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Determination of Essential Oil Composition of *Prangos acaulis* (*DC*) *Bornm* Obtained by Hydrodistillation and Supercritical Fluid Extraction Methods

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Abstract: Chemical composition of the essential oil of the *Prangos acaulis* was extracted by Hydrodistillation (HD) and Supercritical Fluid Extraction (SFE) methods from aerial parts at full flowering stage. Their compositions were identified using GC/MS as the method of analysis. The analyses reveal that samples differ quantitatively and qualitatively. A total of 21 compounds constituting 89.1% of aerial parts oil were in SFE method. The oil obtained by SFE was under condition: pressure 120 bar, temperature 45°C and extraction time 45 min. On the other hand, 26 compounds constituting 98.74% of oil were in HD method. In according to our results, in both extracts, the two compounds present in the biggest quantity were: α-pinene (13.7 versus 22.87% in the SFE and HD oil, respectively) and 3-ethylidene-2-methyl-1-hexen-4-yne (14.3 versus 21.36%).

Key word: *Prangos acaulis*, essential oil, supercritical fluid extraction, hydrodistillation

INTRODUCTION

The genus *Prangos* belongs to the Umbelliferae family consists of about 30 species (Evans, 1989). Fifteen species of the genus *Prangos* are found in Iran, among which five are endemic: *P. gaubae*, *P. crossoptera*, *P. tuberculata*, *P. cheilanthifolia* and *P. cattigonoides* (Mozaffarian, 1996). Some *Prangos* species have been used in the folk medicine as emollient, carminative (Zargari, 1988), anti fungal (Ozcan, 1999), anti oxidant (Çoruh *et al*., 2007), anti bacterial, cytokine, release inhibitor (Tada *et al*., 2002), nutritive (Coşkun *et al*., 2004), anti-HIV (Shikishima *et al*., 2001), tonic, anti flatulent and anti helmintic (Baser *et al*., 2000a; Ulubelen *et al*., 1995) also by Kazerooni *et al*. (2006) reported abortifacient effect of *Prangos ferulacia* on pregnant rats.

The essential oil composition of prangos species: *P. asperula* (Sajjadi and Mehregan, 2003), *P. latiloba* (Akhlaghi and Hashemi, 2006), *P. platychlaena*, *P. uchtritzii* (Uzel *et al*., 2006), *P. heyniae* (Baser *et al*., 2000b), *P. ulopetera* (Mazloomifar *et al*., 2004), *P. acaulis* (Meshkatalsadat and Mirzaei, 2007) were reported previously.

The major components of *P. asperula* fruits were δ-3-carene (16.1%), β-phellandrene (14.7%), α-pinene (10.5%), α-humulene (7.8%), germacrene (5.4%), δ-cadinene (4.2%) and terpinolene (4.0%). The main constituents of the essential oils of different parts of *P. latiloba* were γ-cadinene (30.39%), spathulenol (29.5%), germacrene D (27.79%) and α-pinene (25.47%). The major components of *P. platychlaena* were δ-3-carene (3.39%) and p-cymene (3.38%), while those of *P. uchtritzii* were α-pinene (40.82%), nonene (17.03%), β-phellandrene (11.14%), δ-3-carene (7.39%) and p-cymene (4.90%). The main constituents of the essential oils of *P. heyniae* fruits from two localities Turkey were β-bisabolenal (53.3 and 18%), β-bisabolenol (14.6 and 2.3%) and β-bisabolene (12.1 and 10.1%). The main components of *P. ulopetera* oil from Iran were β-caryophllene (27.1%), caryophyllene oxide (15.9%) and α-pinene (12.4%). The constituents of the essential oils of different parts of *P. acaulis* were α-pinene (34.20, 39.54 and 25.04% in the stems, leaves and flowers oils, respectively), 3-ethylidene-2-methyl-1-hexen-4-yne (56.8, 37.94 and 23.51%) α-terpinene (3.4, 10.9 and 17.26%) and limonene (1.1, 5.21 and 13.64%).

The composition of the oils obtained by steam distillation of the aerial parts and hydrodistillation of seed of *P. uloptera*, growing in Iran, have been reported. The main constituents of the oil of the aerial parts were β-caryophyllene (18.2%), germacrene D (17.2%) and limonene (8.7%) and the major components of seed oil were α-pinene (41.9%) and β-cedrene (4.0%) (Sefidkon and Navaii, 2001). In similar study, reported that the oils from crushed dry aerial parts of *P. acaulis* from Damavand, north of Tehran, in June 2003 during the flowering period contained cis-sesquisabinene hydrate (25.6%) and α-pinene (12.5%) as main constituents (Rustaiyan *et al*., 2006).

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Literature review showed variation between the chemical compositions different *Prangos* species oils from different area.

The essential oils of plants have usually been isolated by either hydrodistillation or solvent extraction. The distillation method has traditionally been applied for the recovery of essential oils from plant materials. One of the disadvantages of the distillation method is that essential oils undergo chemical alterations and the heat sensitive compounds can easily be destroyed. The other method applied for oil recovery from plant materials uses organic solvent extraction, which has limitations with regard to the loss of valuable volatiles during vacuum evaporation of solvent and difficulty in obtaining solvent-free extracts (Assis *et al*., 2000; Lehotay, 1997; Oszagyan *et al*., 1996; Sato *et al*., 1995).

In this study, a supercritical fluid extraction with Carbon dioxide method was used for isolation of the volatile compounds from aerial parts of *P. acaulis* for compared with hydrodistillation method of similar part of plant. The Persian name of the plant is Joshire Kotolei.

Supercritical Fluid Extraction (SFE) is an extraction process using a supercritical fluid as a solvent. When a fluid is taken above its critical temperature (Tc) and critical pressure (Pc), it exists in a condition called the supercritical fluid state. The physical-chemical properties of a fluid in the supercritical state are in between those of a typical gas and liquid. It has the density of a liquid and functions like a liquid solvent, but it diffuses easily like a gas. SFE is a valid alternative for the production of flavors and fragrances from natural materials (Mchugh and Krukonis, 1983).

Carbon dioxide is used as the supercritical fluid because it is a safe, noncombustible, inexpensive, odorless, colorless, tasteless, nontoxic, low critical temperature and pressure (31.06°C and 73.82 bar) and readily available solvent. However to the best of our knowledge, there is no previous report of the SFE of *P. acaulis* aerial part essential oil.

MATERIALS AND METHODS

Plant materials: The fresh plant of *P. acaulis* was collected during full flowering stage from of altitude 600 m Zagros Mountain in the Lorestan state, west of Iran, in July 2006. The plant was identified and authenticated by Dr. H. Amiri at the Department of Biology University of Lorestan. Voucher specimens were deposited in the Herbarium of Research Institute of Forest and Rangeland Tehran.

The dried plant was stored in a dark place. The sample was ground in a blender to produce a fine powder. The average of particle size was 0.5 mm.

Hydrodistillation: The air-dried aerial parts (50 g) of the plant were subjected to hydrodistillation for 4 h in a Clevenger-type apparatus to produce the oil. Were dried over anhydrous sodium sulphate.

Reagent: The carbon dioxide used in this study was supplied by Roham gas Co. (Tehran, Iran) with a purity of 99.7%.

Supercritical fluid extraction: A Pilot Scale system homemade in the SFE mode was used for extraction. The apparatus consist of extractor (V = 30 mL) and separator, equipped with heating jackets, high-pressure pump, heat exchanger, flow-meter and CO_2 tank. Approximately 20 g of ground material was charged in to the extractor. Liquefied CO_2 was continuously pumped with a high-pressure pump. Carbon dioxide enters the extractor where it dissolves the volatile material in the plant and after the temperature and pressure are adjusted to supercritical conditions of about 45°C and 120 bar, respectively, continuous flow of carbon dioxide is initiated. The temperature and pressure regulated and constant level. The CO_2 and the extract then go to the separator where the pressure is below critical by across of the pressure reduction valve thereby allowing the CO_2 to revert to the gas phase and deposit the extract. The CO_2 gas measured with flow-meter.

Analyses of oil: GC-FID analyses of the oil were conducted using a Thermoquest-Finnigan instrument equipped with a DB-1 fused silica column (60 × 0.25 mm i.d., film thickness 0.25 μm). Nitrogen was used as the carrier gas at the constant flow of 1.1 mL min^{-1}. The oven temperature was raised from 60 to 250°C at a rate of 5°C min^{-1}. The injector and detector (FID) temperatures were kept at 250 and 280°C, respectively. GC-MS analysis was carried out on a Thermoquest-Finnigan Trace GC-MS instrument equipped with the same column and temperature programming as mentioned for GC. Transfer line temperature was 250°C. Helium was used as the carrier gas at a flow rate of 1.1 mL min^{-1} with a split ratio equal to 1/50. The constituents of the volatile oils were identified by calculation of their retention indices (Gonzales and Nardillo, 1999) under temperature-programmed conditions for n-alkanes (C_6-C_{24}) and the oil on a DB-1 column under the same conditions. Identification of individual compounds was made by comparison of their mass spectra with those of the internal reference mass spectra

library (Wiley 7.0) or with authentic compounds and confirmed by comparison of their retention indices with authentic compounds or with those of reported in the literature (Adams, 1995). Quantitative data was obtained from FID area percentages without the use of correction factors.

RESULTS

The constituents of the hydrodistillation essential oil and supercritical fluid extracts are show in Table 1. Twenty six compounds which represent about 98.74% of the total composition obtained by hydrodistillation were identified. The major constituents were α-pinene (22.87%), 3-ethylidene-2-methyl-1-hexen-4-yne (21.36%), δ-3-carene (18.15%) and limonene (17.56%).

In present study, we employing these condition for SFE: T = 45°C, P = 120 bar, Extraction time = 45 min and flow rate =1.6 kg h^{-1}. With applied these conditions SFE extracted 21 compounds, represent 89.1% of the total extract composition. The major constituents were α-pinene (13.7%), 3-ethylidene-2-methyl-1-hexen-4-yne (14.3%), octadecane (9.4%) eicosane (8.9%), nonadecan (7.3%), limonene (6.1%) . The total amounts of the essential oils extracted by both HD and SFE were estimated as about 0.32% (w/w) and 0.54%, respectively.

Table 1: Constituents of the HD and the SFE extraction of *P. acaulis*

Compound	RI*	HD (%)	SFE (%)
α-thujene	930	tr	-
α-pinene	943	22.87	13.7
Camphene	952	tr	-
Sabinene	970	1.15	-
β-pinene	977	1.42	-
Myrcene	983	5.31	0.2
α-phellandrene	1002	6.9	1.3
δ-3-carene	1012	18.15	1.8
p-cymene	1015	0.17	-
Limonene	1027	17.56	6.1
z-ocimene	1035	0.1	-
β-terpinene	1050	0.16	-
3-ethylidene-2-methyl-1-hexen-4-yne	1061	21.36	14.3
α-methyl styrene	1070	0.05	-
α -terpinolene	1083	3.21	1.1
m-methyl styrene	1104	0.1	-
Verbenol	1130	tr	-
p_mentha-1,5-dine 8-ol	1146	tr	-
Terpineol	1161	tr	-
β _citronellol	1200	0.02	-
Dodecane	1205	-	1.7
Bornyl acetate	1265	0.03	tr
Tridecane	1299	-	2.4
Tetradecane	1399	-	3.1
Trans-caryophyllene	1418	0.13	1.9
α -humulene	1451	0.02	tr
Ar-curcumene	1465	0.03	-
γ-himachalene	1477	-	tr
Germacrene D	1475	tr	-
β-selinene	1484	tr	-
Pentadecane	1499	-	4.6
Hexadecane	1598	-	3.5
Heptadecane	1698	-	5.3
1,1,3-trimethyl-3-phenylindan	1705	-	2.5
Octadecane	1798	-	9.4
Nonadecan	1898	-	7.3
Eicosane	1997	-	8.9
Total		98.74	89.1

tr = trace <0.01%. *Retention index identification was achieved using retention times and retention indices on a DB-1 capillary Column

DISCUSSION

The volatile concentrate obtained by SFE has a very different appearance with respect to the HD product. In a comparison between HD and SFE methods, there was less number constituents similar in both methods. In according of present results, in both extracts, the two compounds present in the biggest quantity were: α-pinene (13.7 versus 22.87% in the SFE and HD oil, respectively) and 3-ethylidene-2-methyl-1-hexen-4-yne (14.3 versus 21.36%). As shown in Table 1, the SFE extract was poorer in monoterpenes than HD extract. SFE extract exhibit heavier molecular weight compounds than the HD. As can be seen from the Table 1, lighter components, e.g., α-thujene, sabinene, β-pinene, p-cymene and z-ocimene present in HD oil, were not detected in the SFE extract.

Indeed, solvents are classified according to scale of polarity depending on their ability to dissolved polar or non-polar molecules. Carbon dioxide always behaves as a non-polar solvent that selectively dissolved non-polar compounds like fats, essential oil, hydrocarbons. In order to extract polar or oxygenated compounds with SFE, organic solvents have been added as modifiers to decrease polarity (Ashraf-Khorassani and Taylor, 1997; Michael and James, 1996), but in this study HD extraction shows most of essential oil is hydrocarbons component therefore, not used of modifier in SFE method and optimized experimental conditions for $SFCO_2$.

The advantages of supercritical carbon dioxide extraction over hydrodistillation including: shorter extraction period (45 min against 4 h for hydrodistillation) cost (energy cost is fairly higher for performing hydrodistillation than that requested for reading Supercritical Fluid Extraction conditions) and cleaner features (as no great volume of the organic solvent is involved).

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