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Essential oils composition of different *Achillea santolina* L. plant parts growing in Algeria

Tayeb Berramdane[1] · Nadhir Gourine[1] · Abdelghani Zitouni[2] · Isabelle Bombarda[3] · Mohamed Yousfi[1]

Abstract
The essential oils (EOs) of leaves, flowers and stems of *Achillea santolina* L. (Asteraceae) collected at complete flowering stage from Southwest of Algeria, were isolated by hydrodistillation and subsequently analyzed by means of GC and GC/MS. Quantitative and qualitative differences in chemical compositions between the studied parts of this plant were observed. The EOs were rich in oxygenated monoterpenes (65.91–79.94%). The major constituents in the flowers, leaves and stems were: camphor (68.12, 65.17, 55.72%), 1,8-cineole (8.22, 4.77, 0.7%) and α-terpineol (2.84, 5.35, 2.76%). The highest EO yields were obtained for the leaves and the flowers (0.59 and 0.49% "v/w", respectively), whereas, the stems were characterized by very weak yield value (0.05%).

Keywords *Achillea santolina* L. · Essential oils · Leaves · Flowers · Stems · Camphor · Cineole

Introduction

The genus *Achillea* is one of the most important genres of the Asteraceae family and comprises 115 species, which are mainly distributed in Europe, Asia and North Africa (Bremer 1994). *Achillea santolina* is a small shrub grows in Algeria on arid environments and the edge of cultivated lands. There are about five species of *Achillea* which are widely distributed in Algeria; *A. ligustica* All., *A. leptophylla* M.B., *A. odorata* L., *A. santolinoïdes* Lag. and *A. santolina* L. (Quézel et al. 1962).

The aerial parts of different species of the genus *Achillea* are widely used in folk medicine due to various purposes and pharmacological properties in various biological activities, such as, anti-inflammatory (Benedek et al. 2007), antimicrobial (Sökmen et al. 2003; Ünlü et al. 2002), antispasmodic (Yaeesh et al. 2006), antiulcer (Abd-Alla et al. 2016), and antiradical activities (Ardestani and Yazdanparast 2007; Bali et al. 2015). Furthermore, this plant is also used as treatment for cancerous cells (Bali et al. 2015; Ghavami et al. 2010). More specifically for *A. santolina*, the dried aerial parts and flowers of this plant were used traditionally as antidiabetic and as anti-inflammatory. It was also used to relieve pain or dryness of the navel, stomach pain or gas and to relieve the symptoms of common cold (Al-Snafi 2013). Moreover, previous experimental investigations on *A. santolina* confirmed different biological and antioxidant activities of this plant (Al-Awwadi 2010, 2013; Al-Hindawi et al. 1989; Al-Snafi 2013; Ali and Abd El-Moaty 2017; Alkofahi et al. 1996; Ardestani and Yazdanparast 2006, 2007; El-Shazly et al. 2004; Khalil et al. 2009; Khoori et al. 1999; Nenaah 2014; Twaij et al. 1988; Zaringhalam et al. 2010). The volatile oil of *A. santolina* produced insecticidal and insect repellent activities on both domestic flies and honeybees (Mustafa and Al-Khazraji 2008).

To our best knowledge, *A. santolina* growing in Algeria did not exhibit any studies concerning the chemical composition of their essential oil on different plant parts. At the opposite side, and according to literature, there were only few reports (coming from different countries of origin) which investigated the chemical composition of the essential oil of the aerial parts (Bader et al. 2003; Berramdane et al. 2018; Mohamed and Abdelgaleil 2008; Nenaah

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2014; Rahimmalek et al. 2009) or their different parts (El-Shazly et al. 2004; Motavalizadehkakhky et al. 2013a, b). The aim of this study was to investigate difference in amount and chemical composition of flowers, leaves and steams volatile oils from *A. santolina*.

Materials and methods

Plant collection

The aerial parts of *A. santolina* L. (at their full flowering phenological stage) were collected at the end of May 2014 from an area of the high plateaus which belongs to the region of Tousmouline at the wilaya of El-Bayadh. More specifically, the geographical coordinates of the exact location of this region of collection are 33°38′12″N and 0°18′51″E, and its altitudes is 1191 m. The plant was identified by Dr. Seridi abdel-kadir from department of Agronomy, University of Laghouat. Voucher specimens have been deposited in the herbarium of the National Agronomic Institute of Algiers (N.A.I.), Algeria (Herbarium No. P: 11).

Essential oil extraction

The plant material samples were dried in a shade at ambient temperature; subsequently, they were carefully milled; after that they were hydrodistilled for 3 h using a Clevenger type apparatus. The essential oil samples were recovered from the distillate with diethyl-ether solvent and then dried overnight using anhydrous sodium sulfate Na_2SO_4. After filtration the extract solutions, they were reduced at room temperature under light vacuum pressure using rotary evaporator (Rotavap). Finally, the obtained EO samples were stored at (+4 °C) until analysis.

Essential oil analysis

Analysis was carried out by gas chromatography (GC) using two columns and by gas chromatography–mass spectroscopy (GC/MS).

Gas chromatography (GC)

For the first column (ploar), a CP-Varian 3800 gas chromatograph was used with a flame ionization detector (FID), and a UB-Wax fused silica capillary column (60 m × 0.32 mm, 0.25 µm film thickness). Oven temperature was programmed from 50 to 250 °C at a rate of 3 °C·min^{-1} and held at 250 °C for 10 min. Injector and detector temperatures were set at 250 and 260 °C, respectively. Helium was the carrier gas at a flow rate of 1 mL/min. Splitting ratio 1:50.

For the second column (aploar), analytical GC was carried out in a Hewlett-Packard 6890 (Agilent Technologies, Palo Alto, CA, USA) gas chromatograph with a HP GC ChemStation Rev. A.05.04 data handling system, equipped with a single injector and flame ionization detection (FID) system. A graphpak divider (Agilent Technologies, part no. 5021-7148) was used for sampling to fused silica capillary column HP-5 (polydimethylsiloxane 30 m × 0.20 mm i.d., film thickness 0.20 µm). Oven temperature program: 70–220 °C (3 °C·min^{-1}), 220 °C (15 min); injector temperature: 250 °C; carrier gas: helium, adjusted to a linear velocity of 30 cm s^{-1}; splitting ratio 1:40; detectors temperature: 250 °C.

Gas chromatography–mass spectroscopy (GC/MS)

GC–MS was carried out in a Hewlett-Packard 6890 gas chromatograph fitted with a HP-1 fused silica column (polydimethylsiloxane 30 m × 0.25 mm i.d., film thickness 0.25 µm), interfaced with an Hewlett-Packard mass selective detector 5973 (Agilent Technologies) operated by HP Enhanced ChemStation software, version A.03.00. GC parameters as described above; interface temperature: 250 °C; MS source temperature: 230 °C; MS quadrupole temperature: 150 °C; ionization energy: 70 eV; ionization current: 60 µA; scan range: 35–350 units; scans s^{-1}: 4.51.

Components of each EO sample were identified by their linear retention indices on both UB-Wax and SPB-1 columns. Linear retention indices were calculated relative to linear homologous series of *n*-alkanes C_8–C_{24}. The identifications of the components were based on the comparison of their mass spectra with those of Wiley and NIST (National Institute of Standards and Technology) libraries, as well as by comparison of their retention indices with those of the values of a homemade database.

Results and discussion

The essential oils obtained by hydrodistillation from the flowers, the leaves and the stems of *A. santolina* presented strong characteristic odors and yellow colors. The EO yields were varying from moderate to weak values. The maximum yield was recorded for the leaves part with a value of 0.59% v/w, followed by practically similar but lower yield value for the flowers part (0.49%). Alternatively, the stems part yielded a very weak value of EO accounting for only 0.05%. When compared with literature, the obtained yields were lower than reported from Egypt (0.9% for all parts) (El-Shazly et al. 2004) and Iran (0.7; 0.25 and 0.15% for flowers, leaves and stems, respectively) (Motavalizadehkakhky et al. 2013a, b).

The percentage composition of the EOs of the different parts (flowers, leaves and stems) are listed in Table 1. GC and GC/MS analysis of the plant EOs led to the identification and quantification of twenty-eight constituents representing 69.12–89.64% of the total volatile oils. Quantitative and qualitative differences between the EOs of different parts studied were observed.

In the EO extracted from *A. santolina* flowers, 28 compounds were identified, corresponding to 89.64% of the total oil composition. It contained 87.00% of monoterpene derivatives. The oxygenated monoterpenes (79.94%) were prevalent compared to the monoterpene hydrocarbons (7.06%). Furthermore, the oxygenated sesquiterpenes accounted for 1.14% only. The main constituent in the *A. santolina* flowers EO was (68.12%), followed by 1,8-cineole (6.25%). Besides, the most significant minor components were α-terpineol (2.84%) and camphene (2.77%).

In the EO obtained from the leaves part, 19 compounds, comprising 85.43% of the oil, were detected. The sesquiterpenes composed 1.53%, while the monoterpenes made up 83.27% of the oil, of which the monoterpenes hydrocarbons had the most important contribution (79.27%). In addition, camphor (65.17%) was also the main constituent of this oil. The other main compounds were also α-terpineol (5.35%), 1,8-cineole (4.77%) and camphene (1.46%).

The stems EO was constituted by just 9 identified compounds. This EO showed a high percentage of monoterpenes hydrocarbons (65.91%) and oxygenated sesquiterpenes

Table 1 Chemical composition of essential oils from different parts of *A. santolina* L.

No.	Components	RI[a]	RI[b]	GC area %			Identification
				Flowers	Leaves	Stems	
1	Tricyclene	1011	919	0.19	–	–	MS, RI
2	*α*-Pinene	1022	929	1.51	0.92	–	MS, RI
3	Camphene	1061	942	**2.77**	1.46	–	MS, RI
4	*β*-Pinene	1102	970	0.65	0.45	–	MS, RI
5	Sabinene	1117	963	0.34	0.26	–	MS, RI
6	*α*-Terpinene	1184	1008	0.19	0.22	–	MS, RI
7	Limonene	1206	1022	0.40	0.34	–	MS, RI
8	1,8-Cineole	1218	1018	**6.25**	**4.77**	0.70	MS, RI
9	*γ*-Terpinene	1250	1046	0.47	0.35	–	MS, RI
10	(E)-*β*-Ocimene	1277	–	0.24	–	–	MS, RI
11	*p*-Cymene	1287	1011	0.13	–	–	MS, RI
12	*α*-Terpinolene	1299	1076	0.17	–	–	MS, RI
13	*cis*-Sabinene hydrate	1510	1050	0.35	0.28	–	MS, RI
14	*α*-Campholenal	1493	1102	–	–	–	MS, RI
15	Camphor	1518	1118	**68.12**	**65.17**	**55.72**	MS, RI
16	*trans*-Sabinene hydrate	1544	–	0.23	–	–	MS, RI
17	Linalool	1550	1081	–	0.23	**2.04**	MS, RI
18	Pinocarvone	1563	1133	–	–	–	MS, RI
19	*cis-p*-Menth-2-en-1-ol	1577	1105	0.60	0.75	–	MS, RI
20	Bornyl acetate	1592	–	0.66	0.35	–	MS, RI
21	Terpinen-4-ol	1615	1157	1.21	1.40	1.95	MS, RI
22	Myrtenal	1667	1163	–	–	–	MS, RI
23	*α*-Terpineol	1705	–	**2.84**	**5.35**	**2.76**	MS, RI
24	Borneol	1715	1133	0.30	0.28	1.19	MS, RI
25	Myrtenol	1794	1175	0.62	1.32	1.55	MS, RI
26	Cayophyllene oxide	1990	–	1.00	1.30	**2.02**	MS, RI
27	Caryophyllenol II	2230	–	0.14	0.23	–	MS, RI
28	(E,E)-Farnesyl acetate	2279	–	0.26	–	1.19	MS, RI
Essential oil yield % (v/w)				0.49	0.59	0.05	

Compounds are listed in order to their elution on the UB-Wax column

Bold vlaues are refering to the main identified components of the essential oils for each plant part

[a]Retention indices on the UB-Wax column relative to C_8–C_{24} *n*-alkanes

[b]Retention indices on the SPB-1 column column relative to C_8 to C_{24} *n*-alkanes

Table 2 Principal chemical classes in the essential oils from *A. santolina* L. flowers, leaves and stems as a percentage of the total essential oil

Chemical classes	Flowers	Leaves	Stems
Monoterpene hydrocarbons	7.06	4.00	0.00
Oxygen containing monoterpenes	79.94	79.27	65.91
Total Monoterpenes	87.00	83.27	65.91
Sesquiterpenes hydrocarbons	–	–	–
Oxygen containing sesquiterpenes	1.14	1.53	2.02
Esters	1.50	0.63	1.19
Total identified	89.64	85.43	69.12

(24.5%). The oxygenated monoterpenes were the most important derivatives identified, which represented like the two other oils, by camphor (55.72%). The other main compounds of the stems volatile oil were α-terpineol (2.76%), linalool (2.04%) and terpinen-4-ol (1.95%) among others.

The examination of the percentages of classes of the different parts studied (Table 2), reveals some similarities and some differences. The oils were rich in oxygenated monoterpenes (65.91–79.94%), but poorer in monoterpenes hydrocarbons (0.0–7.06%) and oxygenated sequiterpenes (1.14–2.02%). On the other hand, the sequiterpenes hydrocarbons were not detected among the identified components. The esters class was present with low percentages (0.63–1.5%). It was found that a large number of identified components (camphor, 1,8-cineole, camphene, limonene, etc.) were decreasing in the following order flowers > leaves > stems (Table 1). Inversely, there were some components which were exhibiting increasing percentages in the same previous mentioned order (linalool, cayophyllene oxide, terpinen-4-ol, myrtenol and borneol). The main difference between the EO compositions of the stems and both flowers and leaves were the lower content of major compound camphor in the EO of stems and the relatively higher content of the minor component linalool for this same plant part (2.04% *Vs* 0.0 & 0.23%). Finally, the stems EO was also characterized by the absence of many minor components indentified for the EOs obtained from the flowers and the leaves parts.

The comparison of the obtained results by the current investigation with earlier few reports of the same plant species originating from different countries: Egypt (El-Shazly et al. 2004) and Iran (Motavalizadehkakhky et al. 2013a, b) revealed with evidence that our oils were quite different from the others in terms of its major constituents.

As it was mentioned in the following references (Orav et al. 2006; Rahimmalek et al. 2009), the percentage of the total oil and compounds are influenced by the correlate between different conditions of climate, soil, irrigation, age, variety and morphological of the plant, genetic and their geographic origin, effect of biosynthesis or degradation of certain components. On the other hand, there are considerable the accumulation of the EOs and changes in their compounds in vegetative parts during their development and other phytochemical features.

Conclusion

The results of this work gave a new contribution to the specific chemical composition of the essential oils obtained from different parts for *A. santolina* growing in Algeria. The results demonstrated clearly some similarities, but in the mean time spotted some differences in terms of chemical compositions of the studied parts. The results suggests a camphor-rich chemotype of all vegetative parts decreasing in the order (flowers > leaves > stems) for this plant volatile oils that could be used as a base research to find the other probable reasons for phytochemicals use of these specific essential oils.

Compliance with ethical standards

Ethical statement N/A.

Conflict of interest This manuscript described has not been published before; not under consideration for publication anywhere else; and has been approved by all co-authors.

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