

Hazardous Air Pollutants and Volatile Organic Compounds Emitted during Kiln Drying of Southern Pine Lumber to Interior and Export Moisture Specifications

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Abstract

The emission levels of hazardous air pollutants (HAPs) from wood kilns are needed to comply with the Clean Air Act. Softwood lumber, including southern pine (*Pinus taeda*, *Pinus palustris*, *Pinus echinata*, and *Pinus elliottii*), is considered kiln dry when the wood moisture content reaches below 19 percent, but it is sometimes overdried or dried to 8 percent moisture content for export or interior applications. To study HAP emissions when drying to 8 percent moisture content, green lumber was obtained from a local mill, and 12 charges were kiln dried using three schedules: a 99°C elevated schedule and 116°C and 127°C high-temperature schedules. Methanol and formaldehyde were collected and analyzed using the National Council for Air and Stream Improvement (NCASI) 98.01 method, “Chilled Impinger Method for Use at Wood Products Mills to Measure Formaldehyde, Methanol, and Phenol,” where formaldehyde was analyzed via spectrophotometry using acetylacetone and methanol was analyzed via gas chromatography with a flame ionization detector. The high-temperature schedules had significantly greater HAP emissions than the elevated schedule. When drying from 19 to 8 percent moisture content, methanol and formaldehyde emissions increased by an average of 48 and 52 percent, respectively. Volatile organic compounds (VOCs) were also measured using a flame ionization detector by the NCASI VOC method with results similar to previous studies. The methanol, formaldehyde, and total volatile organic emissions were reported according to the Environmental Protection Agency OTM-26 method, “Interim VOC Measurement Protocol for the Wood Products Industry.” The results were slightly higher than the NCASI VOC results because of more accurate quantification of methanol.

Softwood lumber, including southern pine (SP; *Pinus taeda*, *Pinus palustris*, *Pinus echinata*, and *Pinus elliottii*), is considered kiln dry when the moisture content (MC) of the wood reaches below 19 percent. As such, emission studies conducted to date on SP have focused on drying from green to between 15 and 19 percent MC (Ingram et al. 2000, Milota 2006). The Forest Products Department at Mississippi State University has received inquiries into the additional release of hazardous air pollutants (HAPs) and volatile organic compounds (VOCs) when drying SP to 8 percent MC. Lumber dried to this lower moisture content is typically exported and used in interior applications such as millwork and flooring or other niche markets.

Under the Environmental Protection Agency’s (EPA) National Emissions Standards for Hazardous Air Pollutants

(NESHAP) ruling, facilities are a major source of HAPs when emissions exceed 10 tons of a single HAP or 25 tons of total HAPs per year; other facilities are area sources (EPA 1994). A site that is a major source of HAPs must install the maximum achievable control technology

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Forest Prod. J. 61(3):229–234.

Table 1.—Previous studies of hazardous air pollutant emissions from southern pine.

Source ^a	Species ^b	Temperature (°C)	Emission (kg of chemical/m ³) ^c				
			Methanol	Formaldehyde	Acetaldehyde	Acrolein	Propionaldehyde
NCASI (2002)	Loblolly pine	113	0.058	0.0043	0.011	0.0014	0.0005
NCASI (2002)	Loblolly pine	113	0.048	0.0031			
Milota (2006)	SP	113	0.026	0.0007			
Shmulsky and Dahlen (2008)	SP	116		0.0037	0.011	0.002	
Shmulsky and Dahlen (2008)	SP	127		0.0032	0.0092	0.0014	
EPA (2006b)				0.0082	0.016	0.0022	

^a NCASI = National Council for Air and Stream Improvement; EPA = Environmental Protection Agency.

^b Southern pine (SP) encompasses the four major SP species, including loblolly pine (*Pinus taeda*, *Pinus palustris*, *Pinus echinata*, and *Pinus elliottii*).

^c 1 kg/m³ is approximately 4.16 lb/MBF.

(MACT) for any source regulated under a specific MACT ruling. The lumber industry is currently regulated under the Plywood and Composite Wood Product (PCWP) MACT; however, lumber kilns are currently exempt from emission controls (EPA 2006b, 2007b). For sites where a lumber mill is co-located with a composite plant, such as plywood, the combined emissions can create a major source of HAPs at the site, resulting in MACT requirements for the composite press. Of additional interest is the contentious issue of boiler emission controls. The EPA first proposed a Boiler MACT in 2004, which was vacated in 2007 by the District of Columbia Circuit Court of Appeals (EPA 2004, 2011a). The EPA finalized an updated ruling in March 2011, but in May 2011 they stayed the ruling pending judicial review or internal reconsideration of the rules (EPA 2011a, 2011b).

Methanol, formaldehyde, acetaldehyde, acrolein, propionaldehyde, and phenol are listed as “total HAPs” for the PCWP NESHAP ruling (EPA 2006b). These compounds are released as a result of thermal degradation of wood (Otwell et al. 2000). As the MC decreases below 10 percent, the temperature of the lumber surface rises because of the decreased evaporative cooling, which increases thermal degradation (Su et al. 1999). Lignin has a higher potential for releasing formaldehyde than cellulose or hemicelluloses, while some extractives release formaldehyde and others act as scavengers (Schafer and Roffael 2000). Generally, higher temperatures result in greater amounts of methanol and formaldehyde being released (Milota and Mosher 2006). Methanol and formaldehyde emissions are at their highest concentrations during the final stages of drying (Otwell et al. 2000, McDonald et al. 2004).

Table 1 shows previous studies of the HAPs released from SP dried to 19 percent MC. The EPA lists acetaldehyde, acrolein, formaldehyde, and phenol emission values of 0.016, 0.0022, 0.0082, and 0.0024 kg/m³ of lumber dried, respectively (EPA 2006b). In contrast to the EPA, phenol is not released during the kiln drying of lumber according to Milota and Mosher (2008). Based on past research, a typical SP mill will reach 10 tons of methanol released before reaching 25 tons of total HAPs released.

In 2007 the EPA released method OTM-26: “Interim VOC Measurement Protocol for the Wood Products Industry” (EPA 2007a). The method requires VOC permitting for wood products facilities based on total VOCs reported as propane and individual measurements of methanol and formaldehyde. The method allows for exempt VOCs that have negligible photochemical importance to be subtracted from total VOCs; these include acetone,

methane, and ethane (EPA 2009). Terpenes make up the bulk of SP’s total VOC emissions, which are released via three mechanisms: release of terpenes dissolved in surface water, water mobilizing of terpenes from interior to surface via surfactants, and evaporation (Banerjee 2001).

The primary objective of this study was to determine the release of HAPs and VOCs when drying green SP lumber to 8 percent MC using three kiln schedules. To address this, charges of lumber (as required in the *Federal Register*) were dried, and the results are reported herein (EPA 2006b). Methanol and formaldehyde were collected, analyzed, and reported using the National Council for Air and Stream Improvement (NCASI) 98.01 method (NCASI 2010). Although acetaldehyde, acrolein, and propionaldehyde have less than optimal recovery rates with the 98.01 method, they were also collected and analyzed. The VOC emissions were reported according to the NCASI VOC and EPA OTM-26 methods (NCASI 1998, EPA 2007a). A secondary objective of the study was to find a gas chromatography (GC) method capable of separating the compounds of interest without the use of cryogenic cooling.

Materials and Methods

Drying procedure

Green rough-cut SP 51 by 152-mm lumber was obtained from a mill, transported to Mississippi State University, and stored in a cooler at 2°C until drying. To prepare the kiln material, approximately 305 mm of length was removed from every board’s end to eliminate end drying that occurred during storage and handling. Next, a 584-mm-long piece was cut from each lumber section and classified as relatively clear or relatively knotty material and randomly assigned to three kiln temperatures: an elevated 99°C schedule and two high-temperature, 116°C and 127°C, schedules. An approximately 25-mm-long section was cut to estimate initial MC of each board. Each kiln charge contained 15 pieces of lumber. Three runs of relatively clear material and one run of relatively knotty material were dried at each kiln temperature for a total of 12 runs.

The time to dry below 19 and 8 percent was recorded to compare the 19 percent results with previous studies and to determine the increase of emissions when drying to 8 percent MC. The kiln schedules followed that of a commercial lumber kiln and are shown in Table 2. The wet bulb depression was controlled with a mass flow controller via venting action with the quantity of air entering and thus leaving the kiln.

Table 2.—Kiln temperatures, drying times, and wood starting moisture content (MC).

Dry bulb temperature (°C)	Wet bulb temperature (°C)	<19% MC time (h)	<8% MC time (h)	Starting MC (%)
99	77	20	31	81
116	82	14.75	19.5	85
127	85	11	15	83

Air sampling for HAPs

A sample of the kiln air was collected according to the NCASI 98.01 method; the sampling train is shown in Figure 1. Three impingers were filled with approximately 5, 15, and 20 mL of deionized water, respectively. Before and after each run the system flow was measured at the probe tip using a bubble meter; leak checks were conducted prior to each sampling period. The impingers were chilled in an ice bath throughout each sample run. Sampling time was approximately every 2 to 3 hours.

The impinger water from each sampling period was poured into a preweighed polyethylene vial, reweighed, and stored on ice in a dark cooler. A 1-mL aliquot of the sample was transferred to an autosampler vial, capped, and stored in a refrigerator prior to GC analysis. A 2-mL aliquot of the sample was transferred to a sampling vial and stored in a refrigerator until the sample was analyzed for formaldehyde colorimetrically.

Several chemical recoveries were conducted using the sampling train and sampling procedure by two methods: a single injection into the probe tip with a subsequent 3 hours of sampling and injections every 15 minutes into the probe tip over 3 hours to simulate a kiln run. Recovery rates for the method must be ± 30 percent. The target concentration for the compounds was between 1.5 and 3 ppm, with the recovery of the compounds determined based on the mass in the sampling vial versus the expected mass of each compound. Water blanks were collected and analyzed for each kiln run, and kiln blanks with stickers were also conducted and analyzed at each kiln temperature.

Quantification of HAPs by GC

Standards of each compound were prepared by diluting reagent grade compounds with distilled water. The non-formaldehyde standards ranged from 1,000 to 0.5 ppm, with

the calibrating standards for calculation determined based on the range of concentrations found in the samples. Formaldehyde concentration ranged from 0.0 to 7.5 ppm.

The NCASI method recommends a 6 percent cyanopropylphenyl/94 percent dimethyl polysiloxane (DB-624) column connected to a GC equipped with cryogenic cooling. An alternate column was explored to eliminate the use of the cryogenic cooling, which should allow more laboratories to conduct analysis on HAPs released during kiln drying. As such, a Restek Q-bond, 100 percent divinyl benzene porous layer open tubular, 30-m, 0.53-mm inside diameter, 20.0- μ m film column was evaluated for use, and after careful testing the column met the needs of the analysis.

An Agilent 6890N GC with flame ionization detection (FID) and a 7683 autosampler was used for the analysis. A double gooseneck inlet liner was used, and splitless injection volumes were limited to 0.5 μ L to minimize water backflash. Column flow rate was 8.9 mL/min for 36 seconds, was decreased to 4.4 mL/min for 20 minutes, and then was increased to 8.8 mL/min until the column was clean. Operating temperatures for the injection port and FID were 220°C and 280°C, respectively. The oven temperature schedule was 5 minutes hold at 85°C, ramp 5°C/min to 220°C, and hold for 55 minutes or until the column was clean. Calibration curves were developed based on actual chemical concentrations. A sample chromatograph is shown in Figure 2. Water eluted at approximately 2 minutes.

Ethanol, acetone, and 2-propanol are also water soluble and were quantified during the GC analysis. Acetone has minimal photochemical reactivity and can be subtracted from the total VOCs for the OTM-26 method after adjusting to propane based on the response and compound size (EPA 2009). Formaldehyde was analyzed on a Varian Cary 100 Bio Ultraviolet-Visible spectrophotometer according to the NCASI 98.01 procedure (NCASI 2010). Following analysis, the total weight of each compound was determined by scaling up the amount of air collected in the impinger train with the total amount of kiln air exhaust.

Air sampling for total VOC

Emissions sampling was performed according to 40 CFR 60 Appendix A Method 25A, through the use of a direct read FID analyzer (EPA 2006a) and the NCASI “Standard Protocol for VOC Concentration Measurement Method for Use at Small-Scale Kilns” (NCASI 1998). A J.U.M. VE-7 instrument was used to determine the total hydrocarbons in

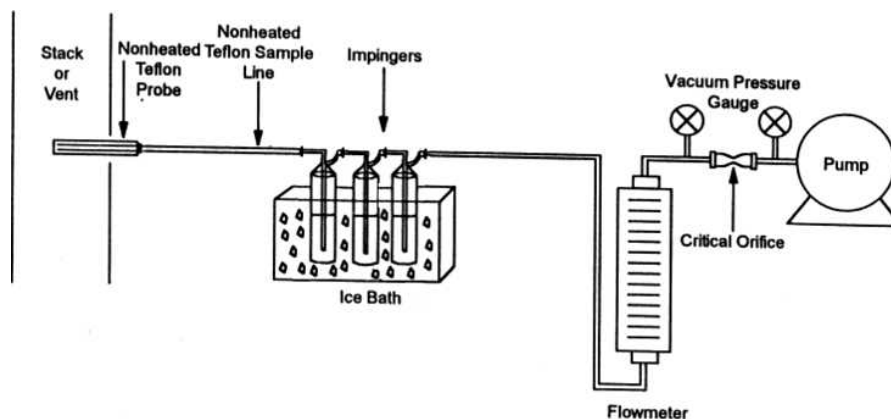


Figure 1.—Hazardous air pollutant sampling train setup (National Council for Air and Stream Improvement 2010).

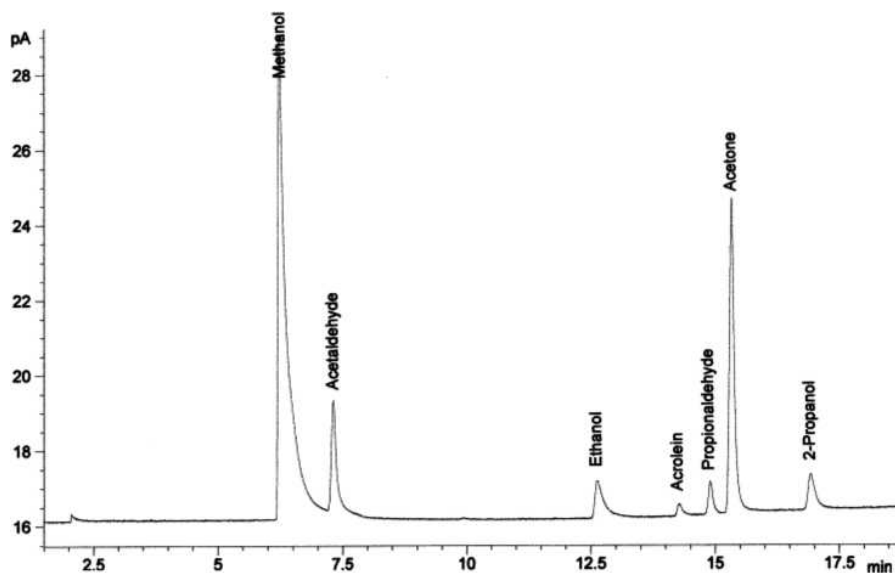


Figure 2.—Sample chromatograph.

the air exhaust of the kiln. The sample air was diluted with clean, dry compressed air heated to approximately 121°C to reduce the gas moisture content to below 20 percent.

Leak checks were performed at the beginning and end of each kiln charge. Calibration of the J.U.M. VE-7 was performed at approximately 3-hour intervals. Ultrahigh-purity zero air was used and middle and span calibration gases were approximately 1,000 and 3,000 ppm volume of propane in air, respectively. Each 3-hour time block was termed a sample run. Emissions data along with wood, moisture, and kiln information were assembled in order to calculate total VOC emissions. To this end, total VOC emissions as carbon per 30-second interval were first calculated according to the NCASI method (1998).

The total VOC data and the individual measurements of methanol, formaldehyde, and acetone were used to calculate the VOC emissions for the EPA OTM-26 method (EPA 2007a). Total VOCs for the OTM-26 method were adjusted from the NCASI VOC method based on the response rates and compound size compared with propane for methanol and acetone.

Results and Discussion

HAP analysis

The GC samples were analyzed immediately to minimize degradation. Chemical recovery depended on the procedure used to calculate recovery and is shown in Table 3.

Table 3.—Chemical recovery of compounds.

Chemical	3 h recovery (%)	Standard every 15 min for 3 h (%)
Methanol	109	107
Formaldehyde	85	95
Acetaldehyde	44	76
Acrolein	10	53
Propionaldehyde	41	75
Ethanol	95	123
Acetone	74	97
2-Propanol	90	103

Methanol and formaldehyde were within acceptable recovery ranges, but the other aldehyde's recoveries generally decreased as sampling time increased. The method of capturing acetaldehyde, propionaldehyde, and acrolein using strictly water in chilled impingers was not suitable; however, the compounds were still recovered in the sampling procedure. The 3-hour recovery for acetaldehyde, propionaldehyde, and acrolein was generally poor, especially for acrolein. However, during the recovery that simulated a kiln run by having injections every 15 minutes for 3 hours, the acetaldehyde, acrolein, and propionaldehyde recoveries improved greatly, although acrolein was still not within ± 30 percent.

The emissions of HAPs from the knotty charges were within 5 percent of the clear charges for each kiln schedule (data not shown). The effect of knots was shown to have had little to no effect on HAP emissions, and so the results for the knotty and clear charges were combined to eliminate redundancies during comparisons between temperatures. There was a small amount of acetone (<0.75 ppm in the impingers) found in the kiln blanks with stickers in the high-temperature schedules.

HAP analysis for final 19 percent MC lumber

The results for the HAPs and other volatiles released when drying SP from green to 19 percent MC with the combined clear and knotty charges grouped together are shown in Table 4. The methanol and formaldehyde emissions from the 116°C and 127°C schedules were significantly higher than the 99°C schedule ($P = 0.0195$, $P = 0.0034$). All three schedules had similar methanol emissions to the NCASI (2002) studies at 113°C. The 116°C and 127°C schedules had similar releases of formaldehyde, and all three schedules had lower levels of acetaldehyde and higher levels of acrolein compared with the NCASI (2002) 113°C and the Shmulsky and Dahlen (2008) study at 116°C and 127°C. All three studies had higher amounts of propionaldehyde than the NCASI (2002) 113°C study. The levels of formaldehyde and acetaldehyde were lower while levels of acrolein were higher compared

Table 4.—Emissions released during southern pine drying from green to 19 percent moisture content.

Chemical	Emission (kg of chemical/m ³) ^a		
	99°C	116°C	127°C
Methanol	0.049 B	0.061 A	0.056 A
Formaldehyde	0.0017 B	0.0034 A	0.0041 A
Acetaldehyde	0.0077	0.0091	0.0077
Acrolein	0.0024	0.0029	0.0029
Propionaldehyde	0.0014	0.0012	0.001
Ethanol	0.031	0.047	0.017
Acetone	0.03	0.03	0.027
2-Propanol	0.0034	0.0031	0.0031

^a 1 kg/m³ is approximately 4.16 lb/MBF. Statistical differences are indicated with different letters.

with the EPA (2006b) levels. Like McDonald et al. (2004), we found that ethanol emissions were greatest at the beginning of the run and declined thereafter.

Based on these results, a SP lumber facility would become a major source of HAPs from their methanol emissions. Provided the kiln schedule, operation, and raw materials were similar, a facility would reach 10 tons of methanol when drying more than 184,000, 149,000, and 155,000 m³ (97.6, 79.1, and 82.0 million board ft [MMBF]) per year for the 99°C, 116°C, and 127°C schedules, respectively.

HAP analysis for final 8 percent MC lumber

The results for the HAPs and other volatiles released when drying SP from green to 8 percent MC with the combined clear and knotty charges grouped together are shown in Table 5. The methanol emissions from the 116°C and 127°C schedules were significantly higher than the 99°C schedule ($P = 0.0032$). As temperature increased, the level of formaldehyde significantly increased ($P < 0.0001$). There was no significant difference with regard to acetaldehyde, while acrolein emissions at 127°C were significantly greater than at 99°C ($P = 0.007$). Propionaldehyde had the opposite trend of methanol where the 99°C schedule had significantly greater emissions than the 116°C and 127°C schedules ($P = 0.05$).

The release of HAPs greatly increased when lumber was dried from 19 to 8 percent MC; increasing by 44 percent overall. This observation was consistent with previous studies on oriented strand board flakes that showed that as

Table 5.—Emissions released during southern pine drying to 8 percent moisture content.

Chemical	Emission (kg of chemical/m ³) ^a		
	99°C (%)	116°C (%)	127°C (%)
Methanol	0.072 (46) B	0.085 (41) A	0.091 (56) A
Formaldehyde	0.0026 (52) C	0.005 (41) B	0.007 (72) A
Acetaldehyde	0.011 (45)	0.012 (33)	0.011 (40)
Acrolein	0.0031 (33) B	0.0036 (29) AB	0.0038 (39) A
Propionaldehyde	0.0022 (50) A	0.0017 (35) B	0.00014 (46) B
Ethanol	0.035 (12)	0.052 (12)	0.021 (24)
Acetone	0.041 (37)	0.037 (25)	0.036 (31)
2-Propanol	0.0046 (36)	0.0041 (27)	0.0041 (38)

^a 1 kg/m³ is approximately 4.16 lb/MBF. Percentages of increases from 19 to 8 percent are presented in parentheses. Statistical differences are indicated with different letters.

the MC of the wood decreases, evaporative cooling of the wood also decreases, which leads to high rates of thermal degradation and thus greater HAP emissions (Su et al. 1999, Otwell et al. 2000). Methanol and formaldehyde were the HAPs most affected by drying to 8 percent MC.

Based on these results, a SP facility would become a major source of HAPs from methanol emissions when drying more than 126,000, 106,000, and 124,000 m³ (66.7, 56.2, and 52.6 MMBF) per year for the 99°C, 116°C, and 127°C schedules, respectively, provided the kiln schedule, operation, and raw material were similar. It is likely that a mill will not dry all of their material to 8 percent, so individual mills will reach the methanol emissions at different volumes based on their drying mixtures.

VOC data

Table 6 lists the average VOC emissions as carbon for the different kiln charges for the NCASI VOC and the EPA OTM-26 method. Also included in Table 3 are data from previous studies that quantified SP VOC emissions from lumber. The relatively clear charges were similar to previous studies on SP lumber. There was no significant difference between the three kiln schedules for the relatively clear charges when drying below 19 or 8 percent MC. The knotty charges for the high-temperature schedules were similar to previous results for knotty southern pine lumber. The knotty charge for the 99°C schedule was lower than previous results and similar to clear lumber values; perhaps the lower kiln temperature factored into this difference.

Drying from 19 to 8 percent MC resulted in an average additional 0.26 kg of VOCs per m³ dried. The greatest VOC increase occurred with the relatively knotty material at 127°C, where an additional 0.45 kg/m³ of VOCs was released. The VOC results from the OTM-26 method are slightly higher than the NCASI VOC method because of more accurate methanol analysis.

Conclusions

The VOC, methanol, and formaldehyde emissions were similar to previous studies with regard to drying below 19 percent MC. The results with regard to HAP release from 19 to 8 percent MC were similar to previous studies that showed that as MC decreased HAPs tended to increase. We had some concerns with lumber degrade when drying from

Table 6.—Volatile organic compounds (VOCs) emitted during drying green southern pine (SP) lumber below 19 and 8 percent moisture content.

Charge	VOCs emitted (kg of VOC/m ³) ^a			
	NCASI		OTM-26	
	19%	8%	19%	8%
99°C relatively clear	0.9	1.09	0.92	1.13
116°C relatively clear	0.95	1.12	0.98	1.17
127°C relatively clear	0.91	1.17	0.95	1.23
SP sapwood (Ingram et al. 2000)	0.91			
SP (Milota 2006)	0.72–0.96			
99°C relatively knotty	0.83	1.06	0.85	1.09
116°C relatively knotty	1.37	1.66	1.4	1.69
127°C relatively knotty	1.36	1.81	1.38	1.85
SP knotty (Ingram et al. 2000)	1.31			

^a 1 kg/m³ is approximately 4.16 lb/MBF. NCASI = National Council for Air and Stream Improvement.

19 to 8 percent MC, but the lumber degrade was minimal for all three temperatures.

The aldehyde stability in water over a 3-hour period is low, particularly for acrolein, and thus the more recent NCASI methods recommend a 1-hour sampling time and use more sampling equipment/derivatization to further capture the aldehydes. Surprisingly, even with the poor aldehyde stability found with the water-only method, acrolein and propionaldehyde emissions were greater than previous studies using the newer methods. The higher levels of acrolein and propionaldehyde may have been the result of analyzing the samples directly following analysis. Perhaps with better recovery of acetaldehyde the results would have been more similar to previous studies.

In all three schedules, regardless of drying to 19 or 8 percent MC, a mill would reach 10 tons of methanol before reaching 25 tons from total HAPs. Currently the HAP ruling treats all chemicals equally with regard to emission limits; however, if the EPA ruling changed such that higher risk compounds are more highly regulated, then SP mills may need to focus more on acrolein emissions.

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