Variation of terpenes in sapwood and heartwood of loblolly pine: Impact on VOC emissions from drying lumber samples

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Abstract

A single loblolly pine tree approximately 45 years in age was harvested for use in this study. Increment core samples from throughout the length of the tree bole were analyzed for terpenes. Lumber samples from different locations within the tree were tested for volatile organic compound (VOC) emissions in a pilot-scale kiln. The concentration of terpenes was also determined in samples from lumber before and after drying. Analysis of terpenes in increment cores gave concentrations of terpenes in heartwood, inner sapwood, and outer sapwood of 1.1 (0.068), 0.29 (0.061) and 0.11 (0.11) percent on a dry weight basis, respectively (standard deviations are shown in parentheses). The measured VOC emissions from heartwood and sapwood lumber samples were 6.21 and 3.27 pounds per dry ton as carbon, respectively. Approximately 50 to 60 percent of total terpenes remained in the lumber samples after drying both heartwood and sapwood samples. The increased VOC emissions from drying lumber sawn from heartwood was due to the higher initial concentrations of terpenes in the heartwood tissue of the loblolly pine.

Due to the potential for volatile organic compound (VOC) emissions from dry kilns to enhance ozone formation and the Environmental Protection Agency (EPA) permitting requirements for old and new kilns, considerable research related to VOC emissions from drying lumber has been reported in recent literature. The monoterpene portion of oleoresin was identified as the major chemical component in VOC emissions from laboratory-scale kilns drying loblolly pine (*Pinus taeda*) lumber samples (Punsuvon 1994, Ingram et al. 2000b). The observation of α-pinene, camphene, β-pinene, myrcene, limonene, and 4-allylanisole as the major chemical components in VOC emissions was consistent with the composition of volatile terpenes reported by Drew and Pylant (1966). These compounds were identified as the major components in turpentine from loblolly pine chips with a measured laboratory yield of 1.0 gal/dry ton (or approximately 7 lb/dry ton).

Studies on the distribution of oleoresin components in the bole of radiata pine (*Pinus radiata*) showed that the highest concentrations occurred in the heartwood near the bottom of the tree and decreased toward the top of the stem (Kininmonth and Whitehouse 1991). The concentration of extractable material was higher in the heartwood than in the sapwood. The composition of the terpenes was very similar to that of southern pine with α-pinene and β-pinene occurring as the major components.

The total concentration of terpenes in sapwood, heartwood, and knot tissue from loblolly pine samples was found to be 0.31, 1.51, and 3.31 percent on an ovendry basis (Ingram et al. 2000a). The VOC emissions from samples of lumber that were sorted to contain a high occurrence of knots was 4.24 pounds per ton as carbon (lbC/ton) on an ovendry basis. Clear sapwood samples from the same tree gave emission values of 2.86 lbC/dry ton. In that study, there was a tendency for knotty lumber samples to give higher emissions of VOC than clear samples; however, the number of samples was too small for statistical comparison.

In studies of full-size kilns, Dallons et al. (1993) measured the amount of VOCs in samples of southern yellow pine lumber before and after drying and reported an emission factor of 2 lbC/thousand board feet (MBF) of lumber. Glass and Elam (1995) reported emission values of 1.7 and 1.4 lbC/MBF of

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lumber for steam-heated and direct-fired kilns, respectively. Results from the comparison of the emission factors for laboratory-scale kilns in different laboratories and full-scale kilns have also been reported (NCASI 2002).

Several other reports in the literature have utilized small-scale kilns to measure VOC emissions from drying lumber samples of other species. The amount of VOC emissions from drying Douglas-fir (*Pseudotsuga menziesii*) lumber in small-scale kilns and large-scale kilns was measured by Lavery and Milota (2000). VOC emissions from drying ponderosa pine (*Pinus ponderosa*) lumber in small-scale and large-scale kilns were also reported (Lavery and Milota 2001).

Results from studies of the effect of lumber dimension, drying schedule, and end-grain effects when drying southern pine in a small-scale kiln were reported by Shmulsky (2000a, 2000b, 2000c). In a recent publication, Fritz et al. (2004) used a pilot-scale and a full-scale kiln to measure VOC emission from drying lumber from a number of inland northwest species. That publication also contains a summary of current literature and a comparison of emission values from much of the previous research in the area of VOC emission from lumber drying. The primary objective of the research reported in this paper was to determine if the variation of terpenes in sapwood and heartwood affected the amount of VOC emissions from kiln-drying lumber. The results reported in this paper contain data from the drying of lumber sawn from a single tree, the distribution of terpenes within the tree, and the loss of terpenes during drying.

Experimental

A loblolly pine approximately 40 to 45 years in age was harvested from the John Starr Memorial Forest, which is located 16 km south of Starkville, Mississippi. The bole of the tree was transported to the Mississippi Forest Products Laboratory after increment cores were collected from positions along the bole that corresponded to tree heights of approximately 0.9 m, 3 m, 6 m, and 9 m. Based on the measured length, each increment core was divided into three sections, which corresponded to heartwood, inner sapwood, and outer sapwood portions of the tree bole at each height. The total terpenes in each increment core section was determined by extraction and gas chromatography (GC) analysis as described in a previous paper (Thompson et al. 2006).

The bole of the tree was divided into three 3-m-long sections and milled into 5.1-cm by 10.2-cm by 58.4-cm boards using a Wood Mizer band saw. Lumber samples were sorted into two sample subsets according to the occurrence of sapwood and heartwood, stacked, and stored under plastic wrap at approximately 4°C. The occurrence of heartwood was visually verified with a heartwood indicator (Koch 1972). The estimated occurrence of heartwood and sapwood resulted in sample sets that were approximately 70 to 90 percent representative of each tissue type. Twenty-five pieces of lumber per kiln charge (about 32 BF) were dried for 18 hours in a laboratory-scale kiln. The drying schedule was similar to a high temperature schedule with the vent open during the time required for the kiln to reach drying temperature (115°C). The laboratory-scale kiln, analytical instrumentation, and experimental procedures were previously described (Ingram et al. 2000b). For these experiments, the total VOC emissions were calculated as pounds of carbon per ovendry ton of wood.

The amount of extractable terpenes in samples of lumber from each kiln charge was determined before and after drying. Soxhlet extraction is a commonly used method for separating extractives from wood samples and was used for this analysis. Small sections were removed from the end of each board before drying and ground through a Wiley mill using a 4-mm screen. The extraction procedure consisted of placing 10 to 20 g of ground wood in a cellulose extraction thimble and covering with a plug of glass wool to prevent floating during extraction. The thimble, containing the sample, was then placed in a Soxhlet extraction apparatus, 20 mL of 1 mg/mL of 1,4-dichlorobenzene was added to monitor recovery efficiency, and the sample was extracted with 180 mL methylene chloride for 16 hours. The final volume of the sample extract was adjusted to 200 mL with additional methylene chloride, and a 0.9-mL aliquot was transferred to 2-mL auto-sampler vials for GC analysis. Prior to GC analysis, 0.10 mL of 1 mg/mL diphenylmethane in methylene chloride was added as an internal standard. The list of target compounds consisted of α-pinene, myrcene, β-pinene, camphene, limonene, and 4-allylanisole. Five different concentrations in the range of 20 to 500 μg/mL were used to establish the response factor for each compound. The percent terpene content was based on the sum of the amount of individual compounds.

To determine the moisture content, a portion of each green lumber sample was dried to constant weight at 105°C. The amount of terpenes obtained from the GC analysis of the sample extract was used to calculate the percent terpenes in each sample on a dry weight basis. After the lumber samples were dried, the same analysis was performed to determine the residual amount of terpenes in each kiln charge after drying.

Table 1. — Percent concentration of terpenes in increment cores from different locations within a single loblolly pine.

Height above ground	Heartwood	Inner sapwood	Outer sapwood
(m)		(%)	
0.9	1.12	0.376	0.277
3	1.23	0.237	0.0508
6	1.08	0.282	0.0567
9	1.09	0.257	0.0567
Average	1.1 (.068)[a]	0.29 (0.61)	0.11 (0.11)

[a]Values in parentheses are standard deviations.

Results

The occurrence and distribution of terpenes in different types of sample tissue was similar to previous observations (**Table 1**). There was not a statistical difference in average total terpenes along the bole of the tree based on the analysis of variance (ANOVA) with a 95 percent confidence interval (SAS 3.2 1999-2001). The average concentration of total terpenes for the four different locations along the bole of the tree in heartwood, inner sapwood, and outer sapwood was 1.1 (0.068), 0.29 (0.061), and 0.11 (0.11) percent, respectively. A comparison of the data using ANOVA software with 95 percent confidence interval indicated that these values are statistically different. Results from ANOVA with a 95 percent confidence interval indicated that there were not statistical differences in the concentration of terpenes in increment core samples relative to different tree heights.

These values are in agreement with results from the previous study where the average concentration of terpenes in the

Table 2. — The average amount of terpenes found in heartwood and sapwood before and after drying was used to estimate the contribution of terpenes to VOC emissions (number of laboratory kiln charges = 6).

	Average terpenes	Calculated emissions as carbon
	-------------------(%)----------------	(lbC/dry ton)
Green sapwood	0.235	1.96
Kiln-dry sapwood	0.125	1.96
Green heartwood	1.52	10.6
Kiln-dry heartwood	0.915	10.6

heartwood, inner sapwood, and outer sapwood was determined for 12 loblolly pines. The 12 trees and the tree used in this experiment were from the same geographical area and were all approximately 40 to 45 years old. The average concentrations of terpenes in the 12 trees for heartwood, inner sapwood, and outer sapwood samples were 2.3 (0.42), 0.77 (0.31), and 0.34 (0.16) percent, respectively (Thompson 2004, Thompson et al. 2006). The average concentrations of total terpenes in each of the three zones were slightly lower in this tree than the averages from the 12 trees in the previous study. Some of this difference is possibly due to the amount in heartwood and sapwood in the different sections of the increment core. More variation than would normally be attributed to the analytical methodology alone was observed with the analysis of samples from the 12 loblolly pines (Thompson et al. 2006). The same trend of increasing concentration of terpenes in heartwood sections of the increment cores was observed in both studies.

The differences in terpene content of the lumber samples were used to estimate the VOC emissions as terpenes for sapwood and heartwood samples. The concentrations of terpenes before and after drying for six kiln charges of sapwood lumber samples and six kiln charges of heartwood samples are shown in **Table 2**. The concentration of total terpenes in green sapwood (ovendry basis) by Soxhlet extraction was 0.24 (0.13) percent and the concentration in the outer sapwood of this same tree by sonication of increment core was 0.11 (0.11) percent. This difference may be due to the better extraction efficiency of the Soxhlet method, actual difference in terpene concentrations, lack of enough samples, or the normal variation encountered with these types of experiments. The percent total terpenes found in lumber samples after kiln-drying was 0.12 (0.11) percent (ovendry basis). The calculated loss of terpenes from lumber samples during drying corresponds to 1.96 lbC/ton of lumber on a dry weight basis.

The concentration of terpenes in heartwood lumber samples was 1.5 (0.93) percent before drying and 0.92 (0.51) percent after drying. This difference corresponds to a loss of terpenes of 10.6 lbC/dry ton. It is important to note that all of the terpenes were not lost during the drying of lumber samples. For sapwood, the amount of terpenes remaining in lumber samples after drying was 53 percent (47% lost) of the original concentration. For heartwood, the amount of terpenes remaining in lumber samples after drying was 60 percent (40% lost). The value of 1.96 lbC/dry ton is in the same range, or slightly lower, than previously measured values for unsorted lumber samples (Ingram et al. 2000a). Values in the range of 9 to 14 lbC/dry ton were previously observed with a limited number of heartwood samples.

VOC emissions were determined for drying duplicate charges of sapwood and heartwood samples from the bottom, middle, and top sections of the loblolly tree bole. There was not a statistically significant difference in VOC emissions among the same type samples from the top, middle, or bottom of the tree bole (**Table 3**). The average VOC emission from six samples of sapwood lumber was 3.3 lbC/dry ton and the average VOC emission from six samples of heartwood lumber was 6.2 lbC/dry ton. ANOVA indicated that the average VOC emissions for samples of sapwood lumber and heartwood lumber were statistically different.

This study was conducted on a single loblolly pine where lumber samples and core samples were collected from different locations within the tree for laboratory tests. The results from these tests suggest the higher concentrations of terpenes in heartwood will contribute to higher VOC emissions during kiln-drying. Additional experimental data are required before a detailed mathematical relation can be developed.

Summary

The results from these experiments indicate that the highest concentration of terpenes occurs in the heartwood tissue of loblolly pine. Not enough samples were analyzed to clearly define a concentration profile through the radius of the tree bole. Lumber samples sawn from the heartwood region of loblolly pine had higher levels of VOC emissions than did lumber samples sawn from the sapwood region and dried by the same procedure. It is likely that some sapwood was present in the lumber sawn from heartwood and equally likely that heartwood was present in the sapwood lumber samples. The mixture of heartwood and sapwood in samples may have affected the VOC emissions from each sample type.

Results from the analysis of increment core samples indicated that the concentration of terpenes was higher in tissue from the center of the tree than in the outside sapwood tissue. The concentration distribution was similar to that observed in previous experiments. There was no variation of terpenes up and down the bole of the tree relative to the samples from along the radius of the tree. There was not a significant statistical difference in VOC emissions from drying samples of lumber taken from different locations in the length of the tree.

Table 3. — Measured VOC emissions from drying heartwood and sapwood lumber samples sawn from different locations within a single loblolly pine.

Sample type	Emissions as carbon	Sample type	Emissions as carbon
	(lbC/dry ton)		(lbC/dry ton)
Bottom sapwood-A[a)]	3.22	Bottom heartwood-A	3.83
Bottom sapwood-B	3.09	Bottom heartwood-B	5.12
Middle sapwood-A	2.67	Middle heartwood-A	4.86
Middle sapwood-B	2.96	Middle heartwood-B	5.12
Top sapwood-A	3.96	Top heartwood-A	9.82
Top sapwood-B	3.72	Top heartwood-B	8.52
Average	3.3 (0.48)[b]	Average	6.2 (2.3)

[a]Capital letters A and B indicate kiln charges.
[b]Values in parentheses are standard deviations.

Too few samples may have been collected and analyzed to statistically observe such a difference. Only one tree was used in this study, consequently, the variation from tree to tree is not known.

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