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Comparative compositions of essential oils of three *Ruta* species growing in different soils

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Abstract
A comparative study on the essential oils of three *Ruta* species (Rutaceae), *Ruta chalepensis*, *Ruta montana* and *Ruta graveolens* growing in different countries has been carried on. The essential oils were extracted by hydrodistillation of the aerial parts. 2-Undecanone (1.79–84.2%), 2-decanone (0.1-11.6%), 2-nonanone (5.2-33.6%) and 2-nonanylacetate (2.8-20.9%) were the main components of the studied essential oils.

Keywords: Ruta chalepensis; *Ruta montana*; *Ruta graveolens*; Rutaceae; essential oil; 2-undecanone; 2-nonanone.

1. Introduction
Ruta species are sources of different classes of natural products with biological activities, including antifungal, antioxidant, phytotoxic, abortive depressant, antidotal and anti-inflammatory activities [1-8]. There are four *Ruta* species and subspecies in Algeria, *R. montana*, *R. chalepensis* subsp. *angustifolia*, *R. chalepensis* subsp. *latifolia* and the Saharian species *R. tubercula* [9;10]. The main purpose of this work is to investigate the composition of essential oils of *R. chalepensis*, *Ruta montana and Ruta graveolens* from several places and their composition. *Ruta chalepensis* L. is a native herb of the mediterranean region [7] but it's widely diffused in many parts of the world, in temperate and tropical countries [1]. *R. chalepensis* is used in traditional medicine of many countries for the treatment of a variety of diseases [11]. *Ruta montana* is used in digestive disorders and helminthiasis [12], it's traditionally known for its abortive and anti-fever effects [13] while *Ruta graveolens* is known for its antiparasitic, stomachic, digestive, vermifuge, emmenagogue and molluscicidal activities [14;15]. In continuation of our works on Rutaceae essential oils [13;16;17], we report here a comparative study of hydrodistilled essential oils composition of *Ruta* species from our group [16;17] and from different places in the world [20-27;29-33].

2. Experimental
Plant material
***Ruta chalepensis*:**
Rc1: Aerial parts of *Ruta chalepensis* subsp. *angustifolia*, collected from Jijel (Eastern Algerian) [18].
Rc2: Aerial parts of *Ruta chalepensis* L., collected from Grarem province of Mila (Eastern Algerian) [17].
Rc3: Aerial erial parts of *Ruta chalepensis* L. subsp. *angustifolia* (Pers.) P. Cout., collected from Boudouaou (Northern Algerian) [19].
Rc4: Aerial parts of *Ruta chalepensis* L., collected from Tunisia [20].
Rc5: Aerial parts of *Ruta chalepensis* L., collected from Turkey [21].
Rc6: Aerial parts of *Ruta chalepensis* L., collected from the North of India [22].
Rc7: Aerial parts of *Ruta chalepensis* L., collected from Iran [23].
Rc8: Aerial parts of *Ruta chalepensis* L., collected from Greece [24].
Rc9: Aerial parts of *Ruta chalepensis* L., collected from Turkey [25].
Rc10: Aerial parts of *Ruta chalepensis* L. collected from Spain [26].
Rc11: Aerial parts of *Ruta chalepensis* L., collected from Portugal [27].

Ruta montana:
Rm1: Aerial parts of *Ruta montana* L., collected from Turkey [21].
Rm2: Aerial parts of *Ruta montana* L., collected from Tipaza (North central Algerian) [28].
Rm3: Aerial parts of *Ruta montana* L., collected from Constantine (Eastern Algerian) [16].

***Ruta graveolens*:**
Rg1: Aerial parts of *Ruta graveolens*, collected from China [29].
Rg2: Aerial parts of *Ruta graveolens* L., collected from Northern Iran [30].
Rg3: Aerial parts of *Ruta graveolens*, collected from Italy [31].
Rg4 : Aerial parts of *Ruta graveolens* L., collected from Malaysia [32].
Rg5 : Aerial parts of *Ruta graveolens* L., collected from Egypt [33].

Extraction
The aerial parts of plants were hydrodistilled in a Clevenger-type apparatus for 3 hours. The isolated oils were dried over anhydrous sodium sulphate and stored at 4-6 °C until tested and analyzed. Respective yields are given in Table 1.

Table 1. Plant material data of *Ruta chalepensis*, *Ruta montana* and *Ruta graveolens.*

Sample code	Region of collection	Reference
Rc1	Jijel (North Eastern Algerian)	[18]
Rc2	Grarem (North Eastern Algerian)	[17]
Rc3	Boudouaou (North Central Algerian)	[19]
Rc4	Tunisia	[20]
Rc5	Turkey	[21]
Rc6	North of India	[22]
Rc7	Iran	[23]
Rc8	Greece	[24]
Rc9	Turkey	[25]
Rc10	-	[26]
Rc11	Portugal	[27]
Rm1	Turkey	[21]
Rm2	Tipaza (North Central Algerian)	[28]
Rm3	Constantine (North Eastern Algerian)	[16]
Rg1	China	[29]
Rg2	North of Iran	[30]
Rg3	Italy	[31]
Rg4	Malaysia	[32]
Rg5	Egypt	[33]

Gas chromatography-mass spectrometry (GC/MS) analyses of essential oils
Rc1, Rm3: GC-MS analyses were performed using a Shimadzu QP5050 mass selective detector using a cross-linked DB5-MS column (40 m × 0.18mm, film thickness 0.18 μm). The oven temperature was programmed as isothermal at 60°C for 5 min, then raised to 275°C at 5°C/min and held at this temperature for 5 min [16;18].

Rc3: GC-MS analyses were carried out using a Shimadzu GC17A chromatograph using fused silica capillary column with stationary phase DB-5. The various parameters fixed for the column are: 30 m x 0.32 mm, 0.25 μm film thickness. The temperature program was 60°C for 3 min then 3°C/ min to 240°C for 3 min; injector 250°C; detector 250°C; N2 was used as carrier gas at a flow rate 1 mL/min in the split mode 1:50, with an injection volume of 0.2 μL. Quantitative data was obtained from electronic integration of area percentages without the use of correction factors. In order to determine retentions indices (RI), a series of n-alkane (C5-C28) mixtures were analyzed under the same operative conditions on DB-5 column; the samples indices were calculated following Van den Dool and Kratz (40). The GC/ MS analysis was performed on a CE-TRACE GC 2000 ThermoFinnigan chromatograph linked to a TRACE MS mass spectrometer using a DB-5 capillary column (30 m x 0.32 mm, 0.25 μm film thickness). It was programmed from 60°C (3min) to 240°C (3min) at 3°C/min with He carrier gas at a flow rate of 1 mL/min and injector heater 250°C. The MS conditions were: EI source, electron energy 70 eV and source temperature 250°C. Acquisition mass range, m/z = 40–450 [19].

Rc4: GC-MS analyzes were carried out using an Agilent 6890 gas chromatograph (Agilent Technologies, Palo Alto, CA, USA) equipped with a HP-5MS Phenyl Methyl Siloxane column (30m×0.25mm i.d.; film thickness 0.25μm). The GC was coupled to a MS (Agilent 5973) with split–splitless injector. The operating conditions were as follows: injector temperature, 300 ◦C; carrier gas, helium with 1.5 ml/min; oven temperature program, 3 min isothermal at 100 ◦C, then at 10 ◦C/min up to 300 ◦C and finally held isothermally for 15 min. The identity of oil components was assigned by comparison of their GC retention times and MS spectra with corresponding data of commercial available standards [20].

3. Results and Discussion
It appears that 2-uncadenone was more abundant in the essential oils of **Rc1**, growing in Jijel (North Eastern Algerian) (83.40%) [18], **Rc4** from Tunisia (77.10%) [20], **Rc5** from Turkey (66.50%) [21], **Rc6** from India (67.80%) [22] **Rc7** from Iran (52.50%) [23] (Table 2). 2-Decanone was identified in the oil of cultivated *R. chalepensis* **Rc2,** grown in Grarem (North Eastern Algerian) (5.56%) and of **Rc4** (8.96) [20], the last one was also mainly characterized by 2-dodecanone (2.37%). The essential oil of **Rc2** was exclusively characterized by the main presence of 2-acetoxytridecane (31.25%) and 2-

acetoxytetradecane (12.06%) which has been not found in any reported essential oil of *Ruta chalepensis*. The essential oil of **Rc5,** from Turkey, contained only 2-uncadenone (66.50%) [21].The composition of the essential oil **Rc8** from Greece was quite different from the previously essential oils, it was mainly represented by 2-methyloctylacetate (44.00%) and β-phellandrene (10.70%) [24]. 2-Decanylacetate (5.60%) [26] was found only in the essential oil of **Rc10** from Spain, which also contained 2-nonylacetate (20.90%), as well as the reported essential oil **Rc6** (2.80-15.30%), and 2-nonanone which was mainly found in the essential oil of **Rc10** (17.20%) and **Rc9** (16.24%) from Turkey [25]. The essential oil of **Rc11,** from Portugal, was distinguished from the other *R. chalepensis* by the presence of methyl nonyl ketone chemotype (90.00%) [27].

Table 2: Percentages of major components (≥5%) of essential oils of *Ruta chelepensis* (Rc1 to Rc11).

Compounds *	**Percentage**										
	Rc1 [18]	**Rc2** [17]	**Rc3** [19]	**Rc4** [20]	**Rc5** [21]	**Rc6** [22]	**Rc7** [23]	**Rc8** [24]	**Rc9** [25]	**Rc10** [26]	**Rc11** [27]
Undecan-2-one	83.40	-	66.49	77.18	66.50	41.30-67.80	52.50	-	66.49	38.10	-
2-Decanone	-	5.56		8.96	-	-	-	-	-	-	-
2-Dodecanone		-		2.37	-	0.10-11.60	-	-	-	-	-
2-Nonanone	-	-	16.24	-	-	5.20-33.60	24.10		16.24	17.20	-
2-Methyloctylacetate	-	-	-	-	-	-	-	44.00	-	-	-
2-Nonylacetate	-	-	-	-	-	2.80-15.30	-	-	-	20.90	-
β-Phellandrene	-	-	-	-	-	-	-	10.70	-	-	-
Methyl nonyl Ketone	-	-	-	-	-	-	-	-	-		90.00
2-Decanyl acetate	-	-	-	-	-	-	-	-	-	5.60	-
2-Acetoxytridecane	-	31.25	-	-	-	-	-	-	-	-	-
2-Acetoxytetradecane	-	12.06	-	-	-	-	-	-	-	-	-

*****The compounds appear according to their RI values order

Few works have been reported on essential oils of *Ruta montana* [14;16;21;28]. Table 3 shows the percentages composition of essential oils of *Ruta montana* from Turkey **Rm1** [21], Tipaza **Rm2** [28] and Constantine **Rm3** [16]. It appears that undecan-2-one has been found to be a chemotype for **Rm1** [20], while the essential oil of **Rm3** is characterized by the main presence of undecan-2-one (37.70%), resorcinol (27.60%) and 2-acetoxytetradecane (9.19%). The oil of **Rm2** Tipaza [28] was ketone (C_6-C_{13}) chemotype (95%). It's interesting to test the anticorrosive activity of the latter because of its high-ketones which can be used in industry [34].

Table 3: Percentages of major components (≥5%) of essential oils of *Ruta montana* (Rm1 to Rm3).

Compounds *	**Percentage**		
	Rm1 [21]	**Rm2** [28]	**Rm3** [16]
Undecan-2-one	84.20	-	37.70
Resorcinol	-	-	27.60
2-Acetoxytetradecane	-	-	9.19
Ketone C_6-C_{13}	-	95.00	-

*****The compounds appear according to their RI values order

Other works have been reported on essential oils of *Ruta graveolens* [29-33]. Table 4 includes the percentages composition of essential oil of *Ruta graveolens* from China (**Rg1)** [29], Iran (**Rg2)** [30], Italy (**Rg3)** [31], Malaysia (**Rg4)** [32] and Egypt (**Rg15)** [33].

Table 4: Percentages of major components of essential oils of *Ruta graveolens* (Rg1 to Rg5).

Compounds*	**Percentage**				
	Rg 1 [29]	**Rg2** [30]	**Rg3** [31]	**Rg4** [32]	**Rg5** [33]
Undecan-2-one	46.15	33.90	46.8	30.73	49.20
2-Nonanone	27.01	8.80	18.80	18.06	24.70
2-Acetyltridecane	12.73	-	-	-	-
1-Dodecanol	-	11.00	-	-	-
Geyrene	-	10.40	-	-	-
2-Heptanolacetate	-	17.50	-	-	-
Sesquitrpenoids	-	13.3	-	-	-
Monoterpenoids	-	4.10	-	-	-
2-Nonylacetate	-	-	-	11.03	-
Xanthotoxin	-	-	-	7.24	-

*****The compounds appear according to their RI values order

It appears that undecan-2-one and 2-nonaone were found to be the main components of all *Ruta graveolens*, while the essential oil of **Rg2** was characterized by the main presence of 1-dodecanol (11.00%), geyrene (10.40%), 2-heptanolacetate (17.50%) and sesquiterpenoids (13.3%). The oil of **Rg4** [32] was mainly represented by 2-nonylacetate (11.30%) and xanthotoxin (7.24%). The essential oil of **Rg1** was exclusively characterized by the main presence of 2-acetoxytridecane (12.73%) which has been not found in any reported essential oil of *Ruta graveolens*.

Conclusion

From this study, it appears that 2-undecanone is the chemotype of 13 from 19 reported essential oils of *Ruta* species. We can conclude that it's a marker of *Ruta* species. The presence of 2-decanone was identified only in *R. chalepensis* of Algeria, collected from Grarem (5.56%) and Boudouaou (8.96%); it has not been found in any reported essential oil of *R. chalepensis* or *R. montana*. In addition, the composition of the essential oil of *R. chalepensis* from Greece was different; it contained 2-methyloctylacetate (44.00%) and β-phellandrene (10.7%) while the oil of *R. chalepensis* from Grarem was mainly characterized by 2-acetoxytridecane (31.25%) and 2-acetoxytetradecane (12.06%). Very differently, the cultivated *R. chalepensis* from Portugal was predominated by methylnonylketone (90.00%). The essential oil of *R. montana* grown in Constantine (Algeria) showed differences with other reported oils of the genus by the presence of resorcinol (27.60%). The 2-acetyltetradecane was found only in essential oils of *R. montana* from Algeria, collected from Grarem (12.06%) and Constantine (9.19%). The cultivated *Ruta graveolens* from China was characterized by the presence of 2-acetoxytridecane (12.73%). The essential oil of *R. graveolens* from Iran showed differences by the presence of 1-dodecanol (11.00%), geyrene (10.40%), 2-heptanolacetate (17.5%) and sesquiterpenoids (13.30%). Exclusively, 2-nonylacetate (11.03%) was found only in the essential oil of *R. graveolens* grown in Malaysia. In regard to the previously reported contents of the essential oils of *Ruta* species, it is interesting to point out some quantitative differences indicating that environmental factors strongly influence the chemical compositions. These chemical differences can be most probably explained by the variability of the plant subspecies and the existence of different chemotypes which change from each region, depending on environmental factors like the soil nature and the climate.

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