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Essential oil combination of three species of *Achillea* growing wild in East Azarbayjan- Iran

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ABSTRACT

Background and aims: The *Achillea* genus has a wide distributional range, and the differences in oil composition may be affected by different environmental factors such as plant genetic type, seasonality, and developmental stage. The aim of this research was to determine the chemical combination of the essential oils derived from flowering aerial parts of *Achillea millefolium, A. biebersteinii and A. wilhelmsii* (family Astreacea) collected from different locations of East-Azarbayjan in Iran.
Methods: In this experimental study, Wild Achillea plants (*A. millefolium, A. biebersteinii and A. wilhelmsii*) were collected from three localities of East-Azarbayjan of Iran during the flowering period. The chemical combination of the isolated oils was examined by gas chromatography-mass spectrometry.
Results: In *A. millefolium,* the major compounds were 1,8-cineole (28.0%), camphor (19.2%), borneol (98.8%) and β-pinene (6.3%). In *A. biebersteinii* the major compounds were α-terpinen (41.42%), 2-carene (13.96%), m-cymene (13.41%) and 1,8-cineole (8.91%). In *A. wilhelmsii* the major compounds were carvacrol (29.2%), linalool (10.3%), 1,8-cineole (11.0%), (E)-nerolidol (8.4%) and borneol (5.04%).
Conclusion: Chemical compounds of essential oils of *Achillea* species were highly variable, which may be due to the differences in their chemical polymorphic structure and environmental conditions.

Keywords: *Achillea*, 1, 8-Cineole, Essential oil, GC/MS, α-Terpinen.

Original article

INTRODUCTION

The genus *Achillea* L. (Asteraceae) is represented by about 115 species found in the Northern Hemisphere, mostly in Europe and Asia, and commonly known as yarrows.[1-3] It has been represented in Iran by nineteen species including seven endemics.[4] The *Achillea* L. species belong to the oldest medicinal plants that are used both for pharmaceutical purposes and in folk medicine. These plants contain a complex of different pharmacological compounds like terpenes, flavonoids, alkaloids, bitters, tannins, lignans, etc.[5] *Achillea* species are diuretic, emmenagogue agents, used for healing wound, curing stomachache, diarrhea and antichloristic antispasmodic, antiseptic and infection preventing properties, and have also been used to reduce sweating and to stop bleeding, amarum, stomachicum, cholagolum and carminativum.[3, 6-12] The *Achillea* genus has a wide distributional range,[13] and the differences in oil composition may be affected by different environmental factors such as plant genetic type, seasonality, and developmental stage, because it is a chemically polymor-phic and perennial plant.[14] Terpenoids (1,8-cineole,

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camphor, borneol, pinenes, artemisia ketone, santolina alcohol, farnesane, caryophyllene and its oxides, cubebene, germacrenes, eudesmol, α-bisabolol and oxides, farnesene, γ-gurjunene, γ-muurolene and chamazulene) are the principle components of *Achillea* essential oils.[3] According to Nemeth's (2005) studies, within the last 15 years, an average of 54 compounds have been identified in samples of different species. Among them, the largest numbers of components (149 compounds) were found in the oils of some *Achillea* species. 1, 8- Cineole, camphor, borneol, α- and β-pinenes are among the five most abundant monoterpene components. Among the monoterpenes, 1, 8-cineole is a major component in *Achillea* species.[15-17] In some *Achillea* species, essential oil components (camphor and borneol) are next ranks.[18-21] Other most detected components are α- and β-pinenes, especially in the *A. millefolium* species [22] Hofmann (1993) also reported that the monoterpenes belonging to the p-menthane, thujane and pinane are the most frequent components of the oils of the Achillea millefolium populations. Sesquiterpenes such as chamazulene, β-caryophyllene and oxide, eudesmol, α-bisabolol as well as its oxides and farnesene, are the most frequently constituents.[3] Some researchers have reported the major constituent of several *Achillea* species as 1,8-cineole, camphor, piperitone and ascaridole in Turkey.[15, 23-25] Chemical composition of the essential oils of five *Achillea* species from Turkey has already been investigated.[6] The essential oils and hexane extracts of *Achillea frarantissima* and *Achillea santolina* from Egypt have been reported.[26] Antimicrobial, antidiabetic, antioxidant, antitumor and spasmolytic effects of different *Achillea* species have been previously reported, [2, 10, 27-30] Essential oil composition of five *Achillea biebersteinii* from central Turkey, their antifungal, and insecticidal activity had also been investigated.[31] Potential activity of the *Achillea wilhelmsii* leaves on bacteria and effects of the leaves essential oil and methanol extract of the *A. wilhelmsii* on the growth of the bacteria were investigated.[32]

METHODS

In this experimental study, plant materials of *Achillea* species were collected from three different places in East-Azarbayjan province of Iran during the flowering period, in June at 2012. The location, the plant species, altitudes and oil yield are listed in Table 1. The collection area has the characteristics of terrestrial climate. The climate is characterized by warm to hot, temperate summer and freezing snowy winters. Aerial parts of *Achillea* samples were dried at room temperature. Flowers and leaves were separated after drying. All samples were hydro-distilled for essential oil by grossly pulverized powdered plant (40 g) using a Clevenger type apparatus for 3 h.

The essential oils were analyzed on a gas chromatograph mass spectrometer (Shimadzu-17A-QP505, Japan). The gas chromatography column was a super CP-Sil 5CB capillary column (50m × 0.32 mm ID, 0.25 μm film thickness). The column oven temperature was set at 70 °C for 1 min, increased to 100°C at a rate of 1.5°C/min, increased to 180 °C at a rate of 4 °C/min, and held at 180 °C for 1 min. Next, it was increased to 200 °C at a rate of 10 °C/min, increased to 250 °C at a rate of 2.5 °C/min, and held at 250 °C for 5 min. Injector and detector temperatures were 280 °C and 300 °C respectively. The gas chromatography mass analysis carried out with the same characteristics as used in gas chromatography. The ionization energy was 70 eV, with a scan time of 1 sec, and a mass range of 40-300 amu. Unknown essential oil was identified by comparing its gas chromatography retention time to that of known compounds, and its mass spectra to known compounds or published spectra.

Table 1: The altitudes of collection localities and essential oil yields of investigated *Achillea* species

Species	Collected area	Altitutes(m)	Oil Rate (%)
A. millefolium	Hashtruod	1735	0.35
A. biebersteinii	Marand	1550	0.45
A. wilhelmsii	Ahar	1391	0.56

RESULTS

A total of 20 components of the essential oil of *Achillea millefolium* flowering aerial parts were identified (Table 2). The main constituents of *A. millefolium* were 1, 8-cineol (28.0%) and camphor (19.2%), followed by borneol (8.8%) and β-pinene (6.3%).

Table 2: Constituents identified from the essential oil of *A. millefolium*, *A. biebersteinii* and *A. wilhelmsi* aerial parts

Compounds	RI*	Species/Constituents (%)		
		A. millefolium	***A. biebersteinii***	***A. wilhelmsii***
α- Thujene	924	0.3	-	0.3
α- Pinene	938	4.8	0.77	3.6
Camphene	950	0.7	0.96	0.8
Sabinene	970	2.6	-	0.9
β- Pinene	977	6.3	0.51	0.8
2- Carene	996	-	13.96	-
α- Terpinen	1008	-	41.42	0.8
m- Cymene	1013	-	13.41	0.9
p- Cymene	1016	3.6	-	-
1, 8- cineol	1026	28.0	8.91	11.0
Terpinene	1052	0.9	0.86	0.8
Terpinolene	1088	-	-	0.8
Linalol	1099	-	0.98	10.3
Hotrienol	1104	-	-	0.6
2- Cyclohexen- 1 a	1123	-	1.01	-
Camphor	1128	19.2	3.62	2.6
Menthone	1142	2.4	-	-
Camphol	1148	-	0.25	-
Borneol	1159	8.8	-	5.4
Terpinen- 4- ol	1166	2.9	-	1.5
α- Terpineol	1178	3.8	3.18	2.2
Myrtenol	1188	1.9	-	0.6
Trans- carveol	1203	0.1	-	-
Piperitone	1228	-	0.96	-
Pulegone	1237	-	-	4.9
Bornyl acetate	1273	0.1	-	0.2
Thymol	1293	-	-	4.6
Carvacrol	1308	-	-	29.2
Geranyl acetate	1384	-	-	0.7
β- Caryophyllene	1418	2.8	-	-
Germacrene D	1479	2.9	2.19	-
(E)- Nerolidol	1569	-	-	8.6
n- Octadecane	1807	0.5	-	
Total identified		93.3	93.79	92.1
Monoterpenoids		86.1	90.63	81.8
Sesquiterpenoids		5.7	2.15	-
Others		1.8	1.01	10.3

** Retention index as determined on a HP-5MS column using the homogous series of n-hydrocarbons;*
a 2-Cyclohexen-1-ol, 1-methyl-4-(1- methylethyl)-trans.

A total of 16 components of the essential oil of *A. biebersteinii* flowering aerial parts were identified (Table 2). The principal compounds in *A. biebersteinii* oil were α-Terpinen (41.42%), 2-carene (13.96), m-cymene (13.41%), 1, 8-cineole (8.91%) and camphor (3.62%). Monoterpenoids represented 14 of the 16 compounds, corresponding to 90.63 of the whole oil, while 1 of 16 constituents were sesquiterpenoids (21.5% of the crude essential oil).

In the essential oils of *A. wilhelmsii* totally 23 components were identified, accounting for 92.1% of the total oil (Table 2). Carvacrol (29.2%), linalool (10/3%) and 1, 8- cineol (11.0%) were the 4 main constituents of A. wilhelmsii essential oil, followed by (E)- nerolidol (8.4%) and borneol (5.04%).

DISCUSSION

The aim of the present study was to determine the chemical composition of the essential oils of *Achillea millefolium*, *A. biebersteinii* and *A. wilhelmsii* from different locations of East-Azarbayjan in Iran.

The aired-dried aerial parts of plants yielded 0.35%, 45% and 0.56% oil, respectively. Chemical composition of essential oils of *Achillea* species was highly variable, which may be due to the differences in their chemical polymorphic structure and environmental conditions.

According to our data 1, 8- cineol (28.0%) and camphor (19.2%) were the main constituents of *A. millefolium*. Previous studies that have determined the chemical composition of *A. millefolium* essential oils also identified high levels of artemisia ketone (4.1% to 12.6%), camphor (6.1% to 24.5%), 1, 8-cineole (11.4% to 40.4%), linalool (0.9% to 9.5%) and borneol (3.2% to 9.2%).[33-35] However, linalyl acetate was only found in trace amounts in other studies of *A. millefolium* essential oil; this difference might be due to the diversity of the plant sources or different essential oil hydrodistillation procedures. Moreover, earlier studies have shown that high quantities of monoterpene hydrocarbons and oxygen sesquiterpenes present in *A. millefolium* essential oils, ranging from 10.4% to 26.9%.[35]

The main constituents in *A. biebersteinii* oil showed some differences from those in the essential oils derived of *A. biebersteinii* from Turkey. For example, the main components of essential oil of *A. biebersteinii* were piperitone (31.06%), eucalyptol (10.98%) and camphor (12.46%).[36, 37]

In another study the main constituents of the oil of the plant collected from Mazandaran province were 1, 8-cineole (7.9%), camphor (6.5%), α-fenchene (5.7%) and santolian triene (5.1%) were four major compounds of the oil of this study.[38]

In the present study *A. wilhelmsii*, carvacrol (29.2%), linalool (10/3%) and 1, 8- cineol (11.0%) were the four main constituents of the essential oil. Nineteen components in the volatile oils of *A. wilhelmsii* from Kerman (Iran) were reported previously in which caryophyllene oxide (12.5%), camphor (9.0%), borneol (6.1%), linalool (5.5%), 1, 8- cineole (3.6%), chrysanthenyl acetate (2.8%) and carvacrol (2.0%) were the main ones.[39] Comparison of two oils shows similarity in chemical composition, but with different percentages. Carvacrol (25.1%, linalool (11.0%), 1, 8- cineole(10.3%) and (E)-nerolidol (9.0%) were present in lower percentage in the oils of *A. wilhelmsii* from Kerman, while caryophyllene oxide the main oxygenated sesquiterpene in the oils of Kerman's present in small amounts in oils of *A. wilhelmsii* from Kazeroon in Fars province. In another study the main

constituents of the oil of the plant collected from Mazandaran province were camphor, borneol and 1, 8- cineole of which borneol and 1, 8- cineole were two major compounds of the oil of this study.[39] 1, 8-cineole, which consistutes 11.0% of the oil has also been reported as the major constituent of the oil of *A. wilhelmsii* from Egypt and Turkey and Mzandaran in Iran,[39, 40] and some other *Achillea* species.[21, 41-43] Camphor (2.6%) and (5.4%) were present as the main components of the oil of *A. tenuifolia*,[44] and *A. kellalensis* from Iran respectively.

CONCLUSION

The essential oil constituents of flowering aerial parts of *Achillea millefolium, A. biebersteinii and A. wilhelmsii* (family Astreacea) collected from different locations of East-Azarbayjan in Iran, were studied. Among the three subjected species, *A. wilhelmsii* has the most abundant oil content (0.56%). In conclusion, our results demonstrate variations in the qualitative and quantitative composition in the oils obtained from the aerial parts of three distinct *Achilleae* species. These differences can probably be attributed to the genetic differences or different geographic or environmental conditions of the plant materials.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interests.

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