Journal of Medicinal Plants and By-products (2014) 1: 47-51

Original Article

Phytochemical Composition of the Essential Oils from Two *Pimpinella* L. species (*P. deverroides* Boiss. and *P. tragium* Vill.) Growing Wild in Iran

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Article History: Received: 01 December 2013/Accepted in revised form: 03 February 2014

Abstract

The experiment was aimed to study the chemical constituents of essential oils from two *Pimpinella* L. species including *P. deverroides* - Boiss, and *P. tragium* Vill. growing wild in Iran. To do this, the essential oil of fruits of two *P. tragium* populations (Sefidkhani and Hesar, Markazi province, Iran) and one population of *P. deverroides* (Kerend, Kermanshah province) were extracted by a Clevenger-type apparatus and subjected to GC and GC/MS analyses. Based on the dry weight, the oil content of *P. deverroides* fruits was calculated as 0.43% and the oil yield of the fruits of *P. tragium* was ranged from 1.19% (Sefidkhani) to 2.23% (Hesar). In total, 42 compounds, constituting about 87.69-95.86% of studied essential oils, were identified. γ-elemene (12.1%), *cis*-muurol-5-en-4α-ol (11.2%) and geijerene (10.1%) were the major components of *P. deverroides* essential oil. Geijerene (33.5%) and γ-elemene (19.4%) were identified as the major components of the essential oil from *P. tragium* fruits collected from Sefidkhani and geijerene (80.1%) constituted the major portion of the essential oil from *P. tragium* fruits collected from Hesar. There were significant differences in the phytochemical composition of examined populations, which, considering constituents of an essential oil determine its flavor and biological activities, enabled selection of favored populations for breeding program and for using in different industries.

Key words: *Pimpinella deverroides* Boiss, *Pimpinella tragium* Vill., Essential oil, γ-Elemene, *cis*-muurol-5-en-4α-ol, geijerene

Introduction

In recent years, there has been a growing interest to study composition and antimicrobial activity of essential oils extracted from different plant taxa. This is due to the great potential of essential oils for use in different industries e.g. food, cosmetics, perfume and medicine. Beside pharmaceutical properties and conferring a pleasant taste or smell, essential oils have the potential to protect stored stuffs from microbial contamination and unwanted oxidative reactions.

The plant family Apiaceae (Umbelliferae) is composed of about 300 to 455 genera and 3000 to 3750 species distributed in the northern hemisphere [1]. The genus *Pimpinella* is included in the subfamily Apioideae and comprises approximately 150 species which are largely found in Europe and Asia extending to China [2]. Anise (*Pimpinella anisum* L.) is a well-known species of this genus and its fruits are widely used for flavouring and extraction of an essential oil which is applied in perfumery and medicine industries.

The genus *Pimpinella* is represented in the flora of Iran by about 22 species including *P. deverroides* and *P. tragium* [3,4]. The first is among the six endemic *Pimpinella* species growing in Iran. Essential oils of *Pimpinella deverroides* plant parts (stem, leaf, inflorescence and seed) from Lorestan province at full flowering and seed maturing stages were analyzed for two years [5]. They reported that the essential oils yield of the stem, leaf, inflorescence and seed of *P. deveroides,* were 0.6%, 2.5% and 7.1% w/w, respectively in 2005 and 1.3%, 2.4% and 7.5% w/w in 2006. Also, the yield

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of seed oils was higher than that of stem plus leaf and inflorescence. The same authors suggested that pregeijerene (35.5%-67%) and Geijerene (8.2%-14.1%) were the major constituents in all oils

The present study was performed to investigate the variations of content and compositions of the fruit (seed) essential oil of two *Pimpinella* species including *P. deverroides* and *P. tragium* growing wild in Iran.

Material and Methods

Plant materials and extraction of essential oils

The mature fruits of two *P. tragium* populations (Sefidkhani and Hesar located central of the country, Markazi province, Iran) and one population of *P. deverroides* (Kerend, located west part of the country, Kermanshah province, Iran) were collected in June to July 2011. Voucher specimens number of two species (*P. deverroides* and *P. tragium*, no. 1260 and 1282, respectively) has been deposited at the Herbarium of Agrculture and Natural Resource Facultyin arak University. Geological information and altitude of sampling area were recorded using GPS. Soil sample of each location also was analyzed for their physicochemical characteristics (Table 1). After collection, the fruits were separately air-dried in the shade at the room temperature and subjected to oil extraction via hydro-distillation method by using a Clevenger-type apparatus for 3 h. Extracted essential oils were dried over anhydrous sodium sulfate and kept in the dark vials at +4 °C prior to analysis.

Essential oils analysis

GC analysis was performed using a Younglin Acm600 gas chromatograph equipped with a flame ionization detector (FID) and a HP-5MS fused silica column (30m × 0.25mm i.d.; film thickness 0.25µm). Helium was used as the carrier gas at a flow rate of 0.8 ml/min. The injector and detector temperatures were kept at 290 °C. Oven temperature program was kept at 50 °C for 5min, then raised to 240 °C at the rate of 3 °C/min then raised to 300 ºC with the ramp of 15 ºC/min and held isothermally for 3 min. GC-MS analysis was carried out by an Agilent 6890 gas chromatograph equipped with the same column as mentioned above and coupled with an Agilent 5973 mass spectrometer. Helium was used as the carrier gas at a flow rate of 0.8 ml/min in split ratio of 1:25. Ionization voltage was 70 eV. Ion source and interface temperatures were 220 °C and 290 °C, respectively. The oven program temperature was the same as GC analysis.

Individual components were identified by comparison of their mass spectra with internal mass spectra library (Wiely 7n.1) and experimental retention indices (RI), which were calculated for all volatile constituents by using a homologous series (C_8 to C_{30}) recorded under the same operating conditions, and comparison with literature data [6].

Results and Discussion

In this study, the fruits of *P. tragium* contained essential oils ranging from 1.19% (Sefidkhani) to 2.23% (Hesar) based on the dry weight (Table 2). Also, the essential oil content of *P. deverroides* fruits was calculated to be 0.43%. In a previous study, the oil content of the fruits of *P. tragium* plants collected from Polour, Tehran province has been reported to be 1.33% [7]. Also, in a study by Askari *et al*. [5]. *P. deverroides* plants collected at two consecutive years from Lorestan province (2005 and 2006) yielded 7.1% and 7.5%, respectively. Moreover, the essential oil composition of *Pimpinella Aurea* DC. and *P. anisum,* has been previously reported by Askari and Sefidkon [7]. The yields of stem plus the leaf, inflorescence and seed oils of *P. aurea* were 0.4%, 1.5%, and 2.0%w/w, respectively.

The chemical profile of analyzed essential oils is shown in table 2. Chemical analyses resulted in identification of forty-two compounds, making about 87.69-95.86% of studied essential oils. γ-elemene (12.1%), *cis*-muurol-5-en-4α-ol (11.2%) and geijerene (10.1%) were identified as the most abundant components of *P. deverroides* essential oil. Besides, linalool (7.1%), α-pinene (6.6%) and 8-epi-dictamnol (5.6%) constituted considerable amount of the fruit essential oil. Geijerene (33.5%) and γ-elemene (19.4%) were also identified as the major components of the essential oil from *P. tragium* fruits collected from Sefidkhani. Similarly, it has been observed that the major portion of the essential oil from *P. tragium* fruits collected from Hesar is composed of geijerene (80.1%).

Previously, Askari and Sefidkon [7] reported β-pinene (25.3%), germacrene B (17.8%) and sabinene (13.6%) as the main components of the seed essential oil of *P. tragium* plants. In the study of Askari *et al*. (2010) on *P. deverroides* plants,

pregeijerene (35.5-66.9%), geijerene (8.2-14.1%), trans-dictamnol (2.3-9.8%) and β-pinene (1.4-7.5%) have been reported as the major constituents of the seed essential oil. Also, study on *P. aurea* essential oils revealed that the major constituents of the stem plus the leaf oil were 1, 8-cineol, limonene (21.4%), viridiflorol (12.8%), α-pinene (11.5%) and kessane (10.5%). Major constituents of the inflorescence oil were viridiflorol (32.5%) and β-bisabolene (29.5%) while main constituents of the seed oil were β-bisabolene (50.8%) and viridiflorol (37.0%) [8]. The total amount of extractable substances or global yield of *P. anisum* seed for the super critical fluid extraction process varied from 3.13-10.67%. The major compound identified and quantified in the extracts was anethol (~90%) [9]. Also, chemical composition of the hydrodistilled essential oils of the aerial parts, inflorescence at flowering stage and seeds at seeding stages of *Pimpinella khorasanica* Engstrand were characterized [10]. They reported that the yields of stem/leaf, inflorescence and seed oils were 0.3 %, 2.4 % and 3.0 % w/w, respectively. The oil analysis confirmed the characterization of thirteen, ten and ten compounds in stem/leaf, inflorescence and seed accounting for 91.3 %, 95.8 % and 91.5 % of the total oil, respectively. Moreover, they showed that the oils was rich in (E)-β-farnesene (48.6 %, 55.3 % and 54.1 %) as the main component in all oils followed by Ar-curcumene (17.8 % and 10.1 %) and foeniculin (6.3 % and 5.3 %) in stem/leaf and inflorescence, respectively [10].

The composition of the essential oil of *Pimpinella serbica* (Vis.) Benth. & Hook.f. ex Drude that occurs in Yugoslavia has been reported. The percentage of essential oil ripe fruits was 2.02-3.25%. About 55% of the essential oil was reported to be made up of sesquiterpenes. The main sesquiterpenes were β-caryophyllene (over 47%) and its isomer α humulene (about 2.5%) [11]. Oil Yield of *Pimpinella aromatica* M.Bieb aerial parts was 6.1%. Main compounds of aerial parts and fruit oils were methyl chavicol (91.1% and 82.6%) and *trans*-anethol (7.2% and 10%) respectively [12, 13]. The oil constituents from the roots, fruits, leaves and stems of *Pimpinella cumbrae* Link gathered in the Canary Island were investigated. The major constituents in the root oil were found to be isokessane (17%), β-dihydroagarofuran (15%), 2-methyl-butyric acid (10%), geijerene (10%) and pregeijerene (7%). In the fruit oil the main components found were α-bisabolol (39%), δ-3-carene (16%) and limonene (8%). In the leaf oil, α-bisabolol (53%) and δ-3-carene (11%) were the predominant constituents. The most important compounds from the stem oil were α-bisabolol (39%), isokessane (10%) and β-dihydroagarofuran (9%). Pseudoisoeugenol esters were also detected in the oils from the roots, fruits and stems [14].

It is well known and documented that the chemical constituents of medicinal and aromatic plants, also their biological activities are influenced by genetic divergence and environmental factors (table 1), also relation between geographic distribution and biochemical composition were previously reported for some medicinal plants, which our present results supported by similar findings [15,16].

This study revealed that high level of phytochemical variability exists among *Pimpinella* species including *P. deverroides* , and *P. tragium* from three populations. However, the population of Hesar (due to its geographical origin) was identified as superior for the high oil content (2.23%) and high geijerene (80.1%) content, which can be used as a talent population for future cultivation, domestication and breeding programs.

Table 1 Edaphic and geological characteristics of the studied populations of *Pimpinella* species from three different location.

Descriptions	Kerend (Kermanshah province)	Sefidkhani (Markazi province)	Hesar (Markazi province)
Longitude (E)	46º 14´ 29.2˝	49º 33´57.6	49º 17´23.9˝
Latitude (N)	34º 11´ 36.4˝	33º 57´55.8˝	33º 59´6˝
Altitude (m)	1812	2967	2361
Collection date	2011.Jul.1	2011.Jul.3	2011.Jun.30
Soil pH	7.2	7.8	8
Soil texture	Clay-Sandy	Clay-Sandy	Sandy-Clay
OC[1]%	1.08	1.12	0.92
TNV[2]%	39	31	35
EC[3](ds/m)	0.79	0.51	0.65

[1,] Organic carbon, [2,] Total neutralized value [3,] Electrical conductivity

Table 2 Composition of essential oils from two *Pimpinella* species, *Pimpinella deveroides and P. tragium* growing wild in Iran.

No.	Compound	R.T	R.I*	*P. deverroides*	*P. tragium*	
				Kerend	Sefidkhani	Hesar
1	*α*-pinene	11.41	932	6.62	1.69	1.04
2	Camphene	11.99	946	0.33	-	-
3	Sabinene	13.31	969	0.87	0.9	0.15
4	*β*-pinene	13.45	974	1.75	0.43	0.28
5	*β*-myrcene	14.25	988	0.79	-	0.75
6	*α*-phellandrene	14.83	1002	0.28	-	0.1
7	*p*-cymene	15.93	1020	2.7	0.5	0.4
8	Limonene	16.14	1024	1.28	0.5	0.48
9	*β-cis*-ocimene	16.78	1032	4.19	0.66	1.7
10	*β-trans*-ocimene	17.17	1044	0.44	-	0.23
11	*γ*-terpinene	17.7	1059	1.9	tr	0.59
12	*α*-terpinolene	19.18	1086	0.28	-	-
13	Linalool	19.75	1095	7.11	-	-
14	*allo*-ocimene	21.32	1128	0.86	4.2	-
15	Cyclohexene, 3,4-diethenyl-3-methyl	21.48	1133	1.73	-	0.57
16	Geijerene	22	1138	10.1	33.46	80.1
17	*p*-methyl-acetophenone	23.84	1179	0.64	0.81	0.56
18	2,4,4-trimethyl-4-vinyl-3-cyclopenten-1-one	28.2	1273	0.23	-	-
19	Isobornyl acetate	28.68	1283	0.62	0.27	0.14
20	Pregeijerene	28.8	1285	0.25	0.2	0.14
21	2,4,5-trimethyl-Benzaldehyde	31.71	1351	tr	-	-
22	Cyclosativene	32.5	1369	0.21	-	-
23	*α*-copaene	32.82	1374	2.06	0.7	0.44
24	8-*epi*- dictamnol	33.08	1379	5.6	2.27	1.42
25	*β*-cubebene	33.4	1387	0.73	0.26	0.16
26	*β*-elemene	33.5	1389	0.6	0.4	0.18
27	*trans*-caryophyllene	34.67	1417	1.95	3.13	1.48
28	*γ*-elemene	35.38	1434	12.08	19.41	2.3
29	*α*-humulene	36.09	1452	0.92	1.27	0.22
30	*α*-amorphene	37.12	1483	1.52	2.5	0.36
31	Germacrene-D	37.3	1484	2.06	1.1	0.12
32	Bicyclogermacrene	37.86	1500	0.6	0.5	0.3
33	*α*-muurolene	37.96	1500	0.45	1.1	0.2
34	*δ*-cadinene	39.01	1522	3.1	3.42	0.7
35	Selina-3,7(11)-diene	39.79	1545	tr	1.87	-
36	Elemol	40.16	1548	tr	0.5	-
37	*Cis*-muurol-5-en-4α-ol	40.52	1559	11.21	0.6	0.35
38	1,5-epoxysalvial-4(14)-ene	40.66	1567	tr	0.85	-
39	Spathulenol	41.17	1577	2.4	2.29	0.17
40	Caryophyllene oxide	41.33	1582	0.9	1.2	0.23
41	*Epi-α*-cadinol	43.51	1638	0.39	0.7	tr
42	*α*-cadinol	43.99	1652	1.32	tr	tr
Total identified (%)				91.07	87.69	95.86
Essential oil content (%)				0.43	1.19	2.23

* Retention indices relative to C_8-C_{30} *n*-alkanes on the HP-5 column. tr < 0.1

** PD: *P. deverroides*; PTS and PTH: *P. tragium* plants from Kerend, Sefidkhani and Hesar, respectively.

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