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Comparative compositions of essential oils of *Thymus* growing in various soils and climates of North Africa

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Abstract
Essential oils compositions of hydrodistilled fresh aerial parts of *Thymus* (Lamiaceae), growing in various soils of Algeria and Morocco with different climates, are compared here. Thymol (17.3–79.1%), carvacrol (4.4-72.4%), α-terpineol (34.4%), *trans*-β-ocimene (0.15-25.8%), *p*-cymene (0.47-19.6%), linalool (6.1-17.2%), viridiflorol (16.12%), γ-terpinene (0.74-14.6%), α-terpinene (0.3-12.3%), *o*-cymene (11.41%), thymol methyl ether (6.73-10.7%), α-pinene (8.75%), camphor (7.5%), nerolidol (6.95%) and borneol (6.1%), were the main components of the studied essential oils.

Keywords: Thymus ciliatus; *Thymus guyonii*; *Thymus numidicus*; Lamiaceae; essential oil; thymol; carvacrol; chemotype.

1. Introduction
Thymus (Zaatar) is one of the most important genera in the Lamiaceae family. *Thymus* belongs to the tribe Mentheae, subfamily Nepetoideae; it includes 300-400 species [1-3]. 'Thyme' is commonly used in the herbalistic sector with a long list of pharmacological and aromatic properties, such as spasmolytic, antiseptic, antitussive and expectorant [4]. In folk medicine, it is popular for stimulating action in all organism functions [5] and also for the antimicrobial activity of the essential oil [6].There is a great number of papers related with *Thymus* essential oils but few of them concern the North African species, *T. ciliatus*, *T. guyonii and T. numidicus* [7-17]. In continuation of our works on Lamiaceae essential oils [7; 8; 12; 13; 15; 16; 18-29], we report here a comparative study of hydrodistilled essential oils compositions of *T. ciliatus*, *T. guyonii and T. numidicus* from our group [7; 8; 12; 13; 15; 16], with those from other groups from Algeria (9; 14; 17) and Morocco [10; 11].

2. Experimental
Plant material
Tc1: Aerial parts of *Thymus ciliatus*, collected from Ain Mlila province of Oum El Bouaghi (Eastern Algerian) [8].
Tc2: Aerial parts of *Thymus ciliatus*, collected from Batna (Eastern Algerian) [7].
Tc3: Aerial aerial parts of *Thymus ciliatus*, collected from Tlemcen (North Western Algerian) [9].
Tc4: Aerial aerial parts of *Thymus ciliatus*, collected from Imilchi (Central Morocco) [10].
Tc5: Aerial aerial parts of *Thymus ciliatus*, collected from Azrou (Central Morocco) [11].
Tc6: Aerial aerial parts of *Thymus ciliatus*, collected from Bir chouhada (Southern Oum El Bouaghi) [12].

Tg1: Aerial parts of *Thymus guyonii*, collected from Zelfana-Province of Ghardaia (Algerian Septentrional Sahara) [13].
Tg2: Aerial parts of *Thymus guyonii*, collected from Djelfa (Haoues mountain, Central Algerian) [14].
Tn1: Aerial parts of *Thymus numidicus* collected from Constantine (North Eastern Algerian) [15].
Tn2: Aerial parts of *T. numidicus* collected from Djebel El-Ouahch, Constantine (North-Eastern Algerian) [16].
Tn3 Aerial parts of *T. numidicus* collected from Azzazga (Yakouren forest, Central Algerian) [14].
Tn4: Aerial parts of *T. numidicus* collected from Kabylie region (Central Algerian) [17].

Extraction

Fresh aerial parts of the collected plants were submitted for three hours to hydrodistillation in a Clevenger-type apparatus. The essential oils obtained were dried over anhydrous sodium sulphate then stored at +4 °C until analyzed. Respective yields are given in Table 1.

Table 1. Plant material data of *Thymus ciliatus, Thymus guyonii and Thymus numidicus* from Algeria and Morocco.

Sample code	Region of collection	Altitude (m)	Oil yield % (w/w)	Reference
Tc1	Ain Mlila (Eastern Algerian)	771	2.0	[8]
Tc2	Batna (Eastern Algerian)	900-1036	2.1	[7]
Tc3	Tlemcen (Northwestern Algerian)	842	5.1	[9]
Tc4	Imilchil (Central Morocco)	1700	1.6	[10]
Tc5	Azrou (Central Morocco)	1278	0.3	[11]
Tc6	Bir chouhada (Southern Oum El Bouaghi)	708	2	[12]
Tg1	Zelfana (Algerian Septentrional Sahara)	355	2	[13]
Tg2	Djelfa (Haoues mountain, Cenral Algerian)	2000	1	[14]
Tn1	Constantine (North Eastern Algerian)	694	2	[15]
Tn2	Djebel El-Ouahch, Constantine (North-Eastern Algeria)	800	2.1	[16]
Tn3	Azzazga (Yakouren forest, North Central Algerian)	1000	2.4	[14]
Tn4	Kabylie region (North Central Algerian)	898	2	[17]

Gas chromatography and mass spectrometry (GC and GC/MS) analyses

Tc1, Tc2, Tc5, Tg2, Tn3, Tn4 and Tc6 were analyzed by GC/MS using a Hewlett Packard 6890 [8; 7; 11; 14; 14; 17; 12]
Tc3 was analyzed by GC using a Perkin-Elmer and ^{13}C– NMR [9]
Tc4 was analyzed with an Agilent GC-MSD 6890/5973 [10]

Tg1 and Tn1 were analysed by GC/MS using a Shimadzu QP5050 [13; 15]
Tn2 GC analyses were performed using a Perkin-Elmer gas chromatograph equipped with two FIDs and GC–MS analyses were performed on a Perkin-Elmer apparatus [16]

3. Results and Discussion

From Table 2, exhibiting the major components (≥5%) of the studied essential oils of *Thymus ciliates*, it appears that the essential oils of *T. ciliatus* collected from Batna (Tc2) and Ain Mlila (Tc1) are thymol chemotype (79.1%, 54.98%) [7; 8] while the essential oil of *T. ciliatus*, collected from Tlemcen (Tc3), is carvacrol chemotype (80.3%) [9]. Thymol (17.3%, 44.2%) was mainly found in essential oils of *T. ciliatus* collected from Imilchil (Tc4) [10] and Azrou (Tc5) [11], respectively. Carvacrol (80.3% and 26.2%) was mainly characterizing the essential oils of Tc3 and Tc4 [9;10], respectively. *p*-Cymene (6.66%, 5.6%, 6.1%, 19.6%) was found to be mainly common to essential oils of Tc1,Tc2,Tc3 and Tc4 [8; 7; 9; 10] respectively. α-Terpinene (12.3%) and *trans*-β-ocimene (25.8%) were mainly found in the essential oil of Tc5 [11]. γ-Terpinene (11.33% and 14.6%), was found to be as a component of essential oils of Tc1 and Tc4, respectively [8, 10]. The composition of the essential oil of Tc6 [12] seems to be different from those of other *Thymus* species particularly with the main presence of viridiflorol which is reported here, for the first time from a *Thymus* oil, but it has been recently reported as a main component of *Salvia officinalis* (Lamaiceae) essential oil [28].

Table 2: Percentages of major components (≥5%) **of essential oils of *Thymus ciliatus* (Tc1-Tc6).**

Compounds*	**Percentage**					
	Tc1 [8]	**Tc2** [7]	**Tc3** [9]	**Tc4** [10]	**Tc5** [11]	**Tc6** [12]
α-Pinene	-	-	-	-	-	8.75
α-Terpinene	-	-	-	-	12.30	-
p-Cymene	6.66	5.60	6.10	19.60	-	-
trans-β-Ocimene	-	-	-	-	25.80	-
γ-Terpinene	11.33	-	-	14.60	-	-
Camphor	-	-	-	-	-	7.50
Thymol	54.98	79.10	-	17.30	44.20	-
Carvacrol	-	-	80.30	26.20	-	-
Nerolidol	-	-	-	-	-	6.95
Viridiflorol	-	-	-	-	-	16.12

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

Table 3 lists the major components the two essential oils of *Thymus guyonii* growing in Djelfa (Central Algerian) and Zelfana (Algerian Septentrional Sahara). Carvacrol is the chemotype (55.55%) [13] of the essential oil of Zelfana (Tg1), while the essential oil of the plant collected from Djelfa (Tg2) is mainly characterizing by *p*-cymene (18.6%) [14]. γ-Terpinene (13%), linalool (6.1%), borneol (6.1%) and thymol methyl ether (10.7%) [14]. Thymol (19.51% and10.9%), was found to be mainly common to the essential oils of Tg1 and Tg2 respectively [13; 14].

Table 3: Percentages of major components (≥5%) **of essential oils of *Thymus guyonii* (Tg1, Tg2).**

Compounds*	**Percentage**	
	Tg1 [13]	**Tg2** [14]
p-Cymene	6.25	18.60
γ-Terpinene	-	13.00
Linalool	-	6.10
Borneol	-	6.10
Thymol methyl ether	-	10.70
Thymol	19.51	10.90
Carvacrol	55.55	-

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

The percentages of major components (≥5) of essential oils of *Thymus numidicus* (Tn1to Tn4) are listed in Table 4. Thymol is the chemotype (68.2 %) [16] of the essential oils growing in Djebel El-Ouahch (Tn2). α-Terpineol (34.4%) was the main component in Tn3 collected from Azzazga (Central Algerian) [14]. Carvacrol (6.02%, 16.9%, 6.8%, 28.1%) was found to be mainly common to the essential oils of Tn1, Tn2, Tn3 and Tn4 [15; 16; 14; 17] respectively. *p*-Cymene (6.78% and 8%) was mainly found in the essential oils of Tn3 and Tn4 respectively [14; 17]. Thymol methyl ether (6.73%) and *o*-cymene (11.41%) were mainly found in the essential oil of Tn1 [15]. γ-Terpinene (10.84% and 8.4%) was the main component of Tn1and Tn3 respectively [15; 14]. Linalool (17.20% and 11.5%) was mainly characterizing the essential oils of Tn1 and Tn2 [15; 16].

Table 4: Percentages of major components (≥5%) **of essential oils of *Thymus numidicus* (Tn1-Tn4).**

Compounds*	**Percentage**			
	Tn1 [15]	**Tn2** [16]	**Tn3** [14]	**Tn4** [17]
o-cymene	11.41	-	-	-
p-Cymene	-	-	6.70	8.00
γ-Terpinene	10.84	-	8.40	-
Linalool	17.20	11.50	-	-
Thymol methyl ether	6.73	-	-	-
α-Terpineol	-	-	34.40	-
Thymol	23.92	68.20	15.00	-
Carvacrol	6.02	16.90	6.80	28.10

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

From the literature review, two couples of components, depending on each other, have been detected in a great number of *Thymus* oils, *p*-cymene/γ-terpinene [14; 30-42] and α-pinene/camphor [43-46]. This is confirmed here in the essential oil of Tc1 and Tc4 by the main presence of the couple *p*-cymene/γ-terpinene (6.66%/11.3% and 19.6%/14.6%, respectively). The couple α-pinene/camphor was mainly found in the essential oil of Tc5 (12.3%/25.8%). The high-content thymol Tc3 (80.30%), Tc2 (79.10%) and Tn2 (68.20%) may be used in anticorrosive tests [51].

Conclusion

The results discussed here revealed significant differences in the composition of the essential oils of different thyme species collected in Eastern, Western and Saharian parts of Algeria and Central Morocco. Among the essential oils of the species Th*ymus ciliatus* collected from three different parts of Algeria (Tc1-Tc3) and two different parts of Morocco (Tc4, Tc5), the species *Thymus guyonii* (Tg1, Tg2) collected from two different parts of Algeria and *Thymus numidicus* (Tn1-Tn4), grown in various regions of Algeria, the essential oils of Tc1, Tc2, Tc5, Tn1 and Tn2 were high thymol –content and poor in carvacrol. However, they were very different from that of Tc3, Tc4, Tg1 and Tn4, which were particularly rich in carvacrol. Differently, Tn3 essential oil was α-terpineol high-content and Tg2 was mainly characterized by *p*-cymene. Exclusively from the genus *Thymus*, Tc6 oil contained viridiflorol as a major component. This component was recently found as a major component of *Salvia officinalis*, belonging to the Lamiaceae family. These differences in the content of the essential oils even in case of one species, collected from different parts, may be due to the nature of soil, climate, altitude which change from each region to another.

References

1. Evans W.C. (1989) Trease and Evans' Pharmacognosy, 13th ed. Bailliere Tindall, London, pp. 216-217.
2. Morales R. (1997) Synopsis of the genus *Thymus* L. in the Mediterranean area. Logascalia **19**, pp. 249-262.
3. Pedersen, J. A., *Biochem. Syst. & Ecol*. 28 (2000) 229.
4. Rasooli, I., Mirmostafa, S. A., *Fitoterapia* 73 (2002) 244.
5. Bellakhdar, J. (1997) Pharmacopée Marocaine, Ibis press, France.
6. Dob, T., Dahmane, D., Benabdelkader, T., Chelghoum, C., *I. J. Arom*. 16 (2006) 95.
7. Kabouche, A., Ghannadi, A., Kabouche, Z., *Nat. Prod. Comp*. 4 (2009) 1251.
8. Ghorab, H., Kabouche, A., Semra, Z., Ghannadi, A., Sajjadi, EB., Touzani, R., Kabouche, Z., *De. Pharm. Lett*. 5 (1) (2013**)** 28.
9. Bousmaha-Marroki, L., Atik-Bekkara, F., Tomi, F., Casanova, J., *J. Essent. Oil Res*. 19 (2007) 490.
10. Alaoui Jamali, C., El Bouzidi, L., Bekkouche, K., Lahcen, H., Markouk, M., Wohlmuth, H., Leach, D., Abbad, A., *Chem. Biodiv*. 9 (2012) 1188.
11. Amarti, F., Satrani, B., Ghanmi,M., Farah, A., Aafi, A., Aarab, L., El Ajjouri, M., Chaouch, A., *Biotechnol. Agr. Soc. Environ.* 14(1) (2010) 14.
13. Lehbili, M., Chibani, S., Kabouche, A., Semra, Z., Smati, F., Abuhamdah, S., Touzani, R., Kabouche, Z., *Der Pharm. Lett*. 5 (2) (2013) 306.
14. Hazzit, M., Baaliouamer, A., Leonor , F. M., Graca M., *J. Agric. Food Chem.* 54 (2006), 6314.
15. Zeghib, A., Laggoune, S., Kabouche, A., Semra, Z., Smati, F., Touzani, R., Kabouche, Z., *Der Pharm. Lett.* 5(3) (2013) 206.
16. Kabouche, A., Kabouche, Z., Bruneau, C., *Flav. Fragr. J.* 20 (2005) 235.
17. Hazzit, M., Baaliouamer, A., *Riv. Ital. EPPOS.* 43 (2007), 11.
18. Ghannadi, A., Sejjadi, E., Kabouche, A., Kabouche, Z., *Z. Naturforsch*. 59c (2004) 187.
19. Touafek, O., Nacer, A., Kabouche, A., Kabouche, Z., Bruneau, C., *Chem. Nat. Comp*. 40 (2004) 28.
20. Kabouche, A., Kabouche, Z. Bruneau, C., *Flav. Fragr. J.* 20 (2005) 235.
21. Kabouche, Z., Boutaghane, N., Laggoune, S., Kabouche, A., Ait-Kaki, Z., Benlabed. K., *Int. J. Aromather*. 15 (2005) 129.
22. Kabouche, A., Touafek O., Nacer, A., Kabouche, Z., Bruneau, C., *J. Essent. Oil Res.* 18 (2006) 175.
23. Kabouche, A., Kabouche, Z., Sajjadi, S.E., Ghannadi, A., *J. Essent. Oil Res.* 19 (2007) 44.
24. Laggoune, S., Kabouche, A., Kabouche Z., El-Azzouny, M. A., *J. Essent. Oil Res.* 21 (2009) 67.
25. Kabouche, A., Ghannadi, A., Kabouche, Z., *Nat. Prod. Commun.* 4(9) (2009) 1251.
26. Bensouici, C., Benmerache, A., Chibani, S., Kabouche, A., Abuhamdah, S., Semra, Z., Kabouche, Z., *Der Pharm. Lett.* 5(2) (2013) 224.
27. Zeghib, A., Laggoune , S., Kabouche, A., Semra, Z., Smati, F, Touzani, R., Kabouche, Z., *Der Pharm. Lett.* 5(3) (2013) 206.

28. Lakhal, H., Ghorab, H., Chibani, S., Kabouche, A., Semra, Z., Smati, F., Abuhamdah, S., Kabouche, Z., *Der Pharm. Lett.* 5(3) (2013) 310.
29. Semra, I., Benmerache, A., Chibani, S., Kabouche, A., Abuhamdah, S., Kabouche, Z., *Der Pharm. Lett.* 5(3) (2013) 381.
30. Salas, J. B., Tellez, Trinidad, R., Pardo, F. M. V., Alonso, M. J. P., Capdevilla, M. A. C., *Acta Bot. Gallica*. 158(2) (2011) 251.
31. Verma, R. S., Padalia, R. C., Chanotiya, C. S., Chauhan, A., *Nat. Prod. Res*. 24(20) (2010)1890.
32. Jia, H. L., Ji, Q. L., Xing, S. L., Zhang, P. H., Zhu, G. L., Wang, X. H., *J. Food Sci*. 75(1) (2010) E59
33. Arraiza, M.P., Andres, M. P., Arrabal, C., Lopez, J. V. *J. Essent. Oil Res*. 21(4), (2009) 360.
34. Hazzit, M., Baaliouamer, A., *J. Essent. Oil Res*. 21(2) (2009) 162.
35. Tzakou, O., Couladis, M., *J. Essent. Oil Res*. 20(5) (2008) 442.
36. Stoilova, I., Bail, S., Buchbauer, G., Krastanov, A., Stoyanova, A., Schmidt, E., Jirovetz, L., *Nat. Prod. Commun*. 3(7) (2008) 1047.
37. Dob,T., Dahmane, D., Benabdelkader, T., Chelghoum, C., *Pharm. Biol*. 44(8) (2006) 607.
38. Raal, A., Arak, E., Orav, A., *Herba Polon.* 51(1/2) (2005) 10.
39. Asllani, U., Toska, V., *J. Essent. Oil Res*. 15(3) (2003) 165.
40. Sefidkon, F., Askari, F., Mirmostafa, S. A., *J. Essent. Oil Res*. 13(3) (2001) 192.
41. Asfaw, N., Storesund, H. J., Skattebol, L., Tonnesen, F., Aasen, A. J., *Flav. Fragr. J*. 15(2) (2000) 123.
42. Kulevanova, S., Ristic, M., Stafilov, T., Matevski, V., *J. Essent. Oil Res*. 10(5) (1998) 537.
43. El Ajjouri, M., Ghanmi, M., Satrani, B., Amarti, F., Rahouti, M., Aafi, A., Ismaili, M . R., Farah, A., *Acta Bot. Gallica*. 157(2) (2010) 285.
44. Yilmaz, G., Telci, I., Kandemir, N., Kaya, N., *Asian J. Chem*. 16(2) (2004) 841.
45. Vila, R., Freixa, B., Canigueral, S., Adzet, T., Tomas, X., Molins, J.J., *Flav. Fragr. J*. 10(6) (1995) 379.
46. Adzet, T., Vila, R., Batllori, X., Ibanez, C., *Flav. Fragr. J*. 4(2) (1989) 63.
47. Morteza-Semnani, K., Rostami, Bahram., Akbarzadeh, M., *J. Essent. Oil Res*. 18(3) (2006) 272.
48. Abousaber, M., Hadjakhoondi, A., Shafiee, A., *J. Essent. Oil Res*. 14(3) (2002)154.
49. Chen, J., Z. K., Cheng, Ch., Shi, B., Zhu, W., *Zhongguo Yaoxue Zazhi* 36(1) (2001) 16.
50. Kasumov, F. Y., *Khim. Prir. Soedin*. 6 (1979) 863.
51. Rekkab, S., Zarrok, H., Salghi, R., Zarrouk, A., Bazzi, L., Hammouti, B., Kabouche, Z., Touzani, R., Zougagh, M., *J. Mater. Environ. Sci.* 3(4) (2012) 613.