radiata pine	212 (100)	140	3.5	0.139	0.005	0.042	NA	NA

¹Measured using NCASI Method 99.02.

²Measured using NCASI Method 98.01.

reports on southern pine lumber dried in commercial and laboratory kilns. The HAPs were measured by Method 99.02, and the results are shown in **Table 1**. This was some of the first work on HAPs from lumber drying and formed a basis for regulations and agency testing requirements. Methanol was the HAP emitted in the largest quantity, and it seemed clear from the results that a producer would reach 10 t/y of methanol before reaching 25 t/y of combined HAPs from lumber kilns. Based on this assumption, most testing of kiln exhaust has been conducted using NCASI Method 98.01 for methanol and formaldehyde and, at least for lumber, the other four MACT HAPs have been largely ignored.

MacDonald et al. (2002) measured the full spectrum of compounds emitted from radiata pine. The methanol emissions from the radiata were lower than the southern pine, however, the drying temperature was lower. Milota and Mosher (2006) demonstrated that there is a strong effect of temperature on methanol emissions. In unpublished work done in 2005, Milota found low levels of HAP emissions from white spruce (**Table 1**). This was consistent with the low starting moisture content.

The present work was initiated after higher than expected levels of acetaldehyde were measured from lumber as it dried. This work had two main objectives. One was to determine the HAP emissions for several species. A second was to determine how the emissions of the lesser reported HAPs, such as acetaldehyde, vary with kiln temperature. The results are significant to many facilities that have based operating permits only on the levels of methanol and formaldehyde emitted. They are also important to facilities that want to use actual measured emission factors rather than Environmental Protection Agency (EPA) estimates.

Procedures

Red alder (*Alnus rubra*), ponderosa pine (*Pinus ponderosa*), white wood (a mix of western pines, fir, and spruce), Douglas-fir (*Pseudotsuga menziesii*), and western hemlock (*Tsuga heterophylla*) were obtained from mills in western Oregon and Washington. White spruce (*Picea glauca*) was obtained from a western Canadian mill. All lumber was sampled by mill personnel, cut to 4-foot (1.22-m) lengths, and wrapped in plastic. Sampling was conducted over an extended time at the mills to assure that pieces from different logs were sampled. The lumber arrived in Corvallis within 48 hours of shipping. The lumber was then stored in either a refrigerator or freezer, depending on the length of time until it would be dried. The red alder lumber was 5/4 random width. The softwoods were 2 by 4 or 2 by 6 dimension lumber.

The small laboratory kiln and procedures described in Milota and Mosher (2006) were used to dry the wood. The kiln is

approximately 1.22 m (4 ft.) on each side. Dry- and wet-bulb temperatures are measured on the entering-air side of the load. The kiln is indirectly heated by steam to maintain the desired dry-bulb temperature. Humidity is controlled by regulating dry compressed air entering the kiln to maintain the desired wet-bulb temperature.

Prior to drying, the lumber was trimmed to 1.12 m (44 in) by removing 50 mm from each end and placed in the kiln on 19-mm-thick stickers (3/4 in). The loads were two to three boards wide, depending on the lumber width, and 10 to 14 courses high, depending on thickness. The conventional temperature drying schedules (<94 °C) were provided by the mills supplying the lumber. The higher temperature drying schedule was selected to match that used in NCASI (2002). The final dry-bulb setting for each schedule is shown in the results. The air velocity was 750 ft/min (3.8 m/s). Each board was weighed prior to and after drying, then oven-dried and reweighed so that the initial and kiln-dry moisture contents could be determined. Drying from green to the final moisture content was accomplished without opening the kiln or other interruptions.

Hydrocarbon measurement

A 1.8 L/min gas sample was withdrawn from the kiln near the exhaust port and directed to a JUM VE7 hydrocarbon analyzer. Heated dilution gas was metered into the hydrocarbon sample gas, if necessary, to lower the gas moisture content to less than 15 percent. All components were heated to prevent the condensation of water or organics. The hydrocarbon analyzer was calibrated every three to six hours by introducing calibration gases (EPA protocol 601 ppm, EPA Protocol 300 ppm, and < 0.1 ppm air) near the probe tip at ambient pressure. The methodology followed is similar to EPA Method 25A (Code of Federal Regulations 1991).

HAP sampling

The sampling train for Method 105 is shown in **Figure 1**. The impingers were in a stirred glycol solution maintained at -1 °C. Prior to each sampling interval, the impingers were lab washed and 15 mL of BHA solution were added to each. The solution contained a stoichiometric excess of O-benzylhydroxylamine hydrochloride for derivitization of the aldehydes to aldehyde oximes. After assembly, the sampling train was checked for leaks by drawing a vacuum. The gas flow rate through the sampling train, 450 to 500 mL/min, was measured using a bubble meter before and after each sampling interval. There were 7 to 28 sampling intervals per kiln charge, each from two to three hours in duration after which the liquid from the impingers was weighed and placed in a vial. The impingers were rinsed with water, then hexane, and these rinses

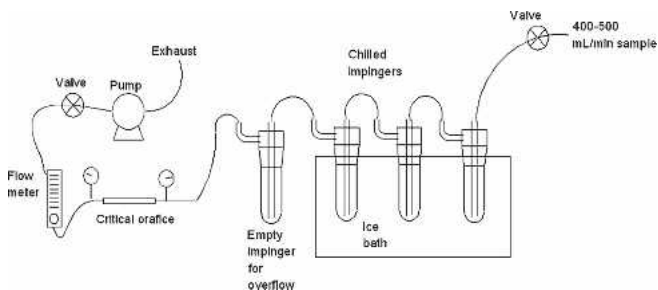


Figure 1. — HAPs sampling train. Three trains were used so that duplicates and recovery spikes could be run.

were added to the vial. It was then refrigerated. Blanks of BHA solution, duplicate samples, and recovery spikes were collected for almost every charge. In the lab the samples were extracted with hexane. The aqueous fraction was analyzed by gas chromatography with a flame ionization detector for methanol and phenol. The hexane fraction was analyzed by gas chromatography with a nitrogen-phosphorous detector for the oximes of formaldehyde, acetaldehyde, propionaldehyde, and acrolein. The complete procedures are described in NCASI (2007).

A kiln blank and a sticker blank were run by bringing the kiln to 82.2 °C over a 30-minute period with no wood or stickers. At this point, a 2-hour Method 105 sample was taken. The kiln was then briefly opened, the stickers normally used for drying were placed in the kiln, and the kiln was closed. Another 2-hour Method 105 sample was then taken. The samples were then analyzed as described above.

Calculations

Hydrocarbon emissions are calculated from the concentration detected by the analyzer and the vent rate of the dryer. The analyzer reading in parts per million is converted to a dry gas concentration (mass/volume) using psychrometric relations and the ideal gas law. This value is then multiplied by the dry gas flow rate (volume/time) and the result integrated over time. The total hydrocarbon values are expressed as carbon (denoted as lb_c) meaning that only the mass of the carbon is used in calculating the hydrocarbon mass in the wood exhaust. No correction is made for the response of the analyzer to oxygenated compounds.

The mass of HAPs in the impingers is determined from the concentrations detected in the water or hexane and quantity of each solvent. The HAPs emitted from the kiln are calculated by scaling up the mass collected in the impingers by the ratio of the gas flow rate through the kiln to the gas flow rate through the impingers. This ratio varies from approximately 20 to 400 depending on the vent rate of the kiln. HAP sampling at the kiln occurred during 60 to 80 percent of the kiln cycle. For the periods between samples, an average emission rate was calculated based on the mass collected during the periods before and after the interval.

Results and discussion

VOC emissions of 1.6 and 3.0 lb_c/mbf were measured from ponderosa pine lumber at conventional and high temperature, respectively (Table 2). The value measured at low temperature compares favorably with 1.42 lb_c/mbf measured previously (Milota 2006b) for drying at 82.2 °C. The value measured at high temperature was within the range of 2.4 to 4.4

reported in NCASI (2002) for loblolly pine dried at 112 °C. For Douglas-fir and western hemlock (Tables 3 and 4), the levels of VOC emissions are similar to those reported in Milota (2006b) and Milota and Mosher (2006), respectively. There are no values for comparison to the VOC emissions for the spruce and white wood (Tables 5 and 6). The 0.17 lb/mbf of VOC emissions from red alder lumber during drying (Table 7) are slightly lower than previously reported (Milota 2006a), 0.2 lb/mbf. However, given the variability in the past work, the present value is not inconsistent. VOC emissions increased to 0.66 at 235 °F. Current industrial practices do not use this high of a temperature; however, red alder can be dried with good quality at high temperature (Kozlik and Boone 1987).

The methanol emissions from ponderosa pine at conventional temperature were lower (0.035 versus 0.065 lb/mbf) and the formaldehyde emissions were similar (0.0027 versus 0.0029 lb/mbf) to those reported in Milota (2006b) for ponderosa pine at 82.2 °C. At high temperature, the methanol and formaldehyde emissions were lower than reported for loblolly pine dried at the same temperature (Table 1), 0.144 compared to 0.22 to 0.24 lb/mbf and 0.009 versus 0.013 to 0.018 lb/mbf, respectively.

The acetaldehyde and propionaldehyde emissions from ponderosa pine were similar to those reported for loblolly pine and the acrolein emissions were higher. The acrolein reported by NCASI was measured using Method 99.02 and the stability of acrolein might result in a low value. Phenol emissions were not detected for ponderosa pine or any other species in this study.

The methanol and formaldehyde emissions from Douglas-fir lumber (Table 3) measured at 76.7 °C (170 °F), 0.024 and 0.0008 lb/mbf, are almost identical to those previously reported at the same temperature (Milota 2006b), 0.023 and 0.0010 lb/mbf. These increase by over a factor of four as the temperature is raised to 112.7 °C (235 °F). The quantity of acetaldehyde emitted was similar to methanol at conventional temperature; however, at high temperature, the acetaldehyde emitted (0.067 lb/mbf) was considerably less than the methanol (0.117 lb/mbf).

The methanol emissions from western hemlock lumber (Table 4) ranged from 0.075 to 0.187 lb/mbf and the formaldehyde emissions from 0.0014 to 0.0045 lb/mbf. These are 10 to 20% lower than predicted by the equation in Milota and Mosher (2006). The equation, however, is based on emissions as measured by Method 98.01. Past work (Milota and Mosher 2008) suggests that Method 105 gives lower results, especially for formaldehyde. The methanol emissions more than double between 82.2 °C and 112.7 °C and the formaldehyde emissions more than triple. The quantity of acetaldehyde emitted was similar to or greater than the methanol at conventional temperature but less than the methanol at the high temperature.

White spruce (Table 5) had lower HAP emissions than the other species; however, the wood was at low initial moisture content. The past results in Table 1 are from the same shipment of spruce; however, the previous work was done using NCASI Method 99.02 during drying at 105 °C. If one adjusts for temperature, it can be seen in Table 6 that the HAP emissions in the present study are somewhat higher than

Table 2. — HAP emissions from ponderosa pine. The last row is the ratio of the emissions at high temperature to lower temperature.

Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
	Initial	Final						
170 (76.7)	82.6	15.0	1.59	0.035	0.0027	0.042	0.0019	0.0017
235 (112.7)	89.1	15.0	3.00	0.144	0.0092	0.028	0.0032	0.0045
ratio				4.11	3.41	0.66	1.68	2.64

Table 3. — HAP emissions from Douglas-fir.

Sample	Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
		Initial	Final						
B	170 (76.7)	56.9	15.0	0.241	0.024	0.0008	0.030	0.0004	0.0005
C	180 (82.2)	43.7	15.0	0.575	0.050	0.0023	0.050	0.0005	0.0009
A	200 (93.3)	64.3	15.0	0.707	0.068	0.0018	0.043	0.0005	0.0009
A	200 (93.3)	59.5	15.0	0.879	0.069	0.0019	0.071	0.0006	0.0004
C	235 (112.7)	47.7	15.0	1.206	0.117	0.0043	0.067	0.0008	0.0012

Table 4. — HAP emissions from western hemlock.

Sample	Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
		Initial	Final						
D	180 (82.2)	102.3	15.0	0.142	0.075	0.0014	0.078	0.0020	0.0012
F	180 (82.2)	93.5	17.5	0.236	0.094	0.0015	0.141	0.0008	0.0012
E	200 (93.3)	83.9	15.0	0.214	0.044	0.0008	0.133	0.0008	0.0024
E	200 (93.3)	98.6	15.0	0.239	0.077	0.0014	0.128	0.0010	0.0011
F	235 (112.7)	81.6	15.0	0.247	--	--	--	--	--
F	235 (112.7)	76.2	15.0	0.226	0.187	0.0045	0.084	0.0014	0.0019

Table 5. — HAP emissions from white spruce. The last row is the ratio of the emissions at high temperature to lower temperature.

Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
	Initial	Final						
180 (82.2)	33.5	15.0	NA	0.025	0.0013	0.036	0.0003	0.0005
235 (112.7)	32.7	15.0	0.11	0.078	0.0044	0.031	0.0007	0.0010
ratio				3.12	3.38	0.86	2.33	2.00

Table 6. — HAP emissions from white wood. The last row is the ratio of the emissions at high temperature to lower temperature.

Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
	Initial	Final						
190 (87.7)	119.2	15.0	1.39	0.074	0.0045	0.144	0.0044	0.0050
235 (112.7)	106.8	15.0	2.31	0.188	0.0101	0.049	0.0043	0.0058
ratio			1.66	2.54	2.24	0.34	0.97	1.16

previously measured; however, the measurement method is different. Again in this data set, the methanol emissions greatly increase with temperature while the acetaldehyde emissions do not.

The exact values for the HAP emissions from the white wood sample are of limited value because it is a mixture of species that can change with log supply. The sample dried was

at least 50 percent ponderosa pine accounting for the relatively high VOC emissions. The important thing to note from this data is the trend for methanol emissions to increase with temperature while acetaldehyde emissions do not (**Table 6**).

The methanol emissions from red alder at the lower temperature, 0.173 lb/mbf, were greater than for many of the softwoods. This may be due to the greater number of methoxy

Table 7. — HAP emissions from red alder. The last row is the ratio of the emissions at high temperature to lower temperature.

Temperature (°F (°C))	MC		VOC	Methanol	Formaldehyde	Acetaldehyde	Propionaldehyde	Acrolein
	Initial	Final						
	(percent)		----- (lb/mbf) -----					
180 (82.2)	102.1	8.0	0.173	0.124	0.0009	0.114	0.0014	0.0011
235 (112.7)	100.1	10.2	0.659	0.416	0.0048	0.129	0.0016	0.0018
ratio				3.55	5.45	1.13	1.14	1.64

groups on the hardwood lignin compared to softwood lignin. Acidic conditions in the wood in the presence of water at the temperatures encountered during drying might cleave these groups. The acetaldehyde was also relatively high for the red alder compared to most of the softwoods. The glucuronoxylan component of the hardwood hemicellulose has a greater number of acetyl groups per monomer unit than the hemicellulose in softwoods. Acid hydrolysis of these groups at the dryer temperature may contribute to acetaldehyde formation. Neither of these suggested mechanisms has been proven, however. As with the softwoods, the HAP emissions increase with temperature. Despite the HAP emissions being higher from the red alder, hardwood mills tend to be smaller than softwood mills and are unlikely to be a major source for HAPs.

All of the HAPs measured in this paper were obtained by NCASI Method 105. The spike recoveries ranged from approximately 60 to 99 percent. The poorest spike recoveries were for the formaldehyde. The best spike recoveries were for methanol (usually > 90%) and acetaldehyde (usually > 80%). While these are satisfactory in the published method for the concentrations measured, they indicate that another method might give higher results for the emissions. A comparison of Method 105 to 98.01 can be found in Milota and Mosher (2008).

The pattern of emissions during the kiln cycle is shown for hemlock at high and conventional temperature in Figure 2. At the conventional temperature (top graph), the rate of emissions decreases with time for all compounds after the initial warm up period. This was generally true for all species tested. Acetaldehyde emissions in particular are high early in the cycle while the temperature is lower. At high temperature (bottom graph), the rate of methanol and formaldehyde emissions increased dramatically as the wood dries. This occurred with every species tested. At high temperature, the rate of emission of the other HAPs do not follow a consistent pattern.

The kiln and sticker blanks had minimal emissions. When the stickers were present, the emissions were similar to or less than less than from the empty kiln for all HAPs except methanol. Methanol was not detected from the empty kiln. We therefore conclude that the stickers are not contributing to the HAPs, except methanol. If the sticker blank had been a 48-hour cycle, the methanol, acetaldehyde, and formaldehyde released would have been approximately 1, 0.01, and 9 percent, respectively, of that emitted during a typical kiln cycle. The formaldehyde value is higher; however, it still constitutes only about 1 to 3 percent of the total HAPs that would be detected during a kiln cycle. Thus, any effect on total HAPs due the kiln and stickers is minor. Also, if the kiln had been run longer prior to testing, the results would likely be lower because the kiln would have had time to bake out. As run, the

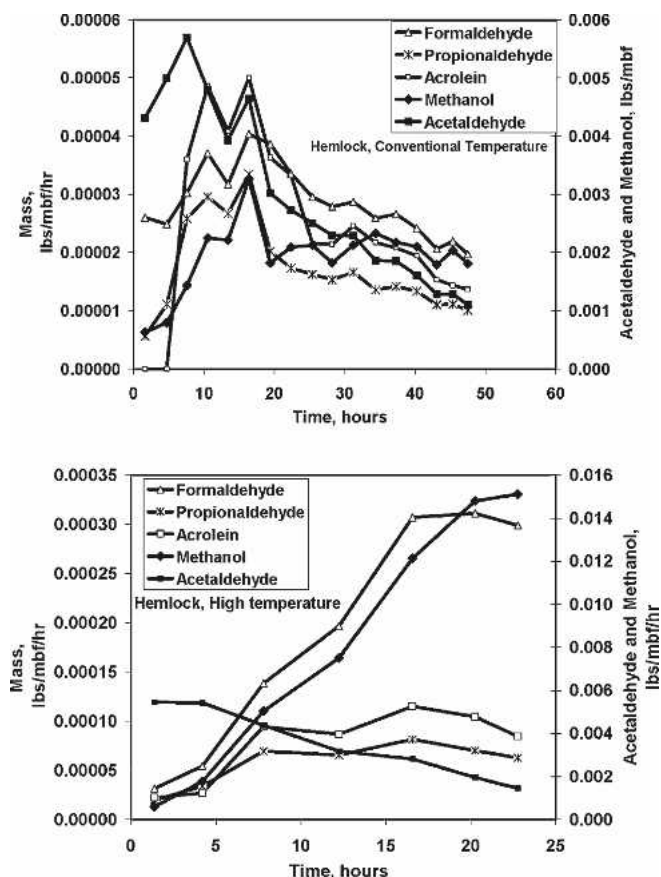


Figure 2. — Rate of HAP emissions (lb/mbf/hr) from hemlock lumber drying at conventional- (upper) and high-temperature (lower) drying.

kiln and sticker blanks represent a worst case. The results suggest that either HAPs are adsorbing onto the kiln walls or absorbing in the wet-bulb water and desorbing or, that residue in the kiln is degrading.

Subpart DDDD (Code of Federal Regulations 2004) contains some emission factors intended to be used with the low-risk option; however, they tend to be more broadly applied by state regulatory agencies. Data in this paper suggest that the acrolein estimate in Subpart DDDD is high by a factor of eight or more and mills could easily justify a lower value by site specific testing. The EPA estimate for acetaldehyde, 0.065 lb/mbf, is low for hemlock and alder and high for the other species tested. Similarly, the estimate for formaldehyde (0.034 lb/mbf) is high in many cases, but low at high temperature. In contrast to the EPA estimate of 0.01 lb/mbf, no phenol was detected.

To date, the industry has not paid much attention to acetaldehyde emissions from lumber drying when deciding if a facility is a major source for HAPs because the methanol emissions from lumber drying were assumed to be so much greater than the other HAPs. This assumption was based on the NCASI (2002) study in which southern pine lumber was dried at a 112.7 °C. As a result, other studies have concentrated on quantifying the methanol. The current research suggests that this is a poor assumption and the acetaldehyde emissions may be at least equal to and often greater than the methanol emissions at conventional kiln temperatures.

Conclusions

HAP emissions vary greatly among species. The hardwood species tested had the highest HAP emissions, probably due to the great number of methoxy groups in hardwood lignin and the higher hemicellulose content and number of acetyl groups.

Overall, HAP emissions increase with temperature; however, not all the HAPs are affected by temperature in the same way. Methanol and formaldehyde emissions increase dramatically with temperature while acetaldehyde emissions may decrease.

At conventional temperatures, the acetaldehyde emissions are on the same order as the methanol emissions and should not be neglected when calculating total HAPs.

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