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Ultrasound pretreatment impact on *Prangos ferulacea* Lindl. and *Satureja macrosiphonia* Bornm. essential oil extraction and comparing their physicochemical and biological properties

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ABSTRACT

The aim of this study was to investigate the impact of ultrasonic pretreatment (US + HD) of *Prangos ferulacea* Lindl. and *Satureja macrosiphonia* Bornm. leaves prior to hydrodistillation (HD). The two plants differed in their secretory structures, internal and external structures, respectively. Extraction kinetics, chemical composition, physical and biological properties of the essential oils was evaluated. Although applying ultrasound pretreatment resulted in a faster extraction process, no significant difference was observed between the yields. Similar main components in essential oils including ρ-cymene, limonene, (E)-β-ocimene, terpinolene, and 2,3,6-trimethylbenzaldehyde for *P. ferulacea* and *cis*-sabinene hydrate, linalool, borneol, and terpinene-4-ol for *S. macrosiphonia* were observed, regardless of applying sonication or not. However, a selective extraction of limonene (*P. ferulacea*) and linalool (*S. macrosiphonia*) was a direct result of 15 min indirect sonicatin of plant materials prior to hydrodistillation. Ultrasonic pretreatment not only had no adverse effect on biological properties of extracted essential oils but also in case of pre-treated *P. ferulacea* improved antioxidant activity. Generally, this study indicates that ultrasonic pretreatment of plant materials resulted in accelerating the essential oil extraction process and producing an essential oil with superior characteristics.

1. Introduction

Prangos ferulacea Lindl. (Apiaceae family) with the Persian name of "Jashir" is grown wildly on mountainous regions of Iran. There are about 50 species in the genus of *Prangos*, which are mainly distributing in the Mediterranean and Middle East regions (Ghahreman, 1995). Different species of *Prangos* are utilized for their emollient, carminative (Zargari, 1990), tonic, anti-flatulent, anthelmintic, antifungal and antibacterial activities (Bouaoun et al., 2007). In Iran, aerial parts of *P. ferulacea* are used as a fodder, also a flavoring additive in yogurt. Presence of coumarin and its derivatives in different species of *Prangos* has been reported (Tada et al., 2002). Extraction of essential oils has been studied from different parts of several *Prangos* species, e.g., fruits of *P. unchtritzii* Boiss. (Baser et al., 2000), and roots of *P. ferulacea* (Sajjadi et al., 2011).

Satureja macrosiphonia Bornm. is one of the species of genus *Satureja* (Lamiaceae family), which contains over 200 species of aromatic herbs and shrubs. The plants are largely distributed across the Mediterranean region, continental Europe, West Asia, North Africa, the Canary Islands and South America; and about 14 species of genus *Satureja* present in the north, northwest and west regions of Iran (Momtaz and Abdollahi, 2008). Different species of *Satureja* are extensively utilized for their analgesic, antiseptic, antimicrobial, antiviral, antioxidant, as well as vasodilatory activities (Sefidkon et al., 2004). Due to the simplicity of cultivation and eminent ethnomedical activity, and in light of their food and pharmaceutical importance, this genus is used worldwide as herbal beverage, spice, food additive and flavoring agent (Zargari, 1990).

There are different technologies to isolate essential oils from different parts of aromatic plants, such as hydro- or steam-distillation that are amongst conventional and the most frequently used methods. However, conventional methods have shown several drawbacks, including long extraction time, degradation of thermally sensitive components, potential loss of volatile components, and high-energy consumption (Golmakani and Rezaei,

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2008). Therefore, there is a need to investigate for alternative techniques to obviate the aforementioned problems of conventional techniques. Various novel technologies have been introduced for essential oil extraction, such as supercritical fluid extraction (Uquiche et al., 2015), microwave-assisted hydrodistillation (Golmakani and Rezaei, 2008), and ohmic-assisted hydrodistillation (Seidi Damyeh et al., 2015).

In order to improve the extraction yield and shorten the extraction time, sono-assisted extraction has been applied by several researchers (Assami et al., 2012; Périno-Issartier et al., 2013; Kowalski et al., 2015; Morsy, 2015). Pretreatment by ultrasound together with size reduction lead to a rapid and more complete extraction of desired components from the plant materials. Mass transfer intensification, cell disruption, improved solvent penetration and capillary effect, high recovery yield and short extraction time are associated with sonication process (Chemat and Zill-e-Huma Khan, 2011). Due to high intensity of sonication particularly when using sonotrode, degradation of flavors may also be occurred (Vinatoru, 2001).

In the present work, influence of ultrasound pretreatment (US + HD) on essential oil extraction from dried leaves of *P. ferulacea* and *S. macrosiphonia* (possessing internal and external secretory structures, respectively), was examined. In addition to extraction time and yield, chemical composition, physical and biological characteristics of the essential oils, and structural changes of the leaves were investigated.

2. Material and methods

2.1. Plant materials

P. ferulacea leaves were collected from Estahban mountainous regions (Fars province, Iran) in early March 2013. The sample was identified, and voucher specimens have been deposited with the Pharmacognosi and herbarium department (UM256-1), School of Pharmacy, Gilan University of Medical Sciences, Rasht, Iran.

S. macrosiphonia leaves were collected from Khoram Abad (Lorestan province, Iran) in October 2013. The sample was identified, and voucher specimens have been deposited with the Herbarium of the Medicinal Plants and Drugs Research Institute (MPH-1590), Shahid Beheshti University, Tehran, Iran.

Herbs were air dried (25–30 °C) and packaged in high-density polyethylene bags. The bags were stored in a dark and cool place until further extraction and analysis. The plant material were grinded (30-mesh sieve) prior to extraction. The 30–40 mesh size is optimal, whereas smaller particles may become slimy during extraction and create some sort of difficulty (Handa, 2008).

2.2. Extraction procedures

2.2.1. Conventional hydrodistillation(HD)

An all-glass Clevenger apparatus, according to the method proposed by British Pharmacopeia (1980), carried out the conventional hydrodistillation (HD). Thirty grams of dried leaves were soaked in 600 ml distilled water for about 15 min at room temperature. This was followed by 180 min of heating on an electric heating mantle to extract the essential oil. Mixture took almost 45 min to come to a complete boil. After boiling stage, during the first 30 min of the process, amount of essential oil extracted was recorded at 5 min intervals, followed by 15 min intervals, and recording during the last 60 min of extraction was at 30 min intervals. The HD continued until no measurable essential oil was extracted. The yield (%) was evaluated by dividing the volume of essential oil extracted (ml) into the weight of dry leaves (g), multiplied by 100. The essential oil was collected, dried under anhydrous sodium sulphate and stored in an amber airtight vial at −18 °C for further analysis. Each extraction was performed at least three times.

2.2.2. Ultrasonic pretreatment prior to hydrodistillation(US + HD)

Ultrasonic-assisted hydrodistillation (US + HD) was performed in a 4 l Sonorex Degitec Bandelin ultrasonic bath (DT103H, Berlin, Germany) with an all-glass Clevenger apparatus. Thirty grams of dried plant materials were immersed in 600 ml distilled water in a one liter round flask connected to a Clevenger apparatus. The flask was placed in an ultrasonic bath (frequency of 35 kHz and effective power of 160 W based on information given by the manufacturer) and was submitted to sonication for 15 min at 30 ± 2 °C. About 10 °C rise in temperature of water in the bath was observed. This was followed by heating the flask using the same electric mantle as in Section 2.2.1 and extraction of essential oil was carried out until no measurable essential oil was extracted. The yield (%) was calculated as in the HD process. The essential oil was collected, dried under anhydrous sodium sulphate and stored in an amber airtight vial at −18 °C for further analysis. The experiments were performed at least three times

2.3. Gas chromatography/mass spectrometry (GC/MS) identification

Determination of essential oil composition was carried out using a gas chromatography (Agilent Technologies 7890A, Santa Clara, California, USA) coupled with mass spectrometry (Agilent Technologies 5975C, USA), operating at 70 eV ionization energy, 0.5 s/scan, and the mass range of 35–400 amu, equipped with a HP-5MS capillary column (phenyl methyl siloxane, 30 m × 0.25 mm ID; 0.25 μm film thickness). The injector and detector temperatures were both 280 °C. Temperature programming was as follows; initial temperature of oven was held at 60 °C for 5 min and then increased by a rate of 3 °C/min until 210 °C and finally increased to 240 °C by 20 °C/min rate. It was then held at 240 °C for 8 min and 30 s.

The carrier gas was helium at flow rate of 1 ml/min, and 1 μl of sample was injected into the GC/MS in the split mode (split ratio: 1/100). Relative percentage data were obtained from electronic integration of peak areas without using correction factor. Then, the MSD ChemStation (G1701EA, E.02.01.1177, Agilent Technologies, Santa Clara, California, USA) software was used to handle mass spectra and chromatograms. Retention indices (RIs) were calculated through retention times (RTs) of C_5–C_{28} *n*-alkanes that were injected after the essential oils under the same chromatographic conditions. The compounds were identified by comparing their mass spectral fragmentation patterns with those stored in the data bank (Wiley/NBS library) and with mass spectra literature data (Adams, 2007). For each compound on chromatogram, percentage of peak area relative to the total peak areas from all compounds was determined and reported as the relative amount of that compound.

2.4. Physical constants

Specific gravity and refractive index of essential oils from *P. ferulacea* and *S. macrosiphonia* samples were measured according to Food Chemical Codex at 25 and 20 °C, respectively (FCC, 1996).

2.5. Antimicrobial activity

2.5.1. Microorganisms

Test microorganisms included the following Gram-positive bacteria: *Bacillus cereus* (ATCC 11778), *Listeria inocua* (ATCC 33090), *Staphylococcus aureus* (ATCC 25923), and Gram-negative bacteria: *Escherichia coli* (ATCC 15224), *Salmonella typhimurium* (ATCC 202026), *Enterobacter aeroginosa* (ATCC 13048). The microorgan-