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Steam distillation extraction kinetics regression models to predict essential oil yield, composition, and bioactivity of chamomile oil

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ABSTRACT

Chamomile (*Matricaria chamomilla* L.) is one of the most widely spread and used medicinal and essential oil crops in the world. Chamomile essential oil is extracted via steam distillation of the inflorescences (flowers). In this study, distillation time (DT) was found to be a crucial determinant of yield and composition of chamomile essential oil, but not of the antioxidant capacity. Essential oil obtained at 30, 60, 90, 120, 180, 240, 360, 480, 600, and 720 min showed significant increase in oil yield with increasing DT, reaching a maximum of 3.1 g oil per 1000 g of flowers at 720 min. The major compounds that were identified and quantified were anethole, β-farnesene, spathulenol, α-bisabolol oxide B, α-bisabolone oxide A, chamazulene, α-bisabolol oxide A, and spiroether. β-farnesene showed a decrease in content with increasing DT, whereas α-bisabolol oxide A, spiroether, and chamazulene rapidly increased up to 240 min, after which it started to plateau showing negligible change. Anethole content showed a steady decrease over time from approximately 2.4% at 30 min to 0.54% at 720 min. Yields of spathulenol, α-bisabolol oxide B, α-bisabolol oxide A, α-bisabolone oxide A, chamazulene, and spiroether essential oil constituents expressed as g/100 g of dried chamomile inflorescences showed a steady increase that was described well by the Michaelis-Menton model. If higher concentrations of α-bisabolol oxide A and chamazulene, and higher oil yields are desired, chamomile flowers must be steam distilled for 480 min. However, if oil with high β-farnesene concentration is desirable, then chamomile flowers should be distilled for 30 min. Distillation time can be used as a modifier of chamomile essential oil yield and composition. The kinetics regression models developed in this study can be utilized to predict essential oil yield, and composition of chamomile oil.

1. Introduction

Chamomile (*Matricaria chamomilla* L. synonym: *Matricaria recutita*) is a member of the Asteraceae family. Chamomile flowers are most commonly used for making chamomile tea known for its calming effect and the essential oil is used in the pharmaceutical and cosmetic industries (Salamon, 2007; Valussi, 2012; Wheatley, 2005). Chamomile flowers and extracts have been extensively used in traditional medicine in many countries, especially in the Mediterranean region. For example, Bulgarian folk medicine has utilized chamomile flower extract against insomnia, hysteria, gastritis, various cystic formations, headache, stomach pain, as wound epithelialization, gas relieve, antispasmodic, and sweating agent (Stojanov, 1973). The bioactivity of essential oils and their effective use in aromatherapy is largely dependent on the composition of major volatile compounds. Cultivation methods, management practices (Zheljazkov and Astatkie, 2011), abiotic factors (Razmjoo et al., 2008), and post-harvest processing (Shahhoseini et al., 2013) are known to largely affect the yields and composition of essential oils. Planting date and seedling age influenced the content and composition of chamomile (Mohammad et al., 2010) whereas irrigation affected the compositions in chamomile (Pirzad et al., 2006). Post-harvest processing parameters like drying methods (Shahhoseini et al., 2013; Pirbalouti et al., 2013) are known to affect essential oil composition and bioactivity. Four different drying methods showed significant effects on composition in Roman chamomile essential oil (Omidbaigi et al., 2004).

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Distillation parameters like processing of plant material and distillation apparatus, along with the specific method and time may additionally affect the essential oil quality (Hosni et al., 2010). Previously, it was found that the duration of the distillation time (DT) significantly affected essential oil yield and composition in peppermint (*Mentha* × *piperita* L.), lemongrass (*Cymbopogon flexuosus* Steud.), and palmarosa (*Cymbopogon martinii* Roxb.) (Cannon et al., 2013), fennel (*Foeniculum vulgare* Mill.) (Zheljazkov et al., 2013a), Rocky Mountain juniper (*Juniperus scopuloum* L.) (Zheljazkov et al., 2013b), sweet sagewort (*Artemisia annua* L.) (Zheljazkov et al., 2013c), lavender (*Lavandula angustifolia* Mill.) (Zheljazkov et al., 2013d), and oregano (*Origanum vulgare* L.) (Zheljazkov et al., 2012a).

Currently, there is no information about how DT would affect the yield and composition of chamomile. The optimal DT for dried chamomile flowers is also unknown. We hypothesized that by changing the duration of the DT of chamomile flowers, we could obtain essential oil with diverse composition and bioactivity and optimize the DT for maximum oil yield. Such standardizations can be commercially utilized by identifying the best suitable DT for desired compositions of essential oils. Therefore, the objective of this study was to evaluate the effect of DT (steam distillation extraction kinetics) on yield, composition, and antioxidant capacity of chamomile essential oil. Furthermore, this study evaluated the essential oil's antimicrobial activities tested against ten bacterial and fungal species.

2. Material and methods

2.1. Plant material

Bulk certified dried chamomile flowers were obtained from Starwest Botanicals (Rancho Cordova, CA). The country of origin was Egypt.

2.2. Steam distillation and distillation time (DT)

The steam distillation study/extraction kinetics experiment was carried out in 2013 at the University of Wyoming, Sheridan Research and Extension Center using a sample of 200 or 250 g of dried flower. The steam DT investigated in this study were 30, 60, 90, 120, 180, 240, 360, 480, 600, and 720 min. These DTs were based on our preliminary studies and literature reports. Also, this spacing of the DT allowed us to develop extraction kinetics regression models that can be utilized to predict chemical composition and oil yield at any specific DT. All DTs were performed in triplicate in a 2 l steam distillation unit as described previously for peppermint, lemongrass, and palmarosa (Cannon et al., 2013), and for lavender inflorescences (Zheljazkov et al., 2013c). Briefly the apparatus includes a 2 l pear shaped flask filled with water on a hotplate and a 2 l bioflask positioned above. The bioflask contains the flowers. The still head is attached to the top of the bioflask and directs the steam to the condenser that allows the co-distilled steam and oil to simultaneously collect and separate in a collector. This system is analogous to large commercial installations; the collector is similar to the Florentine vessel used in commercial installations.

Steam DT of 30, 60, 90, 120, 180, 240, 360, 480, 600, and 720 min were recorded as the times required from the beginning of distillation (the moment the first drop condenses) to the time when distillation was turned off. The distilled oil was separated from water and collected in glass vials equipped with a Teflon lined screw cap. The oils were weighed on an analytical scale and were stored at −5 °C for gas chromatography (GC) identification. Essential oil yield was calculated as grams of oil per weight (g) of 100 g dried flowers.

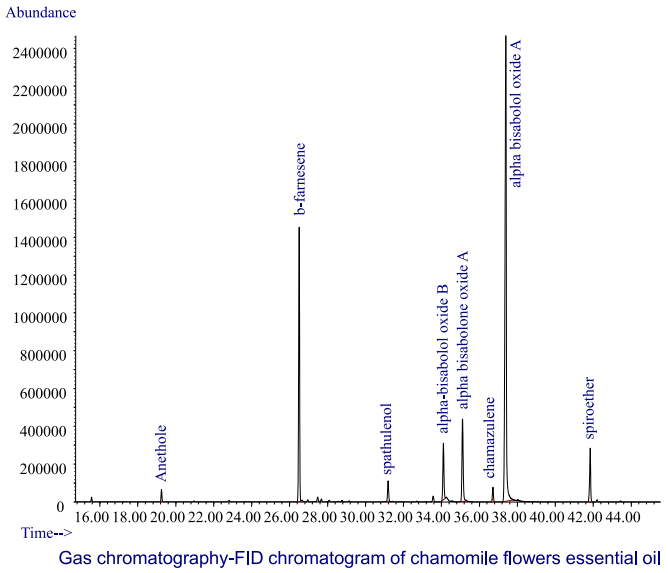

Fig. 1. Gas chromatography-FID chromatogram of chamomile flowers essential oil.

2.3. Gas chromatography–flame ionization detector (FID) quantification of essential oil components

A total of eight constituents were identified and quantified in chamomile flower essential oil (Fig. 1). Oil samples were analyzed by GC-FID on a Agilent CP-3800 GC equipped with a DB-5 fused silica capillary column (30 m × 0.25 mm, with a film thickness of 0.25 μm) operated using the following conditions: injector temperature, 240 °C; column temperature, 60–120 at 3 °C/min, then held at 240 °C at 20 °C/min for 5 min; carrier gas, He; injection volume, 1 μl (split on FID, split ratio 50:1); FID temperature was 300 °C. Compounds anethole, β-farnesene, spathulenol, α-bisabolol oxide B, α-bisabolone oxide A, chamazulene, α-bisabolol oxide A, and spiroether were identified in oil samples by Kovat analysis (Adams, 2007), and comparison of mass spectra with those reported in the NIST mass spectra database. Compounds were quantified by performing area percentage calculations based on the total combined FID area. For example, the area for each reported peak was divided by total integrated area from the FID chromatogram from all reported peaks and multiplied by 100 to arrive at a percentage. The percentage is a peak area percentage relative to all other constituents integrated in the FID chromatogram.

2.4. Antioxidant capacity

The antioxidant capacity of the oil extracts from all DT in three replicates was determined by the oxygen radical absorbance capacity (ORAC) method as described by Huang et al. (2002a,b). Samples of extracted oil were prepared for antioxidant capacity tests by mixing 10 ± 1 mg oil with 1 ml of water and acetone (1:1) with 7% methyl-β-cyclodextrins (w:v). The test was prepared in a 96-well plate by first transferring 25 μl of 74 mM phosphate buffer saline (pH 7.4) to each well. The test sample (25 μl) or Trolox (25 μl) that served as the standard was added to different wells at concentration of 0.2, 0.4, 3.3, 6.5, 10, 13, 25, 50 μg/ml) followed by 150 μl of fluorescein (8.16×10^{-5} mM). The samples were incubated at 37 °C for 10 min, with 3 min of intermittent shaking. After incubation, the reaction was activated by adding 153 mM 2,2′-azobis (2-amidinopropane) hydrochloride (25 μl) to each well. All samples and standards were prepared in 96 well plates and the fluorescence was measured every 1.5 min with a microplate reader set at an excitation and emission wavelength of 485 nm and 520 nm,