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Characterization of volatile organic compound emissions from self-bonded boards resulting from a coriander biorefinery

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ABSTRACT

In this study, the VOC emissions from a self-bonded coriander board originating from a coriander biorefinery, were investigated. These emissions mainly result from the presence of essential oil in the manufactured materials. Firstly, an extensive analysis of the essential oil obtained from French coriander fruits showed the presence of linalool as the major component (72%), with an absolute concentration in the essential oil of 412 g/L. A characterization of the enantiomeric distribution of linalool, which is important in terms of its bioactivity, resulted in an enantiomeric excess of (*S*)-linalool of 77%. Further, the presence of this volatile oil in the coriander boards was confirmed through identification of the terpenoid compounds in the VOC emission profile. The area specific emission rate of linalool and camphor was determined at 125 and 25 $\mu g\, m^{-2}\, h^{-1}$, respectively, at 25 °C and 50% RH, while their emission was found to increase by a factor of 3 with an increase in temperature of 10 °C. The renewable self-bonded boards could thus present potentially interesting alternatives for less sustainable materials in the construction or agricultural industry, where they could provide a significant added value in terms of indoor air quality or storage of food and agricultural products.

1. Introduction

Coriandrum sativum L. presents an annual herb belonging to the Apiaceae (Umbelliferae) family that originates from the Near East and Mediterranean area. It is currently mostly produced in India with an annual production of around 500 000 t and next to its main use as a spice, it has found applications in perfumery and cosmetics (Sharma et al., 2014). Coriander fruits are marked by the presence of both a vegetable oil and an essential oil fraction, the former constituting between 20 and 28% of the fruit, while the latter is present in typically less than 1% (Sahib et al., 2013). The vegetable oil contains petroselinic acid as a key component (around 70%), which presents a rather rare positional isomer of oleic acid and shows anti-inflammatory and anti-aging properties, leading to a recently growing interest in coriander vegetable oil from the food, cosmetic and chemical industry (Alaluf et al., 2002, 2005; Uitterhaegen et al., 2016b). Furthermore, coriander vegetable oil has recently been approved as a Novel Food Ingredient (NFI) and can be obtained efficiently and in high quality through thermomechanical pressing using a twin-screw extruder (European Food Safety Authority (EFSA), 2013; Uitterhaegen et al., 2015). The essential oil fraction is defined by a high linalool content, typically ranging between 60 and 80%, while other important compounds include α-pinene, γ-terpinene, camphor, limonene and geranyl acetate (Sahib et al., 2013). The fragrant oil, exhibiting a characteristic odor of linalool, is widely used in perfumery and the cosmetic industry, and in the food and pharmaceutical industry as a flavoring agent and adjuvant (Burdock and Carabin, 2009). Further, coriander essential oil has been shown to exhibit antioxidant and anti-inflammatory activity and inhibit a broad spectrum of micro-organisms (Beikert et al., 2013; Darughe et al., 2012; Silva et al., 2011).

In view of the increasing industrial interest in coriander vegetable oil, by-products from the oil extraction process will become more important in years to come and their valorization will present a key challenge in the light of sustainable processing and industrial viability. Recently, it has been demonstrated that the press cake resulting from the thermomechanical pressing of coriander fruits can be used for the production of renewable and environmentally friendly materials with a high cost-performance ratio (Uitterhaegen et al., 2016a, 2017a, 2017b). In particular, the deoiled press cake was transformed into binderless boards through thermopressing without the addition of any chemical

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adhesives, which are often toxic and can lead to harmful emissions such as formaldehyde. The self-bonding character of the press cake was mainly attributed to the protein fraction (Uitterhaegen et al., 2017b). These materials were proven viable alternatives for current commercial wood-based and resin-bonded materials such as particleboard (PB), medium-density fiberboard (MDF) and oriented strand board (OSB), the use of which will become increasingly unacceptable considering the depletion of wood resources and recent regulations concerning indoor air quality. Next to this, there is an increasing trend involving the addition of essential oils or essential oil compounds as active agents with a view to obtaining functional materials with antimicrobial, pesticidal and/or antioxidant activities (Hashim et al., 2009; Singh and Chittenden, 2010; Yingprasert et al., 2015). With regard to this, coriander essential oil could present an interesting solution for pest management, the control of bacterial and fungal growth in food or agricultural products or in housing and furniture applications, where mold growth implies significant health risks. Furthermore, its inherent presence in the press cake obtained from thermomechanical oil extraction from coriander fruits would designate an important added value for the materials resulting from the thermopressing of the cake. These materials could then show potential for high-value applications such as functional food packaging for prolonged shelf-life, storage of agricultural products or indoor construction materials for enhanced air quality.

This study presents a full characterization of the essential oil obtained from coriander fruits of French origin, with a clear focus on linalool as the key component. Next to this, a comparison with literature data on coriander essential oil is made in order to assess the variability of the oil composition resulting from the culture region and between different harvesting years. Most research studies on coriander essential oil only report the linalool content. However, as chirality presents a key factor in flavor chemistry and as it has been shown that the chirality of linalool determines not only its sensory profile, but also its biological activities (Aprotosoaie et al., 2014), the enantiomeric distribution of linalool in the coriander essential oil was characterized. This report further aims to evaluate the presence and influence of this essential oil in self-bonded boards produced from the press cake of the twin-screw thermomechanical pressing process of coriander fruits. In particular, it presents a critical analysis of the potential of coriander essential oil for providing an added value to the renewable materials resulting from the biorefinery of coriander fruits. Further, a qualitative determination of volatile organic compound (VOC) emissions from the binderless boards allows justification of their use as furniture or building materials with respect to the indoor air quality. The importance of this research is pronounced by the development of a true coriander biorefinery which includes the sustainable production of coriander vegetable oil and extensive valorization of the process by-products. Therefore, these results could present significant industrial value with a view to recent environmental concerns and the current critical pressure towards sustainability and green processing.

2. Materials and methods

2.1. Materials

Coriander fruits were supplied by GSN Semences (Le Houga, France), consisted of the GSN maintenaire variety and were cultivated in the southwestern part of France. The fruit moisture content was determined according to ISO 665:2000 and was 8.56 ± 0.10% (ISO, 2000). The press cake that was used for the purpose of this study resulted from the vegetable oil extraction process from coriander fruits. For this, a Clextral (Firminy, France) BC 21 twin-screw extruder was used with a screw configuration that was optimized for coriander fruits in a previous study (Uitterhaegen et al., 2015). Further deoiling of the press cake was carried out through a solvent extraction of 5 h with a 1 L Soxhlet apparatus and cyclohexane as the extracting solvent.

2.2. Chemicals

n-Butylbenzene (99.8%), (*R/S*)-linalool (> 99.0%), (*R*)-linalool (> 97.5%), camphor (> 96%), linalyl acetate (> 97%), geranyl acetate (> 99%), limonene (> 99%), nonane (> 99.8%), decane (> 99.8%), undecane (> 99.8%), dodecane (> 99.8%), tridecane (> 99.8%), tetradecane (> 99.8%) and pentadecane (> 99.8%) were obtained from Sigma-Aldrich (Saint Louis, MO, USA). γ-Terpinene (> 97%), *p*-cymene (> 99%) and α-pinene (> 98%) were from Extrasynthèse (Genay, France). All solvents used in this study were purchased from Sigma-Aldrich (Saint Louis, MO, USA) or Merck (Darmstadt, Germany) and were of analytical grade.

2.3. Thermopressing

Self-bonded boards consisting solely of 200 g of deoiled coriander press cake were manufactured through thermopressing. For this, a Pinette Emidecau Industries (Chalon-sur-Saône, France) heated hydraulic press with a 400-ton capacity and a 150 mm × 150 mm aluminum mold was used. All raw materials were dried in a ventilated oven at 50 °C to a moisture content between 3 and 4% prior to thermopressing in order to avoid delamination of the panels. The applied operating conditions were optimized in a previous study (Uitterhaegen et al., 2017b) and include an applied pressure of 21.6 MPa, a mold temperature of 205 °C and a molding time of 300 s. All manufactured materials were conditioned in a climatic chamber (25 °C, 60% RH) for four weeks prior to characterization. The dimensions of the test specimens were 130 × 30 × 6 mm. Before testing, all samples were stored in hermetically sealed aluminum foil bags.

2.4. Essential oil extraction

Essential oils were extracted from 200 g of coriander fruits by hydrodistillation in a Clevenger-type apparatus for 5 h. Milled coriander fruits were introduced to a round bottom flask with 2 L of distilled water to recover the essential oil (1:10 ratio). The distillate was extracted with pentane, which was evaporated with N_2. The oil samples were stored in airtight glassware at 4 °C.

2.5. Essential oil analysis

Gas chromatography analyses of the essential oils were performed with a Thermo Scientific (Thermo Fisher Scientific, Waltham, MA, USA) Trace 1300 gas chromatograph equipped with a Restek (Bellefonte, PA, USA) Rtx-5 capillary column (30 m × 0.25 mm; 0.25 μm film thickness). A Thermo Scientific (Thermo Fisher Scientific, Waltham, MA, USA) ISQ QD mass selective (MS) detector was applied for qualitative analysis, while a flame ionization detector (FID) was used for quantitative analysis. The flow of the carrier gas (helium) was 1.5 mL/min. The oven temperature was programmed from 80 °C (hold 3 min) to 220 °C (hold 2 min) at 4 °C/min. The injector and FID temperature was maintained at 250 °C. Temperatures of the quadrupole and the ion source were 150 and 230 °C, respectively. The MS detector was run in electron impact mode with an electron energy of 70 eV. The mass scan ranged from 35 to 300 *m/z*. For each sample, 1 μL was injected under split mode with a 100:1 ratio. Essential oil components were identified by comparison of their retention indices with those from literature and authentic standards available in our laboratory. The retention indices were calculated by the use of a series of *n*-alkanes (C_9–C_{15}) according to Eq. (1). Further identification was carried out through comparison with library mass spectra (NIST Version 2.0).

$$RI = 100\cdot\frac{t_R(i) - t_R(n)}{t_R(n+1) - t_R(n)} + 100\cdot n \quad (1)$$

RI retention index,
$t_R(i)$ retention time of compound i,