Food Measure (2017) 11:1430–1439
DOI 10.1007/s11694-017-9522-5

ORIGINAL PAPER

Aroma constituents of shade-dried aerial parts of Iranian dill (*Anethum graveolens* L.) and savory (*Satureja sahendica* Bornm.) by solvent-assisted flavor evaporation technique

Armin Amanpour [1], Hasim Kelebek [2], Serkan Selli [1,3]

Received: 23 June 2016 / Accepted: 3 April 2017 / Published online: 29 May 2017

Abstract The aroma profile of shade-dried aerial part from Iranian dill (*Anethum graveolens L.*) and Savory (*Satureja sahendica* Bornm.) plants was analyzed by the gas chromatography–mass spectrometry (GC–MS) and gas chromatography–flame ionization detector (GC–FID). For the first time in these aromatic plants, the solvent-assisted flavour evaporation (SAFE) extraction method with dichloromethane was used prior to GC–MS. A total of 40 and 26 aroma compounds was identified in dill and savory. Dill contained 271.52 μg/g total amount of aroma compounds, which included terpenes (28), aldehydes (3), alcohol (1), acids (3), volatile phenols (3), ketone (1) and norisoprenoid (1). Savory possessed 10,547.16 μg/g total amount of aroma compounds, including terpenes (20), alcohol (1) and volatile phenols (5). Of all aroma compounds detected in both plants, terpenes were quantitatively the most dominant aroma volatiles. In the overall aroma volatiles, α-phellandrene (160.0 μg/g) together with sabinene (26.5 μg/g), D-carvone (16.2 μg/g), DL-limonene (12.3 μg/g) and dill ether (7.8 μg/g) in dill and γ-terpinene (6236.83 μg/g) along with carvacrol (3239.19 μg/g), α-pinene (267.08 μg/g), α-thujene (219.36 μg/g) and β-bisabolene (130.8 μg/g) in savory were the major compounds.

✉ Serkan Selli
sselli@cu.edu.tr

[1] Department of Biotechnology, Institute of Natural and Applied Sciences, Cukurova University, 01330 Adana, Turkey

[2] Department of Food Engineering, Faculty of Engineering and Natural Sciences, Adana Science and Technology University, 01110 Adana, Turkey

[3] Department of Food Engineering, Faculty of Agriculture, Cukurova University, 01330 Adana, Turkey

Keywords Aroma profile · SAFE technique · GC-MS · Dill (*Anethum graveolens L.*) · Savory (*Satureja sahendica* Bornm.)

Introduction

Dill (*Anethum graveolens L.*) is a yearly herb of the Apiaceae family found particularly in regions with warm and tropical climates [1]. Leaves, stems, and fruits of dill are excessively used in different applications of the food industry, especially for their typical taste as well as pleasant and spicy aroma [2], and also for the medical use [1]. It is an important condiment crop from which both herb and seed have been extensively used in all kinds of flavouring including those for baking mixes, sauces, salads, and seafoods [3]. Additionally, the experimental studies demonstrated the antimicrobial, stomachic, antioxidant, carminative, insecticidal properties, and cardiovascular benefits of dill [4].

Savory (*Satureja* L.) is included in the main taxa of the Lamiaceae (Nepetoideae) family involving more than 200 species of aromatic herbs and shrubs, basically distributed in the Asian and Mediterranean zones [5, 6]. Fourteen species are presented in the flora of Iran, in the northern, northwestern, and western districts. Eight of them comprising *Satureja edmondii, Satureja intermedia, Satureja isophylla, Satureja kallarica, Satureja bakhtiarica, Satureja khusstanica* and *Satureja sahendica* are vernacular to Iran [5, 6]. The genus *S. sahendica* in Persian is known as "Marze" and "Marze-Sahandi," respectively [6]. *Satureja sahendica* Bornm. is a habitual, ramified, and rich aromatic herb growing in the western and northwestern areas, particularly in the Sahand Mountains in the northwest of Iran [6]. The volatile oils of the genus *Satureja* is isolated from

their aerial parts possess aromatic and medicinal features [5, 6]. The fresh and dried leaves and vegetative parts of stems have been consumed in many culinary aspects such as stuffings, stews, dishes containing meat and poultry, sausages, and greengroceries [7].

Furthermore, the aerial sections of some *Satureja* species have been used in the folk medicine for curing diarrhea, inflammation of the stomach and intestines, wounds, urinary infections [5], and asthma [6] and as antimicrobial, tonic, antiseptic, flatulent, digestive, diuretic, carminative, and aphrodisiac drugs [6]. Additionally, biological activities entailing antioxidant, antifungal, antibacterial, antidiabetic and anti-inflammatory functions were also evidenced in savory [5].

Aroma is a complicated structure of a big number of low molecular weight volatile compounds whose combination is characteristic to species and often to the variety of plants [7]. These two different plant varieties, dill and savory, are extensively used in the daily diet of the Iranian general population as flavouring agents and spices to improve the flavour of food. Both of them are aromatic and have their own peculiar flavour with multiple volatile compounds in their leaves. Most spices are commonly sold dried because of high water content in the fresh states which leads to intensive deterioration caused by microbial growth and biochemical reactions. Drying processes stabilizes spices microbiologically by reducing the water activity (aw) values under the threshold for microbial development (0.6) [8]. Some volatile compounds vaporize during air-drying, whereas others are somewhat retained, and some oxidation products appear during drying.

When spices are used as marinades or as seasoning for foods, the dried vegetal crop is added directly. The volatile oil composition of both dill and savory has been widely studied [2, 3, 6, 7, 9–16]. The most abundant volatiles determined in dill are terpene compounds [17]. Three main compounds, carvone, α-phellandrene, and limonene, have been detected in dill aroma, but their amounts varied extensively [9]. Within these, α-phellandrene, the main compound of dill aroma greatly contributes to the sensory impression of dill herb [18]. In the case of savory, previous studies in various *Satureja* species indicated the big disagreement among both chemical compositions and their essential oils content.

For instance, the main compounds of the genus *Satureja* were carvacrol (40.8%) and γ-terpinene (26.4%) in *Satureja boissieri* oil from Turkey; pulegone (64.3%) and menthone (20.2%) in *Satureja brownei* oil from Venezuela; piperitone oxide in *Satureja parvifolia* oil from Argentina; γ-terpinene, β-caryophyllene and germacrene D in *Satureja boliviana* oil and menthone and isomenthone in *Satureja brevicalix* oil from Peru; neral, geranial and farnesene in *Satureja punctate*, thymol and carvacrol in *Satureja cuneifolia* oil, Germacrene D in *Satureja coerulea* oil from Turkey; carvacrol and γ-terpinene in *Satureja hortensis* oil and γ-terpinene (42.2%) and carvacrol (31.9%) in *S. sahendica Bornm*. oil from Iran [6, 12]. Volatiles are dependent on many factors, such as plant part, time of harvest, harvested plant part, extraction method, type of cultivar or genotype, geographic origin, storage conditions, and climate [19]. Drying is an effective method that prolongs the shelf life of the product by slowing the growth of microorganisms and preventing biochemical reactions that may impact sensory attributes [20].

There are very few surveys found on the isolation of volatile compounds directly from spices. Concerning the European Pharmacopoeia [21], volatile oils are extracts separated from aromatic plants only by distillation procedures such as hydrodistillation. Further extracts obtained from aromatic plants by other techniques, even entailing volatile compounds, should not be considered as volatile oils [14]. Hydrodistillation (Clevenger and/or steam distillation apparatus) and organic solvent extraction (Soxhlet extractor) have both been extensively applied for the separation of volatile oils from spices as the conventional methods.

Nevertheless, these two extraction methods both tend to degrade the original spice flavour and lose volatile compounds owing to the high temperatures used. In other words, most of the flavour compounds in spices are consisted in their volatile oils and are changed or damaged upon heating or air oxidation [22]. Moreover, new flavours can be progressed from non-volatile precursors by Maillard reaction, carotenoid degradation, and lipid oxidation [23]. These limitations may be overcome using appropriate and novel extraction techniques. Thus, the appropriate extraction technique must be selected with the aim of producing aromatic extracts with odour as close as possible to that of the studied sample.

Under these circumstances, present research reports the results of shade-dried aerial parts of dill (*A. graveolens* L.) and savory (*S. sahendica* Bornm.) aroma profile collected from the city of Tabriz in East Azerbaijan Province, Iran. In this investigation, the solvent-assisted flavour extraction (SAFE) method was chosen as the aroma extraction method. Thanks to low pressure is used in this method, isolation of volatiles at low temperatures such as 40 °C can be achieved, which intercepts the formation of artifacts. It has already demonstrated its reliability for the extraction of volatile compounds in Iranian saffron and golpar spices [24, 25], orange juice [26], and coffee [27].

Gas chromatography (GC) coupled with flame ionization detector (FID) and mass spectrometry (MS) has been applied in quantification and identification of the aroma compounds, respectively.

Materials and methods

Plant material

The aerial parts of both dill (*A. graveolens* L.) and savory (*S. sahendica* Bornm.) were collected from Tabriz at the end of May and September in 2015. The collected materials were dried at room temperature away from sun light, packed into polyethylene bags, and kept at 4 °C for further use.

Isolation of volatile compounds

The isolation of volatiles was conducted in the dichloromethane being an efficacious solvent for the separation of volatiles in fruits and plants [28]. The volatiles exist in the Iranian dill and savory were isolated under vacuum (10^{-3} Pa; Vacuubrand DCP 3000, Wertheim, Germany) using the solvent-assisted flavour evaporation (SAFE) unit (Gläsblaserei Bahr, Manching, Germany). The isolation technique was altered from our earlier research [24]. Briefly, a monotonic powder of shade-dried aerial sections of both samples was separately prepared by applying porcelain mortar at room temperature. Before extraction, 7 g of each powdered sample containing 100 mL of dichloromethane plus 5 µL of 2-octanol as an internal standard were placed into a 500-mL flask. The details of the extraction technique were entirely explained in our earlier research [24, 25]. Each sample was extracted in triplicate. The concentrations of volatiles were calculated according to internal standard.

GC–FID and GC–MS analyses of volatile compounds

An Agilent 6890 GC was equipped with a FID (Wilmington, DE, USA) and an Agilent 5973-network-mass selective detector (MSD). The helium as a carrier gas was used with 1.5 mL/min flow rate. The oven start temperature was 50 °C (1 min), the following gradient was 5 °C/min, and then at a rate of 8 °C/min to 260 °C with a final hold at 260 °C for 5 min (DB-WAX column). GC effluent was split 1:1 among the FID and MSD. A mass spectra in the electron ionization mode were recorded at 70 eV and a mass/charge range of 30–300 amu at 2.0 scan s^{-1} scan rate. The identification of compounds was comprised of the following parameters: retention indices, commercial spectra database (Wiley 6 and NIST 98), an internal library created from our previous studies, and standard reference compounds.

Sensory and statistical analysis of the dill and savory plants and their extracts

Panel

The panel consisted of nine assessors (three females and six males between 25 and 49 years of age) from the Biotechnology Laboratory at the Department of Food Engineering, University of Cukurova. The assessors were familiar with the dill and savory aroma and also formerly trained in the scent distinction and sensory assessment methods, and had the experiences in the GC–MS.

Preparation and presentation of the samples

Various ways could be applied to estimate the representativeness of the aroma of odorous extracts belonging to this kind of study. In the current essay, a cardboard smelling strip (reference 7140 BPSI, Granger-Veyron, Lyas, France) was used to investigate the representativeness of the extracts acquired using the SAFE extraction technique. These cardboards have already evidenced positive outcomes for the representativeness test in the juice of orange [28] and Iranian saffron spice [24]. As a reference, a 1 g of each powdered sample was inserted in a 25 mL brown coded flask. Odorous extracts of samples acquired using the SAFE technique were adsorbed onto the cardboard. The detailed procedure was given in our earlier investigation [24].

Descriptive analysis

Two different lists separately for each plant such as nine descriptors composed of fresh, green, herbal, citrusy, minty, woody, dill, spicy and oily for dill and other nine descriptors consisted of fresh, green, herbal, citrusy, minty, woody, balsamic, floral and earthy for savory that describe their characteristic aroma were defined by the trained and expert panelists and subsequently applied to describe their extracts. More details are available in our previous study [24].

Statistical analysis

The statistical method used for the sensorial data analysis was an independent-samples analysis of variance (*t* test) to compare the sensory profile of each extract obtained from the SAFE method with that of their original samples. All the data in this experiment are presented as the average of nine replicates. Statistical analysis was

performed using SPSS statistics software version 22.0 (SPSS Inc., Chicago, Illinois, USA).

Results and discussion

Sensory analysis

Odor sensory profiles

The aromatic extracts of both plants isolated by the SAFE technique were compared to the reference samples, dill and savory plants, by the nine panelists. From the panelist group point of view, the outcomes of the similarity and intensity evaluation of the odorous isolation evidenced an extract representing the characteristic aroma of the plants, when a droplet of the aroma extract was vaporized on a strip of smelling paper. Figures 1 and 2 displays the two different principal intensity groupings of the both original samples and their extracts depicted on a spider graph applying nine descriptors. Each rating was separately applied by the panelists to depict the characteristic odor of the shade-dried aerial parts of the dill and savory plants and their extracts.

To the best of our knowledge, the descriptive analysis of these both samples were used for the first time to depict their characteristic odour. As can be demonstrated in Figs. 1 and 2, it appears that the sensory profile of each extract obtained from the SAFE method is analogous to that of their original samples. No variations were statistically detected between the aromatic extract using the SAFE isolation technique and original sample for the nine descriptors in both samples ($p < 0.01$). Of descriptors in the

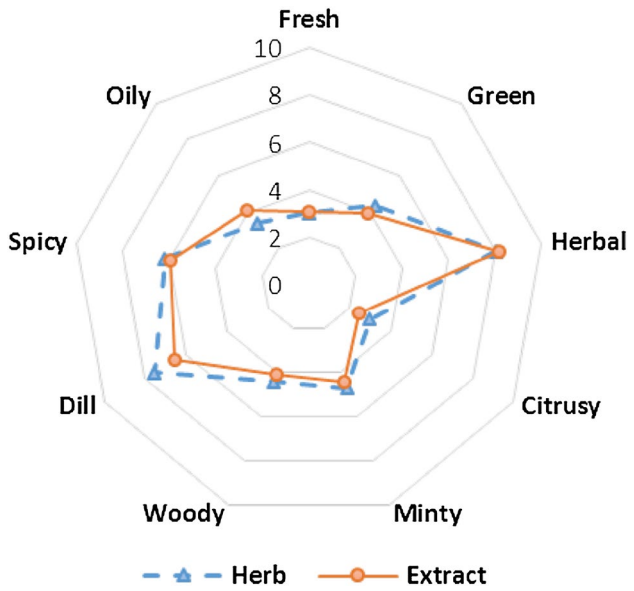

Fig. 1 Odour sensory features of the dill plant and its extract

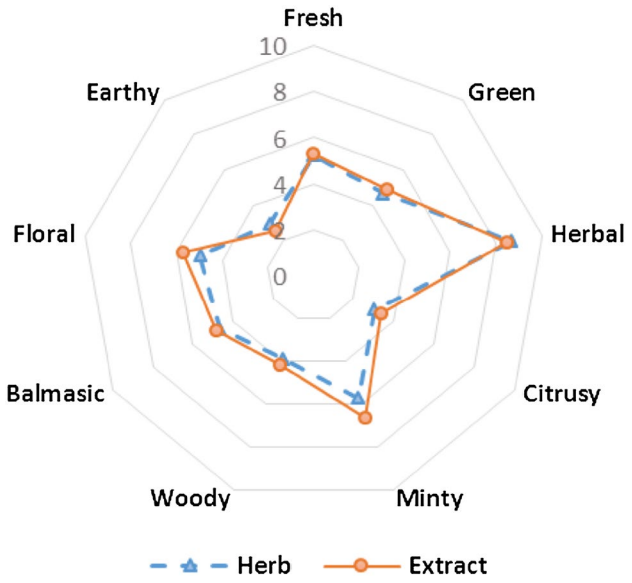

Fig. 2 Odour sensory features of the savory plant and its extract

dill sample (Fig. 1), herbal, dill and spicy aroma descriptors had the largest scores and the rest of the descriptors with the lowest scores were proved. In the case of the savory sample (Fig. 2), fresh, herbal, minty and floral odor descriptors with the highest scores and green, citrusy, woody, balsamic and earthy notes with the lowest scores were evidenced.

Aroma compositions of dill and savory

The volatile compounds identified in the shade-dried aerial parts of the dill and savory plants and linear retention index values on the DB-Wax column for these compounds were presented in Table 1. Mean values (µg/g) of the GC analyses of triplicate extractions and standard deviations were reported. A total of 40 and 26 compounds were identified and quantified in the dill and savory (Fig. 3), most of which have already been identified by previous studies in both different countries and diverse parts of dill and savory plant' volatile oil [2, 3, 6, 7, 9–16]. Dill contained 271.52 µg/g of the total amount of volatile compounds, which included terpenes (28), aldehydes (3), alcohol (1), acids (3), volatile phenol (3), ketone (1) and norisoprenoid (1).

Of all volatile compounds detected in the dill, terpenes were quantified as the most prevailing volatile compounds, followed by the acids (isovaleric, hexanoic and octanoic acid), aldehydes (nonanal, benzaldehyde and anisaldehyde), volatile phenols (isoanethole, anethol and isoeugenol), an alcohol (benzyl alcohol), a ketone (bornyl acetate) and a norisoprenoid (β-ionone epoxide). Savory contained 10,547.16 µg/g of the total concentration of volatile compounds, comprising the terpenes (20), an alcohol (1) and

Table 1 Aroma compounds of the shade-dried aerial parts of the Iranian dill (*A. graveolens L.*) and savory (*S. sahendica* Bornm.) using the SAFE technique

No.	Volatile compounds	LRI[a]	Concentration[b] (mean ± SD)		Identification[c]
			Dill	Savory	
1	α-Pinene	1012	4.49 ± 0.62	267.08 ± 0.08	LRI, MS, Std
2	α-Thujene	1017	nd	219.36 ± 0.95	LRI, MS, Std
3	β-Pinene	1100	0.66 ± 0.04	82.34 ± 0.32	LRI, MS, Std
4	*(+)*-2-Carene	1110	nd	82.04 ± 1.6	LRI, MS, tent
5	*(+)*-4-Carene	1128	nd	22.85 ± 0.48	LRI, MS, tent
6	α-Phellandrene	1167	160.0 ± 5.24	nd	LRI, MS, Std
7	α-Terpinene	1178	1.24 ± 0.01	nd	LRI, MS, Std
8	DL-Limonene	1186	12.3 ± 1.56	nd	LRI, MS, Std
9	Sabinene	1231	26.5 ± 3.24	nd	LRI, MS, Std
10	γ-Terpinene	1246	1.69 ± 0.06	6236.83 ± 2.5	LRI, MS, Std
11	*(E)*-β-Ocimene	1279	0.26 ± 0.03	nd	LRI, MS, Std
12	α-Terpinolene	1284	0.72 ± 0.06	10.34 ± 0.52	LRI, MS, Std
13	Nonanal	1388	0.30 ± 0.05	nd	LRI, MS, Std
14	α-Ocimene	1404	nd	1.06 ± 0.69	LRI, MS, tent
15	1-Octen-3-ol	1445	nd	9.72 ± 1.27	LRI, MS, Std
16	*(E)*-Sabinene hydrate	1460	0.82 ± 0.14	17.89 ± 1.29	LRI, MS, Std
17	*(Z)*-Sabinene hydrate	1466	nd	23.1 ± 1.69	LRI, MS, Std
18	L-Menthone	1474	2.62 ± 1.92	nd	LRI, MS, Std
19	Benzaldehyde	1516	0.40 ± 0.05	nd	LRI, MS, Std
20	Dill ether	1521	7.80 ± 0.35	nd	LRI, MS, tent
21	α-Copaene	1526	0.70 ± 0.38	nd	LRI, MS, Std
22	Isopulegone	1540	0.30 ± 0.04	nd	LRI, MS, tent
23	Bornyl acetate	1544	0.44 ± 0.08	nd	LRI, MS, tent
24	Dihydrocarvone	1549	1.03 ± 0.14	nd	LRI, MS, Std
25	Calarene	1554	1.82 ± 0.91	nd	LRI, MS, Std
26	1-Terpineol	1575	0.43 ± 0.06	nd	LRI, MS, Std
27	4-Terpineol	1579	nd	7.54 ± 0.51	LRI, MS, Std
28	Methylcarvacrol	1590	nd	11.07 ± 0.3	LRI, MS, tent
29	*(E)*-Caryophyllene	1596	nd	98.4 ± 1.48	LRI, MS, Std
30	Aromadendrene	1627	nd	22.57 ± 0.3	LRI, MS, tent
31	α-Caryophyllene	1632	nd	5.75 ± 0.48	LRI, MS, Std
32	Pulegone	1644	6.67 ± 0.56	nd	LRI, MS, tent
33	Isoborneol	1664	nd	10.7 ± 0.32	LRI, MS, Std
34	Isovaleric acid	1665	1.62 ± 0.61	nd	LRI, MS, Std
35	Isoanethole	1670	2.06 ± 0.14	nd	LRI, MS, tent
36	Carvotanacetone	1683	0.45 ± 0.16	nd	LRI, MS, tent
37	δ-Elemene	1688	1.00 ± 0.30	nd	LRI, MS, Std
38	α-Terpineol	1691	nd	12.8 ± 1.47	LRI, MS, Std
39	β-Bisabolene	1720	nd	130.8 ± 1.93	LRI, MS, Std
40	D-Carvone	1726	16.2 ± 2.21	nd	LRI, MS, Std
41	β-Cubebene	1761	1.19 ± 1.26	nd	LRI, MS, Std
42	β-sesquiphellandrene	1762	nd	1.55 ± 0.12	LRI, MS, tent
43	*(Z)*-γ-Bisabolene	1767	nd	5 ± 0.04	LRI, MS, tent
44	α-Curcumene	1773	0.60 ± 0.11	nd	LRI, MS, tent
45	Anethol	1833	0.17 ± 0.01	nd	LRI, MS, Std
46	Hexanoic acid	1840	3.22 ± 0.90	nd	LRI, MS, Std
47	Benzyl alcohol	1870	0.49 ± 0.14	nd	LRI, MS, Std
48	Carvacrol acetate	1880	nd	3.47 ± 0.67	LRI, MS, Std
49	β-Ionone epoxide	1957	3.82 ± 1.32	nd	LRI, MS, tent

Table 1 (continued)

No.	Volatile compounds	LRI[a]	Concentration[b] (mean ± SD)		Identification[c]
			Dill	Savory	
50	Anisaldehyde	2023	0.16 ± 0.05	nd	LRI, MS, Std
51	*ρ*-cresol	2078	nd	0.26 ± 0.08	LRI, MS, Std
52	Octanoic acid	2083	0.40 ± 0.12	nd	LRI, MS, Std
53	Eugenol	2169	nd	0.25 ± 0.16	LRI, MS, Std
54	Isoeugenol	2180	0.18 ± 0.19	nd	LRI, MS, Std
55	Thymol	2188	0.48 ± 0.18	25.2 ± 0.3	LRI, MS, Std
56	Carvacrol	2206	0.45 ± 0.16	3239.19 ± 1.44	LRI, MS, Std
57	*ρ*-Cumenol	2211	0.94 ± 0.84	nd	LRI, MS, Std
58	Myristicin	2216	0.43 ± 0.23	nd	LRI, MS, tent
59	Dill apiole	2226	6.66 ± 2.77	nd	LRI, MS, tent
	TOTAL		**271.52**	**10547.16**	

[a]LRI—Retention indices on DB-WAX column

[b]Concentration—Mean values based on three repetitions as μg/g; *nd* not determined

[c]Identification: Methods of identification; *LRI* linear retention index, *MS tent*. tentatively identified by MS, *Std* chemical standard; When only MS or LRI is available for the identification of a compounds, it must be considered as an attempt of identification

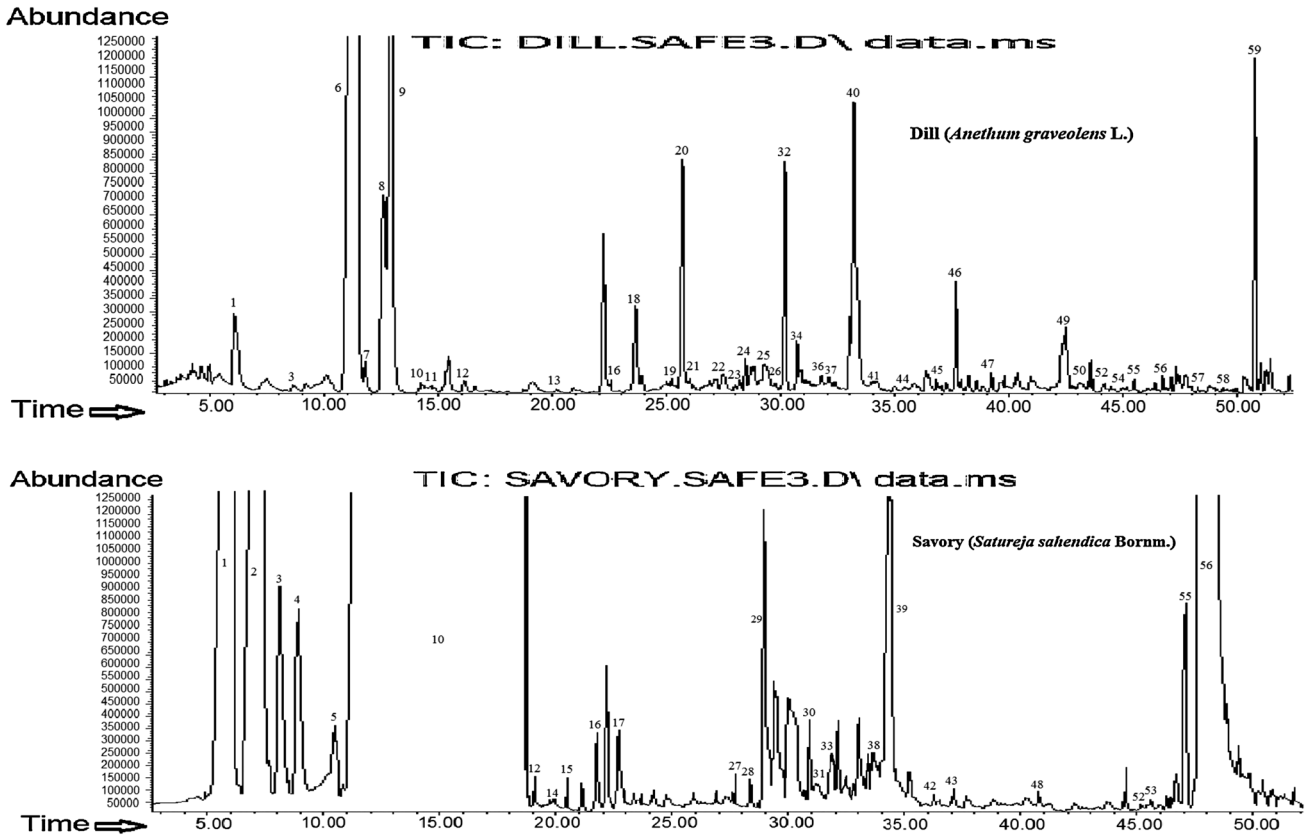

Fig. 3 The gas chromatography-mass spectrometry (GC–MS) chromatograms of Dill and Savory (peak numbers refer to aroma compounds represented in Table 1)

volatile phenols (5). The most prevailing volatile compounds in savory extracts were terpenes, followed by volatile phenols (methylcarvacrol, carvacrol acetate, *ρ*-cresol, eugenol and thymol) and an alcohol (1-octen-3-ol). Aromatic plants are mostly used in the folk medicine, and volatile oils and volatile compounds isolated from them are extensively used as the antioxidants and anti-inflammatory agents. Several anti-inflammatory, digestive, antinecrotic, neuroprotective, and hepatoprotective drugs have recently been demonstrated to possess an antioxidant and/or radical-scavenging mechanism as a section of their activity [29].

In the overall aromatic compounds of the dill in this study, the *α*-phellandrene (160.0 µg/g) is the major compound together with the sabinene (26.5 µg/g), D-carvone (16.2 µg/g), DL-limonene (12.3 µg/g), dill ether (7.80 µg/g), pulegone (6.67 µg/g), dill apiole (6.66 µg/g), *α*-pinene (4.49 µg/g), *β*-ionone epoxide (3.82 µg/g) and hexanoic acid (3.22 µg/g). In general, the *α*-phellandrene, D-carvone and DL-limonene are impact compounds of many essential dill oils and are responsible especially for the aroma and biological effects [10]. The most abundant volatiles determined in the dill are terpenes [17]. *α*-Phellandrene, the main compound of dill aroma greatly contributes to the sensory impression of the dill herb [18].

In our research, terpenes were also quantitatively (258.4 µg/g) the leading volatile compounds in the dill, representing 95.2% of the total volatile profile analyzed. The amount of the *α*-phellandrene (58.93%) shown in Table 1 is much higher than that reported in the Iranian dill essential oil (19.1%) [30]. Of all aroma compounds discovered in the savory, the *γ*-terpinene (6236.83 µg/g) is the major compound along with the carvacrol (3239.19 µg/g), *α*-pinene (267.08 µg/g), *α*-thujene (219.36 µg/g) and *β*-bisabolene (130.8 µg/g). In an earlier study, the investigated *S. sahendica* Bornm volatile oil using the hydrodistillation extraction method was detected to be rich in terpenes with substantial contents of the *γ*-terpinene and carvacrol, and they contained approximately 77% of the determined compounds and were the most predominant compounds of the oil [6]. *γ*-Terpinene as a volatile oil indicates antimicrobial characters against different human pathogens [31]. The antioxidant, anti-inflammatory, and anti-proliferative activities of *γ*-terpinene is also investigated [32]. There are diverse agents that can affect the volatile compounds during the drying process; for instance, the temperature reached, the interaction among volatiles and water vapor, and the hydrophobic nature of volatiles [33].

Terpenes

Terpenes quantitatively represent the main class of aroma compounds in both tested samples, accounting for 95.18 and 68.8% of the total aroma compounds determined in the dill and savory. Plenty of the terpene compounds are direct products of terpene synthases, while others are formed through changes of the primary terpene skeletons made by terpene synthases by the hydroxylation, dehydrogenation, acylation, and other reactions [34].

α-Pinene together with the *β*-pinene, *α*-phellandrene, *α*-terpinene, DL-limonene, sabinene, *γ*-terpinene, *(E)-β*-ocimene, *α*-terpinolene, L-menthone, *(E)*-sabinene hydrate, dill ether, *α*-copaene, isopulegone, dihydrocarvone, calarene, 1-terpineol, pulegone, carvotanacetone, *δ*-elemene, D-carvone, *β*-cubebene, *α*-curcumene, thymol, carvacrol, *ρ*-cumenol, myristicin and dill apiole were determined in the dill sample. Most of these terpenes were identified in the dill from different countries in previous studies [1, 3, 9, 35, 36]. Among the higher terpenes, *α*-phellandrene (160 µg/g) was detected as the highest proportion in the dill aromatic extract, followed by the sabinene (26.5 µg/g), D-carvone (16.2 µg/g), DL-limonene (12.3 µg/g), and dill ether (7.8 µg/g). α-Phellandrene also was the main compound of all the three studied fresh dill cultivars (Dura, Dukat and Mammut) grown at Sahalahti of Finland in 1983 and 1984 using a modified Soxhlet technique [35] and dill leaves from Latvia using the solid phase micro-extraction technique [1]. The proportion of α-phellandrene (58.93%) was approximately at the identical concentration level (57.10%) as detected earlier in microwave vacuum dried dill stems [1], and much higher than the proportions of 19.12 and 14.68% revealed in aerial parts using the hydrodistillation [30] and fresh leaves by the ultrasound-assisted extraction [36].

The relative extent of the α-phellandrene from the total aroma content in the three different cultivars of the dill, Dura, Dukat and Mammut, grown at two diverse regions in Finland, Sahalahti and Viikki, was comparatively constant between 40–60% and 30–50%, respectively [35]. *α*-Phellandrene gives a typical dill flavor and also fragrant and fresh notes [3]. As can be indicated in Fig. 1, the dill aroma descriptor was also distinguished in the highest score by the panel. Fresh dill aroma is composed of synergistic communication contributed mainly by α-phellandrene with an altering efficacy from dill ether [1]. Three fundamental compounds, carvone, *α*-phellandrene, and limonene, have been ascertained in the dill aroma, but their contents differed extensively, belonging to the diverse agents such as the geographical origin, ripening degree and growth situations.

Additionally, the extraction technique could also influence the volatile oil amount and composition [35]. Dill seed oil is distinguished by a high extent of carvone and limonene [2–6], while the aerial part oil of the herb consists, in addition to carvone and limonene, remarkable values of α-phellandrene [9, 35]. Carvone reveals caraway-like and cooling notes in the dill herb volatile oil [3]. According

to the geographical origin, the content of carvone in the herb volatile oil has been evidenced to differ from 6 to 44%, while its content in the dill seed volatile oil is more constant at 40–55% [9]. The amount of the limonene as another important terpene compound in dill leaves is more than in its stems and it liberates lemon and mint aroma [1].

α-Pinene along with the *α*-thujene, *β*-pinene, *(+)*-2-carene, *(+)*-4-carene, *γ*-terpinene, *α*-terpinolene, *α*-ocimene, 1-octen-3-ol, *(E)*-sabinene hydrate, *(Z)*-sabinene hydrate, 4-terpineol, methylcarvacrol, *(E)*-caryophyllene, aromadendrene, *α*-caryophyllene, *α*-terpineol, isoborneol, *β*-bisabolene, *β*-sesquiphellandrene, *(Z)*-*γ*-bisabolene, carvacrol acetate, *ρ*-cresol, eugenol, thymol and carvacrol were detected in the savory as terpene compounds. Most of these were discovered in the savory from various countries and species in earlier investigations [6, 7, 11–16]. Generally, volatile oils have mainly a cytotoxic activity because of the presence of thymol, carvacrol, α-pinene and *ρ*-cymene compounds [36]. *γ*-Terpinene (6236.83 µg/g) (59.13%) as indicated in Table 1 was the most abundant compound in savory samples followed by carvacrol (3239.19 µg/g) (30.71%), *α*-pinene (267.08 µg/g) (2.53%), *α*-thujene (219.36 µg/g) (2.08%) and *β*-bisabolene (130.8 µg/g) (1.24%).

The identical results were approximately evidenced in an earlier research on air-dried aerial parts of savory from the Maragheh region in the Northwest Iran using the hydro distillation technique. Particularly, γ-terpinene (42.2%) and carvacrol (31.9%) consisted of approximately 77% of the distinguished compounds and were the most prevailing compounds of the volatile oil [6]. Disagreements among chemical combinations and amounts of various *Satureja* species volatile oils were also indicated concerning the literature review. Carvacrol (40.8%) and *γ*-terpinene (26.4%) in *S. boissieri* volatile oil from Turkey; pulegone (64.3%) and menthone (20.2%) in *S. brownie* from Venezuela; *γ*-terpinene, *β*-caryophyllene and germacrene D in *S. boliviana* and piperitone oxide in *S. parvifolia* from Argentina; germacrene D in *S. coerulea* from Turkey and carvacrol and *γ*-terpinene in *S. hortensis* from Iran were characterized as conquering compounds [7].

Considering the isolation technique in the volatile oil of *S. sahendica*, thymol (37.2%), *ρ*-cymene (32.6%) and *γ*-terpinene (11.5%) via the hydrodistillation technique and the similar compounds, but with various contents, thymol (66.0%), *ρ*-cymene (20.3%) and *γ*-terpinene (3.6%) by steam distillation technique were detected as the main compounds [11]. Additionally, *α*-pinene, *β*-pinene, *α*-thujene and *β*-myrcene in the steam distilled volatile oil was not observed [11]. In another study on *Satureja hortensis* volatile oil, the outcomes demonstrated that the hydrodistillation revealed the largest content of carvacrol (46.0%), while the steam distillation released the lowest content of carvacrol (12.3%) and the highest content of γ-terpinene (70.4%) [13]. Carvacrol (52.2–62.0%), thymol (8.6–11.0%), *ρ*-cymene (6.9–12.8%), *γ*-terpinene (6.4–9.4%) and *β*-bisabolene (2.0–2.7%) were the principal compounds in *Satureja montana* using hydrodistillation. Supercritical fluid extraction volatile oil contains of the similar major compounds but with diverse contents as carvacrol (41.7–64.5%), thymol (6.0–11.3%), *ρ*-cymene (6.0–17.8%), *γ*-terpinene (2.3–6.0%) and *β*-bisabolene (2.2–3.5%) [16].

According to the different parts of *S. sahendica* volatile oil, thymol (31.5%), *γ*-terpinene (29.33%) and *ρ*-cymene (23.48%) in the cluster volatile oil and *ρ*-cymene (44.88%), thymol (28.22%) and *γ*-terpinene (10.07%) in the leaf and stem volatile oil were shown as the principal compounds [37]. On the basis of different localities, thymol (19.6–41.6%), *ρ*-cymene (32.5–54.9%) and *γ*-terpinene (1–12.8%) were the major compounds in *S. sahendica* volatile oil from eight localities [7]. Based on the drying method in *S. hortensis* volatile oil extracted by the hydrodistillation, carvacrol (46.0%), *γ*-terpinene (37.7%), *ρ*-cymene (4.2%) and *α*-terpinene (3.1%) in shade-dried aerial parts, carvacrol (46.8%), *γ*-terpinene (39.4%), *ρ*-cymene (4.4%) and *α*-terpinene (3.3%) in sun-dried aerial parts and carvacrol (48.1%), *γ*-terpinene (38.4%), *ρ*-cymene (3.5%) and *α*-terpinene (3.4%) in oven-dried aerial parts at 45 °C were the major compounds [13]. The main compounds of *Satureja horvatii* volatile oil in accordance with the growing regions were thymol (63.7%), *γ*-terpinene (7.5%), carvacrol methyl ether (4.9%), carvacrol (4.7%), and *ρ*-cymene (4.5%) in Orjenske Lokve, Mt. Orjen, carvacrol (68.1%), *ρ*-cymene (8.3%) and *γ*-terpinene (5.8%) in Mt. Lovcen [15]. *γ*-Terpinene with the oily, woody, terpene lemon/lime, tropical and herbal, and carvacrol with the spicy, woody, camphor and thymol aroma characters were proved in *Satureja myrtifolia* aerial parts from the Lebanon origin [38].

Conclusion

The current investigation was allocated to define the aroma compounds in the shade-dried aerial parts of the Iranian dill (*A. graveolens L.*) and savory (*S. sahendica* Bornm.) plants. Fourty and twenty six aroma volatiles, in total, were specified in the dill and savory, respectively, using the GC–MS and GC–FID. The SAFE isolation technique gave highly representative aromatic extract in both analyzed samples. Dill with 271.52 µg/g aroma compounds consisted of the terpenes (28), aldehydes (3), an alcohol (1), acids (3), volatile phenols (3), a ketone (1) and norisoprenoid (1) and savory with 10547.16 µg/g aroma compounds composed of the terpenes (20), an alcohol (1) and volatile phenols (5). From all aroma compounds found in both samples,

terpenes were quantitatively the most prevailing. Within these compounds, α-phellandrene (160.0 μg/g) followed by sabinene (26.5 μg/g), D-carvone (16.2 μg/g), DL-limonene (12.3 μg/g) and dill ether (7.8 μg/g) in dill and γ-terpinene (6236.83 μg/g) followed by carvacrol (3239.19 μg/g), α-pinene (267.08 μg/g), α-thujene (219.36 μg/g) and β-bisabolene (130.8 μg/g) in savory were quantitatively demonstrated as the principal compounds.

References

1. Z. Kruma, R. Galoburda, M. Sabovics, I. Gramatina, I. Skudra, I. Dabina-Bicka, Aroma composition of microwave vacuum dried dill (*Anethum graveolens* L.) stems. Proc. Food Sci. **1**, 1338–1343 (2011)
2. L. Jirovetz, G. Buchbauer, A.S. Stoyanova, E.V. Georgiev, A. Damianova, Composition, quality control, and antimicrobial activity of the essential oil of long-time stored dill (*Anethum graveolens* L.) seeds from Bulgaria. J. Agric. Food Chem. **51**, 3854–3857 (2003)
3. J.A. Pino, A. Rosado, I. Goire, E. Roncal, Evaluation of flavor characteristic compounds in dill herb essential oil by sensory analysis and gas chromatography. J. Agric. Food Chem. **43**, 1307–1309 (1995)
4. A. Ortan, M.L. Popescu, A.L. Gaita, C. Dinu-Pirvu, Gh.. Campeanu, Contributions to the pharmacognostical study on *Anethum graveolens*, dill (*Apiaceae*). Rom. Biotechnol. Lett. **14**, 4342–4348 (2009)
5. M. Yousefzadi, A. Riahi-Madvar, J. Hadian, F. Rezaee, R. Rafiee, In vitro cytotoxic and antimicrobial activity of essential oil from *Satureja sahendica*. Environ. Toxicol. Chem. **94**, 1735–1745 (2012)
6. M.B. Hassanpouraghdam, M.S. Shalamzari, M.A. Aazami, A.M. Shoja, γ-Terpinene and carvacrol rich volatile oil of *Satureja sahendica* Bornm. from Maragheh district in Northwest Iran. Chemija. **20**, 186–189 (2009)
7. F. Sefidkon, Z. Jamzad, M. Mirza, Chemical variation in the essential oil of *Satureja sahendica* from Iran. Food Chem. **88**, 325–328 (2004)
8. M. Usai, M. Marchetti, M. Foddai, A. Del Caro, R. Desogus, I. Sanna, A. Piga, Influence of different stabilizing operations and storage time on the composition of essential oil of thyme (*Thymus officinalis* L.) and rosemary (*Rosmarinus officinalis* L.). LWT: Food. Sci. Technol. 44, 244–249 (2011)
9. R. Huopalahti, R.R. Linko, Composition and content of aroma compounds in dill, *Anethum graveolens* L., at three different growth stages. J. Agric. Food Chem. **31**, 331–333 (1983)
10. L. Jirovetz, G. Buchbauer, A. Nikiforov, Composition of the essential oil of dill, celery, and parsley from Estonia/Eestis kasvatatud tilli, selleri ja peterselli eeterliku oli koostis. Ernahrung/Nutrition. **18**, 534–536 (1994)
11. A.R. Gohari, M.S. Nourbakhsh, Saeidnia, Comparative investigation of the volatile oils of satureja sahendica bornm., extracted via steam and hydro distillations. J. Essent. Oil Bear. Plant **14**, 751–754 (2011)
12. F. Sefidkon, A. Akbarinia, Essential oil content and composition of *Satureja sahendica* Bornm. at different stages of plant growth. J. Essent. Oil Res. **21**, 112–114 (2009)
13. F. Sefidkon, K. Abbasi, G. Bakhshi Khaniki, Influence of drying and extraction methods on yield and chemical composition of the essential oil of *Satureja hortensis*. Food Chem. **99**, 19–23 (2006)
14. C. Grosso, J.A. Coelho, J.S. Urieta, A.M.F. Palavra, J.G. Barros, Herbicidal activity of volatiles from coriander, winter savory, cotton lavender, and thyme isolated by hydrodistillation and supercritical fluid extraction. J. Agric. Food Chem. **58**, 11007–11013 (2010)
15. B. Lakusic, M. Ristic, V. Slavkovska, M. Milenkovic, D. Lakusic, Environmental and seasonal impacts on the chemical composition of *Satureja horvatii Silic* (*Lamiaceae*) essential oils. Chem. Biodivers. **8**, 483–493 (2011)
16. C. Grosso, A.C. Figueiredo, J. Burillo, A.M. Mainar, J.S. Urieta, J.G. Barroso, J.A. Coelho, A.M.F. Palavra, Enrichment of the thymoquinone content in volatile oil from *Satureja montana* using supercritical fluid extraction. J. Sep. Sci. **32**, 328–334 (2009)
17. P.J. Delaquis, K. Stanich, B. Girard, G. Mazza, Antimicrobial activity of individual and mixed fractions of dill, cilantro, coriander and eucalyptus essential oils. Int. J. Food Microbiol. **74**, 101–109 (2002)
18. M.H. Boelens, Spices and condiments II. in *Volatile Compounds in Food and Beverages*, ed. by H. Marsee (Marcel Deker, New York, 1991)
19. M.A. Mitu, D. Lupuliasa, C.E. Dinu-Pirvu, F.S. Rădulescu, D.S. Miron, L. Vlaia, Ketoconazole in topical pharmaceutical formulations. The influence of the receptor media on the in vitro diffusion kinetics. Farmacia. 59, 358–366 (2011)
20. M.C. Diaz-Maroto, M.S. Perez Coello, M.A. Gonzalez Vinas, M.D. Cabezudo, Influence of drying on the flavor quality of spearmint (*Mentha spicata* L.). J. Agric. Food Chem. **51**, 1265–1269 (2003)
21. Council of Europe (COE), European Directorate for the Quality of Medicines, *European Pharmacopoeia*, 6th edn. (United States Pharmacopeial Convention Inc., Strasbourg, 2007)
22. K. Hirasa, M. Takemasa, *Spice Science and Technology*. (CRC Press, Boca Raton, 1998)
23. R. A Flath, H. Sugisawa, R. Teranishi, Introduction – Problems in Flavour Research. in *Food Flavours: Biology and Chemistry*, ed. by C. Fisher, T.R. Scott, (Royal Society of Chemistry, London, 2006), pp. P001–P004
24. A. Amanpour, A.S. Sonmezdag, H. Kelebek, S. Selli, GC-MS-olfactometric characterization of the most aroma-active components in a representative aromatic extract from Iranian saffron (*Crocus sativus* L). Food Chem **182**, 251–256 (2015)
25. A. Amanpour, H. Kelebek, S. Selli, Aroma components of Iranian dried *Heracleum persicum* fruit (golpar) using solvent-assisted flavour evaporation technique. J. Food Nutr. Res. 55, 141–147 (2016)
26. A. Beuttner, P. Schieberle, Changes in the concentrations of key orange odorants induced by mastication of orange segments and orange juice. Abstr. Pap. Am. Chem. Soc. **218**, U47–U48 (1999)
27. C. Sanz, M. Czerny, C. Cid, P. Schieberle, Comparison of potent odorants in a filtered coffee brew and in an instant coffee beverage by aroma extract dilution analysis (AEDA). Eur. Food. Res. Technol. **214**, 299–302 (2002)
28. H. Kelebek, S. Selli, Determination of volatile, phenolic, organic acid and sugar components in a Turkish cv. Dortyol (*Citrus sinensis* L. Osbeck) orange juice. J. Sci. Food Agric. **91**, 1855–1862 (2011)
29. M.G. Repetto, S.F.L. Braz, Antioxidant properties of natural compounds used in popular medicine for gastric ulcers. J. Med. Biol. Res **35**, 523–534 (2002)
30. M. Kazemi, Effect of *Carum copticum* essential oil on growth and aflatoxin formation by *Aspergillus* strains. Nat. Prod. Res. **29**, 1065–1068 (2015)
31. T.S. Pereira, J.R. de Sant Anna, E.L. Silva, A.L. Pinheiro, M.A.A. de Castro-Prado, In vitro genotoxicity of *Melaleuca*

alternifolia essential oil in human lymphocytes. J. Ethnopharmacol. **151**, 852–857 (2014)
32. B. Bayala, I.H.N. Bassole, Ch. Gnoula, R. Nebie, A. Yonli, L. Morel, G. Figueredo, J.B. Nikiema, J.M.A. Lobaccaro, J. Simpore, Chemical composition, antioxidant, anti-inflammatory and anti-proliferative activities of essential oils of plants from Burkina Faso. PloS ONE **9**, 92122 (2014)
33. A. Figiel, A. Szumny, A. Gutierrez-Ortiz, A.A. Carbonell-Barrachina, Composition of oregano essential oil (*Origanum vulgare*) as affected by drying method. J. Food Eng. **98**, 240–247 (2010)
34. N. Dudareva, E. Pichersky, J. Gershenzon, Biochemistry of plant volatiles. Plant Physiol. **135**, 1893–1902 (2004)
35. R. Huopalahti, The content and composition of aroma compounds in three different cultivars of dill, *Anethum graveolens* L. Z. Lebensm. Unters. Forsch. **181**, 92–96 (1985)
36. M. Stan, M.L. Soran, C. Varodi, I. Lung, L. Copolovici, C. Marutoiu. Extraction and GC determination of volatile aroma compounds from extracts of three plant species of the Apiaceae family. AIP Conf. Proc. **1565**, 75–78 (2013)
37. A. Tabatabaei-Raisi, A. Delazar, A. Khalighi, B. Kaviani, D. hashemabadi, Variability of Essential oils of various parts of *Satureja sahendica* Bornm.and their antioxidant activity. Int. J. Bot. **4**, 245–248 (2008)
38. B. Zebib, M.E.L. Beyrouthy, C. Safi, O. Merah, Chemical composition of the essential oil of *Satureja myrtifolia* (Boiss. & Hohen.) from Lebanon. J. Essent. Oil Bear. Plants **18**, 248–254 (2015)