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Chemotypes investigation of essential oils of Chamomile herbs : A short review

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Abstract
"Chamomile" herbs are shared by the following genus, *Anthemis*, *Chamaemelum*, *Cladanthus* and *Matricaria* (Asteraceae). They are used as ornemental and medicinal plants around the world. Essential oils of Chamomile are traditionally used as analgesic. Our investigation on chemotypes of essential oils of *Chamomile*, worldwide distributed, showed the main presence of *cis*-*β*-farnesene (0.9-65.9%), spathulenol (1.3-19.4%), *α*-bisabolol oxide B (0.1-25.1%), *α*-bisabololone oxide A (0.1-29.9%), *α*-bisabolol (0.1-56.9%), chamazulene (0.2-25.5%) and *α*-bisabolol oxide A (0.1-55.9%).

Keywords: Asteraceae, chemotype, *Anthemis, Chamaemelum*, *Cladanthus*, *Matricaria*, *Chamomile.*

1. Introduction
Plants named "*Chamomile*", in Arabic "*babounj*", belong to the genus *Anthemis, Chamaemelum, Cladanthus* and *Matricaria* (Asteraceae) [1]. Chamomile has been recommended for a variety of healing applications and is included in the FDA's (US Food and Drug administration) GRAS (generally recognized as safe) list [2]. There has been an increase interest in the use of natural substances instead of synthetic chemicals [3], due to the fact that many herbal medicines are free from side effects and seen as 'green' so that's advisable to cultivate medicinal plants because of the possibility to better control quality of the target bioactive components [4]. The species *Matricaria chamomilla* (German chamomile), *Anthemis nobilis* syn. *Chamaemelum nobilis* (Roman chamomile) and *Cladanthus mixtus*.L (Moroccan Chamomile) are usually referred to the common name '*Chamomile*' [5; 6]. Nowadays, chamomile is a highly favored and much-used medicinal plant in folk medicine throughout the world as an antispasmodic [7]. Essential oils of chamomile herbs have been reported for their anti-inflammatory, antiseptic, healing, stimulative, carminative, spasmolytic and sedative activities [8; 9]. In continuation of our works on Asteraceae, we report here a compilative investigation of the chemotypes of essential oils of "Chamomile" herbs from Asteraceae family.

2. Material and Methods
Plant material
The investigated samples data are presented in table 1.

Extraction
The essentials oils were obtained by hydrodistillation, for 3 hours, using a Clevenger-type apparatus, then dried with anhydrous sodium sulfate and stored at 4°C, until analyzed by GC and GC/MS.

Table 1 Plant material data of studied *chamomile* essential oil

Code	Species (aerial parts)	Locality	Ref
Ch1	*Anthemis nobilis* L.	Sậo Paulo (Brazil)	[10]
Ch2	*Anthemis nobilis* L. ("White-headed" variety)	Casola Valsenio (Italy)	[11]
Ch3	*Anthemis nobilis* L.	Casola Valsenio (Italy)	[11]
Ch4	*Anthemis nobilis* L.	Chaffant-saint-jurson (France)	[12]
Ch5	*Chamaemelum nobile* L. (Roman chamomile)	Anjou (France)	[13]
Ch6	*Chamaemelum nobile* L.	Firoozabad (Iran)	[14]
Ch7	*Chamaemelum nobile* L.	Podgorica (Montenegro)	[15]
Ch8	*Cladanthus mixtus* L. Chevall	Bouznika (Morocco)	[16]
Ch9	*Cladanthus mixtus* L. Chevall	Oujda (Morocco)	[16]
Ch10	*Cladanthus mixtus* L. Chevall	Chefchouane (Morocco)	[16]
Ch11	*Cladanthus mixtus* L. Chevall	Sidi Allal Albahraoui (Morocco)	[16]
Ch12	*Matricaria chamomilla* L. (German chamomile)	Kenitra (Morocco)	[17]
Ch13	*Matricaria chamomilla* L.	Fayoum (Egypt)	[18]
Ch14	*Matricaria chamomilla* L.	Neyshabur (Iran)	[19]
Ch15	*Matricaria chamomilla* L.	Fars (Iran)	[20]
Ch16	*Matricaria chamomilla* L.	Golestan (Iran)	[21]
Ch17	*Matricaria chamomilla* L.	Kazeroon (Iran)	[21]
Ch18	*Matricaria chamomilla* L.	Hamedan (Iran)	[21]
Ch19	*Matricaria chamomilla* L.	Tahran (Iran)	[21]
Ch20	*Matricaria chamomilla* L.	Tahran (Iran)	[22]
Ch21	*Matricaria chamomilla* L.	Meshehad (Iran)	[23]
Ch22	*Matricaria chamomilla* L.	Isfahan (Iran)	[24]
Ch23	*Matricaria recutita* L. Rauschert (German chamomile)	Cairo (Egypt)	[25]
Ch24	*Matricaria recutita* L. Rauschert	Tartu (Estonia)	[26]
Ch25	*Matricaria recutita* L. Rauschert	Loodusravi (Estonia)	[27]
Ch26	*Matricaria recutita* L. Rauschert	Teekanne (Germany)	[27]
Ch27	*Matricaria recutita* L. Rauschert	Wilken Tee (Germany)	[27]
Ch28	*Matricaria recutita* L. Rauschert	Wallerstein (Germany)	[28]
Ch29	*Matricaria recutita* L. Rauschert	Isfahan (Iran)	[29]
Ch30	*Matricaria recutita* L. Rauschert	Laplanta (Latvia)	[27]
Ch31	*Matricaria recutita* L. Rauschert	Tea Trader (Latvia)	[27]
Ch32	*Matricaria recutita* L. Rauschert	Švenčionių vaistažolės (Lithuania)	[27]
Ch33	*Matricaria recutita* L. Rauschert	Rimi order (The Neatherlands)	[27]
Ch34	*Matricaria recutita* L. Rauschert	Belin (Poland)	[27]
Ch35	*Matricaria recutita* L. Rauschert	Eko Natura (Poland)	[27]
Ch36	*Matricaria recutita* L. Rauschert	Radawiec (Poland)	[30]
Ch37	*Matricaria recutita* L. Rauschert	Bigelow (USA)	[27]
Ch38	*Matricaria suaveolens* L.	Harjumaa,Virla (Estonia)	[26]
Ch39	*Matricaria pubescens* (Desf.) Sch. Bip.	Ghardaia (Algeria)	[31]

3. Discussion

From Table 2, concerning chamomile herbs related to *Anthemis* genus (*A. nobilis*), the essential oils of Italian chamomile (**Ch2**, **Ch3)** were mainly characterized by n-butylangelate hexenylacetate (34.2%, 14.5%), 2-methylbutyl-2-ethylbutyrate (7.3%, 9.2%) and isoamyl angelate (22.8%, 19.4%). The latter was mainly found in chamomile growing in France (**Ch4**) (7.6%) together with the esters of angelic acid (isobutyl: 24.5%, 2-butenyl:

7.3 % and 2-methyl butyl: 17.4%) and estragol (5%). However, the chemical composition of the essential oil of Brazilian chamomile (**Ch1)** is quite different with the main presence of 1,8-cineole (14%), γ-muurolene (8.3%), *trans*-geraniol (6.7%) and *β*-bisabolene (5%).

Table 2. Percentages of major components (≥5%) of essential oils of *chamomile of Anthemis* genus

Compounds*	Ch1	Ch2	Ch3	Ch4
1,8-Cineole	14.0	-	-	-
n-Butyl angelate hexenyl acetate	-	34.2	14.5	-
2-Methyl butyl-2-ethyl butyrate	-	7.3	9.2	-
Isoamyl angelate	-	22.8	19.4	7.6
trans-Geraniol	6.7	-	-	-
ϒ-Muurolene	8.3	-	-	-
β-Bisabolene	5.0	-	-	-
Isobutyl angelate	-	-	-	24.5
2-Butenyl angelate	-	-	-	7.3
2-Methyl butyl angelate	-	-	-	17.4
Estragol	-	-	-	5.0

***** Compounds listed according to crescent RI order.

From Table 3, concerning the major components of the essential oils of chamomile related to *Chamaemelum* genus (*C. nobile*), the oils of herbs collected from Anjou (France) and Podgorica (Montenegro) (**Ch5, Ch7)** were characterized by the predominence of the esters isobutylangelate (35.9-38.5%), isobutylisobutanoate (6%), isoamylangelate (18%) and 2-methylbutyl angelate (0.1-20.3%). However, the main constituents of the roman chamomile oil from Iran (**Ch6)** were chamazulene (19.9%), α-bisabololone oxide A (21.6%) and α-bisabolol (20.9%).

Table 3. Major components (≥ 5%) of essential oils of *chamomile of Chamaemelum* genus

Compounds	Ch5	Ch6	Ch7
Chamazulene	-	19.9	-
α-Bisabololone oxide A	-	21.6	-
α-Bisabolol	-	20.9	-
Isobutyl isobutanoate	6.0	-	-
Isobutyl angelate	35.9	-	38.5
2-methylbutyl angelate	-	-	20.3
Isoamyl angelate	18.0	-	-

***** Compounds listed according to crescent RI order

From Table 4, related to *Cladanthus* genus (*C. mixtus* from Morocco), the ester, 2-methyl-2-*trans*-butenyl methacrylate (34.0 %) characterized the chamomile growing in Bouznika (**Ch8**). However, the sesquiterpene *trans-β*-farnesene (43.0%, 18.0%) was the chemotype of the chamomile growing in Oujda and Chefchouane (**Ch9, Ch10**). In addition, the latter contained 2-tridecanone (16.0%) and chamazulene (5.0%). The essential oil of the chamomile growing in Sidi Allal Albahraoui (**Ch11**) was characterized by santolina alcohol (17.0%) together with 1,8-cineole (12.0%).

Table 5, concerning the major components of essential oils of chamomile related to the genus *Matricari*a (*M. chamomilla*), shows that the couple *α*-bisabolol oxide B/*α*-bisabolol oxide A (5.0%-9.9%) / (2.2%-53.4%) seems to be a chemotype of Iranian herbs **Ch14, Ch16, Ch21, Ch22** and Egyptian chamomile **Ch13.**

Chamazulene (0.9%-25.2%) and *cis-β*-farnesene (2.7-65.9 %) were detected in most *Matricaria chamomilla* herbs. Whereas, 1,8-cineole (9.2%), anethole (6.2 %), *trans-β*-farnesene (5.2 %), *α*-farnesene (8.7 %) and galaxolide (6.3%) were mainly found only in the essential oils of **Ch12, Ch13, Ch14, Ch16** and **Ch12**, respectively.

Table 4. Percentages of major components (≥5%) of essential oils of *chamomile* of *Cladanthus* genus

Compounds*	Ch8	Ch9	Ch10	Ch11
Santolina alcohol	-	-	-	17.0
β-Myrcene	-	-	10.5	-
1,8-Cineole	-	-	-	12.0
Camphor	-	-	-	-
ar-Curcumene	-	-	-	-
2-Tridecanone	-	-	16.0	-
trans-β-Farnesene	-	43.0	18.0	-
Chamazulene	-	-	5.0	-
2-Methyl-2-*trans*-butenyl methacrylate	34.0	-	-	-

*****Compounds listed according to crescent RI order

Table 5. Percentages of major components of essential oils of *Chamomilla (M. chamomilla)*

Compounds*	Ch12	Ch13	Ch14	Ch15	Ch16	Ch17	Ch18	Ch19	Ch20	Ch21	Ch22
1,8-Cineole	9.2	-	-	-	-	-	-	-	-	-	-
Anethole	-	6.2	-	-	-	-	-	-	-	-	-
cis-β-farnesene	12.5	34.6	-	7.1	24.2	11.5	65.9	4.4	-	2.7	-
trans-β-Farnesene	-	-	5.2	-	-	-	-	-	-	-	-
α-Farnesene	-	-	-	-	8.7	-	-	-	-	-	-
Spathulenol	-	-	9.4	0.2	0.4	-	-	-	-	0.8	-
α-Bisabolol oxide B	-	8.8	7.0	-	5.0	-	-	-	-	9.9	9.4
α-Bisabololone oxide A	-	7.4	10.0	-	0.2	-	-	-	-	5.2	-
α-Bisabolol	-	-	5.0	56.9	7.3	7.3	-	-	-	-	-
trans-trans-Farnesol	-	-	-	15.6	-	17.0	39.7	66.0	-	-	-
Chamazulene	25.2	0.9	4.2	2.2	-	2.6	-	-	21.0	4.3	-
α-Bisabolol oxide A	-	32.5	21.5	2.2	10.2	-	-	-	41.0	53.4	25.0
Guaiazulene	-	-	-	-	10.6	10.6	17.0	16.2	-	-	-
Galaxolide	6.3	-	-	-	-	-	-	-	-	-	-
Spiroether	-	8.6	-	-	-	-	-	-	8.0	-	-

***** Compounds listed according to crescent RI order

From Table 6**,** concerning the major components of essential oils of *chamomile* of *Matricaria, (M. suaveolens, M. pubescens; M. recutita*) it appears that the couple α-bisabolol oxide B/α-bisabolol oxide A (0.2-25.1%)/(0.1-55.9%) is the chemotype of *M. recutita* oils **Ch23** to **Ch37,** whereas Z-ene-yne-dicycloether (3.6-.37.2%) was detected in the most studied chamomile herbs.

From our investigation, it appears that there are various chemotypes of chamomile essential oils which are responsible of several biological properties:

cis-β-Farnesene (0.9-15.9%) characterized the Estonian **Ch25** and American chamomile **Ch37**. Spathulenol (1.3 -19.4%) was detected in most reported essential oils of the three *Matricaria* species whereas geranyl isovalerate (22.9% and 8.2%) mainly characterized the Estonian and Algerian chamomile herbs **(Ch38 and Ch39)**, respectively. *trans-β*-farnesene (15.6 %), *α*-cadinol (12.9%) and Isochrysanthemic ethyl ester (26.5%) were found only in the essential oil of the Algerian chamomile **(Ch39)** also called "Chamomile" in Morocco, as major components.

Table 6. Percentages of major components of essential oils of *chamomile (M. recutita, M. suaveolens, M. pubescens)*

Compounds*	**Ch23**	**Ch24**	**Ch25**	**Ch26**	**Ch27**	**Ch28**	**Ch29**	**Ch30**	**Ch31**	**Ch32**	**Ch33**	**Ch34**	**Ch35**	**Ch36**	**Ch37**	**Ch38**	**Ch39**
Artemisia alcohol	-	-	0.1	0.2	0.1	-	-	0.2	0.3	0.2	0.1	0.2	0.2	-	7.8	-	-
cis-β-farnesene	-	-	10.4	5.9	4.5	15.9	-	3.4	0.9	2.6	6.1	1.9	6.4	-	10.9	-	-
trans-β-farnesene	-	2.3	-	-	-	-	1.2	-	-	-	-	-	-	-	-	15.6	-
Spathulenol	-	2.4	3.5	2.1	2.1	2.5	1.3	2.0	3.6	2.1	2.1	5.4	5.4	-	2.4	1.4	19.4
Geranyl isovalerate	-	0.3	-	-	-	-	-	-	-	-	-	-	-	-	-	22.9	8.2
α-Bisabolol oxide B	9.4	9.9	9.7	20.8	22.5	17.1	6.6	25.1	23.2	8.4	10.6	21.5	21.9	-	1.2	0.2	-
α-Bisabololone oxide A	-	13.9	8.8	3.3	3.2	5.0	29.9	4.9	10.0	3.8	4.7	11.1	11.4	-	0.1	0.6	-
α-Bisabolol	-	5.6	-	-	-	6.8	-	-	-	-	-	-	-	-	-	0.1	-
Chamazulene	-	4.7	3.4	1.5	2.6	4.2	2.3	2.0	3.8	0.6	1.4	4.3	6.7	24.8	0.2	6.5	-
α-Bisabolol oxide A	25.0	39.4	22.5	29.1	23.8	28.0	53.6	13.9	15.4	55.9	49.1	13.4	19.8	31.7	0.1	0.1	-
α-Cadinol	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	12.9
Isochrysanthemic ethyl ester	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	26.5
Z-enyne-dicycloether	-	11.5	12.6	12.1	12.7	-	-	5.5	3.6	14.0	10.0	6.0	10.6	-	25.8	37.2	-
Hexadecanoic acid	-	-	2.4	0.4	1.2	-	-	6.4	9.2	0.3	1.3	7.6	2.1	-	23.0	-	-
linoleic acid	-	-	1.2	-	-	-	-	11.9	0.9	-	0.1	0.1	0.2	-	-	-	-
Polyacetylene	-	12.5	-	-	-	-	-	-	-	-	-	-	-	-	-	38.3	

***** Compounds listed according to crescent RI order

Firstly, the essential oils composition of French (**Ch2, Ch3)** and Italian **(Ch5)** *Anthemis* and *Chamaemelum* genus was characterized by the chemotype isoamyl angelate (18.0%-22.8%) that was responsible for genotoxic properties [32].

Secondly, Iranian *Chamaemelum* (**Ch6)** and *Matricaria chamomilla* were mainly represented by sesquierpene compounds such as *α*-bisabolol (0.1-56.9%), bisabololone oxide A (0.1-21.6%) and chamazulene (0.9-25.2%) that had high therapeutic effects as sedative, anti-inflammatory and antiphlogistic [33].

Thirdly, essential oils of *Matricaria* (*M. recutita, M. suaveolens, M. pubescens*) were characterized by several chemotypes, *α*-bisabolol (0.1-6.8%), bisabolol oxide B (0.1-25.1%) / A (0.1-55.9%), bisabolone oxide A (0.1-29.9%), *cis β*-farnesene (0.9-15.9%), spathulenol (0.4-19.4%), *Z*-enyne-dicycloether (3.6-.37.2%) which are antilisteric, antiepileptic, mitogenic, spasmolytic, insecticidal and antimicrobial agents [34].

Finaly, the composition of essential oils of *Cladanthus* herbs, collected from the same regions (Morocco), was quite different [16]. However, 1,8-cineole which was mainly found in *Cladanthus* (**Ch11)** from Sidi Allal Elbahraoui (Morocco) (12.0%), the brazilian *Anthemis* (**Ch1)** (14.0%) and *M. chamomilla* (**Ch12)** from Kenitra (Morocco) (9.2%), was reported to be responsible of insecticidal properties [35].

Besides their therapeutic properties, chamomile herbs, rich with α-bisabolol oxide A, α-bisabolol and β-farnesene, may be tested as a green corrosion inhibitors [36].

Conclusion

Our investigation on chemotypes of essential oils of *Chamomile*, shared by various genus, *Anthemis, Chamaemelum, Cladanthus* and *Matricaria* (Asteraceae), worldwide distributed, shows the presence of several chemotypes wich are different from a genus to another one. *cis-β*-Farnesene, spathulenol, *α*-bisabolol oxide B, *α*-bisabololone oxide A, *α*-bisabolol, chamazulene and *α*-bisabolol oxide A, isoamyl angelate, 1,8-cineole, were the main components of reported chamomile essentials oils. This change in the composition of essential oils, even those obtained from the same species growing in different regions, is due to the environmental factors like soil nature, climate and collecting period of plants. Eventhough, this difference has a good impact on the biodiversity including the pharmacological properties.

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