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# The effects of paints and moisture content on the indoor air emissions from pinewood (*Pinus sylvestris*) boards

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## Abstract

The emissions of volatile organic compounds (VOCs) from building materials may significantly contribute to indoor air pollution, and VOCs have been associated with odor annoyance and adverse health effects. Wood materials together with coatings are commonly used indoors for furniture and large surfaces such as walls, floors, and ceilings. This leads to high surface-to-volume ratios, and therefore, these materials may participate remarkably to the VOC levels of indoor environment. We studied emissions of VOCs and carbonyl compounds from pinewood (*Pinus sylvestris*) boards of 10% and 16% moisture contents (MC) with three paints using small-scale test chambers (27 L). The emissions from uncoated pinewood and paints (on a glass substrate) were tested as references. The 28-day experiment showed that the VOC emissions from uncoated pinewood were lower from sample with 16% MC. Painted pinewood samples showed lower emissions compared to paints on glass substrate. Additionally, paints on 16% MC pinewood exhibited lower emissions than on drier 10% MC wood. The emissions from painted pinewood samples were dominated by paint-based compounds, but the share of wood-based compounds increased over time. However, we noticed differences between the paints, and wood-based emissions were clearly higher with the most permeable paint.

## KEYWORDS

emissions, interior paints, moisture content, pinewood, VOC, wood materials

## Practical implications

- The results of this study provide new data on the development of VOC emissions from painted pinewood boards, which can be used to minimize the indoor exposure.
- During the first 28 days, the emissions from painted pinewood boards are dominated by paint-based compounds, and the painting drastically changes the emission profile compared to uncoated boards.
- Increased moisture content reduced the VOC emissions from both uncoated and painted pinewood boards.

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## 1 | INTRODUCTION

Volatile organic compounds (VOCs) and aldehydes emitted from different building materials are a possible cause of poor indoor air quality, and their indoor concentrations can be frequently higher compared to corresponding outdoor concentrations.<sup>1,2</sup> Several VOCs and aldehydes have been associated with odor annoyance and adverse health effects such as allergic and asthma symptoms, airway inflammation, and sensory irritation.<sup>2,3</sup> Additionally, possible harmful effects of individual compounds and mixtures of different chemicals are not evaluated thoroughly.<sup>4-6</sup> By contrast, exposure to some VOCs emitted from wood has been connected with beneficial health effects, such as psychological and physiological relaxation.<sup>7,8</sup>

Wood is widely used in buildings and indoor surfaces, and as a furniture material. The use of wood and wood-based materials in construction is further increasing because of environmental aspects, such as recyclability and biosourcing.<sup>9</sup> Different wood species emit a wide variety of VOCs, mainly terpenes, aldehydes and organic acids from softwoods, and carboxylic compounds, alcohols, and organic acids from hardwoods.<sup>10</sup> Emissions from softwoods are substantially larger than from hardwoods due to the presence of volatile terpenes.<sup>11</sup> In the Finnish classification for material emissions, building materials are classified in three categories (M1, M2, and M3) based on their chemical emissions.<sup>12</sup> Unprocessed solid softwood materials are considered to be equivalent with materials of the least emitting M1 class without any testing.<sup>12</sup> However, it is well known that the emissions, especially from freshly dried wood, can be higher than the requirements for other materials.<sup>13,14</sup>

Wood surfaces in indoors can be left uncoated. However, to improve performance and lifetime of the surfaces they are often treated with different coatings, such as paint, lacquer, wax, or oil. Coatings used on surface often reduce the emissions from the wood substrate while the coating itself additionally emits different chemicals.<sup>15,16</sup> Emission rates and emitted compounds from coatings vary significantly depending on type and content of resins and other agents used in the coating,<sup>17</sup> indicating the importance of analyzing and studying different coatings before installing.<sup>18</sup>

Together with coating, wood forms a simple composite material. In emission testing of these kind of materials, substrates and coatings usually come from different manufacturers and, thus, are tested separately. Coatings are commonly tested on an inert, non-porous substrate, such as glass plate, but in real-life environments they are used on various materials, and it has been noticed that the material affects the emission behavior of the coating.<sup>19</sup> Several studies have shown that porous substrates have a significant effect on time profile of VOC emissions by reducing the initial peak concentrations.<sup>17,19-22</sup> Additionally, porous substrates have indicated to prolong the emission decay process.<sup>20,21</sup> Coatings' effect on emissions from substrate material is less studied subject probably because emissions from substrates are substantially smaller compared to emissions from coatings.<sup>23</sup> However, coatings' emissions decrease

rapidly,<sup>4</sup> and substrates' emissions might become more dominant over time.

Moisture may enable, alter, or promote chemical reactions in materials,<sup>24</sup> and increasing ambient relative humidity (RH) has been found to have significant effect on emissions from water-borne coatings.<sup>24-27</sup> Wood is a hygroscopic material, so it can store and release moisture when RH of surrounding air changes.<sup>10</sup> Therefore, the moisture content (MC) of wood varies according to its environment. Additionally, wood species have naturally different moisture contents,<sup>10</sup> and the moisture content of the wood product depends on its drying process and purpose of use. For indoor and structural use, wood's MC should be below 20% to avoid fungal degradation and excess movement from moisture variation.<sup>28</sup> Coatings significantly affect the moisture exchange between wood and indoor environment, and the effect varies depending on the properties of the coating.<sup>29</sup> In uncoated wood, it has been suggested that reduced MC below 10% increases VOC emissions because of increased temperature from decreased evaporative cooling, and additionally, lower MC promotes wood degradation process causing the formation of VOC compounds, such as aldehydes and methanol.<sup>11</sup> Moreover, wet wood is less exposed to oxidation which results in decreased secondary emissions.<sup>11</sup> Conversely, some studies have reported higher VOC emissions from wood with higher MC.<sup>30</sup> However, little is known about how the MC of the substrate affects emissions from the overlying coating.

The main objective of this study was to investigate the effect of different water-borne paints on the emissions of pinewood substrates with two moisture contents and how the MC itself affects the emissions from uncoated wood. Additionally, we investigated how the substrate and its MC affect the emissions of the paints.

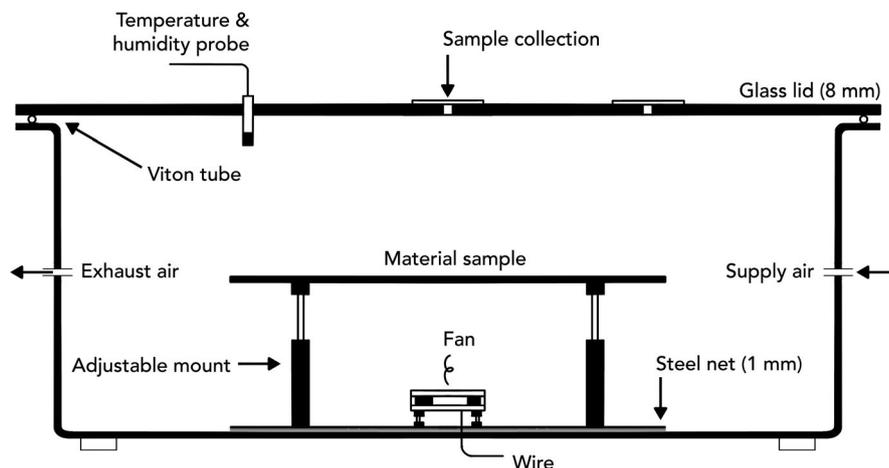
## 2 | MATERIALS AND METHODS

### 2.1 | Chamber apparatus and instrumentations

The materials' emissions were tested using six identical small-scale (27 L) stainless-steel chambers (Figure 1). The chambers had 8-mm glass lids sealed with Viton tubes. Measurements were performed from the three apertures present in the glass lid: one to measure humidity and temperature data (HMP 44L, Vaisala Oyj, Vantaa, Finland), one to collect the emission samples (Tenax TA and DNPH samples), and one aperture remained unused. When not in use, the apertures were covered with a glass plate to avoid pressure drop and emission loss.

The material samples were placed inside the chambers on adjustable steel mounts. The distance between the surface of the sample and the bottom of the chamber was adjusted to 95 mm to guarantee identical air velocity at the top surface of the samples. The adjustable mounts were placed in the center of the chamber on a 1 mm stainless-steel wire net (surface area 220 × 280 mm<sup>2</sup> with a mesh size of 1.0 × 1.0 mm<sup>2</sup>). The proper mixing and air velocity inside

**FIGURE 1** Configuration of the environmental test chamber designed by Wirtanen.<sup>24</sup>



the chamber were regulated with a fan placed in the middle of the stainless-steel wire net.

Teflon tubing simultaneously supplied the six chambers with cleaned and humidified air. The air was supplied from the laboratory compressed air outlet, and it was cleaned and dehumidified with oil separator, moisture extractor, activated carbon filter, series of particulate filters, zero-air generator, and pressure and flow velocity regulators connected in series. The relative humidity (RH) of supply air was maintained at 50% by combining dry air with water-saturated air before entering the chambers. Conditions in the chambers are specified in Table 1. The air flow was regularly checked with an air flow meter (Flowmeter 7000, Ellutia Ltd, Ely, UK).

The chambers were placed in an air-conditioned laboratory (+21°C). Before testing, the chambers were rinsed with methanol and distilled water. The loose metal parts were heated at 400°C before introducing them into the chambers.

## 2.2 | Test materials

The procedure of the emission test was based on ISO 16000-9, ISO 16000-6, ISO 16000-3, and Finnish M1 protocol for emission classification of building materials.<sup>31-34</sup> The chamber tests consisted of evaluating the volatile organic compounds (VOCs) and carbonyl emissions of uncoated pinewood, painted pinewood, and paints (on glass substrate). Surface areas of the test specimens (pinewood, glass substrate) were selected to represent the wall surfaces of the European reference room (CEN/TS 16516), which resulted in loading factors of 1 m<sup>2</sup>/m<sup>3</sup> inside the chambers.<sup>35</sup>

The planed pinewood (Scots pine, *Pinus sylvestris*) boards (15 × 145 × 3300 mm) were purchased from a local hardware store. Scots pine was selected because of its availability and prevalence as construction material. The boards had been packed by the manufacturer two months earlier. To achieve as homogenous samples as possible, wood samples that did not contain knots or other irregularities were cut from one board selected for the testing. After cutting, the wood samples were stored in a weather chamber made of stainless steel at 20 ± 1°C and 50 ± 5% or 80 ± 5% of

**TABLE 1** Test conditions of the chambers.

| Test condition                                   | Small-scale chamber (27 L)   |
|--|------------------------------|
| Sample area (m <sup>2</sup> )                    | 0.03                         |
| Volume (m <sup>3</sup> )                         | 0.03                         |
| Loading factor (m <sup>2</sup> /m <sup>3</sup> ) | 1                            |
| Temperature (°C)                                 | 21 ± 2°C                     |
| Relative humidity (%)                            | 50 ± 5% (chamber supply air) |
| Air flow rate (mL/min)                           | 225 ± 5%                     |
| Air exchange rate (h <sup>-1</sup> )             | 0.5                          |
| Air velocity on sample surface (m/s)             | 0.1–0.3                      |

relative humidity (RH) to reach the moisture equilibrium of surrounding environment, which resulted in samples with moisture content (MC) of around 10% or 16%, respectively. All samples were placed in the weather chamber at the same time to avoid any contamination. The samples with 10% MC were tested first, and after removing these samples from the weather chamber, its RH was set to 80%. Therefore, the samples with 16% MC remained in the chamber approximately one month longer than the dryer samples. After both moisturizing periods, one sample was selected to be dried in 100°C oven, and the MC was calculated from the weight loss of the sample.

Three water-borne indoor paints were selected as coating materials (Table 2). In addition to uncoated wood samples, all paints were tested on a wood substrate of both moisture contents, and on a glass substrate that had the same surface area as the wood substrates.

To minimize edge emissions, bottom and edges of the pinewood samples were taped with low emission self-adhesive aluminum tape. Two layers of paints (following the products instructions) were applied on the pinewood samples and reference samples (glass substrate) using foamed plastic paint rollers. Target weight for each paint was calculated according to paint's density and the manufacturer's specifications (m<sup>2</sup>/L). The test samples were introduced in the chambers immediately after applying the second layer of paint.

TABLE 2 Materials and their main characteristics

| Material | Main characteristic                          | Finnish classification <sup>a</sup> | European classification <sup>36</sup> |
|----------|--|-------------------------------------|---------------------------------------|
| Pinewood | Planed wood board, 145 × 194 mm <sup>2</sup> |                                     |                                       |
| Glass    | Glass plate, 145 × 194 mm <sup>2</sup>       |                                     |                                       |
| Paint 1  | Sample paint for timber walls                | N/A                                 | N/A                                   |
| Paint 2  | Furniture paint                              | M1                                  | Cat A/d VOC max 130 g/L               |
| Paint 3  | Interior paint                               | M1                                  | Cat A/a VOC max 30 g/L                |

<sup>a</sup>The paints have M1 classification, except Paint 1 which was a recently formulated sample product. The limit for the area specific emission rate of total volatile organic compounds (TVOC) for M1 products is below 200  $\mu\text{g}/\text{m}^2\text{h}$  after 28 days.<sup>12</sup>

The unopened paint cans were stored in the laboratory room until the application of the paints. After use, the cans were carefully closed to avoid variations between tests.

### 2.3 | VOC and carbonyl sampling

Two series of tests were performed for 28 days. First, the 10% MC pinewood samples were tested, and then the 16% MC pinewood samples. Both VOC and carbonyl samples were collected from the central aperture of the glass lid (right above the sample).

The VOCs were actively sampled at 150 mL/min on Tenax TA sorbent tube using a pump (GilAir Plus, Gilian). The samples were collected from the center of the emission chamber above the tested material sample on days 1, 3, 7, 14, and 28 with a sampling volume varying from 0.45 to 6 L, depending on the sampling day. The 0.45 L sample was used on day 1 when high emissions from the paints were expected, and the sampling volume was increased on each sampling day to detect smaller concentrations emitting from the wood itself. Therefore, it is possible that some compounds remained undetected during the first sampling days. The Tenax TA sorbents were analyzed using Thermal Desorption-Gas Chromatography-Mass Spectrometry (TD-GC-MS) and HP-5MS column (50 m × 0.2 mm × 0.33  $\mu\text{m}$ , Agilent technologies). The compounds were identified using a 50 compounds standard (Sigma Aldrich) and NIST 2011-mass spectrometry database. The results were quantified as toluene equivalents. Limits of detection (LOD) for toluene were calculated as  $\text{SER}_a$  values for each sampling day: 2.92  $\mu\text{g}/\text{m}^2\text{h}$  (day 1), 2.19  $\mu\text{g}/\text{m}^2\text{h}$  (day 3), 1.10  $\mu\text{g}/\text{m}^2\text{h}$  (day 7), 0.38  $\mu\text{g}/\text{m}^2\text{h}$  (day 14), and 0.22  $\mu\text{g}/\text{m}^2\text{h}$  (day 28). Additionally, specific LOD values were calculated for  $\alpha$ -pinene,  $\beta$ -pinene, D-limonene, nonanal, and decanal. The LOD is available in the Table S1 as  $\mu\text{g}/\text{m}^2\text{h}$  and  $\mu\text{g}/\text{m}^3$ .

To determine carbonyl compounds, air samples were collected into DNPH cartridges (Sep-Pak XPOsure, Waters) using a pump (GilAir Plus, Gilian) at 150 ml/min. During the first test series, sampling volume was 10 liters. Sampling volume was increased to 15 L in the second series to achieve more accurate results. The samples were collected on days 1, 3, and 28 and analyzed with high-performance

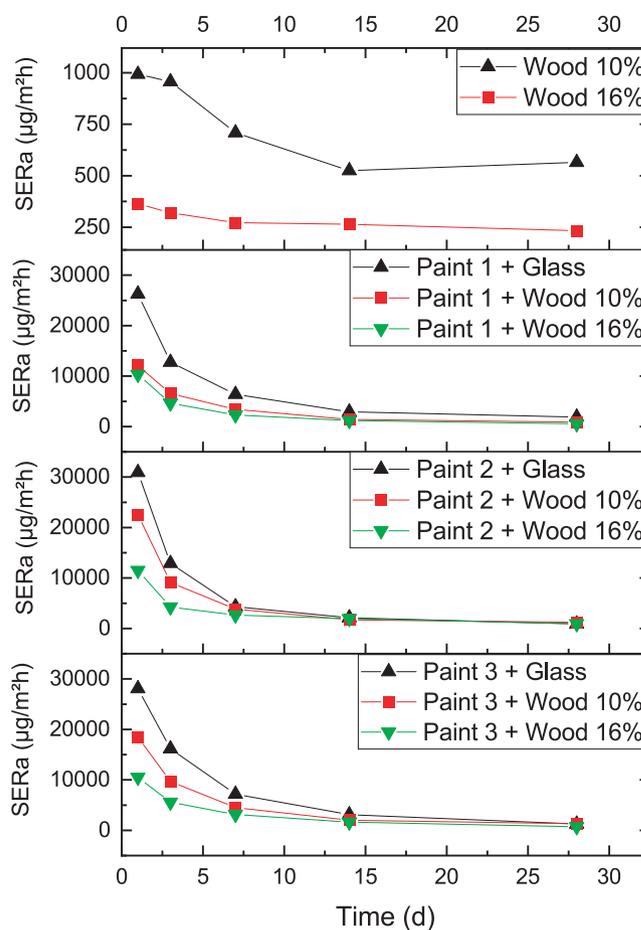


FIGURE 2 The area-specific emission rates of TVOC compounds from all materials and combinations.

liquid chromatography (HPLC) according to ISO 16000-3.<sup>33</sup> In our study, the analyzed compounds included formaldehyde and acetaldehyde. The LOD for formaldehyde was 7  $\mu\text{g}/\text{m}^3$  for 10 L sample and 3  $\mu\text{g}/\text{m}^3$  for 15 L sample, and calculated LOD  $\text{SER}_a$  ( $\mu\text{g}/\text{m}^2\text{h}$ ) was 3.5 and 1.5, respectively. For acetaldehyde, the LOD was 14  $\mu\text{g}/\text{m}^3$  for 10 L sample and 7  $\mu\text{g}/\text{m}^3$  for 15 L sample, and calculated LOD  $\text{SER}_a$  ( $\mu\text{g}/\text{m}^2\text{h}$ ) was 7 and 3.5, respectively.

### 3 | RESULTS

#### 3.1 | TVOC emissions of uncoated and painted samples

Background concentrations of VOCs in empty chambers were between 20 and 31  $\mu\text{g}/\text{m}^3$ , which partly exceeded the limit value of 20  $\mu\text{g}/\text{m}^3$  given in ISO 16000-9.<sup>31</sup> However, concentrations of single compounds were  $<2 \mu\text{g}/\text{m}^3$ , and thus, the background concentrations were not subtracted from the results of the emissions tests presented in the article.

The area-specific emission rates (SERa) of total volatile organic compounds (TVOC) from all studied materials and combinations throughout the study are presented in Figure 2. The emission rates of the uncoated wood samples were remarkably lower compared to the painted samples at the beginning of the testing period. Wood with 10% MC had significantly higher emissions, and the emission rates were two to three times higher than from wood with 16% MC on each sampling day. The emission rates of wood with 16% MC decreased throughout the tests, while wood with 10% MC had a slight increase from day 14 to day 28. On day 28, the emission rate for wood with 10% MC was 43% smaller than on day 1, while the emission rate of wood with 16% MC decreased by 36%.

TVOC profiles of the painted samples showed high initial emissions, and the emission rates approximately halved between each consecutive sampling day. Substrate's effect was significant on the emission rate. Paints on glass plates had higher emission rates than paints on wood substrates; except on day 28, emissions from Paint 2 were smaller on glass plate than on wood substrates. In addition, on day 28, emissions of Paint 3 were marginally higher on wood with 10% MC compared to glass plate. The effect of the woods' MC was clearly visible. Paints 2 and 3 had almost two times higher initial emission rates on dryer wood substrates compared to the moister substrates, and similar trend was observable throughout the test. The same trend was also visible between the wood substrates of Paint 1, but the difference of initial emissions was more moderate. The emission rates decreased substantially during the experiment, and the emission rates on day 1 were 12 to 35 times larger than on day 28, varying between different paints and substrates.

#### 3.2 | VOC emissions of uncoated pinewood samples

The most abundant chemical groups in uncoated wood's emissions were terpenes and aldehydes, followed by monatomic alcohols (Table 3). Wood with 10% MC had higher emission rates in all

TABLE 3 The area-specific emission rates of the compounds from the most abundant chemical groups from uncoated wood samples.

| Compound                  | CAS        | Wood 10% SER, [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |                 |                 |                |                | Wood 16% SER, [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |       |       |                |                |
|---------------------------|------------|---|-----------------|-----------------|----------------|----------------|---|-------|-------|----------------|----------------|
|                           |            | Day 1   | Day 3           | Day 7           | Day 14         | Day 28         | Day 1   | Day 3 | Day 7 | Day 14         | Day 28         |
| <b>Aldehydes</b>          |            | 171   | 203             | 155             | 166            | 222            | 153   | 139   | 126   | 126            | 122            |
| Pentanal                  | 110-62-3   | 27  | 25              | 25              | 28             | 38             | 15  | 15    | 18    | 20             | 20             |
| Hexanal                   | 66-25-1    | 109   | 101             | 106             | 123            | 168            | 113   | 102   | 93    | 93             | 89             |
| Heptanal                  | 111-71-7   | 3   | 4               | 2               | 2              | 2              | <LOD  | 2     | 2     | 2              | 2              |
| Octanal                   | 124-13-0   | 7   | 10              | 5               | 4              | 5              | 7   | 5     | 4     | 4              | 4              |
| Nonanal                   | 124-19-6   | 14  | 41              | 10              | 6              | 6              | 8   | 8     | 5     | 5              | 5              |
| Decanal                   | 112-31-2   | 10  | 22              | 6               | 3              | 2              | 7   | 6     | 3     | 2              | 2              |
| <b>Terpenes</b>           |            | 323   | 314             | 276             | 185            | 168            | 46  | 55    | 57    | 59             | 46             |
| $\alpha$ -pinene          | 7785-26-4  | 186   | 171             | 137             | 80             | 67             | 26  | 29    | 28    | 25             | 18             |
| Camphene                  | 79-92-5    | <LOD  | 3               | 3               | <2             | <2             | -   | <LOD  | <LOD  | <2             | <2             |
| Sabinene                  | 3387-41-5  | -   | <LOD            | 2               | -              | -              | -   | -     | -     | -              | -              |
| $\beta$ -pinene           | 127-91-3   | 10  | 11              | 8               | 6              | 5              | -   | <LOD  | <2    | <2             | <2             |
| $\beta$ -myrcene          | 123-35-3   | 8   | 9               | 7               | 6              | 6              | -   | -     | -     | -              | -              |
| 3-carene                  | 13466-78-9 | 97  | 100             | 98              | 74             | 70             | 17  | 20    | 24    | 27             | 23             |
| D-limonene                | 5989-27-5  | 20  | 19              | 22              | 18             | 18             | 3   | 4     | 4     | 5              | 4              |
| <b>Monatomic alcohols</b> |            | 41  | 41              | 40              | 36             | 47             | 18  | 21    | 15    | 13             | 14             |
| 1-pentanol                | 71-41-0    | 28  | 27              | 29              | 27             | 39             | 9   | 10    | 10    | 10             | 11             |
| 2-ethyl-1-hexanol         | 104-76-7   | 13 <sup>a</sup>                                     | 14 <sup>a</sup> | 11 <sup>a</sup> | 9 <sup>a</sup> | 8 <sup>a</sup> | 9   | 11    | 5     | 3 <sup>a</sup> | 3 <sup>a</sup> |
| <b>TVOC</b>               |            | 992   | 957             | 708             | 525            | 565            | 364   | 320   | 272   | 264            | 233            |

<sup>a</sup>Co-elution of 2-ethyl-1-hexanol and p-cymene.

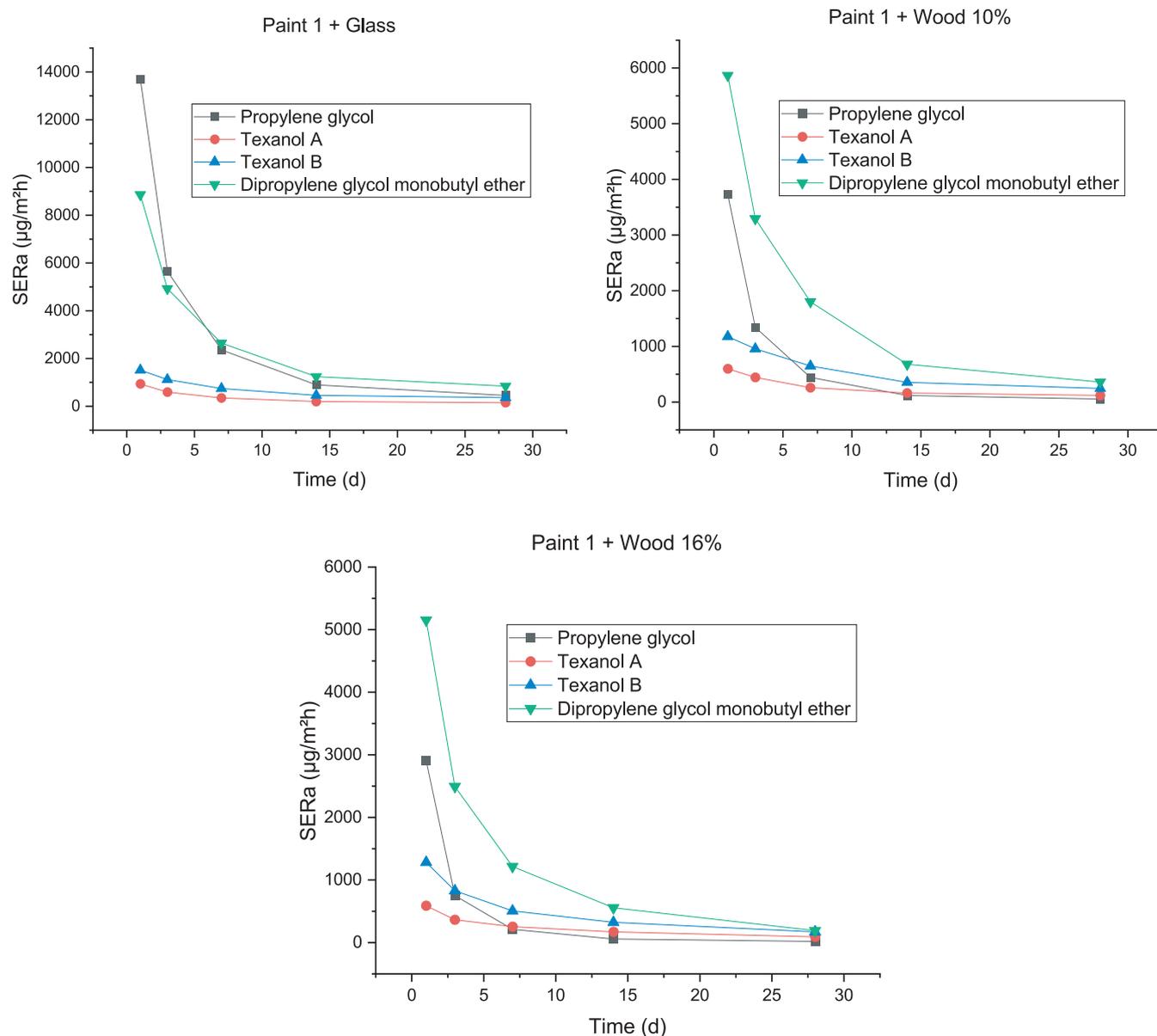


FIGURE 3 The area-specific emissions rates of the main compounds from Paint 1 on different surfaces.

chemical groups. Identified terpene compounds covered 30%–39% of TVOC emissions of wood with 10% MC, and 17%–39% of emissions were aldehydes. Share of aldehydes was 42%–53% in the TVOC emissions of wood with 16% MC, while terpenes had a smaller part with 13%–22%. In both wood samples, the main compounds from the terpene group were  $\alpha$ -pinene and 3-carene, and hexanal had the highest emissions of aldehydes followed by pentanal. However, emission rates of these compounds differed between the samples. Initial hexanal emissions were nearly the same from both samples, but the emissions from wood with 10% MC increased during the research period, while the emissions from wood with 16% MC slightly decreased. Terpene emissions were notably higher from wood with 10% MC and the emission rates decreased throughout the testing period. For wood with 16% MC, the emission rates of terpenes were approximately the same in each sampling. Additionally,

a greater variation of different terpene compounds was found in the emissions of wood with 10% MC.

### 3.3 | VOC emissions of painted samples

On different substrates, all paints showed high initial emissions of propylene glycol (CAS 57-55-6), Texanol A (CAS 74367-33-2), and Texanol B (CAS 74367-34-3). Additionally, Paints 1 and 3 had high dipropylene glycol monobutyl ether (CAS 29911-28-2) emissions, while the emissions were fractional from Paint 2. The emission rates of these main compounds from the three paints on different substrates are presented in Figures 3-5. The main compounds covered 95%–98% of the total emissions from all paints on day 1, and 78%–93% on day 28. The effect of substrate on propylene

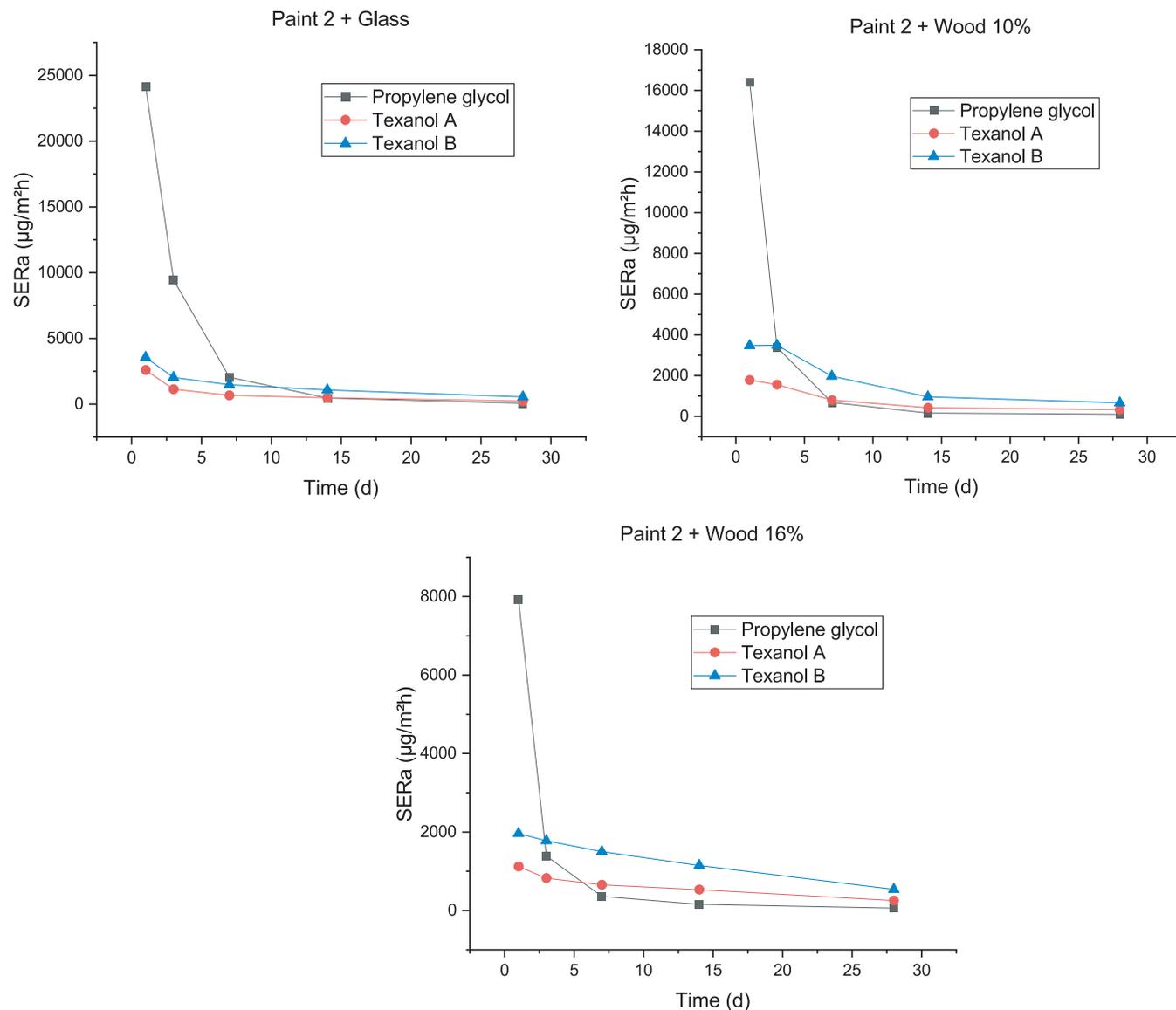


FIGURE 4 The area-specific emissions rates of the main compounds from Paint2 on different surfaces.

glycol emissions was significant with all paints, and the emission rates were clearly lower on wood substrates with one exception. On day 28, the emission rate from Paint 2 on glass substrate was on the same level with wood with 16% MC and smaller than from wood with 10% MC. All paints on wood samples with 10% MC had higher propylene glycol emissions than on wood samples with 16% MC. In addition, propylene glycol's proportions of TVOC emissions from Paints 1 and 3 on the wood substrate were smaller compared to glass plates. However, on day 1 and 28, propylene glycol's share of TVOC emissions from Paint 2 was approximately the same on all substrates.

Emissions of Texanol A and B showed more variation between different paints and substrates. Texanol B had higher emission rates than Texanol A from all paints on all substrates, and the initial emission rates were significantly smaller compared to propylene glycol. The emission rates of both compounds from Paint 1 on glass were higher compared to wood substrates. Paints 2 and 3 had the

lowest emission rates for Texanols on wood with 16% MC while the emission profiles from other substrates resembled each other. Even though Texanol emissions decreased throughout the tests, their share of TVOC emissions increased toward the end of the test period. On day 28, over 80% of TVOC emissions from Paint 2 on all substrates were Texanols.

The emission rates of dipropylene glycol monobutyl ether on day 1 from Paints 1 and 3 on glass plates were lower than those of propylene glycol, but the emissions decreased less over time. On wood substrates, the emission rates from Paint 1 were clearly lower compared to the glass plate, but the time-emission profiles were somewhat similar between the wood substrates. On day 1, dipropylene glycol monobutyl ether's emission rate from Paint 3 was slightly higher on wood with 10% MC than on glass plate, but otherwise, the emission rates were lower from both paints on wood substrates than on glass plate. Additionally, the emissions from both paints were the smallest on wood with 16% MC.

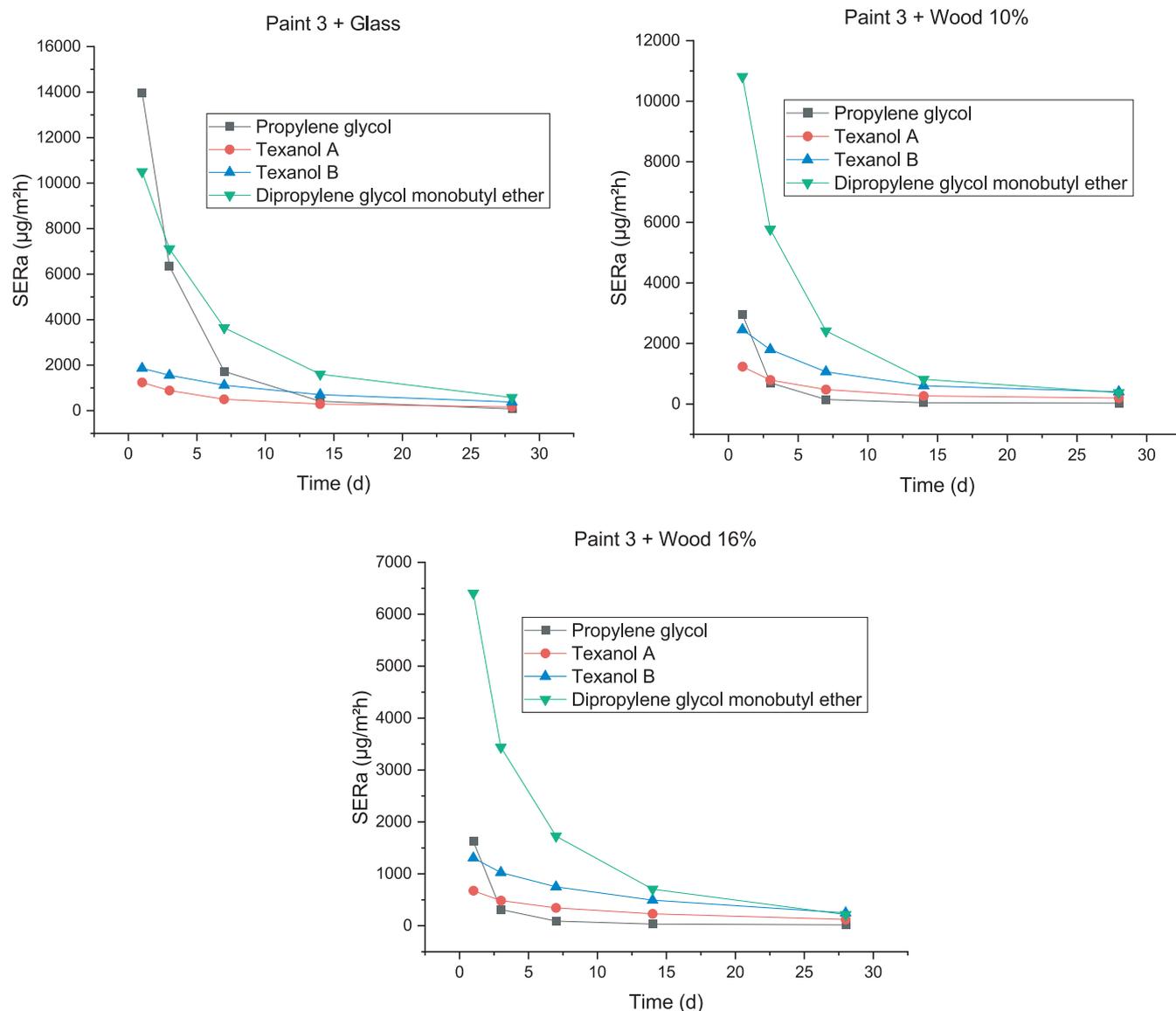


FIGURE 5 The area-specific emissions rates of the main compounds from Paint 3 on different surfaces.

### 3.4 | Pinewood-based VOC emissions from painted samples

Several compounds were identified from emissions of paints on wood substrates that were detected from uncoated wood emissions but not from paints on glass plates (Table 4). These compounds include terpenes (mainly  $\alpha$ -pinene,  $\beta$ -pinene, 3-carene, D-limonene), aldehydes (pentanal, hexanal, heptanal), and alcohols (1-pentanol). Additionally, some compounds such as aldehydes (octanal, nonanal, decanal) and alcohols (2-ethyl-1-hexanol) were found from emissions of both uncoated wood and paints on glass plates. They were also present in most of the air samples taken from the coated wood samples in somewhat elevated concentrations compared to painted glass samples. However, it was difficult to determine whether they originate from the wood or the paints, and therefore, they were left out of this section.

Same as with the uncoated wood samples, paints on wood with 10% MC exhibited higher emissions compared to the wood samples with 16% MC. Emission rates of the individual wood-based compounds from samples with Paints 1 and 2 were up to  $13 \mu\text{g}/\text{m}^2 \text{h}$ , which is significantly less than from uncoated wood samples. Wood samples with Paint 3 had higher emission rates with a maximum value of  $90 \mu\text{g}/\text{m}^2 \text{h}$  for hexanal. Emission rates of terpene compounds seemed to increase throughout the testing, and the highest concentrations were measured on day 28. 1-pentanol emissions slightly decreased over time while aldehydes exhibited more variation.

Share of these wood-based compounds of TVOC emissions increased toward day 28 with all paints on both wood substrates. During the first two sampling days, the shares were between 0 and 1%. On day 28, wood-based emissions were more moderate from substrates with Paints 1 and 2, between 2 and 7% of TVOC

TABLE 4 The area-specific emission rates of wood-based compounds from painted wood samples.

| Compound         | CAS        | Wood 10% SER <sub>a</sub> [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |       |                |                |                | Wood 16% SER <sub>a</sub> [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |       |       |        |                |
|------------------|------------|---|-------|----------------|----------------|----------------|---|-------|-------|--------|----------------|
|                  |            | Day 1   | Day 3 | Day 7          | Day 14         | Day 28         | Day 1   | Day 3 | Day 7 | Day 14 | Day 28         |
| Paint 1          |            |   |       |                |                |                |   |       |       |        |                |
| Pentanal         | 110-62-3   | <LOD  | 2     | 3              | 3 <sup>a</sup> | 3 <sup>a</sup> | -   | -     | <LOD  | <2     | 2 <sup>a</sup> |
| Hexanal          | 66-25-1    | 9   | 6     | 6              | 6              | 8              | <LOD  | 3     | 3     | 5      | 7              |
| 1-pentanol       | 71-41-0    | 15  | 3     | 2              | 2              | 2              | 9   | <LOD  | <LOD  | <2     | <2             |
| $\alpha$ -pinene | 7785-26-4  | 7   | 6     | 7              | 8              | 9              | 3   | 3     | 5     | 8      | 13             |
| 3-carene         | 13466-78-9 | 6   | 4     | 3              | 3              | 4              | <LOD  | 2     | 3     | 5      | 10             |
| D-limonene       | 5989-27-5  | 7   | 3     | <2             | <2             | <2             | -   | -     | -     | <2     | 2              |
| Paint 2          |            |   |       |                |                |                |   |       |       |        |                |
| Pentanal         | 110-62-3   | 4   | 4     | 3 <sup>a</sup> | 3 <sup>a</sup> | 3              | -   | <LOD  | <LOD  | <2     | <2             |
| Hexanal          | 66-25-1    | -   | 4     | 6              | 5              | 8              | -   | -     | <2    | 2      | 3              |
| 1-pentanol       | 71-41-0    | -   | 8     | 7              | 5              | 6              | -   | 3     | 3     | 3      | 2              |
| $\alpha$ -pinene | 7785-26-4  | 8   | 8     | 10             | 11             | 12             | <LOD  | <LOD  | 2     | 4      | 4              |
| 3-carene         | 13466-78-9 | 10  | 8     | 10             | 9              | 10             | 4   | 2     | 3     | 3      | 4              |
| D-limonene       | 5989-27-5  | 5   | 5     | 5              | 4              | 4              | 2   | 2     | <2    | <2     | <2             |
| Paint 3          |            |   |       |                |                |                |   |       |       |        |                |
| Pentanal         | 110-62-3   | 23  | 11    | 10             | 11             | 13             | 6   | 4     | 5     | 6      | 8              |
| Hexanal          | 66-25-1    | 90  | 46    | 35             | 36             | 47             | 25  | 28    | 31    | 34     | 35             |
| Heptanal         | 111-71-7   | <LOD  | <LOD  | <LOD           | <2             | <2             | -   | -     | <LOD  | <2     | <2             |
| 1-pentanol       | 71-41-0    | 23  | 11    | 8              | 7              | 8              | 5   | 4     | 4     | 3      | 3              |
| $\alpha$ -pinene | 7785-26-4  | 24  | 24    | 45             | 53             | 60             | 7   | 10    | 14    | 15     | 15             |
| $\beta$ -pinene  | 127-91-3   | -   | -     | 2              | 3              | 3              | -   | -     | <LOD  | <2     | <2             |
| $\beta$ -myrcene | 123-35-3   | 4   | 4     | 3              | 3              | 3              | -   | -     | -     | -      | -              |
| 3-carene         | 13466-78-9 | 20  | 16    | 21             | 29             | 37             | 6   | 6     | 9     | 10     | 12             |
| D-limonene       | 5989-27-5  | 11  | 6     | 7              | 9              | 11             | -   | <LOD  | <2    | <2     | 2              |

<sup>a</sup>Co-elution of heptane and pentanal.

TABLE 5 The area-specific emission rates of formaldehyde and acetaldehyde from all tested materials and combinations.

| Specimen                        | Formaldehyde [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |       |        | Acetaldehyde [ $\mu\text{g}/\text{m}^2 \text{h}$ ] |       |        |
|---------------------------------|--|-------|--------|--|-------|--------|
|                                 | Day 1  | Day 3 | Day 28 | Day 1  | Day 3 | Day 28 |
| Wood 10%                        | 5  | 7     | 9      | 9  | 14    | 17     |
| Wood 16% <sup>a</sup>           | 6  | 10    | 14     | 12   | 24    | 20     |
| Paint 1 + Glass <sup>a</sup>    | 7  | 2     | 4      | 7  | 4     | <LOD   |
| Paint 1 + Wood 10%              | 4  | <LOD  | <LOD   | 16   | 7     | <LOD   |
| Paint 1 + Wood 16% <sup>a</sup> | 7  | <LOD  | 3      | 25   | 5     | 9      |
| Paint 2 + Glass                 | 4  | 5     | 4      | 10   | <LOD  | <LOD   |
| Paint 2 + Wood 10%              | <LOD   | <LOD  | 4      | <LOD   | <LOD  | <LOD   |
| Paint 2 + Wood 16% <sup>a</sup> | 4  | 4     | 2      | 6  | <LOD  | <LOD   |
| Paint 3 + Glass                 | 7  | <LOD  | 4      | <LOD   | <LOD  | <LOD   |
| Paint 3 + Wood 10%              | <LOD   | <LOD  | 5      | <LOD   | <LOD  | 11     |
| Paint 3 + Wood 16% <sup>a</sup> | 3  | 2     | 3      | 5  | 6     | 21     |

<sup>a</sup>Sampling volume was 15 L instead of 10 L.

emissions. From samples coated with Paint 3, the share was 14% on wood with 10% MC and 11% on wood with 16% MC.

### 3.5 | Formaldehyde and acetaldehyde emissions

Emissions of formaldehyde and acetaldehyde were studied separately because of the highly volatile nature of these compounds they cannot be collected and analyzed using Tenax TA adsorbents. Emission rates of formaldehyde and acetaldehyde are presented in Table 5. Uncoated wood samples with higher moisture content showed higher emission rates for both compounds throughout the testing period. The emission rates increased slightly over time, except wood with 16% MC had higher emissions for acetaldehyde on day 3 than on day 28.

In general, the emission rates of formaldehyde were low and partly under the limit of detection. On day 1, the emissions from painted glass and uncoated wood samples tended to be slightly higher than those from painted wood samples. On days 3 and 28, uncoated wood samples yielded the highest emissions while the emission rates from painted samples were low and closer to each other.

The emission rates of acetaldehyde had more variation between different samples. On day 1, wood samples coated with Paint 1 showed higher emissions compared to uncoated wood samples and Paint 1 on glass, and wood samples coated with Paint 2 had the opposite results. Emission rates from samples coated with Paint 3 were close or under the limit of detection. On days 3 and 28, emissions from samples coated with Paint 1 decreased, and emissions from samples with Paint 2 were under the limit of detection. Acetaldehyde emissions from wood samples coated with Paint 3 increased over time, and on day 28, Paint 3 on wood with 16% MC showed slightly higher emission rate than uncoated wood sample with 16% MC. Emission rates from Paint 3 on glass were under the limit of detection on all days.

## 4 | DISCUSSION

As expected, the emission behavior of uncoated wood samples differed significantly from the painted samples. The emission rates in the beginning of the testing period were considerable but clearly lower than from the painted samples. TVOC emissions decreased moderately throughout the experiments, and the emission levels were comparable with previous results.<sup>14</sup> Additionally, the main compounds identified from the emissions were consistent with the VOC data from the literature.<sup>13-15,37-39</sup> However, we noticed some differences between the wood samples of two different MCs. Emissions from wood with 10% MC were consistently higher than from wood with 16% MC. Wood with 10% MC had higher emission rates in all chemical groups, and especially terpene emissions were clearly lower from the wood with higher MC. Wood with 16% MC was kept in the environmental chamber approximately one month longer before testing than wood with 10% MC, so one possible explanation is that some of these terpenes may have emitted during that time. In a

previous study, it has been noticed that even a storage time of two weeks reduces terpene compounds' emissions drastically.<sup>13</sup> In our study, the emission rates of terpenes from wood with 10% MC approximately halved during the 28-day experiment, and the emissions were almost on the same level already after two weeks.

The emissions of certain chemical groups depend on different mechanisms, such as diffusion and chemical interactions as well as chemical distribution in wood. Hexanal and the other aldehydes are formed from degradation processes of wood (from the oxidative degradation of natural lipids present in wood).<sup>40</sup> This explains the steady emission of aldehydes at both MCs in a moisture independent manner, whereas terpenes are found in resin located in parenchyma cells or resin canals.<sup>41</sup> Wood contains also fatty acid surfactants, such as oleic acid, which promote hydrophobic compounds, such as terpenes, to dissolve to water and transfer in wood.<sup>42</sup> In lower moisture content (MC 10%), higher terpene concentration is dissolved in water and higher volatile loss of terpenes dissolved in surface water occurs than in case of MC 16%. Same phenomenon is seen in wood drying process where terpenes like  $\alpha$ -pinene are emitted through evaporation when the wood is almost dry.<sup>39</sup>

It should be noted that the moisture contents of all wood samples presumably did not remain steady throughout the experiment. Before testing, the wood samples with 10% MC were stored in the environmental chamber in humidity and temperature conditions that were approximately same as they were inside the test chambers. The wood samples with 16% MC were kept in 80% RH before introducing them into the test chambers with 50% RH. Therefore, while the wood samples with 10% MC remained in moisture equilibrium through the testing, the wood samples with 16% MC desorbed humidity until the moisture equilibrium with the test chamber conditions was reached. Lin et al<sup>43</sup> suggested that in higher surrounding RH, VOC emissions increase because of reduced evaporation of water from wood. Alternatively, lower RH increases water evaporation, which absorbs energy and prevents the evaporation of VOCs. Therefore, when the moister wood samples were placed in the test chambers, water evaporating from wood might have decreased the VOC emissions. However, this should result in higher emissions after the moisture equilibrium has been reached but the TVOC emissions from wood with 16% MC decreased throughout the study. Additionally, previous studies indicate that moisture cycles reduce and modify VOC emissions, especially terpene emissions, from different wood species, which is in line with our results.<sup>44</sup>

TVOC time-emission profiles of the painted samples showed high initial emissions that decreased rapidly, which was in line with the results from previously published studies.<sup>4,20,45,46</sup> It is known that these initial emissions are controlled by rapid evaporation that reaches the emission peak a few hours after the paint has been applied.<sup>20</sup> The emissions are quickly reduced after the peak, and while the paint dries, the emission process transforms from evaporation-controlled stage to internal diffusion-controlled stage.<sup>20</sup> In this study, the TVOC emission rates reduced throughout the 28-day experiment. On average, the percentage of the decay was strongest between the first measurements (day 1 and day 3) but the percentage remained close to 50% between each consecutive sampling. On

day 28, the emission rates from the painted samples were 8% or below of the emissions on day 1. However, the TVOC emission rates were still significantly higher than the limit values of Finnish M1 material emission classification.<sup>12</sup> In M1 classification, it is instructed to apply decorative coatings as a single layer if multiple layers are not mandatory according to the manufacturer's technical information.<sup>34</sup> In this study, we used double layers of paints to achieve results that are more representative of real-life situations. This may have caused slower drying of the paint, and together with greater total mass of paint, it has probably resulted in higher emission rates on day 28.

The main compounds identified from the emissions of painted samples were propylene glycol, Texanol A and Texanol B. Additionally, Paints 1 and 3 had significant dipropylene glycol monobutyl ether emissions. Propylene glycol had the highest initial emission rates from all paints on glass plates, and on wood substrates from Paint 2. However, its emissions decreased drastically over time. Dipropylene glycol monobutyl ether was dominant in the emissions from Paints 1 and 3 on wood substrates. Wieslander & Norbäck<sup>47</sup> had similar findings in their study where they found out that the most abundant compounds in the emissions from water-based paints were propylene glycol, glycol ethers, and Texanol.

The effect of the wood substrate was significant especially during the first test week, and all paints showed clearly higher emission rates on glass plates than on wood. The wood substrate had the largest impact on propylene glycol's emissions, which were significantly lower from paints on wood substrates while the emissions of other main compounds remained closer to each other on different substrates. On day 1, propylene glycol's emission rates from paints on wood substrates were 32%–88% lower compared to glass substrates, and the lower levels mostly remained throughout the study. It has been previously noticed that porous substrates, such as wood, act like a "sponge" that hinder the initial peak emissions from the overlying coating.<sup>17,19,20</sup> Additionally, Li et al.<sup>20</sup> suggested that the aforementioned "sponge" effect prolongs the VOC emissions compared to a coating on an inert substrate, such as glass or metal plate. However, we did not observe this in our study, and after 28 days, the emission rates were higher or approximately same from paints on glass substrate than on wood substrates. Longer experiment would be needed to observe how this effect develops. Furthermore, Corsi & Lin<sup>48</sup> proposed that the emissions retardation of porous substrate might occur because of sorptive interactions between polar compounds and the substrate instead of liquid paint "soaking" into porous substrate.

Moreover, we noticed that the emission rates from paints on wood samples with 16% MC were lower compared to wood with 10% MC as a substrate. Substrate's MC did not affect any compound specifically, and the emission rates of the main compounds decreased evenly between the wood substrates. Their shares of TVOC emissions were approximately on the same level in all paints on both wood substrates throughout the study. It is possible that the moisture from the moister substrate may have postponed the drying process of the overlying paint and decelerated the emissions. It could be assumed that this would result in higher emissions over time, but this did not happen during the 28-day experiment.

Coating's effect on the emissions from the underlying wood substrate is less studied, but it is generally believed that the coating significantly decreases the emissions from the wood.<sup>11,15,37,49</sup> In our study, paint-based compounds dominated the emissions from painted wood samples throughout the experiment, but the share of wood-based compounds increased toward the end of the research period. Emission rates of wood-based terpene compounds increased over time, while aldehydes had more variation. The MC of the wood substrate had similar effect with both coated and uncoated wood samples, and the painted wood samples with 16% MC showed lower terpene emissions. Additionally, we observed differences between the paints. Paint 3 clearly had the highest emission rates of wood-based compounds, and the emissions from Paints 1 and 2 were more similar with each other. According to the manufacturer, Paint 3 has the highest water vapor permeability of their indoor acrylate paints. Therefore, it presumably allows more polar VOCs from the wood to evaporate to the surrounding air.

We studied the emissions of acetaldehyde and formaldehyde separately because they can not be collected with Tenax TA adsorbents, but they are known to be emitted from pinewood.<sup>11</sup> Both compounds had low emission rates from all samples, which were partly under the limit of detection. Therefore, presented differences between samples and sampling days may not be of great significance. Formaldehyde emissions from the uncoated wood samples increased over time, and after day 1, the emission rates were higher compared to coated wood and glass samples. Pibiri et al.<sup>50</sup> had similar results in their study, where they found out that paints applied on particleboards substantially hinder formaldehyde emissions from the substrate. Additionally, the emission rates were higher from the wood with higher MC. This is in line with earlier studies, where it has been noticed that higher MC promotes formaldehyde emissions from wood.<sup>51–54</sup> Moreover, it has been shown by experimental and theoretical studies that emission rate of formaldehyde is positively related to both temperature and humidity.<sup>55</sup> According to experiments done by Irle et al.,<sup>54</sup> wood's moisture content increases the formaldehyde diffusion. Additionally, MC may influence the mobility of formaldehyde in wood.<sup>50</sup> Therefore, diffusion in dry wood should be much slower than in wet wood, which might explain formaldehyde emission differences between MC 10% and 16%. Formaldehyde emissions from wood are proposed to originate from hydrolytic degradation of wood components, such as cellulose, hemicelluloses, and lignin as well as its extractives.<sup>56</sup> Therefore, higher moisture content could increase the hydrolytic degradation process and result in higher formaldehyde emissions. However, Irle et al.<sup>54</sup> suggested recently that degradation processes should be independent of moisture content, and at least 50% of the formaldehyde emitted from *Pinus radiata* is due to prior adsorption. Other possible explanation for increased formaldehyde emissions with higher MC is that because formaldehyde is water soluble, there is more formaldehyde available to be emitted. Similarly with formaldehyde emissions, uncoated wood had steadier acetaldehyde emissions. Additionally, coating seemed to increase the emission rates, but more variation occurred.

## 5 | LIMITATIONS AND FUTURE WORK

The main limitations for generalization of the results of the present study concern the following aspects. First, it is well known that the emissions from wood are influenced by several factors, such as the growing location of the tree and growth conditions, time of harvest, drying temperature and process, and handling of the timber.<sup>57</sup> Additionally, variation within an individual tree can be substantial, especially between heartwood and sapwood.<sup>10</sup> Therefore, the samples that were included in the study represent only an individual pine tree, and thus, the emission rates do not necessarily show the average emissions of pinewoods. Further, duplicates of all material samples studied would have helped to verify findings of our study. However, our study used wood samples cut from a single board with rather consistent emission results, so it is possible to use these results to estimate how the MC of wood and different coatings affect the overall emission when the emissions of the components are known.

Moisture contents for both conditions (10% and 16%) were determined using one sample in each case. For more accurate estimation, duplicate samples could have been used. However, the calculated moisture contents of the dried wood samples were similar with estimates provided by The Finnish Timber Council Puuinfo Ltd for pinewood at the same temperature and humidity conditions.

Additionally, the results of formaldehyde and acetaldehyde emissions were close or under the limits of detection. Therefore, these results should not be generalized, and they should be used with care. Larger sample volumes and duplicate samples are recommended in the future work.

In future work, it would be of great interest to follow how the effect of different MCs, their changes due to surrounding air, and coating develop on a longer time span. We noticed that the emissions from wood under the paints started to increase moderately during the 28-day experiment, but still the paints remained as the dominant emissions sources. As the service life of such material combinations can be several years, it would be interesting to know whether and when these ratios change and how the coating affects the emissions compared to those of uncoated wood. Additionally, it is suggested that porous substrates postpone emissions of coatings. With longer timespan, it would be possible to observe whether the wood substrate and higher MC result in prolonged emissions.

## 6 | CONCLUSIONS

We recognized some differences between the emissions from uncoated wood samples of different MC. Wood with 10% MC showed clearly higher emission rates of VOCs, and especially the emissions of terpene compounds were higher than in wood with 16% MC. However, the differences in the terpene emissions are probably explained with longer moisture stabilization time before

the emission tests of wood with 16% MC. Conversely, the moister wood sample had higher emission rates for formaldehyde and acetaldehyde.

Substrate's effect on the emissions from paints was remarkable. Particularly during the first week of testing, the TVOC emission rates from paints on wood substrates were substantially lower than on inert glass plate, and the lower rates mostly remained throughout the experiment. The difference was mainly caused by lower propylene glycol emissions. Additionally, we noticed that paints on wood samples with 16% MC had lower emission rates compared to wood samples with 10% MC.

The emissions from painted wood samples were dominated by paint-based emissions, but the share of wood-based emissions increased over time. However, we noticed some differences between the paints, and between wood substrates of different MC. Paint 3 exhibited clearly higher emission rates from the underlying wood substrates than the other paints. Similarly with the uncoated wood samples, paints on the moister 16% wood substrates had lower emission rates compared to paints on wood with 10% MC.

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### CONFLICT OF INTEREST

The authors declare no conflict of interests. The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; and in the decision to publish the results.

### AUTHOR CONTRIBUTIONS

TA took primary responsibility for planning the work, operating the test setup, collecting the literature, and writing the original draft of the manuscript. EC planned the chemical sampling and analyzed the samples as well as reviewed and edited the final version of the manuscript. LS prepared the material samples and took the chemical samples. RM, PP, and HS instructed and supervised the research process as well as reviewed and edited the final version of the manuscript.

### DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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## SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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