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VOC and carbonyl compound emissions of a fiberboard resulting from a coriander biorefinery: comparison with two commercial wood-based building materials

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Abstract
Indoor air quality is a major public health issue. It is related to the choice of construction materials and associated with VOC emissions. Two wood-based commercial panels were tested: a medium-density fiberboard (MDF) and a chipboard (CH), and they were compared to a material produced from a coriander biorefinery (COR). Indicators chosen to compare the materials were physical properties (density, bending properties, surface hardness, thickness swelling, and water absorption) and VOC emissions. Emissions were evaluated in an environmental chamber at 23 °C, 31 °C, and 36 °C, and during 28 days. Carbonyl emissions on day 1 at 23 °C were 74, 146, and 35 μg m^{-2} h^{-1}, respectively, for MDF, CH, and COR. Terpenic emissions were 12, 185, and 37 μg m^{-2} h^{-1}, respectively. Higher temperature resulted in higher emissions which decreased over time, except for formaldehyde. VOC emissions depended largely on material and temperature. Formaldehyde emission was 300 to 600 times lower for coriander boards (< 0.2 μg m^{-2} h^{-1}), making them significantly more environmentally friendly materials in comparison with MDF and chipboard. These results highlight the interest of coriander by-products as raw materials for producing fiberboards with low impact on indoor air quality.

Keywords VOC emissions · Formaldehyde · Coriander · Self-bonded fiberboards · MDF · Chipboard

Introduction

Indoor air quality (IAQ) is an increasingly important health concern for many types of buildings: industrial, commercial, hospital, residential, etc. (Poulhet et al. 2014; Marć et al. 2016; Baurès et al. 2018; Militello-Hourigan and Miller, 2018). Most people spend more than 90% of their time in confined spaces (Cincinelli and Martellini 2017). IAQ has therefore an important impact on health and quality of life. Different pollutants including particulate matter and gaseous compounds are involved. They may enter buildings with outside air or may be generated internally via combustion devices, decoration products (paints, floor coatings, varnishes, etc.), human activity (smoking, cleaning products, cooking, etc.), or furniture and building materials. Volatile organic compounds (VOCs) have been studied in indoor air, particularly formaldehyde, due to its mutagenic and carcinogenic character (IARC 2006; Salthammer 2015). Short-term exposure to formaldehyde and VOCs causes acute diseases, such as headache, asthma symptoms, and tiredness, all described as sick building syndromes (Molhave 1989; Wolkoff 2018; Vuokko et al. 2019).

The traditional way of material selection for building construction was primarily based on factors such as cost, usage properties (i.e., mechanical properties, fire-resistance, and resistance to water over time), esthetic aspects, availability and durability, and more recently on health aspects. Wood-based panels such as oriented strand board (OSB), particleboard, chipboard (CH), plywood panel, and medium-density fiberboard (MDF) are widely used for furniture manufacturing and building products. They have numerous environmental benefits. However, they also have some environmental issues. Wood composites bonded with urea-formaldehyde (UF)-type

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adhesives have been identified as a significant source of indoor formaldehyde emissions (Salem et al. 2012; da Silva et al. 2017). Meanwhile, the wood itself has been shown to generate significant levels of formaldehyde when exposed to certain conditions during the composite panel manufacturing process (Böhm et al. 2012).

Binderless boards based on press cake and straw residues from coriander (*Coriandrum sativum*), manufactured through thermopressing (or hot pressing), and chemical adhesive free, have recently been developed (Uitterhaegen et al. 2016, 2017a, 2017b). Firstly, the press cake originating from the thermo-mechanical pressing of coriander fruits is useful for the manufacturing of renewable particleboards revealing a low carbon impact and having an interesting cost to performance ratio (Uitterhaegen et al. 2016, 2017b). The self-bonding ability of the press cake was principally due to its protein fraction whose thermoplastic behavior and adhesive capacity were revealed during the extraction of oil from fruits inside the twin-screw extruder-press owing to the mechanical shear applied to the solid material (Uitterhaegen et al. 2016, 2017b). The VOC emissions from such particleboards were then investigated, and these emissions mainly resulted from the presence of residual essential oil from the coriander fruits inside the manufactured materials (Uitterhaegen et al. 2018). As expected, linalool, which represents 72% of the essential oil from fruits, was obviously the major emitted compound from coriander boards. These renewable materials could, therefore, present a potential interest for the agricultural or building industries, providing benefits in (i) the quality of indoor air, or (ii) the storage of agricultural and food products, due to the bioactivity of such essential oil compounds. Secondly, the coriander straw could also be added to the defatted press cake before molding, thus allowing the manufacture of renewable binderless fiberboards (Uitterhaegen et al. 2017a). This allowed the simultaneous valorization of both the crop residue and the by-product generated by the twin-screw process. On the one hand, the press cake operated, due to its protein fraction, as a natural binder. On the other hand, the straw operated as a mechanical reinforcement. It was also evidenced that a pre-treatment with twin-screw extrusion to refine the straw before mixing it with the press cake enhanced fiber morphology and improved the performance of the fiberboards, especially for resistance to water and, less strongly, for flexural properties. The optimal fiberboard, obtained without adding any chemical adhesive, revealed promising mechanical properties and low sensitivity to water. Thus, it could appear as a sustainable substitute for wood-based materials commercially available, e.g., chipboard (CH), medium-density fiberboard (MDF), and oriented strand board (OSB), which contribute to the depletion of wood resources and can cause a reduction in the indoor air quality. However, the optimized coriander-based fiberboard made from cake and extrusion-refined straw was not yet characterized for its VOC emissions. Conducting such analyses is of great interest to facilitate its future entry into the market.

For the characterization of VOC emissions from building materials, to strengthen skills in risk assessment and to develop low emission products, several methods are currently used (Marć et al. 2012), including emission test chambers (Liang et al. 2016), micro-chambers (Kang et al. 2012), and the Field and Laboratory Emission Cell (Nicolle et al. 2009). They can be coupled to online analytical techniques such as PTR-MS (Majchrzak et al. 2018) and SIFT-MS (Vitola Pasetto et al. 2019), but most often a preconcentration technique is applied using a sorbent cartridge filled with sorbent(s) or a solid-phase microextraction (SPME) fiber. Thermodesorption of VOCs is then conducted, and the emitted compounds are after analyzed by gas chromatography (GC) coupled with mass spectrometry (MS) and/or flame ionization detection (FID) (Martinez et al. 2014; Gross et al. 2017; Jiang et al. 2017; Uitterhaegen et al. 2018). For carbonyl compounds, a derivatization of DNPH (dinitrophenylhydrazine) cartridges followed by HPLC-UV analysis is usually conducted (Huang et al. 2016; Harb et al. 2018). The experimental measurements are commonly conducted in specific test conditions (Salem et al. 2012; Que et al., 2013; Son et al. 2013). Some empirical predictive models and theoretical models have also been developed (Liu et al. 2013; Xiong et al. 2013; Plaisance et al. 2014; Liu et al. 2015).

This work aims to compare three materials through their physical properties and VOC emissions, in particular two wood-based commercial panels, one medium-density fiberboard (MDF) and a chipboard (CH), and a self-bonded fiberboard resulting from a coriander biorefinery and composed of a mixture of deoiled cake as natural binder and extrusion-refined straw as mechanical reinforcement (COR). VOC emissions were determined by the use of a test chamber to monitor the temporal emissions of VOCs and to investigate the correlation between emissions and temperature. The work concludes on the relevance of this new type of material as opposed to commercial materials in relation to their usage properties and VOC emissions.

Materials and methods

Materials

Two different wood-based commercial panels were tested in this study: a medium-density fiberboard (MDF) and a chipboard (CH). They were obtained from Mr. Bricolage (Tarbes, France). Traditionally, MDF panels are made from wood waste, which is first shredded into chips and then undergoes a steam pre-treatment for enhanced properties. A formaldehyde-based synthetic resin is then added before drying, hot pressing, cooling, conditioning, and cutting to the

final size. MDF boards used in this study were 800 mm long, 400 mm wide, and 10 mm thick.

Chipboards are made of waste from the woodworking industry and include sawdust and thinning wood. These different fragmented wood materials are mixed with a formaldehyde-based synthetic resin before being hot pressed, cut, and sanded. Chipboards used in this study were 800 mm long, 400 mm wide, and 10 mm thick.

A binderless fiberboard resulting from a coriander biorefinery (COR) was also tested. This experimental binderless material was largely described in a previous study (Uitterhaegen et al. 2017a). It was made from an extrusion-refined coriander straw and a deoiled coriander press cake, thus presenting a fully renewable material and at the same time ensuring the valorization of a crop residue and a process by-product. The protein-based press cake acts as a natural binder inside the board owing to the thermoplastic behavior of its protein fraction during thermopressing. In parallel, lignocellulosic fibers from the straw act as a reinforcement. Immediately after hot pressing, 150 mm × 150 mm square boards were subjected to an additional heat treatment (10 min, 200 °C) in order to improve their water resistance (Uitterhaegen et al. 2017a). They were then wrapped inside aluminum foil and sealed in polyethylene plastic bags until testing.

Density of materials

The three materials used in this study were first equilibrated in a climatic chamber (60% RH, 25 °C) for 3 weeks in order to assess their properties using equilibrated materials. Thickness and mean apparent density were determined from four 30-mm-wide test specimens. Their thickness was measured at three points and their length at two points, with a 0.01-mm resolution digital sliding caliper. Thickness and length mean values were recorded to calculate the specimen volume, and test specimens were all weighed to calculate their density. The thickness (t) and mean apparent density (d) of materials were the mean values of measurements made on the four test specimens.

Bending properties

The four 30-mm-wide test specimens used for density measurement were also used to determine their flexural properties according to the ISO 16978:2003 standard (ISO 2003a). Bending tests were performed using an Instron 33R4204 (USA) universal testing machine fitted with a 500-N load cell, and the 3-point bending technique. The test speed was 2 mm min ^{1} with 80-mm grip separation. The load was applied equidistant from the two supports, and the loading direction was perpendicular to the upper face of the test specimen. Properties covered breaking load (F), flexural strength at break (σ_f), and elastic modulus (E_f). All determinations were carried out four times, i.e., for each of the four test specimens.

Shore D surface hardness

The Shore D surface hardness of the materials was assessed using a Bareiss (Germany) durometer according to the ISO 868:2003 standard (ISO 2003b). The indentation direction was perpendicular to the upper face of the fiberboard. All determinations were carried out 48 times for each fiberboard (24 times for each board side).

Thickness swelling and water absorption

Four 50 mm × 50-mm samples were used to determine thickness swelling (TS) and water absorption (WA) of the materials. To evaluate these parameters, the samples were submerged in distilled water at 25 °C for 24 h. TS was determined according to the ISO 16983:2003 standard (ISO 2003c) and thickness of each sample was measured at four points, midway along each side 10 mm from the edge, before and after soaking in distilled water. Each sample was also weighed to an accuracy of 0.01 g to determine WA values.

Morphological characterizations of materials

Firstly, the three materials were observed using a Nikon SMZ1500 (Japan) binocular magnifier. Then, the tomography technique was used to analyze them. A RX Solutions EasyTom (France) 3D X-ray laboratory tomography device was used to generate the volume images. A 93-kV tension and a 282-μA intensity were the acquisition parameters chosen. The 70 mm × 70-mm specimens were positioned on a rotation stage. At each angular step of 0.25°, 1440 projections of transmitted X-ray intensity field were recorded through an X-ray detector using a flat panel detector (1920 × 1536 pixels). For each projection, ten different images were recorded at the same angular position, and they were then averaged. Using a filtered back-projection algorithm, it was then possible to reconstruct a volume image from all radiographies, reflecting the variations of the linear attenuation coefficient in the sample. Local differences in density inside the studied material resulted in a specific distribution of gray levels in the 3D images. The microstructure of the studied material was thus represented in three dimensions. The same 55-μm voxel size was used to reconstruct each volume image. The Scilab (France) software was then used to determine the porosity rates from the tomographic slices.

Evaluation of VOC emissions

Quantitative determination of the VOC emissions was conducted by the use of dynamic headspace analysis coupled to

gas chromatography, as described by Uitterhaegen et al. (2018) and according to the ISO 16000-6:2011 and ISO 16000-10:2006 standards (ISO 2006a, 2006b, 2011). Table 1 lists the test conditions. The cell was washed with a basic soap, rinsed with tap water, and then with Milli-Q water. It was dried, closed, and flushed with air for 24 h. Then, a cell blank was made. The material was subsequently installed in the cell. After 24-h stabilization, air was sampled at days 1, 2, 3, and 28 using Tenax TA® tubes and DNPH cartridges. Area-specific emission rates (SER) were calculated according to Eq. (1).

$$\mathrm{SER} = \frac{(m - m_0)}{V} \cdot \frac{q}{S} \quad (1)$$

SER	area-specific emission rate (μg m $^{-2}$ h $^{-1}$)
m	mass of VOCs in the sorbent tube/cartridge (μg)
m_0	reference value of the glass cell (μg)
V	sampled volume (m^3)
q	air flow (m^3 h $^{-1}$)
S	sample surface (m^2)

Analytical conditions

VOCs were collected using sorbent tubes packed with Tenax TA® (300 mg; 60–80 mesh; TERA environnement, France). They were subsequently thermodesorbed and analyzed using a gas chromatograph (Perkin Elmer auto system XL, USA) with a flame ionization detector (FID) and equipped with an automated desorption device (TurboMatrix, ATD, PE). Analytical conditions were according to those reported by Uitterhaegen et al. (2018).

Carbonyl compounds were sampled on a dinitrophenylhydrazine (DNPH) silica-gel cartridge (350 mg; 60/100 mesh; Supelco, USA). They were extracted with 5 mL of acetonitrile. Analyses were carried out on an Ultimate 3000 high-performance liquid chromatography system (UHPLC+, ThermoFisher Scientific, USA) equipped with a diode array detector. Separation was achieved using a Supelcosil LC-18 column (25 cm, 4.6 mm, 5 μm) (Supelco, USA) at a constant flow rate of 1.5 mL min $^{-1}$, using a water-acetonitrile gradient. For the first 9 min, the acetonitrile content was 70%, and then it increased to 100% within 8 min. Subsequently, the acetonitrile content was decreased to 70% which was held for 11 min. The column was thermostated at 30 °C. The sample injection volume was 20 μL. DNPH derivatives of the target substances were quantified at 350 nm.

Quality control and quality assurance

Quantification was based on external calibration curves obtained from liquid standards. Stock solutions and daughter solutions were prepared from commercial standards: α-pinene (> 99%, Sigma), toluene (> 99%, Carlo Erba), cyclohexane (> 99.5%, Sigma), and linalool (> 99%, Fluka) by dilution in methanol. Five to 100 μL of these solutions were then injected on Tenax TA® cartridges. The latter were swept with nitrogen to remove the solvent (about 15 min at a flow rate of 70 mL min $^{-1}$) and analyzed by GC-FID under the conditions mentioned in the "Analytical conditions" section. Monoterpenes were quantified using the α-pinene response coefficient. Other compounds were quantified using the theory of effective carbon number (Jorgensen et al. 1990; Dettmer-Wilde and Engewald 2014).

Commercial hydrazone standard mixing solutions (Carb Carbonyl DNPH Mix 1 and Carb Method 1004 DNPH Mix 2; Supelco, Sigma-Aldrich) were used as standard solutions to study formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, crotonaldehyde, methacrolein, 2-butanone, butyraldehyde, benzaldehyde, pentanal, *m*-tolualdehyde, and hexanal. For the calibration curves, standard solutions were diluted in acetonitrile to create concentrations ranging from

Table 1 Test conditions used for the quantitative determination of the VOC emissions

Test conditions	
Sample area (m^2)	0.0254
Volume (L)	0.580
Loading factor (area of sample/volume, m^2/m^3)	43.79
Air change rate (h $^{-1}$)	31
Air supply (mL/min)	300
Equilibration time	Sampling after 24 h
Temperature	23 ± 1 °C/31 ± 1 °C/36 ± 1 °C
Humidity	50 ± 1%
Inlet	High purity air
Sampling flow (total volume sampling)	VOCs, 195 mL$_N$/min (5.85 L)
	Aldehydes, 210 mL$_N$/min (31.5 L)

0.18 to 12 μg mL ^{1} and analyzed by HPLC-DAD. The calibration curves used were linear over the concentration ranges.

The sampled volumes were selected in such a way as to optimize the signal/threshold ratio without exceeding the breakthrough volumes of each compound (Simon et al. 1995). The limit of detection (LOD) and limit of quantification (LOQ), defined as the mass corresponding to a signal-to-noise ratio of three and ten, respectively, were estimated from the chromatogram of standards at the lowest calibration level used for each compound. Limit of detection (LOD SER) and limit of quantification (LOQ SER) are also presented for emission rates (Table 2). Qualitative HPLC analysis was made by injection of authentic standards of the compounds through the procedure described in the "Analytical conditions" section. Qualitative GC-FID analysis was made via a SPME sampling analyzed by GC-MS. The retention indices were calculated by the use of a series of *n*-alkanes (C6-C16). Further identification was carried out through comparison with library mass spectra (NIST Version 2.0) (Uitterhaegen et al. 2018).

All experiments were conducted in duplicate. Reproducibility was assessed from the study of 5 samples of each material at day 1 and at 23 °C.

Results and discussion

Physical characteristics of materials

The photographs at different magnifications of the three tested materials are presented in Table 3. These images show the relatively smooth surface of the coriander panels, comparable to that of commercial MDF panels. However, due to the high temperature applied during hot pressing of the COR panels, some dark discoloration can be observed in certain areas. Even though this mainly results from the relatively small dimensions of the manufactured panels, it should be taken into account when considering these materials for commercial purposes such as furniture.

Because the internal porosity of the materials could possibly influence their VOC emissions, the latter was also estimated from the tomography radiographies. Porosity was 4.4 10 ^{4}% (i.e., negligible) for MDF, 5.4% for CH, and 1.2% for COR. Porosity was therefore intermediate for COR. Conversely, it was significantly higher for CH, and this result can be explained by the rather coarse size of wood waste particles traditionally used in the manufacture of chipboards in industry. Their stacking leaves voids between them that the added synthetic resin cannot fully fill.

When comparing the materials in terms of bending properties, COR is the panel that revealed the highest value for flexural strength at break, i.e., 29.1 MPa instead of 20.7 MPa for MDF and 10.2 MPa for CH (Table 4). The same tendency was also observed for the elastic moduli. The coriander-based fiberboard was thus the most resistant panel in bending. However, it has to be mentioned here that the three tested materials did not have the same thickness, COR being much thinner than the two others (5.5-mm thickness instead of around 10 mm for MDF and CH).

In real usage conditions, for having an equivalent breaking load (i.e., 534 N to compare with MDF, and 257 N to compare with CH) and from the flexural strength at break values in Table 4, COR would be thinner than the two tested commercial materials: around 8.6 mm instead of 10.2 mm (i.e., − 16%) and around 5.9 mm instead of 10.0 mm (i.e., − 41%), respectively. Therefore, even if COR shows a much higher density than CH, its reduced thickness would result in roughly the same weight per unit surface of material. On the contrary, the 8.6-mm-thick COR material would be heavier than the MDF material (+ 71% for the same material surface).

The higher density for COR resulted from the absence of synthetic resin, which required the use of hot pressing at high temperature and pressure to mobilize the protein-based binder from coriander cake during molding (Tajuddin et al. 2016). Such density contributed to the highest Shore D surface hardness value for COR. In parallel, the water resistance of COR after 24-h immersion was relatively good, showing a value of 24% for both TS and WA. In fact, WA was minimal for COR, and its thickness swelling value was close to the best one (i.e., 20% for MDF). Such good results for COR boards can be attributed to the additional heat treatment (10 min, 200 °C) conducted after hot pressing (Uitterhaegen et al. 2017a).

Looking at the physical properties of COR, the latter thus compare quite well to the two commercial wood-based materials tested in this study, i.e., MDF and CH. According to ISO 16895-2:2010 standard, the COR material complies with international ISO requirements for general purpose medium density fiberboard (MDF), as well as furniture grade and load-bearing MDF for use in dry conditions. When looking at the ISO 16895-1:2008 standard, this would allow it to be used for carcasses, furniture, cabinets, domestic flooring, shelving, and general construction.

Emission pattern of materials

The compounds that were identified and quantified in the emissions of the materials were essentially terpenic and carbonyl compounds, mainly aldehydes (Table 5). The main VOCs emitted from MDF and CH at 23 °C and 50% RH were formaldehyde and linalool, whereas COR emitted mainly acetaldehyde and linalool. Acetaldehyde, formaldehyde, acetone, pentanal, and hexanal were carbonyl compounds involved in the emissions from MDF and CH but only acetaldehyde and nonanal were present in COR with values superior to the limits of quantification. It should be noted that acetaldehyde and linalool were the only two VOCs present at levels

Table 2 Standard curves, linearity ranges, and limits of detection/quantification for all analyzed compounds, i.e., carbonyl and monoterpenic compounds

	Y = ax + b	R^2	Linear range (ng)	LOD (ng)	LOQ (ng)	LOD SER (µg m $^{-2}$ h $^{-1}$)	LOQ SER (µg m $^{-2}$ h $^{-1}$)
Formaldehyde	$Y = 1.5313\,x - 0.2504$	0.995	0.28 to 116.7	0.08	0.28	0.14 [(a)]	0.46 [(a)]
Acetaldehyde	$Y = 1.4603\,x - 0.2039$	0.988	0.33 to 52.3	0.10	0.33	0.22 [(a)]	0.75 [(a)]
Acrolein	$Y = 1.3275\,x - 0.082$	0.999	0.40 to 38.6	0.12	0.40	0.33 [(a)]	1.09 [(a)]
Acetone	$Y = 1.078\,x + 0.0427$	0.991	0.49 to 35.6	0.15	0.49	0.41 [(a)]	1 36 [(a)]
Propionaldehyde	$Y = 1.2637\,x + 0.0183$	0.983	0.44 to 35.1	0.13	0.44	0.36 [(a)]	1 21 [(a)]
Crotonaldehyde	$Y = 1.0941\,x - 0.1051$	0.999	0.63 to 32.8	0.19	0.63	0.60 [(a)]	2.01 [(a)]
Methacrolein	$Y = 1.3508\,x - 0.1605$	0.998	0.52 to 33.4	0.16	0.52	0.50 [(a)]	1.66 [(a)]
2 Butanone							
Butyraldehyde	$Y = 1.2928\,x + 0.0854$	0.997	0.58 to 25.0	0.17	0.58	0.57 [(a)]	1.90 [(a)]
Benzaldehyde	$Y = 0.3015\,x + 0.0088$	0.999	2.32 to 25.1	0.70	2.32	2.96 [(a)]	9.86 [(a)]
Valeraldehyde	$Y = 0.8876\,x - 0.0415$	0.998	1.08 to 35.3	0.33	1.08	1.20 [(a)]	3.98 [(a)]
m Tolualdehyde	$Y = 0.8797\,x - 0.0259$	0.999	1.09 to 22.7	0.33	1.09	1.49 [(a)]	4.96 [(a)]
Hexaldehyde	$Y = 1.0951\,x - 0.0495$	0.999	1.18 to 25.3	0.36	1.18	1.45 [(a)]	4.83 [(a)]
Linalool	$Y = 643.6\,x + 14{,}389$	0.992	0.4 to 1200	0.16	0.41	0.02 [(b)]	0.05 [(b)]
α Pinene	$Y = 771.2\,x - 11.8$	0.998	0.4 to 1100	0.16	0.41	0.02 [(b)]	0.05 [(b)]
Toluene	$Y = 561.2\,x - 1587.9$	0.972	0.6 to 525	0.16	0.57	0.02 [(b)]	0.07 [(b)]
Cyclohexane	$Y = 400.1\,x + 496.1$	0.993	0.5 to 700	0.16	0.50	0.02 [(b)]	0.06 [(b)]

[(a)] For a sampling volume of 31.5 L (DNPH)

[(b)] For a sampling volume of 5.85 L (Tenax TA)

Table 3 Photographs of the three tested materials at different magnifications

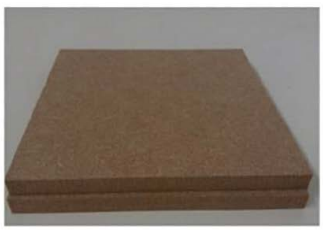
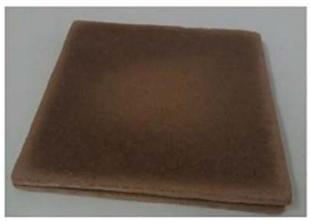
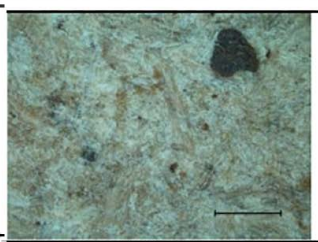
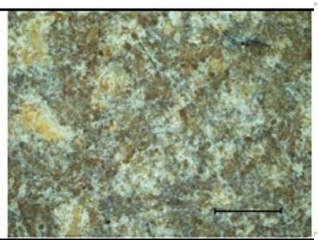
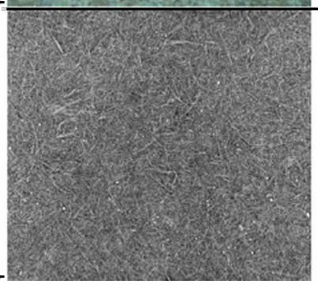
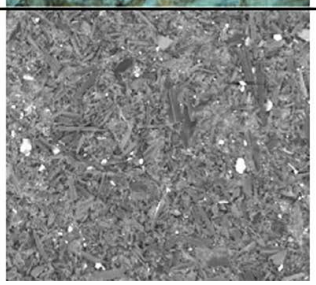
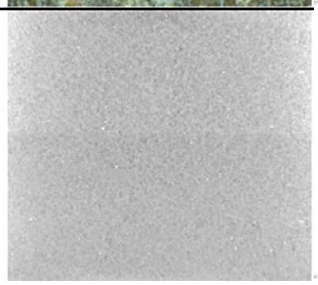
Tested material	MDF	CH	COR
×1 (squares of 10 cm side)			
Binocular magnifier (⊢—⊣ 1 mm)			
3D X-ray tomograph (resolution of 55 µm per pixel, the material on the photograph corresponding to a square of 70 mm side)			

Table 4 Thickness, density, bending properties, and water sensitivity of the three tested materials

Tested material	MDF	CH	COR*
t (mm)	10.16 ± 0.05	10.03 ± 0.01	5.53 ± 0.19
d (kg/m^3)	589 ± 4	710 ± 7	1195 ± 13
F (N)	534.3 ± 51.8	257.0 ± 37.6	222.5 ± 27.9
σ_f (MPa)	20.7 ± 2.1	10 2 ± 1.5	29.1 ± 3.7
E_f (GPa)	1.5 ± 0.1	1.4 ± 0.1	3.9 ± 0.4
Shore D (°)	63.0 ± 3.5	65 5 ± 4.1	81.1 ± 1.3
TS (%)	20 ± 1	25 ± 1	24 ± 4
WA (%)	51 ± 5	58 ± 3	24 ± 2

*Uitterhaegen et al. 2017a

above the quantification limits common to the three materials studied.

Among the three panels, CH showed the highest emission rate of carbonyl compounds and terpenic compounds with 145 μg m $^{-2}$ h $^{-1}$ and 184 μg m $^{-2}$ h $^{-1}$, respectively. Total emissions from MDF and COR were approximately 80 μg m $^{-2}$ h $^{-1}$ with different carbonyl/terpenic compounds distributions, i.e., 85/15 for MDF and 50/50 for COR. Thus, despite an almost non-existent internal porosity, MDF revealed significant VOC emissions. On the contrary, although having a significantly higher internal porosity, COR did not emit more VOCs, indicating that this morphological parameter does not solely explain the emission of VOCs. The chemicals that are present inside these different materials also need to be taken into account. COR was the only material for which formaldehyde emissions were below LOD. In contrast, the coriander-based board was the main emitter of acetaldehyde, which accounts for about 50% of the material's emissions, with an emission rate of 31 μg m $^{-2}$ h $^{-1}$. The maximum emission rate in linalool was measured for chipboard and reached 136 μg m $^{-2}$ h $^{-1}$.

In a previous study, it was demonstrated that the predominance of linalool and presence of camphor in the VOC emissions from COR is in accordance with the obtained composition of the essential oil from coriander fruits, where it represented 72%, while camphor was also identified as a significant compound at 5% of the essential oil (Uitterhaegen et al. 2018). An emission rate of 125 μg m $^{-2}$ h $^{-1}$ (25 °C and 50% RH) was obtained for linalool in that previous study whereas it is only 23 μg m $^{-2}$ h $^{-1}$ for this study, i.e., five times lower. The reason is that different coriander-based materials were used between both studies. In the first one, it was a particleboard obtained from the only coriander press cake whereas the material from the present study was a fiberboard consisting of a mixture of press cake and extrusion-refined coriander straw. In addition, the panel manufacturing process, i.e., the applied conditions for hot pressing, were different between the two studies. The emission profiles of the raw materials, i.e., of the coriander press cake and straw, were also examined and showed that

Table 5 Area specific emission rates (μg m $^{-2}$ h $^{-1}$) at day 1 (23 ± 1 °C and 50 ± 1% RH)

	MDF	CH	COR
Carbonyl compounds			
Formaldehyde	42.09	83.87	< 0.14
Acetaldehyde	2.27	6.29	31.39
Acrolein	< 0.3	< 0.3	< 0.3
Acetone	2.68	34.26	< 0.4
Propionaldehyde	< 0.36	0.78	< 0.36
Crotonaldehyde	n.d.	n.d.	n.d.
Methacrolein	n.d.	n.d.	n.d.
2 Butanone/butyraldehyde	< 0.57	1.23	< 0.57
Benzaldehyde	< 2.96	6.41	< 2.96
Pentanal	2.59	1.20	< 1.20
m Tolualdehyde	n.d.	n.d.	n.d.
Hexanal	10.02	13.16	< 1.45
Heptanal	10.35	n.d.	< 0.10
Octanal	< 0.10	< 0.10	n.d.
Nonanal	< 0.10	< 0.10	4.06
Decanal	< 0.10	< 0.10	< 0.10
Sum	74.2	145.6	34.9
Terpenic compounds			
α Pinene	< 0.02	11.78	< 0.02
Camphene	< 0.02	< 0.02	< 0.02
β Pinene	< 0.02	< 0.02	< 0.02
Δ3 Carene	< 0.02	< 0.02	< 0.02
Limonene	< 0.02	< 0.02	< 0.02
p Cymene	< 0.02	< 0.02	4.41
Linalool	12.25	135.75	23.16
Camphor	< 0.02	36.97	9.11
Borneol	< 0.02	0.04	< 0.02
α Terpineol	n.d.	n.d.	< 0.02
Longifolene	< 0.02	< 0.02	n.d.
α Muurolene	< 0.02	< 0.02	n.d.
β Copaene	< 0.02	< 0.02	n.d.
Trans calamenene	< 0.02	< 0.02	n.d.
Sum	12.4	184.6	36.8
Other compounds			
Toluene	< 0.07	< 0.07	< 0.02
Cyclohexane	n.d.	n.d.	< 0.02

n.d. not detected for any temperature (23 °C, 31 °C, or 36 °C)

linalool was the main emitted compound at 31 °C and 50% RH, with an emission rate of 9 and 12 μg g_{DW} $^{-1}$ h $^{-1}$ for the press cake and the straw, respectively. Further, acetaldehyde was also detected and quantified with emissions of 0.5 and 0.4 μg g_{DW} $^{-1}$ h $^{-1}$, respectively. However, no trace of formaldehyde was detected in the cake or the straw, which explains why it was also not detected in the coriander panel (COR).

The manufacturing process of commercial engineered wood products typically involves the use of synthetic resins such as urea-formaldehyde (UF) resins for bonding of the panels. When such chemical binders are applied, the resulting materials inevitably show significant emissions of non-terpenoid VOCs, notably formaldehyde. A study comparing the emission profiles of lumber products (i.e., wood materials without any synthetic resins) and resin-bonded wood materials such as PB and MDF, found that terpenes made up more than 90% of the total VOC emissions for lumber materials, while this was less than 30% for the glued materials (Son et al. 2013).

Aldehydes have been detected not only from composite wood products (Yrieix et al. 2010; Que et al., 2013; Cheng et al. 2015) but also from lumber (Kirkeskov et al. 2009; Böhm et al. 2012) and during the manufacture of composite panels (Wang and Gardner, 1999; He et al. 2012). Formaldehyde is the most studied aldehyde because it is one of the most ubiquitous and priority indoor pollutants. Lumber products emit small amounts of formaldehyde while MDF and PB which are made with urea-formaldehyde (UF) resin (particleboard, plywood, MDF, and paneling) or with phenol-formaldehyde (PF) resin (softwood plywood, OSB) (Kelly et al. 1999) are the strongest emitters. The detected formaldehyde is the result of degradation/hydrolysis of the bonding resin. In addition, the hot pressing of wood, particularly in the production of wood composites, can sometimes generate significant "native" (i.e., wood-based) formaldehyde emissions, even in the absence of adhesives (Birkeland et al. 2010).

Hexanal is also known to be emitted from wood-based panels such as particleboard, at relatively high concentrations (Baumann et al. 2000; Jiang et al. 2017). For example, Baumann et al. (2000) found that hexanal was the most abundant VOC emitted from particleboards made from different tree species (e.g., hardwood, pines, and Douglas-fir), followed by pentanal, and the total concentration of these two aldehydes typically accounted for more than half of the measured total volatile organic compounds. It is generally believed that hexanal and other straight-chain aldehydes in wood-based panels are the oxidation/degradation products of wood components (e.g., fatty acids).

Temporal evolution of emissions

The evolution of the VOC emission rate is shown in Fig. 1 for each of the materials for day 1, day 2, day 3, and day 28. For most compounds, there is a decrease in their emission as a function of exposure time. For example, the emissions of acetone and linalool decreased by about 73% and 99%, respectively, for CH after 28 days. In the same way, emissions of acetaldehyde and camphor decreased by 85% and 73% after 28 days for COR. In addition, although acetaldehyde is a toxic compound (classified as a group 1 carcinogen by the International Agency for Research on Cancer), it was only emitted at the beginning of the service life of the coriander-based fiberboard (COR), and its emission rate became negligible beyond 1 month. The variations were less important for pentanal and hexanal from the MDF material, with decreases of 17% and 10%, respectively. Looking specifically at formaldehyde emissions, the latter remained quite stable over time for MDF and CH. A 2% variation was measured for MDF, and a 15% variation for CH. Formaldehyde remained the predominant emitted compound at day 28 for these two commercial materials. It has to be noted that the specific emission rate (SER) of formaldehyde was below the detection limit (0.14 $\mu g\ m^{-2}\ h^{-1}$) for COR regardless of the sampling day. On the contrary, it was much higher for the two commercial wood-based materials: 78 $\mu g\ m^{-2}\ h^{-1}$ for CH, and 42 $\mu g\ m^{-2}\ h^{-1}$ for MDF, as an average of the first 3 days.

The total emission rates, calculated from an average of quantified SER for days 1 to 3 (D 1–3), and for day 28 (D 28), were respectively 292 and 76 $\mu g\ m^{-2}\ h^{-1}$ for CH, 56 and 5 $\mu g\ m^{-2}\ h^{-1}$ for COR, and 71 and 51 $\mu g\ m^{-2}\ h^{-1}$ for MDF. These variations represented a decrease of 74%, 91%, and 28%, respectively, for the three tested materials.

Chipboard was thus the largest source of VOC emissions, with emission rates about 5 times higher than those of MDF and COR. At day 28, the emissions for CH were of the same order of magnitude as those obtained in the first days for the two other tested panels. For MDF, the total emission rate over the first 3 days is of the same order of magnitude as for COR.

Previous studies also mentioned a reduction of VOC emissions from different types of materials over time. For example, Yrieix et al. (2010) observed an average emission reduction level of 40% for PB after 28 days compared to the initial emissions of α-pinene, β-pinene, pentanal, and hexanal. The decrease was estimated at 62% for acetaldehyde while it was only 6% for formaldehyde. Son et al. (2013) studied specifically the emissions of monoterpenes from PB and MDF materials. β-Pinene and Δ3-carene were the two main monoterpenes identified, and a decrease of a factor of 6 and 3.5, respectively, was observed after 14 days for PB while it was 54 and 3, respectively, for MDF. Lastly, Da Silva et al. (2017) observed a decrease of formaldehyde emissions of a factor of 1.6 after 28 days for an MDF panel.

Impact of temperature on emissions

The increase in temperature of the material environment (at 31 °C or at 36 °C) led not only to an increase in the emission rate of each compound detected at ambient temperature (23 °C) but also to the detection of other compounds which should have been present at a quantity below the detection limit at 23 °C (Fig. 2). An increase of temperature indeed influences the emission rate by decreasing the partition coefficient (K) at the material/air interface and by increasing the

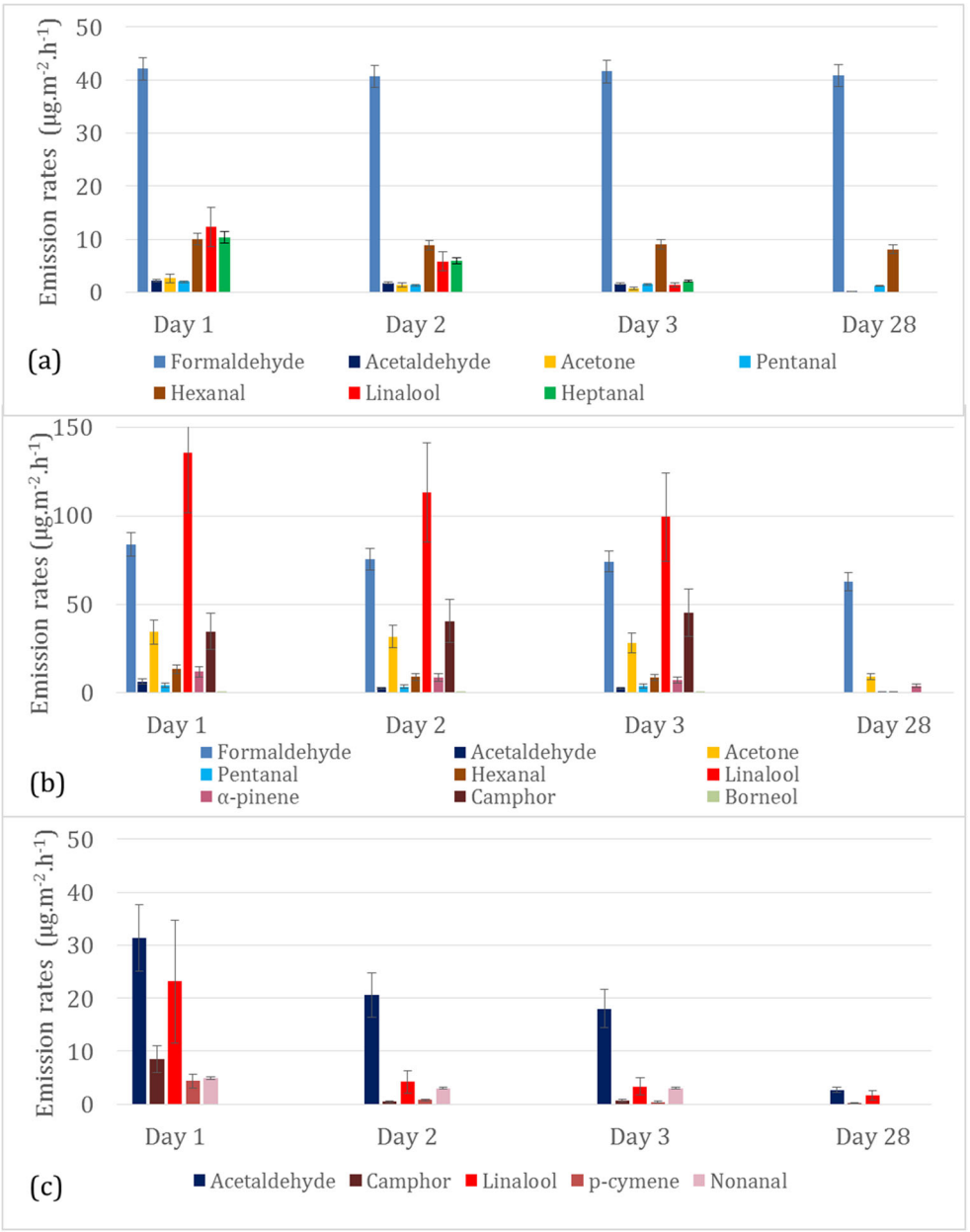


Fig. 1 Specific emission rates measured at day 1, day 2, day 3, and day 28 for the three tested materials (23 ± 1 °C and 50 ± 1% RH): **a** MDF, **b** CH, and **c** COR

diffusion of the compound within the material, as previously observed by Zhang et al. (2007) for PB and MDF emissions. The initial material-phase concentration also determines the emission profile for a given chamber configuration. The temperature dependence of VOC vapor pressures can be expressed through the Antoine equation: $\log P = a + \frac{b}{c+T}$, where P is the vapor pressure of the compound of interest (atm) and T is the absolute temperature (K). a, b, and c are positive parameters with $b = \Delta H_{vap}/R$, where R is the gas constant and ΔH_{vap} is the heat of vaporization.

Thus, toluene, β-pinene, and *o*-cymene had no contribution to the emissions of CH at 23 °C whereas they represented 13% of the total emissions at 36 °C. It was the same for acetone/propionaldehyde/butyraldehyde and for propionaldehyde/octanal/nonanal/camphor, whose emissions at 36 °C contributed to 8% of the total emissions of COR and MDF, respectively. At day 1, the total emission rates measured at 36 °C were approximately 2800 µg m ^{2} h ^{1} for CH, 800 µg m ^{2} h ^{1} for COR, and 2100 µg m ^{2} h ^{1} for MDF, instead of 324, 72, and 81 µg m ^{2} h ^{1} at 23 °C, respectively. At 36 °C, the number of VOCs emitted and detected was 15 for CH, 12 for MDF, and 11 for COR. The proportion of carbonyl compounds relative to terpenoids also increased with temperature in the range of [0.8–2.1] for CH, [1.1–2.5] for COR, and [5.8–10.9] for MDF. The obtained results thus suggest that the emission of carbonyl compounds, aldehydes in particular, is much more sensitive to temperature change as compared to the emission of terpenoids. For COR, whatever

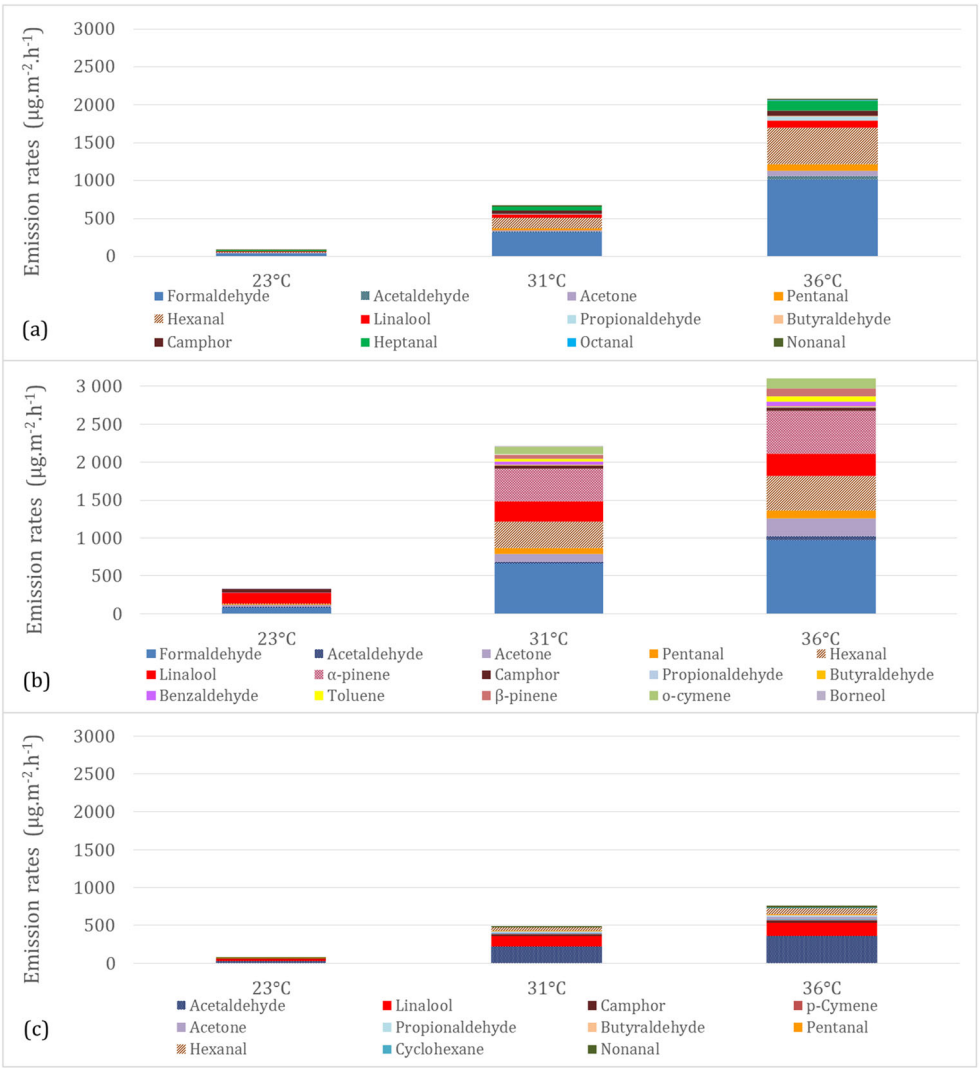


Fig. 2 Impact of temperature on the specific emission rates for the three tested materials (day 1, 50 ± 1% RH): **a** MDF, **b** CH, and **c** COR

the temperature, formaldehyde emissions always remained below the limit of quantification.

Next to the volatiles resulting from the presence of essential oil inside the coriander material (COR), cyclohexane was also detected as a VOC emitted from COR. The origin of this compound lies within the additional deoiling process of the press cake prior to thermopressing, which consists of a cyclohexane solvent extraction. This residual oil extraction was carried out in order to avoid the formation of defects inside the boards during thermopressing, due to the possible expression of residual vegetable oil from cake under high pressure (Uitterhaegen et al. 2017b). The emission of cyclohexane was less than the limit of detection at 23 °C (< 0.02 µg m $^{-2}$ h $^{-1}$), while it was 7.0 ± 1.0 µg m $^{-2}$ h $^{-1}$ at 36 °C and 50% RH.

It is well known that the emission of formaldehyde is especially affected by temperature which enhances the degradation of UF and PF resins inside commercial wood-based materials. The evolution of the emission rate as a function of temperature is presented for the three most representative VOCs, i.e., especially formaldehyde but also acetaldehyde and linalool (Fig. 3).

Conclusion

The fiberboards resulting from a coriander biorefinery and evaluated in this study present performance characteristics that are comparable to those of commercial glued wood panels such as MDF and CH. The VOC emissions of the three materials are mainly composed of carbonyl and terpenic compounds and are temperature and exposure time dependent. Higher temperatures resulted in higher emission rates for the experimental domain (from 23 to 36 °C). The emissions decreased over time, except for formaldehyde. The total emission rate was less important for COR than for MDF and CH with values of 72, 87, and 330 µg m $^{-2}$ h $^{-1}$, respectively, at 23 °C and 50% RH for day 1. Coriander fiberboard emissions are formaldehyde-free (< 0.2 µg m $^{-2}$ h $^{-1}$), which is contrary to

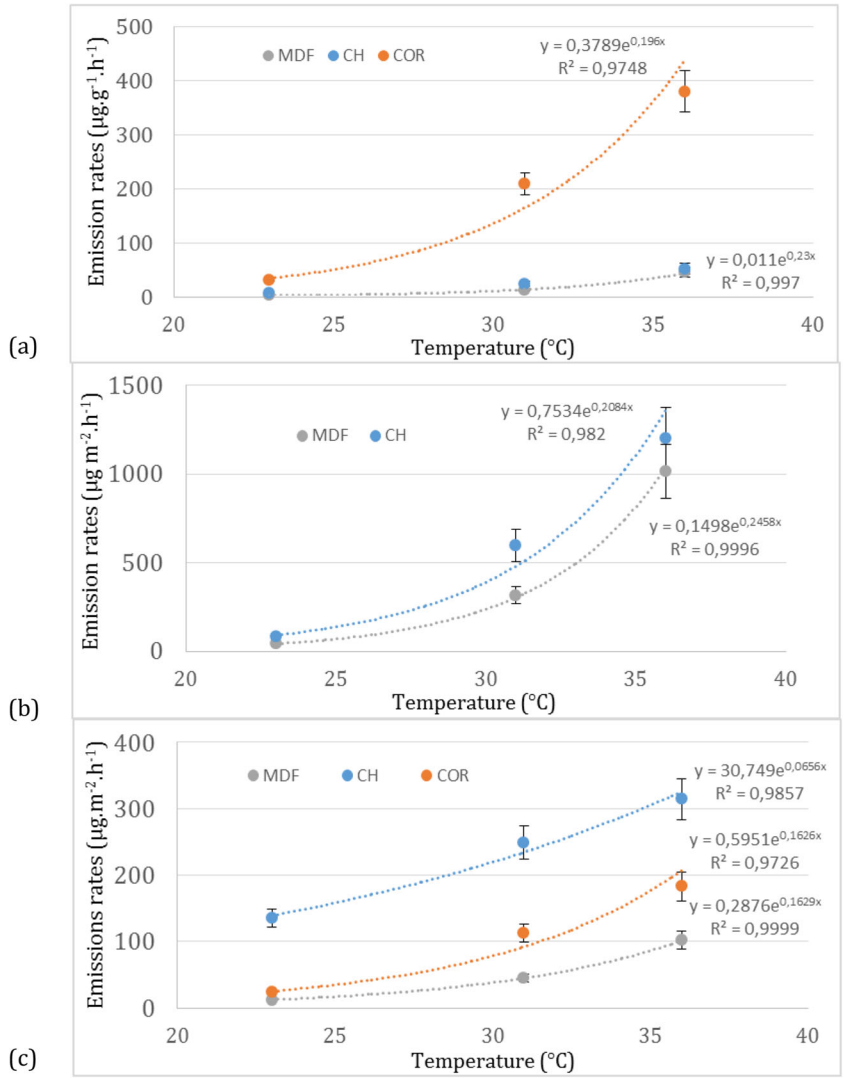


Fig. 3 Evolution of specific emission rates as a function of material temperature and material type for three volatile organic compounds: **a** acetaldehyde, **b** formaldehyde, and **c** linalool

MDF and CH for which the formaldehyde emissions are of 42 and 84 µg m ^{2} h ^{1}. Fiberboards from coriander could thus present a viable and competitive alternative to glued wood panels with a view to sustainability and the reduction of VOC emissions.

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