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Volatile organic compounds emitted from hardwood drying as a function of processing parameters

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ABSTRACT: During the drying of wood, volatile organic compounds are emitted. These emissions contribute, in the presence of nitrogen oxides and sunlight, to the formation of ground level ozone and other harmful photo-oxidants. Emissions of volatile organic compounds from the drying of birch sawdust in a spouted bed were analyzed with a flame ionization detector and with a gas chromatograph-mass pectrometer. A D-optimal model of the emissions showed that the emissions increased exponentially with decreasing sawdust moisture content and that the final sawdust moisture content was influencing emissions about twice as much as the inlet drying medium temperature and the month of logging. At inlet temperatures of 140-170 °C, the emissions increased steeply when the moisture content of the sawdust reached 10%, whereas an inlet temperature of 200 °C caused a surge of thermal degradation products at 15% moisture content. The results of this study should help to reduce the emissions of volatile hydrocarbons during the drying of hardwood sawdust and wood chips.

Key words: *Sawdust, wood, volatile, hydrocarbon, organic, birch, model*

INTRODUCTION

Reduction of industrial emissions of volatile organic compounds (VOCs) is a part of a global strategy to reduce pollution due to tropospheric ozone. VOCs, in the presence of NO_x, cause the formation of ozone and other photochemical oxidants, which irritate the respiratory system (Burnett *et al.*, 1999; Krupnick *et al.*, 1990), cause distress for asthmatics (Levy *et al.*, 2001; Peden, 2002) and obstruct the photosynthesis of plants (Heath, 1980; Laurence and Weinstein, 1981). Hardwood is predominantly used for furniture, carpentry and floors. This requires drying to suitable moisture contents (Table 1) (Thomassen, 1998). Birch cutter shavings are used for heating, mostly as briquettes. In Sweden, the hardwood sawdust from large sawmills is currently used for heating in its undried state (Martinsson, 2003), whereas small sawmills often sell it as animal bedding. However, hardwood sawdust can be used for the production of pellets (Munter, 2004). Wood pellets are usually produced from sawdust with moisture content ranging from 6% to 15%. The Swedish wood pellets industry mostly uses conifer sawdust although some producers mix hardwood into it (Olsson, 2001). Hardwood has been mentioned as one of the several "new" raw materials for pellets to be used as the demand for wood pellets exceeds the amount of available softwood sawdust (Martinsson, 2003). High temperatures in wood cause thermal degradation of wood tissue. The wood temperatures necessary to cause thermal degradation have been reported to be 130 °C (Broege *et al.*, 1996), 200 °C (Bridgwater, *et al.*, 1995) and within the range of 130-160 °C (Otwell *et al.*, 2000). In hardwoods, thermal degradation products are the only VOCs emitted during drying (Banerjee, 2001). Examples of typical thermal degradation products are methanol, formaldehyde (Barry and Corneau, 2006; Su *et al.*, 1999), acetaldehyde (McDonald *et al.*, 1995), pentanal, hexanal (Otwell *et al.*, 2000) formic acid, acetic acid, alcohols and furfurals (Bridgwater *et al.*, 1995). There are studies published on thermic degradation emissons from lumber drying (McDonald *et al.*, 2002; Milota, 2006), rotary drum drying of fragmented wood (Banerjee *et al.*, 2006) and drying in a ceramic tube furnace (Otwell *et al.*, 2000; Su *et al.*, 1999). The results from these studies are not necessarily transferable to the drying of sawdust in spouted/fluidized beds. Different temperature ranges are used and a spouted

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bed is much gentler on the dried material than a drum dryer is, with lower risk for overdrying of fines - a factor that has a large influence on thermic degradation (Banerjee *et al.*, 2006; Su *et al.*, 1999). It is important to establish if VOC emissions from hardwood spouted bed drying are sufficiently large to warrant mandatory installment of equipment to reduce emissions. This would incure with additional costs. To estimate the amount of emissions correctly during various drying schemes, it is necessary to know how the emissions change with the used wood and with important drying parameters. A model of the VOC emissions' dependence on the inlet temperature, final sawdust moisture content and time of logging gives this predictability, which is important both to regulative bodies and to the wood industry.

MATERIALS AND METHODS

Birch sawdust was dried in a spouted bed dryer in continuous operation using superheated steam with maximum steam temperatures of 140 °C, 170 °C or 200 °C. The final moisture contents studied were 25% (the fiber saturation point, where the water in the lumen has been removed but not the water in the wood cell walls) and low sawdust moisture contents in the interval 4-9%. Sawdust with a moisture content of 4% is used by the particle board industry, whereas sawdust with a final moisture content of 9% can be used by the wood pellets industry. The sawdust used in this study was collected during summer and winter. The season can be of importance, since the extractive content differs over the year, with the nutrient reserve of fatty acids and fats stored in marrow streaks being at its peak in autumn and depleted in spring. The experimental design (Table 2) can be seen as a three-level full factorial design (equivalent to a CCF design in two factors) with replicate center-point for December sawdust, combined with a two-level fractional factorial design with one center-point for June sawdust. A D-optimal approach was used due to the irregular experimental region (4% MC at 140 °C was unattainable) and the mixed full fractional and fractional factorial design. The drying medium was analyzed for solvent soluble substances in order to determine whether, in case of birch wood, the emitted VOCs are entirely made up of thermal degradation products or whether biogenic substances are also emitted. This was done for runs with high inlet temperature and low sawdust final moisture content, where biogenic emissions would be most likely to occur. Furthermore, undried sawdust was analyzed for solvent soluble substances in order to compare these with the solvent soluble substances in the drying medium.

Table 1: Suitable moisture contents for different applications of hardwood in a temperate climate (Thomassen, 1998)

Furniture in houses with central heating	5-8%
Furniture in summer cottages	9-12%
Doors and floors in houses with central heating	6-8%
Inner doors and floors in summer cottages	10-13%
Outer doors and windows	12-15%
Outdoor furniture	14-18%
Storage without risk of fungal growth	<20%

Birch (*Betula pendula*) was used in the experiments. The sawdust was taken from a sawmill located in central Sweden that processes timber grown in the region. The sawdust was transported and stored in sacks of woven polypropylene at room temperature and was used for tests within a week. One batch was collected in December and one in June. The sawdust moisture content was 42% wet basis (72% dry basis) in December and 35% wet basis (54% dry basis) in June. Undried birch normally has 40% wet basis (% wb) moisture content.

The drying system used for the experiments is a laboratory scale spouted bed dryer located at the University of Karlstad (the KaU dryer). The system is schematically depicted in Fig. 1. A spouted bed resembles a fluidized bed; the difference being that in a fluidized bed the drying material moves randomly, whereas in a spouted bed the material moves upwards at the center of the drying tower and falls down at the periphery.

Table 2: Experimental design. Experiments were conducted at four final sawdust moisture contents (4, 6.5, 9 or 25% wet basis) at three temperatures (140, 170 or 200 °C) with sawdust logged in June (Jun.) or December (Dec.) and with a replicate center point for December sawdust

	4 % wt b		6.5 % wt b		9 % wt b		25 % wt b	
200 °C	Dec.	Jun.	Dec.		Dec.	Jun.	Dec.	Jun.
170 °C	Dec.		Dec. x 2	Jun.	Dec.		Dec.	
140 °C			Dec.	Jun.	Dec.	Jun.	Dec.	

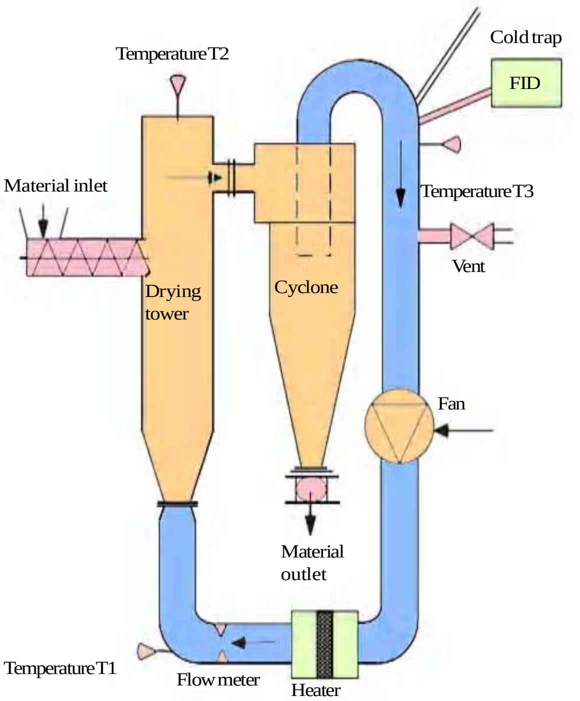

Fig. 1: The spouted bed superheated steam dryer (KaU dryer) used for the experiments described in this study

A spouted bed has high coefficients for heat and mass transfer and relatively high fluid speeds. Spouted/ fluidized bed is a common drying technique, used by many pellets producers. Recirculation of the drying medium is becoming increasingly more popular due to the need for energy conservation. The sawdust was continually fed into the top of the drying tower, where it met a stream of superheated steam (the drying medium) at atmospheric pressure. The material was dried as it circulated in the drying tower. After drying, it was transported with the flow of steam into the cyclone, where it was separated and fed out of the drying system by a rotary vane feeder. The drying medium was reheated and returned to the drying tower. Although the dryer continuously keeps sawdust in various stages of drying in the drying tower, the sawdust transported to the cyclone has small moisture content variation. Sawdust particles encounter the inlet temperature at the bottom of the drying tower. Overheating of fines is unlikely, as they are swiftly removed to the cyclone. The control system is designed to keep the steam flow, the inlet temperature T1 (before the drying tower) and the tower temperature T2 (after the drying tower) at set levels. There is an established correlation between the temperature after the drying tower in this spouted bed and the final moisture content of the dried sawdust (Berghel and Renstrom, 2004). The final sawdust moisture content is also dependent on the initial sawdust moisture content. To achieve a certain final moisture content, a material with high initial moisture content must be dried with T2 set at a higher temperature than is the case for a material with low initial moisture content (Persson and Berghel, 2005). The temperatures of the drying medium were 140 °C, 170 °C or 200 °C at the drying tower inlet, whereas the temperatures after the drying tower ranged from 110-130 °C for the sawdust produced in December and barely reached above 100 °C for the sawdust produced in June. Due to lower inlet moisture content, the June sawdust had 1.5-1.9 times higher than the December sawdust, regarding dry substance flow. The final sawdust moisture content was stable during drying runs and the values obtained were close to the target values.

The VOC emissions were continuously measured with a flame ionization detector (FID, JUM VE7). The gas sample was withdrawn via a short length of pipe after the drying tower. All lines were heated to prevent condensation. The FID was calibrated with a test gas of 900 ppm methane in synthetic air purchased from AGA. The concentration of hydrocarbons was measured as methane equivalents. The zero gas was nitrogen. All data presented on total hydrocarbon concentrations constitute mean values from drying runs of 30 min under steady state conditions. The concentration of hydrocarbons varied within ±5% of the median value. There are some problems with the use of FID: moisture in the sampled gas can suppress the FID response by 20% of the actual concentration for gas moisture contents between 40% and 60% (Milota and Lavery, 1999) and the response for methanol and formaldehyde is low (Otwell *et al.*, 2000). However, FID is a measurement method used extensively in the hydrocarbon emitting industry, accepted by regulative bodies, and adequate for detecting responses to various changes in the drying parameters. Solvent soluble emissions in the drying medium were captured by leading a drying medium sample stream to a dry ice trap (-78 °C), causing water and volatiles to form a solid. The captured volatiles were then dissolved in dichloromethane and analyzed using a GC-MS. The VOC emissions measured by FID were quantified by the method devised by Granström

(Granström, 2005; Granström, 2003). Undried sawdust was extracted with acetone in a Soxhlet® extractor for 8 h The acetone extract was diluted with water, followed by a dual extraction with isooctane. The isooctane was analyzed using a GC-MS. This method is described in more detail elsewhere (Ståhl *et al.*, 2004). The extracted substances were quantified as octane. The GC (Perkin Elmer AutoSystem) was fitted using an unpolar capillary column (J&W Scientific, DB5-MS, 30 m x 0.25 mm). The temperature program was set at 45 °C for 4 min., 10 °C/min., 80 °C for 8 min, 15 °C/min, 250 °C for 0-15 min The MS (Perkin-Elmer Q mass 910) had a m/e of 20-300. The sawdust's dry mass (DM) was determined as the weight after drying in air at 103 ± 2 °C until constant weight was obtained in accordance with the standard SS 187170 (SIS, 1997). The moisture content of the sawdust was calculated as wet basis.

RESULTS AND DISCUSSION

The emissions from birch sawdust steeply increased with decreasing moisture content at low moisture contents. A steep increase in emissions was seen at 10% sawdust moisture content for inlet temperatures of 140-170 °C, whereas the thermal degradation increased at about 15% of moisture content when the inlet temperature was 200 °C (Fig. 2). The inlet temperature had a considerable effect when the sawdust moisture content was below the fiber saturation point with emissions increasing steeply from 170 °C to 200 °C. For sawdust that was still evaporatively cooled,

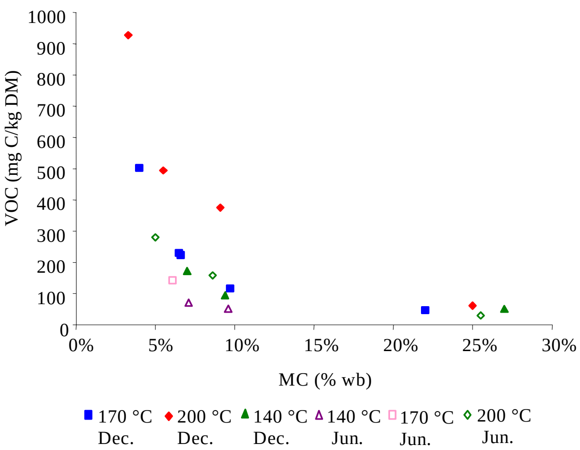

Fig. 2: VOC emitted from birch sawdust during drying (mg C per kg dry matter) at different final sawdust moisture contents (MC, as % wet basis) and inlet temperatures of 140, 170 or 200 °C, using sawdust logged in June or December

the inlet temperature had no effect. The emissions were by far larger for wood sawn in December than in June. If drying is terminated at 10% wood moisture content, the emissions would be approximately 100 mgC/kg DM at inlet temperatures of 140-170 °C and 400 mgC/kg DM at an inlet temperature of 200 °C. At most, about 1g C/kg DM was emitted-this at a final moisture content, this was more relevant for the production of particle board than for wood pellets. The VOC response, as a function of inlet temperature (Temp), final moisture content (MC) and month (M), was modeled with a quadratic D-optimal model fitted with multiple linear regression (MLR). The model showed excellent correlation with logarithmic VOC emissions (R^2=0.988; Q^2=0.973; R^2 adj=0.982). The importance of the variables modeled, regarding the VOC emissions, is seen in Fig. 3. All the examined factors were significant. The final sawdust moisture content had the greatest effect on the VOC emissions as measured by FID. The month had a slightly greater effect than the inlet drying medium temperature. There is interaction between temperature and moisture content. The interaction terms Temp×M and MC×M proved to be insignificant and were thus removed from the model. The emission response equations, expressed in terms of actual factors, are as follows for December and June, respectively:

$$\log VOC = 3{,}052 - (0{,}0113)\,Temp - (0{,}0661)\,MC + (0{,}152)\,M - (0{,}000285)\,Temp \times MC + (0{,}0000603)\,Temp^2 + (0{,}00242)\,MC^2$$

$$\log VOC = 3{,}052 - (0{,}0113)\,Temp - (0{,}0661)\,MC - (0{,}152)\,M - (0{,}000285)\,Temp \times MC + (0{,}0000603)\,Temp^2 + (0{,}00242)\,MC^2$$

where, *temp* equals the inlet drying medium temperature, *MC* equals the final sawdust moisture content, M equals the month in which the tree was felled and sawn. The relationship between the VOC emissions and the three factors is illustrated in Fig. 4. The increase of emissions at low moisture contents and high inlet temperatures is confirmed. The same amount of emissions found at 10% MC and 140 °C is found at 16% MC and 200 °C. Undried sawdust was analyzed for solvent soluble substances and the results were compared with the substances in the drying medium. Undried birch contains oxygenated hydrocarbons with low retention times (Fig. 5a).

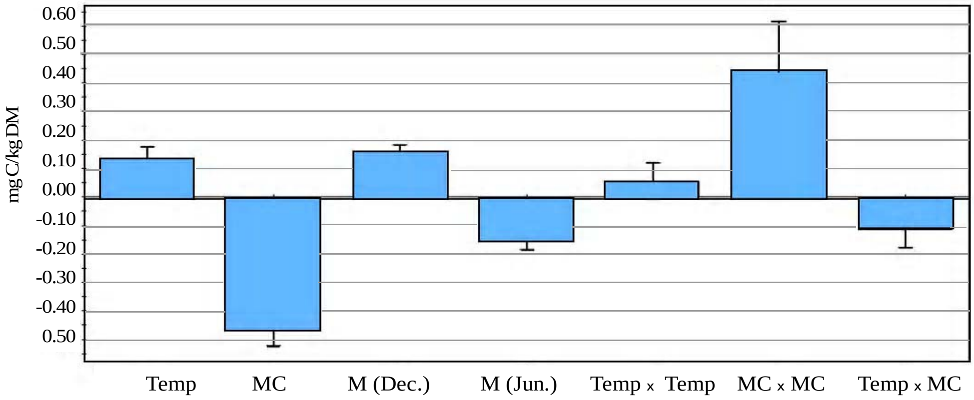

Fig. 3: Scaled and centered coefficients with confidence intervals (p 0.05). Scaled and centered factors are reduced in range to a common scale, regardless of their magnitude.
Temp=inlet drying medium temperature, MC=final sawdust moisture content, M=month of logging

These low-boiling biogenic substances in birch are not found in the drying medium (Fig. 5b), that is, they are not emitted during drying at the applied conditions but remain in the sawdust throughout the drying process without contributing to the VOC emissions. Pentanal and hexanal were notably absent from the VOC emissions. The peak shown in Fig. 5b with a retention time of 5.0 has a mass spectrum and a retention time consistent with butylacetate. The content of low-boiling oxygenated hydrocarbons is almost double in undried birch in December compared with the same in June (Table 3).

There is a low level of VOC emissions at high sawdust moisture contents. A wood temperature of 100 °C is thus sufficient for thermal degradation although the process is more pronounced at higher temperatures. During medium and high temperature drying, the temperature in the drying wood increases to the boiling point of water at the applied pressure (Johansson *et al.*, 1997) and it remains there until the wood is almost dry (<10% moisture content) due to the cooling effect of the evaporating water. Subsequently, the evaporative cooling decreases and the wood temperature increases to match the drying medium temperature. In these experiments, the increase of the wood temperature to approach the temperature of the drying medium (observed as increased thermal degradation) happened at 10% moisture content for inlet temperatures of 140 and 170 °C and at 15% moisture content when the inlet temperature was 200 °C. Thus, the partially wet surface of the sawdust at

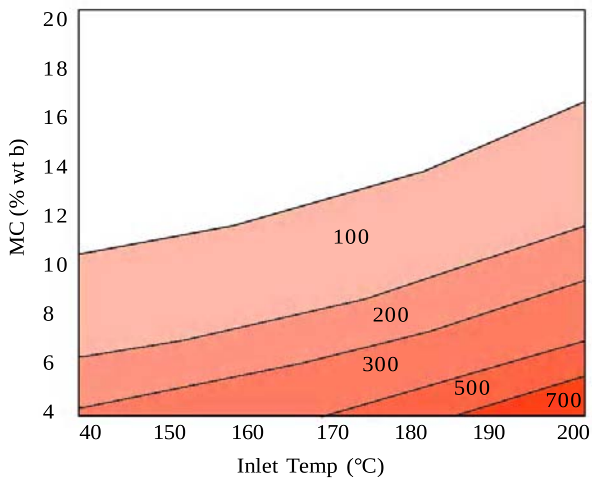

Fig. 4. Modeled VOC emissions from birch sawdust during drying, as a function of final sawdust moisture content (MC, as % wet basis) and inlet temperature (as °C). The emission values shown are given in mgC/kg odw dry matter and represent December sawdust

15% moisture content prevents major thermal degradation at inlet temperatures as high as 170 °C, but is insufficient when the drying medium is 200 °C. The relationship between emissions and inlet temperatures in this study is different from the linear relationship found for a rotary dryer using the inlet temperature interval 440-600 °C, that is, temperatures sufficient to produce charring (Banerjee *et al.*, 2006). The temperature necessary to cause considerable amounts of thermal degradation products depended on the sawdust moisture content. Wood at a later stage

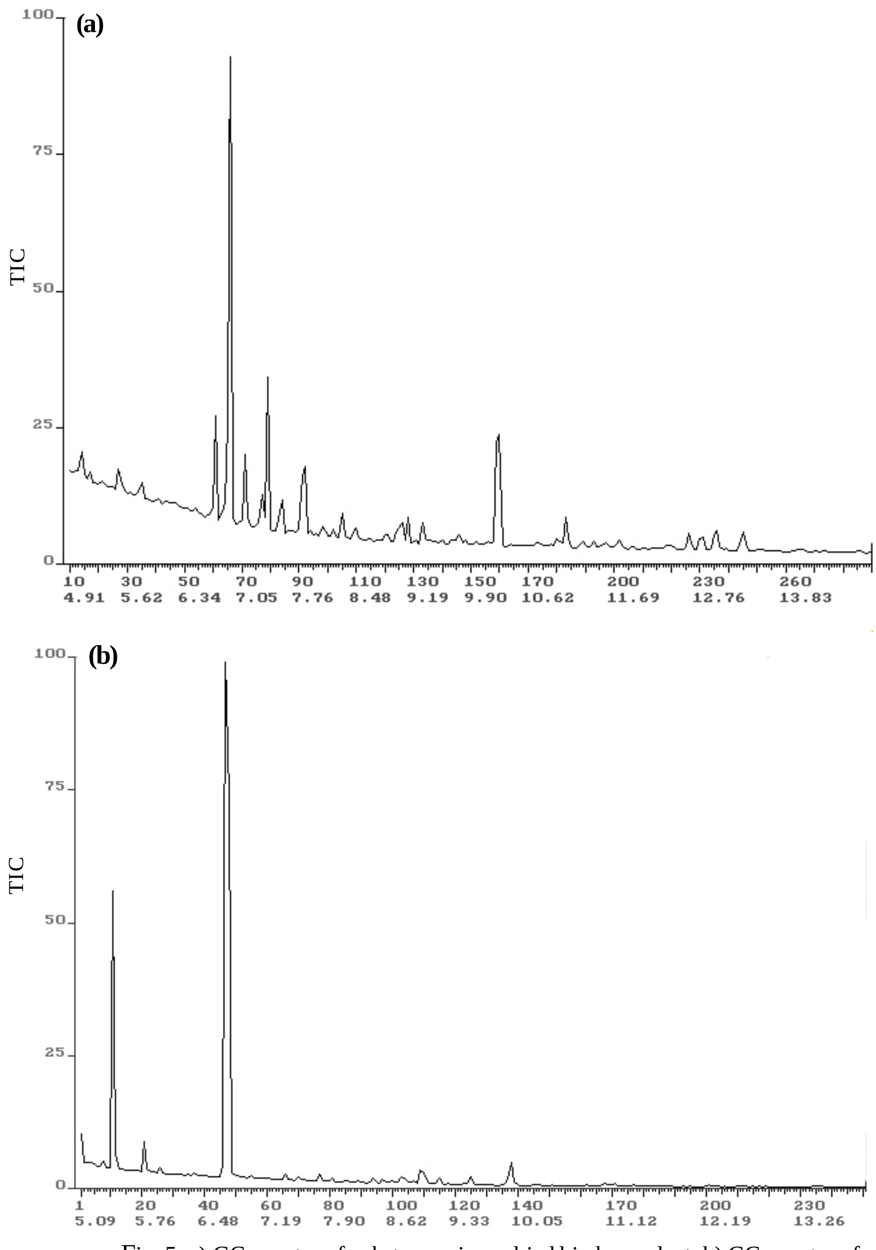

Fig. 5: a) GC-spectra of substances in undried birch sawdust. b) GC-spectra of substances emitted from birch sawdust during drying

Table 3: Compounds extracted with acetone from undried birch, logged in Dec. or Jun., calculated as mg C_8H_{18} per kg odw**

Ret.*	4.8	6.7	6.9	7.1	7.3	7.6	7.8	8.2	9.1	9.3	10.2	11.8	13.3	Sum
Dec	4.6	0.63	3.5	0.46	0.97	0.24	0.70	0.22	0.22	0.09	1.2	0.23	0.18	13
Jun	2.3	0.21	1.8	0.41	0.67	0.13	0.82	0.09	0.04	0.00	0.54	0.00	0.21	7

*Ret.=GC retention time
**odw: On dry weight

of drying, when the wood moisture content is low, is the most sensitive to the temperature of the surrounding medium. The effect of temperature on the VOC emissions is in agreement with the results from a study (Otwell *et al.*) in which the amount of wood degradation products was reported small at 130 and 160 °C and large at 200 °C. As for the effect of sawdust moisture content, the surge in wood degradation products occurred at lower moisture contents in that study. It is important to determine the moisture content at which the temperature in sawdust rises so that thermal degradation products form, as the drying should be stopped before the emission peak. This is to avoid unnecessary emissions to the environment. Sawdust was intentionally taken from the same mill and region and chosen to represent wood harvested during summer and winter. The VOC emissions were greater for wood logged in December than in June. This is probably due to a shorter drying time for the June sawdust. The lower initial moisture content in the June sawdust resulted in a shorter drying time and a lower temperature in the drying tower (the reasons for this is described by Persson and Berghel (Persson and Berghel, 2005). The lower drying tower temperature should not be considered as the cause of the difference in emissions between the December and June sawdust, as these temperatures are much lower than the inlet temperature, which would therefore be more important. Also, the December sawdust had higher emissions even at 25% MC, where evaporative cooling prevails. While in softwoods a low initial moisture content would imply a loss of biogenic VOC due to some previous drying, this is not the case for hardwoods where the emitted VOC is thermal degradation products. The wood contained more low-boiling extractives in December than in June (which would be consistent with the pattern for a nutrient reserve), but as these substances were not released, they did not influence the VOC emissions. Based on these two samples only, it cannot be concluded that thermal degradation emissions are greater for wood logged in the winter although it is likely that the initial sawdust moisture content, manifested in the required drying time, is of importance. The specific compounds emitted would conceivably be of interest in the US due to the MACT rule (regulating emissions of methanol, formaldehyde, acetaldehyde, propionaldehyde, phenol and acrolein), but this study focuses on total VOC, since limits on the VOC emissions are the primary public health concern in most parts of the world. It can be noted, however, that there was no detectable pentanal or hexanal in the drying medium. These compounds have been found when wood is dried in a ceramic tube (Otwell *et al.*, 2000; Su *et al.*, 1999), but not in emissions from industrial drying (Otwell *et al.*, 2000). The absence of these aldehydes in the field was suggested to be due to decomposition caused by the steeper temperature and moisture gradients in industrial dryers. However, pentanal and hexanal are absent in this study although the drying process is relatively benign to the wood. Their absence could be due to the use of fresh sawdust, assuming that previous studies have found pentanal and hexanal formed by oxidation of biogenic fatty acids. Such oxidation is known to happen in damaged wood that is exposed to light (Arshadi and Gref, 2005; Manninen *et al.*, 2002).

In conclusion, The amounts of the VOC emitted during drying of birch in a spouted bed are strongly related to the final sawdust moisture content. The effect varies with the inlet temperature. An inlet temperature of 140–170 °C makes it possible to dry to a moisture content of 10% before a great increase in thermal degradation products occur, whereas an inlet temperature of 200 °C will cause a surge of thermal degradation products at a moisture content of 15%. For the sawdust which is dried only to the fiber saturation point, the inlet temperature has no effect. The month of logging may have an effect on the VOC emissions, although the larger emissions from the December sawdust, compared with the June sawdust in this study, can also be attributed to different inlet sawdust moisture contents. Birch contains low-boiling oxygenated hydrocarbons that are not emitted during drying at the applied conditions. They remain in the sawdust throughout the drying process. There are almost twice as much of these low-boiling oxygenated hydrocarbons in undried birch in December as is the case in June. A D-optimal model of the VOC emissions showed that all the factors examined were significant and that some of the factors interacted. The emissions increased exponentially, with the final sawdust moisture content having about twice the effect on emissions as had the inlet drying medium temperature and the month of logging.

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