Wood processing as a source of terpene emissions compared to natural sources

K. M. Granström
Department of Energy, Environmental and Building Technology, Karlstad University, Sweden

Abstract

The aim of this paper is to analyse the importance of terpene emissions from forestry and the wood industry and relate these anthropogenic emissions to the natural terpene emissions from undisturbed forests. Biogenic volatile organic compounds are emitted naturally from trees and other plants. Boreal forests are dominated by terpene-emitting tree species. Anthropogenic terpene emissions occur due to drying, machining and tooling of wood. This is a cause of concern with regulatory authorities faced with expanding production of biofuels, e.g. wood pellets. Anthropogenic terpene emissions are of current importance as the use of biofuels has increased considerably over the recent years. The most important environmental effects of terpene emissions are due to the formation of photo-oxidants and atmospheric aerosols. Photo-oxidants cause forest and crop damage, and are harmful to humans. Carbonaceous aerosols have an impact on climate change. Furthermore, most sesquiterpenes are used in the chemical communication systems of various insects. There are also issues concerning the working environment in the wood industry. Environmental effects of terpene emissions are analysed with focus on three key aspects: the total amount of emissions, the concentration in air and the types of terpenes emitted. The reported concentrations of monoterpenes are in most cases below the occupational exposure limits, with drum barking and pellets production from non-dried sawdust close to the limit. Anthropogenically emitted terpenes have a higher proportion of sesquiterpenes than naturally emitted terpenes have, especially when the processing of wood involves elevated temperatures.
Keywords: terpenes, VOC, spruce, pine, sawmill, drying, wood, biofuel, pellets, sawdust.

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1 Introduction

There is a growing concern about terpene emissions from the wood industry. Terpene emissions contribute to several different environmental problems, and have an impact on work environment. Anthropogenic emissions are studied in order to estimate effects on local atmospheric chemistry, assess the suitability of different process procedures, and see where efforts should be focused in order to decrease emissions. They should be seen in relation to the natural emissions where environmental effects are concerned. I focus on the situation in the northern hemisphere, the European boreal region, where the main species are Scots pine, Norway spruce and Silver birch. Biofuel in this region is almost exclusively made from spruce and pine.

1.1 Terpenes

Terpenes are primarily found in resin the heartwood and sapwood of softwoods (conifers), and in the sapwood of hardwoods (deciduous trees). Terpenes are also stored in special cells in the needles of conifers and leaves of deciduous trees. In softwoods, the terpene group consists mostly of monoterpenes ($C_{10}H_{16}$), sesquiterpenes ($C_{15}H_{24}$) and diterpenes ($C_{20}H_{32}$), whereas triterpenes and sterols dominate in hardwoods. Terpenes have several protective functions in plants. They neutralize ozone that would disturb photosynthesis. They repel many insects and herbivores (terpenes attract both beetles and their predators). They also lower the viscosity of resin, making it possible for resin to flow to a damaged part of the plant. There the volatile mono- and sesquiterpenes are emitted to air and the non-volatile diterpenoic resin acids are left as a hydrophobic cover, protecting the tree from further damage.

1.2 Environmental issues

Terpenes contribute in the presence of nitrogen oxides and light to the formation of ground level ozone, radicals, aldehydes, peroxides and other potentially harmful photo-oxidants. Ozone and other photo-oxidants cause forest and crop damage, and are harmful to humans as they cause irritation in the respiratory tract and in sensitive parts of the lungs. The impact of VOC on ozone chemistry is frequently expressed in terms of VOC/NO_x ratios, but the true impact of VOC emissions depends not so much on the total amount of VOC but on the reactivity of the VOC species with respect to OH Sillman [1]. Terpenes are especially reactive and have a large impact relative to their ambient concentration

Terpenes contribute to the forming of aerosols, which contribute to cloud formation (Tunved *et al* [2]), which have an effect on global warming.

1.3 Work environment issues

Exposure limits are set by authorities for monoterpenes and turpentine (a mixture of predominantly monoterpenes, used as solvent). Some examples are given in table 1.

Table 1: Occupational exposure limits for terpenes and turpentine SWEA [3], HSE [4]. TWA=Time Weighted Average.

Country (Body)	Substance	Concentration (ppm)	Concentration (mg/m^3)
UK (HSC)	Turpentine	100 ppm TWA 8 hr 150 ppm TWA 15 min	570 mg/m^3 TWA 8 hr 850 mg/m^3 TWA 15 min
Sweden (SWEA)	Turpentine, monoterpenes	25 ppm TWA 8 hr 50 ppm TWA 15 min	150 mg/m^3 TWA 8 hr 300 mg/m^3 TWA 15 min
USA (OSHA)	Turpentine, α-pinene, β-pinene, 3-carene	100 ppm TWA 8 hr	570 mg/m^3 TWA 8 hr
USA (ACGIH)		20 ppm TWA 8 hr	120 mg/m^3 TWA 8 hr
Norway	Turpentine, 3-carene	25 ppm	150 mg/m^3

2 Anthropogenic terpene emissions

Each step in the processing of trees into products is an opportunity for terpenes to be released. The total emitted amount from a factory's production is the factor most often regulated in permission by authorities to start or increase production. It is therefore of major importance to both regulatory bodies and industry.

2.1 Thinning and logging

The monoterpene concentration in air near the harvester during logging of both Scots Pine and Norway spruce are 1.0–1.5 mg/m^3 (Strömvall and Petersson [5]). Four weeks after thinning, the concentration in air is 50 μg/m^3. This should be compared to the background level of 1–10 μg/m^3.

2.2 Storage

Norway Spruce wood chips in heaps outdoors in a temperature of 12°C emit at most 64 mg terpene/m^3 (Axelsson *et al* [6]). In wood chips the terpene content decrease rapidly, with 4% loss in the first week (Marutzky [7]). Storage of roundwood unsurprisingly causes less terpene emissions than storage of chips. Turpentine yield from Norway spruce and Scot pine has been reported as 23% lower if pulpwood is stored as chips instead of as roundwood (Strömvall and Petersson [26]).

2.3 Barking and sawing

The concentration of monoterpenes in air during single-log barking of Norway Spruce and Scots pine timber are 5–20 mg/m^3, and during drum barking of pulpwood 50–100 mg/m^3 (Strömvall and Petersson [8]). Measurements in Swedish sawmills have shown terpene concentrations of 50–550 mg/m^3 in air (Lundberg [9]). The higher concentrations were found in older studies before working conditions had received much attention.

Drum barking of Norway spruce cause emissions of 53 g/m^3 produced chips; the average emission for terpenes during sawing of Scots pine was 153 g/m^3 board and for Norway spruce 25 g/m^3 board (Svedberg and Paulsson [10]). With these data, the terpene emissions from a model medium sized sawmill, producing 100 000 m^3 board annually, would be for spruce 1.4 tonnes from barking and 2.5 tonnes from sawing, and for pine 8.3 tonnes from barking and 15.3 tonnes from sawing.

2.4 Drying

Spruce and pine meant for building purposes are generally dried to a moisture content of 18% (dry basis), and for furniture 10–15%.

Norway spruce has a low resin content, as resin is produced by the tree when needed, and thus low terpene emissions when processed. Scots pine produces a constant supply of resin and thus higher terpene emissions. Judging from lumber drying data, the terpene amounts in Scots pine are similar to that of Douglas-fir and Radiata pine.

Table 2: Emissions during drying of boards.

Tree	**Max temp (°C)**	**Monoterpenes (mg/kg odw)**	**Ref**
Douglas-fir	commercial dryer	315	Lavery and Milota [11]
Norway spruce	60–66	<50	Broege *et al* [12]
Ponderosa pine	commercial dryer	1590	Lavery and Milota [13]
Radiata pine	120–140	120–200	McDonald and Wastney [14]
Scots pine	65	210–380	Broege *et al* [12]

Using emission data in table 2, a sawmill drying 100 000 m^3 boards annually in a low temperature dryer would have emissions from the dryer of at most 14 tonnes monoterpenes if the boards are Scots pine, but only 2 tonnes if the boards are Norway spruce.

Fragmented wood is often dried in higher temperatures than boards are. When flue gas is used as drying medium, terpenes will mix with combustion gases, e.g., NO_x. Emissions during commercial drying of sawdust have been measured in exhaust air to 400-800 mg/kg odw (based on Nyrén [15], Münter *et al* [16], Ek *et al* [17]). Using these emission data, a medium sized pellets producer drying sawdust to produce 50 000 tonnes pellets per year, would have emissions from the dryer of 20–40 tonnes terpenes annually. This estimation from emission data is lower than calculations from terpene content in wood before and after drying. Commercially sawn sawdust tend to have a monoterpene content of about 0.5–4 g/kg odw in its undried state, of which 50–80% are emitted during drying (Ståhl *et al* [18], Marutzky [19]). Using sawdust with 2 g/kg odw would cause about 50-75 tonnes terpenes to be emitted from the sawdust.

The choice of end moisture content and drying technique heavily influence the terpene emissions from the dryer (Ståhl *et al* [18], Granström [20]). Part of the drying gas in Nyrén [15] is recirculated to the burner where terpenes are destroyed.

2.5 Pelleting

Cutter shavings and sawdust of a moisture content of 8–12% (dry basis) are preferred materials for production of wood pellets.

Dry sawdust contain 0.3–1.1 mg/kg odw terpenes, and during the production of pellets, 70–95% is released (Ståhl *et al* [18]). Using these data, pellets production of 50 000 tonnes would emit 10–50 tonnes terpenes, depending on whether the sawdust terpene content are in the lower or higher range.

Peak terpene concentration in the air in two pellet plants using dried sawdust has been measured to 23 mg/m^3 and 15 mg/m^3, respectively, and in one pellet plant using non-dried sawdust to 119 mg/m^3 (Davila [21]). Data of workers personal exposure to monoterpenes range from 0.64 to 28 mg/m^3 (Edman *et al* [22]).

3 Total amount

There are large natural emissions of biogenic volatile hydrocarbons from trees. Boreal forests are dominated by terpene-emitting tree species. Estimates of global natural monoterpene emissions, done after 1990, range from 127 to 147 TgC/yr (Kesselmeier and Staudt [23]). Monoterpene emissions from forests in Europe have been estimated to 3.8 Tg/yr (Simpson *et al* [24]). Emissions from forests in Sweden is about 0.4 Tg/yr (Janson [25]), which can be compared to the anthropogenic emissions from forestry of 0.1 Tg/yr and forest industry of 0.05 Tg/yr (Strömvall and Petersson [26]).

4 Types of terpenes emitted

The terpene amount and composition in trees are different in bark, sapwood, hardwood, and needles or leaves.

4.1 Anthropogenic emissions

Monoterpene emissions from the industrial processing of wood reflect the composition in the wood. The terpene content in Norway spruce wood is dominated by α-pinene, β-pinene and 3-carene (often in an approximate relation of 3:2:1). The wood has little sesquiterpenes, about 15 mg/kg odw, with longifolene the dominating compound. Bark from Norway spruce has a much higher percentage of myrcene and β-phellandrene (Strömvall and Petersson [8]) than the wood, and about twenty times more sesquiterpenes, mostly β-caryophyllene but also longifolene, α-humulene, α-farnesene, β-farnesene, longipinene, α-cedrene and α-bisabolene (Martin *et al* [27]). In Scots pine wood, α-pinene and 3-carene dominate, followed by β-pinene. Sesquiterpenes in Scots pine wood are mostly muurolenes, with minor amounts of copaene, β-copaene and β-ylangene (Westfelt [28]).

About 20% of the terpenes released during drying of Norway spruce sawdust are sesquiterpenes (Granström [29]).

4.2 Natural emissions

Monoterpene emissions from trees reflect the composition in the needles or leaves. The relative amounts of the monoterpenes in natural emissions from Norway spruce and Scots pine vary widely, though the monoterpenes emissions are usually dominated by α-pinene, β-pinene and 3-carene.

Monoterpenoids and sesquiterpenoids have been identified as major defensive emissions from Norway Spruce foliage, dominated by the oxygenated monoterpene (2)-linalool and the sesquiterpenes α-bisabolene and β-farnesene. Based on precedents in other species, the induced emission of terpenes from Norway spruce foliage may have ecological and physiological significance (Martin *et al* [30]).

5 Discussion

5.1 Work environment

Measurements of terpenes in air regarding workers exposure have focused on monoterpenes. Sesquiterpenes should be measured and added to the total exposure load. This sum of monoterpenes and sesquiterpenes can then be compared to the exposure limit for turpentine. This would be especially important for studies of emissions from bark.

5.2 Emissions from production

Storage of wood chips means considerable evaporation of terpenes. Storage would be a way to lower the terpene content in wood that is to be further processed. However, undried wood chips are susceptible to mould and insect attacks. Storage of dry chips would not have this disadvantage, but there would be emissions during the drying stage.

Estimated emitted amount for barking, sawing and drying of Norway spruce and Scots pine for a model sawmill is shown in table 3. The lower range is for Norway spruce, the higher for Scots pine. The amount actually emitted from sawdust dryers are similar to the amounts emitted during pellets production, although emissions during drying could be considerably larger as most of the terpenes in undried wood are emitted during the drying process.

Table 3: Monoterpene emissions related to a model sawmill producing 100 000 m^3 board.

Process	Tonnes
Barking	1–8
Sawing	2–15
Lumber drying	2–14

Emissions at the pellets press depend on the amount of terpenes in the sawdust. The variation in terpene content in sawdust used industrially is large, which is important for pellets producers. If a choice is made to reduce emissions at a dryer by terminating drying at higher sawdust moisture content, this high moisture sawdust will increase emissions as the press. It is possible to clean emissions from dryers, for example by returning a part of the exhaust to a boiler. In US, natural gas is used to burn of terpenes from dryer exhaust, but that cause increased NO_x emissions and costs. Pellets presses have less defined exhausts.

5.3 Concentration in air

The concentration of monoterpenes in emission plumes caused by anthropogenic activities is typically 10–1000 times higher than the background level in conifer forests. Comparing this disturbance to herbivory, where 10% damaged foliage will emit about 2 times more monoterpenes, which is sufficient to increase local tropospheric ozone production and suppress local hydroxyl radical concentrations (Litvak *et al* [31]), it is clear that anthropogenic emissions are larger than the natural variation and will alter regional atmospheric chemistry for several weeks.

5.4 Types of terpenes emitted

Sesquiterpenes that are especially active as defensive terpenes are present in Norway spruce bark. Considering that emissions from drying of wood sawdust had a higher proportion of sesquiterpenes than are present in stem wood, drying of bark would likely cause biologically significant emissions of defensive terpenes.

6 Conclusions

Drum barking and pellets factories using non-dried sawdust can cause monoterpene concentrations at or close to the lower occupational exposure limits. Sesquiterpenes should be included in the total terpene exposure load.

Sawing and lumber drying are equally important terpene emitting processes. Emissions during pellets production can be as important as emissions from sawdust drying, although emissions during drying can also be considerably larger depending on drying technique.

The importance of natural versus anthropogenic emissions depends on the environmental issue. Climate change is global. The importance of the terpene contribution depends on the total amount of emitted terpenes. Natural sources of terpenes would therefore be more important than anthropogenic sources. Formation of photo-oxidants is a regional problem. Terpenes contribute in accordance with their concentration. Hence anthropogenic emissions would be more important.. Effects on insects are local, with bark drying an interesting anthropogenic source.

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