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ESSENTIAL OIL COMPOSITION OF SOME PLANTS OF FAMILY ZYGOPHYLLACEAE AND EUPHORBIACEAE

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Abstract

Our objectives were to find out the chemical constituents of some selected plants of family Zygophyllaceae and Euphorbiaceae collected from Peshawar and Attock Hills during 2009, by GC/MS. The oil obtained from three analysed plants of family Zygophyllaceae showed that oxygenated monoterpenes were the highest (90.99%) in *Tribulus terrestris,* followed by *Fagonia cretica* (89.94%) and the lowest (36.01%) found in *Peganum harmala*. *Peganum harmala* had maximum esters (11.58%) followed by *Tribulus terrestris* (5.8%) and *Fagonia cretica* (5.5%). Monoterpenes hydrocarbons were the highest (1.22%) in *Fagonia cretica* followed by *Peganum harmala* and absent in *Tribulus terrestris*. Sesquiterpenes hydrocarbons were maximum (11.01%) in *Peganum harmala* and absent in *Tribulus terrestris*. The analysis of essential oils revealed that *Fagonia cretica* oils had 17 compounds that constituted 100% of the oil composition. Oxygenated monoterpenes (89.94%), were a major group of compounds. *Peganum harmala* oil had 18 compounds. There were 10 compounds in *Tribulus terrestris* oil that consisted 100% of the total oil composition. Eight compounds were identified in *Chrozophora tinctoria* oils giving complete oil composition. It had oxygenated monoterpenes (86.93%), constituting 2(4H) - Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy (50.718%). *Ricinus communis* . oil had 8 compounds with 100% of the oil composition. The present study exhibited that phytochemical attributes and chemical composition of the studied plants have potential uses for food, pharmaceutical and cosmetic industry in future. Detailed research work on the antioxidant principles and biological activities of the studied plants is further recommended.

Key words: Medicinal plants, Essential oil composition, Zygophyllaceae, Euphorbiaceae.

Introduction

Fagonia cretica L., is found in dry habitat throughout Pakistan. It is also called Azghakhi, in Khyber Pakhtunkhwa. Abdel-Khaliq *et al*., (2000) reported flavonoids and triterpenoids saponins from *Fagonia cretica*. Alam (2010) detected saponins, tannins, unsaturated sterols, alkaloids, cardiac and cyanogenic glycosides, coumarins, chlorides and sulphates in *Fagonia* species. Anil *et al*., (2012) reported four flavonoid compounds in *F. indica*. Saeed *et al*., (1999) stated that folkloric medicinal literature claimed that *Fagonia cretica* is an anticarcinogen.

Peganum harmala L. Wild rue (English), vern. names, Harmal, Spalani (Pashto). Shah & Khan (2006) stated that *Peganum harmala* seeds are antiseptic, and used in the treatment of asthma, paralysis, gastrointestinal, urinary problems, epilepsy and menstrual disorders. *Peganum harmala* seeds are anthelmintic especially for tape worms, for hemorrhoids and baldness. They are brain tonic and used along with olive oil for ear problems (Qureshi *et al.,* 2007). Arshad *et al*., (2008) reported flavonoids and anthraquinons from *Peganum harmala*. Bukhari *et al*., (2008) and Sarpeleh *et al.,* (2009) isolated harmine, harmaline, harmal, harmalol, tetrahydroharmine and tetrahydroharmol from seeds. Aziz *et al*., (2010) and Nenaah (2010) reported β-carboline alkaloids and a novel carboline alkaloid from aerial parts of *P. harmala* and it showed antibacterial activity against *Streptococcus pyogenes*. Farouk *et al*., (2009) and Khan *et al*., (2011) detected alkaloids, saponins, tannins, anthraquinones, flavonoids, flavons, flavonols, chalcones, terpenoids, phlobatannins and cardiac glycosides in *P. harmala*.

Tribulus terrestris L., Caltrop, Puncture vine (English), vern. name: Bhakra, It is common in sandy soils of waste places and cultivated fields.Little information is available on the oil composition of *T. terrestris*.

Chrozophora tinctoria (L.) Raf. is known as dyers-croton (Delazar *et al*., 2006). It is used to treat warts while leaves are used in chest burning in Kadhi areas of Khushab (Qureshi *et al*., 2011). Delazar *et al*., (2006) reported flavonoids, alkaloids, coumarins, chromones, xanthones, phenylpropanoid glycosides and flavonoids, acylated flavone glucoside, named chrozophorin from *C. tinctoria*. The aerial parts of *C. tinctoria* had flavonoids 1, 3 and 4, and plant contained acacetin 7-*O*-rutinoside and flavonoid 5 a novel natural product. It also had flavonoids 2-5 (apigenin derivatives with glycosylation at C-7.

Qureshi *et al.,* (2009) stated that *Ricinus communis* L. is an erect, single-stemmed or much-branched shrubby or tree like plant and having English name: Castor Seed; Erand (Hindko, Gujri), Arand (Punjabi, Urdu) and Haran (Sindhi). Castor oil is given in constipation before and after child birth to mother. Kensa & Yasmin (2011) stated that oil and seeds are used for warts. Ricinoleic acid has been served in contraceptive jellies. Castor oil softens and lubricates the skin. A branded gel Gastro oil is useful against dermatitis and protective in eczema and dermatitis (Rizvi, 2007). Kensa & Yasmin (2011) detected tannins, saponins, alkaloids, phenols, flavonoids, sterols, resins, and ricin in *R. communis*. Darmanin *et al*., (2009) identified monoterpenoids, 1, 8-cineole, camphor, pinene and caryophyllene in *R. communis* leaves. Kadri *et al*., (2011) reported thujone, 1,8-cineole, pinene, camphor and camphene in the essential oils of *R. communis*. No reports are available on the detailed physicochemical features of

the oils of the presently studied plants from Pakistan. An attempt was made to undertake a thorough investigation of the essential oils of these plants.

Materials and Methods

Collection of plant samples: The aerial parts of *Fagonia cretica* L., *Peganum harmala* L., *Tribulus terrestris* L., *Chrozophora tinctoria* (L.) Raf. and *Ricinus communis* L.,were collected from Peshawar and Attock Hills during 2009 and air-dried for about one week. The dried powdered samples were stored in plastic bags for oil extraction. Voucher specimens were deposited in Botany Department, University of Peshawar, Pakistan.

Dilute approximately 40 mg of oil samples, weighed accurately up to 0.1 mg, with 2 ml of dichloromethane and filtered through 0.45 µm membrane filter and injected 1 µl to GC-MS using auto injection system. The obtained essential oil from the aerial parts of *F. cretica*, *P. harmala*, *T. terrestris*, *C. tinctoria* and *R. communis* was analyzed using Shimadzu Model QP 2010 plus GC-MS. The separation was performed on a column (30 m × id 0.25 mm, thickness 0.25 µm 95% Dimethyl-5% diphenyl polysilphenylene; DB-5MS, Agilent technologies, USA) using Helium as the carrier gas with a splitting ratio of 1:50. The injector and the interface temperature were 240ºC and 240ºC, respectively. The *m/z* range was 40-500. The relative amounts (RA) of individual constituents of the oil are expressed as the percent peak area relative to the total peak area (Essien *et al*., 2008). The oil analysis was carried out at PCSIR Laboratory, Peshawar, Pakistan.

Results and Discussion

The identified oil components from the aerial parts of investigated plants are summarized in Table 1.

Table 1. Comparative percentage of various constituents in some plants of family Zygophyllaceae and Euphorbiaceae.

S. #	Family	Zygophyllaceae			Euphorbiaceae	
	Compounds	***Fagonia*** *cretica*	***Peganum harmala***	***Tribulus terrestris***	***Chrozophora tinctoria***	***Ricinus communis***
1.	Beta Phellandrene	0.337	0.0	0.0	0.0	0.0
2.	o-Cymene	6.166	0.0	0.0	0.0	0.0
3.	Cineole	5.088	0.0	0.0	0.0	0.0
4.	Alcanfor	63.925	28.24	0.0	0.0	0.0
5.	n-Octyl acetate	5.541	6.671	5.811	0.0	0.0
6.	Trans-Geraniol	1.884	2.753	0.0	0.0	0.0
7.	Alpha –Citral	1.638	2.602	0.0	0.0	0.0
8.	Geraniol-acetate	1.229	2.635	0.0	0.0	0.0
9.	Eugenyl methy	1.748	9.674	5.473	3.692	0.0
10.	Alpha Curcumene	1.084	2.135	0.0	0.0	0.0
11.	Alpha-Limonene diepoxide	0.801	0.0	0.0	0.0	0.0
12.	Cyclohexyl Ketone	3.464	6.663	14.768	13.742	3.745
13.	2(4H)-Benzofuranone,5,6,7,7a tetrahydro-4,4,7a-trimethy	2.19	7.175	38.194	50.718	77.645
14.	Citronellyl propionate	1.142	2.259	0.0	0.0	0.0
15.	Alpha Cedrol	0.082	1.433	0.979	3.497	2.384
16.	Elemicin	0.636	3.669	7.162	8.558	2.947
17.	Capillin	3.046	13.176	23.556	9.948	3.156
18.	p-Anisaldehyde	0.0	0.380	0.0	0.0	0.0
19.	6-Isopropyl-3,6-dimethyl-2-cyclohexane-1-one	0.0	0.545	0.0	0.0	0.0
20.	Germacrene D	0.0	0.208	0.0	0.0	0.0
21.	1H-Cycloprop[e]azulen-7-ol,decahydro-1,1,7-trimethyl	0.0	8.671	0.0	9.845	8.504
22.	Caryophyllene oxide	0.0	0.581	0.0	0.0	0.0
23.	Z,E)-Farnesol	0.0	0.523	0.0	0.0	0.0
24.	Phenol,2-methyl-5-(1-methylethyl)-	0.0	0.0	1.839	0.0	0.0
25.	Beta Farnesene	0.0	0.0	1.698	0.0	0.0
26.	Delta-cadinol	0.0	0.0	0.0	0.0	0.0
27.	Beta Iraldeine	0.0	0.0	0.0	0.0	1.136
28.	P-cymene	0.0	0.0	0.0	0.0	0.481

A. Zygophyllaceae

Percentage of major groups of constituents

Oxygenated monoterpene: Oxygenated monoterpenes have antispasmodic, sedative and tranquilizing action (Gyawali & Kyong-Su Kim, 2012). They have antimicrobial, food preservatives and natural antioxidants properties (Kotan *et al*., 2008; Kordali *et al*., 2009; Gundidza *et al.,* 2009; Abu-Darwish *et al*., 2012). They have antitumoral activity and prevented mammary, lung, skin, liver and stomach cancers in rat models (Fayed, 2009). Judpentiene & Mockute (2004) stated 1,8-cineole helps in cold and other hardships of the Himalayan region and higher amounts are toxic. They are widely used in the flavoring agents (Chang & Kim, 2008).

In *Fagonia aretica* oils, 17 compounds were identified constituting 100% of the oil composition (Tables 1 & 3). Oxygenated monoterpenes (89.94%), were a major group of compounds and it consisted of 2(4H)-Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy, alcanfor, O-cymene, cineole, trans-geraniol, eugenol methy and α-citral and esters had lower amounts. Other chemical groups had less than 1.22% components (Table 2). Several workers found low oxygenated monoterpenes in different medicinal plants **(**Bozin *et al*., 2008; Sukari *et al*., 2008; Moradalizadeh *et al*., 2012**)** while *F. aretica* showed increseased percentage of oxygenated monoterpenes. Several researchers **(**Sonibare & Effiong, 2008; Kanjilal & Kotoky, 2010) reported less alcanfor in different medicinal plants. Ebadollahi *et al*., (2010) also reported poor alcanfor in *Lavendula stoechas* and *Eucalyptus globulus* as compared to *F. aretica* and *P. harmala*. Earlier workers (Fayed, 2009; Chun-Di Hu *et al*., 2010; Zhang *et al*., 2011) found high amount of trans-geraniol in *Pelargonium graveolens*, *Mentha arvensis* and *Osmanthus fragrans*. These results are not in line with the present study.

Peganum harmala oils had 18 compounds constituting 99.8% of the total oil composition (Tables 1 & 4). These oils consisted of oxygenated monoterpenes 36.01%, with major components as alcanfor 28.244% and capillin 13.176%. Esters were 11.58% (Table 2). Other chemical groups were lower than 11.01% in the oil.

There were 10 compounds in *T. terrestris* oils that consisted 100% of the total oil composition (Tables 1 & 5). It consisted of oxygenated monoterpenes (90.99%), having 2(4H)-Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy 38.194% and capallin 23.556% and cyclohexyl ketone 14.768% Esters were 5.8% (Table 2). Other chemical groups shared less than 5.8% components in the oil. Several workers have reported variation of 2(4H)- Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy(volatile terpenes) in different medicinal plants (Wang *et al*., 2006; Akhbari *et al.,* 2011; Akhol & Yilmazer, 2011; Chalannava, 2011; Ramasubramaniaraja, 2011). These findings agree with the present inferences. All these studies found less 2(4H) - Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy as compared to *F. cretica, P. harmala*, *T. terrestris.* Abirami & Rajendran (2011) found α-Amyrin (63.73%) in *T. terrestris* and found long chain hydrocarbons and eight minor components. This disagrees with the present findings.

Essential oils rich in alcohols, aldehydes, phenols, esters, sesquiterpenes and ketones had antimicrobial and spasmolytic effect (Derwich *et al*., 2004; Hammer & Carson, 2011).

Table 2. Composition of essential oil of some selected plants of family Zygophyllaceae and Euphorbiaceae.

Constituents	Zygophyllaceae			Euphorbiaceae	
Major groups	***Fagonia*** *cretica*	***Peganum harmala***	***Tribulus terrestris***	***Chrozophora tinctoria***	***Ricinus communis***
a. Oxygenated monoterpenes (%)	89.94	36.01	90.99	86.93	88.63
b. Monoterpenes hydrocarbons (%)	1.22	1.01	0.0	3.5	2.86
c. Sesquiterpenes hydrocarbons (%)	1.08	11.01	1.7	9.85	0.0
d. Oxygenated sesquiterpenes (%)	0.082	2.54	1.5	3.5	2.4
e. Esters (%)	5.5	11.58	5.8	0.0	0.0
Total oil (%)	100	99.8	100	100	100
Number of compounds	17	18	10	7	8

Table 3. Components, their retention time, concentration (%) and peak area of *Fagonia cretica* oil.

Name of components	R.Time	Conc. (%)	Area
Beta-Phellandrene	12.32	0.337	4574
o-Cymene	12.56	6.166	83682
Cineole	12.90	5.088	69046
Alcanfor	18.17	63.925	867546
n-Octyl acetate	20.35	5.541	75205
Trans-geraniol	21.61	1.884	25566
Alpha-citral	21.61	1.683	22231
Geraniol acetate	24.12	1.229	16675
Eugenyl methy	23.94	1.748	23722
Alpha-curcumene	25.21	1.084	14706
Alpha- Limonene Diepoxide	25.69	0.801	10875
Cyclohexyl ketone	25.69	3.464	47013
2(4H)- Benzofuranone, 5,6,7,7a-tetrahydro -4,4,7a-trimeth	26.02	2.190	29717
Citronellyl propionate	26.44	1.142	15493
Citronellyl Cedrol	27.43	0.082	1116
Elemicin	27.43	0.636	8630
Capillin	27.51	3.046	41335

Table 4. Components, their retention time, concentration (%) and peak area of *Peganum harmala* oil.

Name of components	R.Time	Conc. (%)	Area
Alcanfor	18.17	28.244	254248
n-Octyl acetate	20.35	6.671	60052
Trans-geraniol	21.60	2.753	24783
P- anisaldehyde	21.09	0.380	3418
Alpha-citral	21.60	2.602	23425
6 isopropyl- 3,6 dimethyl – 2- cyclohexen- 1 one	22.33	0.545	4910
Geraniol acetate	24.11	2.635	23724
Eugenyl methy	23.94	9.674	87080
Alpha- curcumene	25.21	2.135	19223
Germacrene D	25.14	0.208	18730
Cyclohexyl ketone	25.69	6.663	59983
2(4H)- Benzofurancone,5,6,7,7 a- tetrahydro - 4,4,7a - trimeth	26.02	7.175	64592
Citronellyl propionate	26.44	2.259	20335
IH-cycloprop{e} azulen- 7- ol, decahydro- 1,1,7 trimethyl -	26.67	8.671	78051
Caryophyllene oxide	27.06	0.581	5233
Alpha cedrol	27.15	1.433	12902
Elemicin	27.43	3.669	33032
Capillin	27.50	13.176	118608
(Z,E) farnesol	28.21	0.523	4710

Table 5. Components, their retention time, concentration (%) and peak area of *Tribulus terristris* oil.

Name	R.Time	Conc. (%)	Area
n-Octyl acetate	20.12	5.811	11856
Phenol, 2-methyl-5-(1-methlethyl)-	22.00	1.839	3751
Eugenyl methy	23.93	5.473	11166
Beta-farnesene	24.82	1.698	3464
Cyclohexyl ketone	25.68	14.768	30129
2(4H)- Benzofurancone,5,6,7,7 a- tetrahydro - 4,4,7a – trimeth	26.01	38.194	77919
Alpha cedrol	27.14	0.979	1997
Elemicin	27.42	7.162	14611
Capillin	27.50	23.556	48056
Delta-Cadinol	27.43	0.521	1062

Comparing the chemical groups in three tested plants of family Zygophyllaceae, it was found that oxygenated monoterpenes were the highest (90.99%) in *T. terrestris*, followed by *F. cretica* (89.94%) and the lowest (36.01%) occurred in *P. harmala* (Table 2). These findings agree with (Judpentiene & Mockute, 2004; Al-Qudah *et al*., 2010) who reported less amount of oxygenated monoterpenes and sesquiterpenes hydrocarbons in oils of *Dittricha viscosa* and *Artemisia absinthium*. *Peganum harmala* had maximum esters followed by *T. terrestris* and *F. cretica*. Monoterpenes hydrocarbons were maximum in *F. cretica* followed by *P. harmala* and absent in *T. terrestris*. Sesquiterpenes hydrocarbons were highest in *P. harmala* and absent in *T. terrestris* (Table 2). The variations in occurrence of these sesquiterpenes might be used to differentiate the respective oils.

Euphorbiaceae: Eight compounds were identified in *Chrozophora tinctoria* oil giving complete oil composition (Tables 1 & 6). It had oxygenated monoterpenes 86.93%, comprising 2(4H)- Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy 50.718%, cyclohexyl ketone 13.742%, capallin 9.948%. Esters were absent (Table 2).

Ricinus communis oil had 8 compounds with 100% of the oil composition. (Tables 1 & 7). The oxygenated monoterpenes were 88.63%, and it consisted of 2(4H)-Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy 77.6%, this value was the highest individual component in the present study. Esters were absent (Table 2). Other chemical groups had less than 2.4% components in the oil.

Several workers observed variation of 2(4H) - Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy in different plants (Akhbari *et al*., 2011; Ramasubramaniaraja, 2011). That agrees with the present results. These findings reported less 2(4H)- Benzofuranone, 5, 6, 7, 7a tetrahydro-4, 4, 7a-trimethy as compared to *C. tinctoria* and *R. communis.*

Comparing the chemical groups in two plants of family Euphorbiaceae it was observed that oil consisted of oxygenated monoterpenes and it was highest (88.63%) in *R. communis* and lowest (86.93%) in *C. tinctoria*. Monoterpenes hydrocarbons were highest (3.5%) in *C. tinctoria* than in *R. communis* (2.86%) (Table 2). These

findings agree with Kadri *et al.,* (2011) who also reported increased monoterpenes hydrocarbons and oxygenated monoterpenes in oils of *R. communis*. *Chrozophora tinctoria* had 9.85% sesquiterpenes hydrocarbons and were absent in *R. communis*. Moreno *et al*., (2012) also reported sesquiterpenes hydrocarbons in essential oil of *Croton heterocalyx* (Euphorbiaceae). *Chrozophora tinctoria* had more (3.5%) oxygenated sesquiterpenes than in *R. communis* (2.4%) (Table 2). Similar results were also reported by Lima *et al*., (2009) and Patricia *et al*., (2011) who studied volatile constituents in oils of *Acalypha ornata* and *Julocroton triqueter*.

The variations in the percentage of chemical composition of essential oils can be explained by the differences in the geographic origins, climate, nature of soil, solar radiation, age of the plant and part of the plants. All these factors involve the activation or inactivation of certain enzymatic groups, leading to the predominance of a particular biosynthetic pathway **(**Khadhri *et al*., 2011; Neves & Camara, 2012).

The knowledge of the chemical composition of an essential oil is of key importance for determining its potential industrial or medicinal applications. The literature shows that there are no reports on the detailed physicochemical features of the oils of the studied plants from Pakistan.

In the present study all the plants were found to be a rich source of oxygenated monoterpenes representing 2(4H)-Benzofuranone, 5, 6, 7,7a tetrahydro-4, 4,7a-trimethy as the major constituent. This study revealed that phytochemical attributes and chemical composition of the studied plants have potential uses for food, pharmaceutical and cosmetic applications in future. Detailed research work on the antioxidant principles and biological activities of the studied plants is further recommended.

Table 6. Components, their retention time, concentration (%) and peak area of *Chrozophora tinctoria* oil.

Name	R.Time	Conc. (%)	Area
Eugenyl methy	23.95	3.692	1646
Cyclohexyl ketone	25.69	13.742	6126
2(4H)- Benzofuranone,5,6,7,7 a- tetrahydro - 4,4,7a – trimeth	26.01	50.718	22610
IH – cycloprop{e} azulen - 7- ol, decahydro - 1,1,7 trimethyl -	26.66	9.845	4389
Alpha cedrol	27.14	3.497	1559
Elemicin	27.43	8.558	3815
Capillin	27.52	9.948	4435

Table 7. Components, their retention time, concentration (%) and peak area of *Ricinus communis* oil.

Name	R.Time	Conc. (%)	Area
Cyclohexyl ketone	25.69	3.745	3357
2(4H)- Benzofuranone,5,6,7,7 a- tetrahydro - 4,4,7a – trimeth	26.01	77.645	69595
IH – cycloprop{e} azulen – 7- ol, decahydro – 1,1,7 trimethyl -	26.67	8.504	7622
Alpha cedrol	27.15	2.384	2137
Elemicin	27.43	2.949	2643
Capillin	27.53	3.156	2829
Beta-Iraldeine	3.62	1.136	1018
p-Cimene	32.48	0.481	431

Conclusion

Our findings suggest that the essential oils from five plants could have potential in the pharmaceutical, cosmetics industries and aroma therapy. Research is needed to prove this. Other pharmacological activities e.g., anticancer, antidiabetic, antilashmanial, antiinflammatory and analgesic etc can be carried out in future to assess these plants therapeutic potential.

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