Article

Chemistry of essential oils of Citrus species

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Abstract

Essential oils from genus ***Citrus*** are natural flavouring materials of commercial importance. Worldwide demand of essential oils, especially ***Citrus*** oils, has increased during the past few years. The chemistry of volatile oils of ***Citrus*** species like ***Citrus aurantifolia, C. aurantium, C. bergamia, C. hystrix, C. jambhiri, C. limon, C. paradisi, C. medica, C. reticulata, C. sinensis, C. unshiu*** and little known species like ***C. elementina, C. flaviculpus, C. grandis, C. ichangensis, C. indica, C. inflata, C. junos, C. sudachi, C. tachibana*** and ***C. tamurana*** have been reviewed here. A considerable difference in chemical composition of a particular species is observed, which may be due to the effect of soil or geographical regions or environment. It can help in the isolation of a particular high valued component from corresponding species and from specified geographical area.

Introduction

There are a great number of ***Citrus*** varieties; most of them are evergreen aromatic shrubs, which are widely distributed throughout the tropical and temperate regions of the world. They have pleasant aroma, taste and are a good source of Vitamin C. The genus is believed to be native to subtropical and tropical regions of Asia and Malaysian archipelago. Most of the species are cultivated in India for their fruits, viz. ***C. aurantifolia*** (lime), ***C. aurantium*** (sour or bitter orange), ***C. limon*** (lemon), ***C. maxima*** (pummelo), ***C. reticulata*** (mandarin) and ***C. sinensis*** (sweet orange). These species produce volatile oils from their leaves, stem, fruit and fruit peel, which contain a number of chemical compounds mostly mono and sesquiterpenoids. These volatile oils have been applied in perfumery industry, to inhibit the growth of various pathogenic micro-organisms, in food preservation and in aromatherapy. In order to investigate the structure activity relationship, it is necessary to know the chemical composition of these volatile oils. The objective of our ongoing investigation is to establish a chemical basis for subsequent ethnobotanical evaluations of oil constituents as potential source for beneficial uses. A great number of studies on ***Citrus*** essential oils has been reported. Shaw[1] presented a comprehensive review in 1979 and very recently Sawamura[2] has written on cold pressed volatile oils of ***Citrus*** spp. In view of commercial value and wide applications of ***Citrus*** essential oil in flavouring industry, researchers require detailed information especially about the chemistry of the essential oils. Keeping this in view and in continuation of our research program[3-14], we have reviewed here the chemistry of various samples and species of ***Citrus*** oils.

Extraction of volatile oil

Most of the ***Citrus*** leaf, peel and fruit (whole), which are reviewed here, have been collected from different countries of the world like Chinese, Cuban, Brazilian, Andalusian, Sicilian, Japanese, Indian, Mexican, Korean, Libyan, etc. They are also from different cultivars and species.

The plant materials were cut into small pieces, washed and hydrodistilled under Clevenger's apparatus[66] to yield the

volatile oils. Some *Citrus* oils were obtained by supercritical CO_2 extraction method and cold pressed techniques. The chemistry of some solvent extracted volatile oils has also been reviewed here.

Chemical investigation

Investigations on chemical composition of most of the volatile oils were undertaken by various authors using GC and GC-MS techniques. The other techniques used are Kovat's index from packed capillary column, retention time data from GC, HPLC, GC-MS; capillary GC, HRGC-FID, GC-MS, HRGC-MS and IR, C^{13} NMR. The identification of components was done by co-injection with authentic samples and also by comparing their mass spectra with those available in literature.

Chemical composition and utilization

The chemical composition of various species of ***Citrus*** obtained from various sources are as follows:

Citrus aurantium Linn. (Bitter orange, Hindi- *Khatta*)

Bitter orange is said to be indigenous to India, mostly cultivated in Guntur district (Andhra Pradesh). It includes a number of varieties, which are mostly hybrid in origin and used in preparation of confection, marmalades, liqueurs and other drinks. It produces essential oil from its leaves and peel of fruits whose major components are listed in Table 1. A number of samples of bitter orange leaf oil was analysed[15-20] by GC and GC-MS and found rich in linalool/linalyl acetate. Lin *et al*[21] investigated two Chinese bitter orange oil by GC-MS and retention index studies, which indicated linalyl acetate and in another limonene as major components. A comparative chemical investigation on leaf and peel oil of bitter orange showed[22] linalyl acetate in leaf oil and limonene in peel oil as major component. Limonene was also reported to be a major component in commercial sample[23] and peel oil obtained at different storage condition[24]. Dugo *et al*[25] determined the variation in chemical composition of a number of cold pressed bitter orange oils. The monoterpene hydrocarbons (limonene) in Cuban bitter orange oil was also reported[26] as major components. Boelens and Jimenez[27] compared the chemical composition of Andalucian (Spanish) with Sicilian (Italian) bitter orange oil and found limonene as major component. Fakim *et al*[28] investigated leaf oil (Mauritius origin) by capillary GC and GC-MS which gave linalool as major component. Very recently, Song *et al*[29] and Sawamura[2] examined cold pressed daidai peel oil by GC and GC-MS and found limonene as main component. These results show linalool/ linalyl acetate as major component in leaf oil. However, limonene was found in the peel oil of this plant.

Citrus aurantium

Citrus aurantifolia (Christm.) Swingle (Lime, Hindi- *Kaghzi nimbu*)

It is distributed in the most parts of tropical region and used for flavouring jams, jellies, marmalades and alcoholic drinks. It gives well-known lime oil on distillation or solvent extraction. Chemistry of various samples of these oils is listed in Table 2. Kamiyama and Amaha[15] analysed chemical composition of Japanese lime leaf oil and found geranial, neral and limonene as major components. Shaw[1] reported geranial followed by limonene as major constituents. The GC-MS analysis of lime leaf oil from different origins[30-32], showed presence of limonene in high content. Koketsu *et al*[33] examined the chemical composition of a number of samples of lime oil of Brazilian origin, using Kovat's indices from packed capillary column and reported limonene/β–pinene as major components. The GC and retention time data analysis of lime oil from India [34-35] and Cuba[36] showed high content of limonene. Clark and Chamblee[37] undertook GC and GC-MS analysis of lime oil and reported limonene as major component. A high content of terpene hydrocarbons (86.66%) was also reported[38] in a Mexican lime oil. Recently Jiwajinda *et al*[39] analysed lime oil from Thailand and found six furan-ocoumarins. These studies concluded the presence of limonene, geranial and neral as major components and it also seems that there occurs a large difference in chemical composition of lime oil from different origins.

Citrus aurantifolia

Table 1: Major constituents (%) of essential oils of *Citrus aurantium*

β-Pinene	Limonene	Linalool	Linalyl acetate	Geraniol	Sabinene	Terpinene	Nerol	α–Terpineol	Myrcene	β–Ocimene	Ref.
3.0	0.4	38.4	49.8	-	0.5	-	-	1.0	1.0	1.5	15
-	3.5-10	26-30	30-46	-	-	-	-	3-7	-	-	18
0.48-2.51	0.14-0.58	57.14-67.16	13.59-23.03	1.07-2.13	0.13-0.55	-	1.13-4.33	3.02-5.87	0.51-1.28	1.01-2.56	16
1.57	1.05	26.62	50.01	2.24	0.30	-	0.95	5.10	1.96	1.81	17
Tr	0.78	11.72	44.08	0.98	-	-	Tr	1.26	1.02	0.30	21
2.5	4.0	24.1	45.5	1.8	0.4	-	0.8	5.2	2.5	2.6	20
2.4	4.4	17.0	56.8	2.3	0.4	-	0.8	3.7	1.5	1.6	22
0.7-1.7	0.7-1.1	19.9-26.9	46.3-55.0	2.0-3.5	0.4	-	1.0-1.5	4.6-7.6	1.3-5.6	1.1	20
9.4	0.9	36.0	35.4	-	-	-	-	-	-	-	19
0.6	81.3	-	-	-	-	7.3	-	-	-	-	26
0.46	93.5	0.23	-	-	-	-	-	-	-	-	23
0.2	92.2	0.34	0.75	0.02	-	-	-	-	-	-	24
0.14	93.2	0.24	0.28	0.02	-	-	-	-	-	-	24
0.13	94.5	0.20	0.23	0.02	-	-	-	-	-	-	24
0.63-1.2	0.20-0.33	0.64-1.17	-	-	-	-	-	-	-	-	25
0.09	96.41	0.16	0.64	-	-	-	tr	0.03	1.91	-	2
0.29	94.34	0.15	0.284	0.01	0.137	0.121	0.005	0.567	1.806	-	27
0.9	92.43	0.371	0.775	0.015	0.274	0.103	0.01	0.061	2.073	-	27
0.5	93.6	0.1	-	-	0.2	0.3	-	0.2	1.8	-	22
-	-	66.10	-	0.99	-	-	-	0.30	tr	tr	28
0.4	92.5	0.2	1.4	tr	0.2	-	-	0.1	2.2	0.3	29

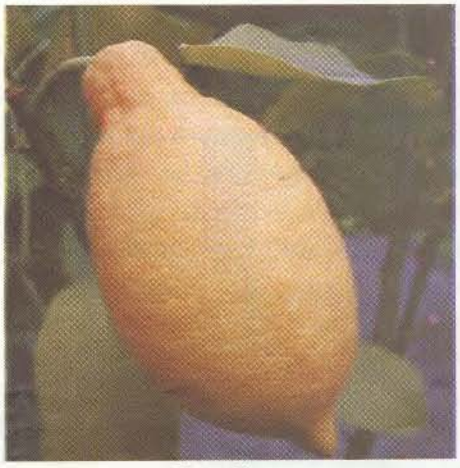
Table 2: Major constituents (%) of *Citrus aurantifolia* (lime) oil

α-Pinene	β-Pinene	Sabinene	Limonene	γ-Terpinene	p-Cymene	1,8-Cineole	Nerol	Geranial	Geraniol	Reference
4.3-5.0	14.6-16.0	0.9-1.4	51.5-59.7	1.3-8.5	1.2-10.4	-	0.5-1.2	2.2-3.9	tr	33
2.0	3.6	-	32.0	11.3	6.8	1.4	2.3	3.8	-	36
1.9	11.6	-	60.4	17.6	0.1	1.8	16.6	20.7	4.0	31
1.1	4.9	-	47.3	-	6.8	-	-	2.9	-	34
1.4	6.8	-	49.4	6.0	2.6	-	-	1.5	0.3	34
0.2	0.4	-	35.9	8.6	-	-	-	4.0	0.5	34
1.05	1.41	tr	45.26	10.97	2.39	4.52	-	-	-	24
0.3	0.3	2.3	24.7	0.3	tr	-	23.8	40.0	0.6	1
-	-	-	21.6	-	-	-	12.4	-	17.4	30
0.61	5.83	-	33.76	-	-	-	-	21.54	3.95	32
2.48	-	3.37	49.20	8.04	-	-	1.88	2.89	0.04	37
2.57	-	3.43	57.8	0.61	-	-	-	-	-	38
1.91	9.61	0.07	46.57	11.11	1.88	-	0.03	0.07	2.01	35

Citrus limon (Linn.) Burm. f. (Lemon, Hindi-*Nimbu*)

It is a native of North West regions of India and is cultivated in home gardens in U.P., Maharashtra, Tamil Nadu and Karnataka. It is widely used in the preparation of lemonade, squash and home made sherbet. It is used in culinary preparations like lemon pies, lemon cakes, lemon ices and in flavouring for candies, jellies, jams and marmalades.

Chemistry of the leaf and peel oil of this plant is reported in Table 3. The lemon leaf oil was found to contain limonene as major component[18, 22, 40-43]. Kumiyama and Amaha[15] examined methyl chloride extract of lemon leaf of Japanese origin throughout a season using GC as method of analysis and geranial was reported as main component. An Argentinean lemon oil analysed[44] by capillary GC showed presence of limonene, γ-terpinene and β-pinene as major components. Limonene and β-pinene are also reported[45] as main components in the lemon leaf oil analysed by GC and GC-MS techniques. Two Russian cultivars (Georgian and Monacello leaf oil) were analysed[46] and found to contain limonene, β-pinene, neral and geranial as main components. Crescimanno *et al*[47] compared the chemical composition of four samples of lemon leaf oils from Italy, which were rich in geranial. In addition Barth *et al*[48] used supercritical CO_2 extraction to separate lemon oil into four fraction – terpene-rich fraction, deterpenified oil, wax-rich fraction and residual fraction. The HPLC and GC-MS analysis[38] of one of the Italian samples showed the presence of mono and sesquiterpenes. A number of samples of lemon oil from three different areas of United State was analysed[24] by GC and found to contain limonene as main component. The commercial[49] lemon oil analysed by GC-MS technique showed limonene as major component. Ayedoun *et al*[50] examined the lemon oil (limonene rich) using capillary GC and GC-MS. Recently, Miyake *et al*[51] isolated two glucosylflavones namely 6,8-di-C-β-glucodiosmin and 6-C-β-glucosyldiosmin from peel of lemon fruits. The same author (Miyake *et al*[52]) has also isolated coumarins namely 8-geranyloxypsolaren, 5- geranyloxypsolaren (bergamottin) and 5-geranyloxy 7-methoxy coumarin by



Citrus limon

Table 3: Major constituents (%) of *Citrus limon* (Lemon) oil

α-Pinene	β-Pinene	Limonene	γ-Terpinene	Linalool	Geraniol	Geranial	Neral	Reference
2.1-3.6	12.1-14.4	22.2-28.9	2.1-3.5	1.7-3.2	1.3-2.8	24.2-29.6	16-18.2	15
-	-	30	-	24	-	-	-	18
1.6	11.4	70.2	7.5	0.1	-	1.0	0.5	44
1.5	18.7	38.6	0.2	1.2	1.6	9.7	5.9	40
1.38	17.1	25.9	0.26	1.6	1.4	11.1	1.42	45
0.84-1.35	13.32-18.43	22.71-26.07	0.21-0.28	0.89-1.03	-	12.03-14.79	12.03-14.79	46
1.66-2.23	3.97-5.22	14.59-16.29	1.48-1.91	-	12.44-15.04	-	21.48-25.32	47
2.4	12.6	67.3	7.6	0.1	-	0.8	0.9	22
0.15	0.1	38.2	0.5	8.0	3.0	15.20	10.20	41
2.5	13.9	64.6	10.2	-	0.06	1.13	0.61	42
1.38	9.98	71.52	9.66	0.41	-	0.82	0.54	48
1.5-5.0	6.0-14.0	60.0-80.0	6.0-12.0	0.2	0.1	2.0	-	43
4.2	18.5	70.4	11.8	-	-	-	-	50
1.5-1.7	2.8-10.6	58.4-74.3	7.3-8.1	-	-	-	-	49
tr	-	90.6	tr	1.95	-	1.58	0.92	24
1.58	8.908	70.36	0.17	0.114	0.045	0.948	0.572	24
2.08	12.172	67.57	0.18	0.85	0.269	1.68	1.227	24

spectroscopic analysis. Dellacassa *et al*[53] analysed the composition of 31 Uruguayan lemon oils using HRGC- FID and GC-MS and also studied the enantiomeric distribution of α-pinene, sabinene, limonene, linalool, terpinen-4-ol and α-terpineol. These studies showed that limonene, β-pinene, neral, geranial and γ-terpinene are the major components in lemon oils.

Citrus paradisi Macf. (Grape fruit)

It is reported to have originated in Western India and is used as breakfast fruit. ***C. paradisi*** is a rich source of vitamin C and has good medicinal value. Its regular use builds up resistance to common colds and wound infections. From a taxonomical standpoint grape fruit is known to be a stabilised hybrid between pummelo or shaddock (***C. grandis***, also called ***C. maxima***) and the sweet orange (***C.sinensis***).

The chemistry of this oil is listed in Table 4. A number of varieties of grape fruit is known which have different chemical composition. Attaway *et al*[54] examined Duncan marsh cultivars and a grape fruit grown in Florida using a combination techniques including mass spectroscopy and found sabinene as major component. The leaf oil of ***C. paradisi*** from Japan[1] and Taiwan[40] was analysed using GC analysis and was found to contain sabinene as major component. A high content of limonene in Argentinean grape fruit was also reported[44] using GC studies. The limonene rich grape fruit oils were also analysed by various workers[22, 55-59]. Ekundayo *et al*[60] examined the Nigerian ***C. paradisi*** leaf oil using GC and GC-MS and found p-cymene, terpinen-4–ol, linalool and limonene as major components. A sample of leaf oil from Mauritius analysed[28] by capillary GC and GC-MS, contained 45 components with linalool and terpinen-4-ol as major components. In

Citrus paradisi

Table 4: Major constituents (%) of *Citrus paradisi* (Grape fruit) oil

α-Pinene	Sabinene	Limonene	γ-Terpinene	Linalool	Terpen-4-ol	Citronellol	p-Cymene	Reference
1.6-2.7	42.0-59.0	1.6-3.3	1.2-3.8	5.9-24.0	1.4-14.0	-	-	54
2.4	18.5	6.4	12.0	4.4	0.8	-	1.4	1
0.53-0.54	0.84-0.86	91.8-93.1	0.13-0.22	0.079-0.23	-	-		44
2.98	61.91	4.38	0.82	3.33	099	-	0.05	40
3.09	2.49	11.37	-	12.78	17.02	-	21.32	60
0.52-0.58	0.38-0.40	92.9-93.8	0.09-0.01	0.08-0.09	-	-	tr	55
0.42-0.47	0.27-0.60	76.14-83.11	0.08-0.12	0.17-0.30	0.24-0.4	-	tr	56
0.33	0.36	92.16	0.01	0.34	-	-	-	24
-	-	0.14	-	22.93	20.00	8.6		28
-	-	89.53-95.34	-	-	-	-	-	60
0.15	0.81	88.5	0.05	0.13	0.20	-	0.4	58
0.45	0.4	92.5	0.07	0.08	0.12		0.1	59

1985, Correa *et al*[61] analysed the chemical compositions of Cuban grape fruit oil using a combination of techniques and results indicated the presence of 89.4% hydrocarbons (α-pinene, limonene, sabinene, myrcene, γ-terpinene, etc.) and 9.4% oxygenated compounds (geraniol, nerol, cetronellol, α–terpineol, octanol, linalool, etc). Various natural products like coumarins and limonoids as well as four new acridone-coumarine dimers are also isolated[62-63] and characterised from different part of the plant. On the basis of these studies, it may be concluded that the grape fruit oil contains limonene, sabinene, linalool, terpinen-4-ol and p-cymene as major components. It may also be concluded that oils obtained from different origins have different compositions.

Citrus sinensis (Linn.) Osbeck (Sweet orange, Hindi-*Musambi*)

It is a native of China and India and is cultivated in subtropical regions. Its fruits are sweet, juicy, nutritious and highly esteemed as dessert fruits. The leaves and peel of the fruit produces essential oil, which contains different chemical composition as reported in Table 5.

Table 5: Major constituents (%) of *Citrus sinensis* (Sweet orange) oil

α-Pinene	β-Pinene	Sabinene	Limonene	Linalool	Linalyl acetate	Geranial	cis-Piperitol	Reference
0.35	0.37	0.21	92.2	0.46	-	0.1	-	44
2.19-2.79	1.50-1.74	41.8-54.75	2.53-5.57	3.9-8.55	0.11-0.16	0.04-0.06	-	40
0.2	-	0.2	52.0	15.8	-	3.5	-	70
1.48-3.09	1.28-2.49	16.03-29.48	3.71-7.38	9.67-17.59	0.0-0.26	0.69-7.9	-	73
0.88-1.75	1.06-1.30	15.81-32.58	1.25-2.96	5.13-20.92	-	-	-	65
-	-	-	0.2	28.0	0.1	6.5	-	67
0.13-0.32	0.93-1.83	0.518-1.62	55.12-86.57	-	-	-	-	68
0.1	0.5	-	94.8	-	-	-	-	69
1.6-2.6	-	52-58	2.3-6.4	1.6-16.0	-	0.5-2.5	-	66
2.7-3.1	4.5-5.7	42.4-51.0	2.6-4.9	8.7-14.5	tr	1.9-2.5	-	1
0.35	0.01	0.21	96.19	0.35	tr	tr	-	71
0.41	tr	0.37	96.57	0.12	tr	tr	-	71
0.94	0.32	-	-	1.65	-	1.04	26.42	28

Various samples of ***C. sinensis*** oil from Nigeria[64], Israel[65] and China[40] were examined using various techniques and found sabinene as major component. Sabinene as main component was also found[1] in two different samples of sweet orange leaf oil. The leaf oil of 'Hamlin' cultivar of sweet orange grown in Florida was examined[66] throughout the year using high efficiency packed column GC and also reported sabinene as main component. Moyler *et al*[67] examined a commercial sweet orange oil and found linalool as major component. A limonene-rich sweet orange oil was analysed by Cappello *et al*[44] from Argentina using GC and retention time studies and by Usai *et al*[68] using GC-MS technique. Singh *et al*[69] have reported a high content of limonene (94.8%) in Indian sweet orange peel oil using GC-MS techniques. We have also found that the oil and its major components as potent fungicides against fungal pathogens of sugarcane. Limonene was also reported[70] as major component in a sweet orange peel oil obtained from a local market of Libya, using GC-MS. A very interesting and different result was reported[28] in a ***C. sinensis*** leaf oil from Mauritius using capillary GC and GC-MS and found piperitol as major component. Very recently, Mitiku *et al*[71] analysed cold pressed fruit peel essential oils of two cultivars of sweet orange (Valencia and Hamlin) by GC and GC-MS, and found high content (>96%) of limonene. These results showed presence of sabinene, linalool and limonene as major component in sweet orange essential oil and the chemistry of oil varies with its origin.

Citrus reticulata Blanco (Mandarin or Tangerine, Hindi-*Santara*)

It is native of China and is widely cultivated in all subtropical regions. ***C. reticulata*** is the most valued commercial orange, used for production of orange juice. The leaves and peel of fruit produce useful essential oils. The chemistry of these volatile oils obtained from different parts and also from different varieties is listed in Table 6.

Vermin[18] determined a 50% content of monoterpene hydrocarbon along with limonene as a major component in Mandarin petitgrain oil. In another study[66], linalool was reported as the main component in leaf oil from 'Dancy' cultivar grown in Florida, analysed by high efficiency packed column. Kumiyama and Amaha[15] examined Mandarin leaf oil of Japanese origin and found linalool and sabinene as major components. Linalool was also reported[72] as major component in their different cultivars. A more different Mandarin leaf oil of Kinnow cultivar was analysed[73] and found rich in δ-3-carene. Kekelidze *et al*[74] compared the composition of Mandarin leaf oil which was rich in p-cymene. Nigerian mandarin leaf oil analysed [75] by combination of GC and GC-MS, showed terpinene as major component. Recently,

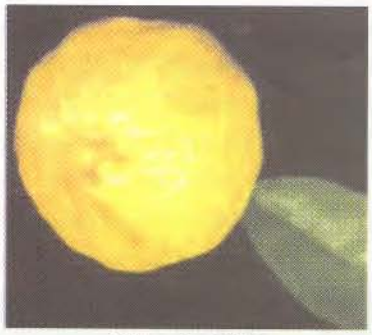
Table 6: Major constituents (%) of Mandarin or Tangerine leaf oil (*Citrus reticulata*)

Linalool	Limonene	Ocimene	α-Pinene	δ-3-Carene	Myrcene	β-Caryophyllene	Terpinene	β-Pinene	Ref.
2.00	35.00	-	-	-	-	-	-	-	18
2-78	0.8-1.9	2.8-8.2	1.1-2.7	-	0.3-0.6	-	0.1-0.3	1.4-2.6	66
59.2	1.2	2.1	1.3	tr	2.2	0.2	0.8	7.4	1
21.82	3.35	5.75	1.52	47.36	3.31	5.08	-	0.02	73
0-35	1-6	0-5	3-5	0-10	0-2	tr	-	tr	72
3.4-15.2	2.8-4.8	-	1.3-2.1	-	0.3-0.7	0.6-10.2	0.1-0.3	5.8-20.2	74
9.55	3.63	1.74	3.9	-	0.63	-	20.15	4.71	75
0.11	4.91	0.04	1.34	-	0.31	0.14	12.75	0.72	76
45.12	1.00	-	1.83	-	0.34	5.44	4.68	1.51	76
50.73	0.39	0.10	1.31	-	0.11	0.24	3.32	1.18	76
55.1	0.23	2.31	-	-	0.25	2.00	2.19	0.11	76
25.17	0.68	-	1.70	-	-	0.16	1.45	0.02	76
30.34	0.73	-	1.28	-	0.54	0.09	-	1.33	76
24.8	2.88	4.16	0.98	-	2.00	0.30	-	0.62	76

Fleisher *et al*[76] analysed lab distilled leaf oil from seven different mandarin cultivars using GC-MS and indicated linalool as major component in most of them. Very recently, Verzera *et al*[77] analysed Nova and Satsuma mandarin oil by HRGC/MS and reported β-pinene, sabinene, limonene, linalool and α-terpineol as major components. These investigations showed presence of β-pinene, sabinene, limonene, linalool, terpinene, p-cymene as major components and chemical composition varies with the varieties and with change of soil conditions.

Citrus jambhiri Lushington (Jamberi or Rough Lemon)

It is also known as 'Jambhiri' or Rough lemon and is said to be native of Northern India. The leaf oil of this plant is not produced commercially and this plant is rarely used for fruit production. The rough lemon leaf oil examined by Attaway *et al*[57] was found to contain α-pinene, β-pinene, sabinene, myrcene, δ-3-carene, limonene, β-ocimene, p-cymene, terpinolene, citronellal, decanal, linalool, menthone, terpinen-4-ol, β-caryophyllene, neral, geranial, neryl acetate and nerol. In 1969, Scora *et al*[78] compared the chemical composition of various leaf oils of a number of cultivars of rough lemon using GC. Lund *et al*[79] examined steam distilled rough lemon leaf oil of Florida origin using analytical and preparative GC, IR and MS techniques. The author also examined the methylene chloride extract of aqueous phase that was separated from the oil during steam distillation. Agarwal *et al*[80] studied the effect of seasonal variation on the composition of rough lemon leaf oil produced in India and found limonene as well as sabinene as major components. Recently, Nemec and Lund[81] compared the composition of leaf oils of rough lemon grown in phosphorus deficient soils, soils treated with a phytoalexin and phosphorus deficient soil treated with phosphorus (440 ppm). These studies showed that this oil contains limonene, sabinene, myrcene, linalool, citronellal and neral as major constituents (Table 7).



Citrus jambhiri

Table 7: Major constituents (%) of *Citrus jambhiri* (Jamberi or Rough lemon) oil

Sabinene	Limonene	γ-Terpinene	p-Cymene	β-Ocimene	Myrcene	Isopulegol	Linalool	Citronellal	Neral	Reference
7.4	32.4	7.4	3.3	7.3	1.2	4.4	1.3	0.8	0.91	79
0.4	1.3	-	-	-	-	0.23	4.0	0.14	2.7	79
24.1-34.4	26.7-41.0	-	-	-	1.1-2.4	-	6.9-17.2	5.0-9.1	1.2-3.6	80
-	13.05	3.17	-	-	16.66	-	34.07	14.54	8.14	78
-	13.07	4.06	-	-	15.61	-	25.59	14.63	8.43	78
-	17.29	4.16	-	-	16.57	-	10.84	17.02	13.11	78
-	20.17	4.36-	-	-	16.52	-	11.54	16.4	11.88	78
-	22.92	4.31	-	-	14.66	-	10.61	10.50	11.29	78
-	20.12	4.77	-	-	16.30	-	9.76	11.40	12.30	78
-	17.75	4.15	.		18.89	-	12.66	12.86	11.29	78
-	22.94	5.12	-		20.55	-	9.37	11.37	10.57	78
-	16.20	5.75	-	-	24.04	-	13.44	14.81	10.42	78
-	24.00	-	-	0.13	-	-	-	-	-	81
1.18	18.4	-	0.51	1.66	-	0.57	1.59	6.7	6.5	81
10.2	29.0	-	0.25	3.4	0.1	0.33	1.9	6.7	4.0	81

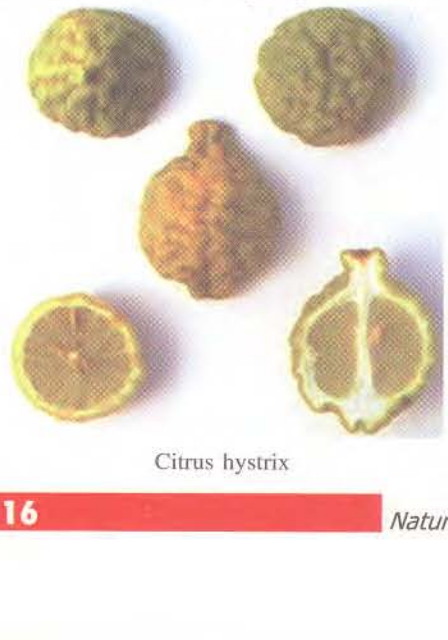
Citrus bergamia Risso & Poit. (Bergamot)

The bergamot leaf oil is only encountered on very rare occasions. Peyron[82] reported that main consituents of bergamot leaf oil were linalyl acetate, linalool, limonene, α-terpineol, β-pinene, γ-terpinene, geranyl acetate and neryl acetate. These constituents are arranged in decreasing order of their amount but no quantitative data was presented. Ortiz *et al*[16] examined the composition of leaf oil of bergamot grown in California and was found to contain linalool as main component. Cheng and Lee[40] examined Taiwanese sample of bergamot leaf oil using a number of pre-fractionation techniques prior to GC and found linalyl acetate as major component. Recently, Poiana *et al*[83] isolated ***C. bergamia*** (peel) oil by supercritical CO_2 extraction method and found bergaptene in higher percentage. These studies showed that ***C. bergamia*** leaf oil contains linalyl acetate and linalool as major components (Table 8).

Citrus hystrix DC. (Combava petitgrain)

In Comoro and Indian ocean, it is known as Combava petitgrain while in other parts of the world, it is known as Makrut lime or Swangi. Murakami *et al*[84] isolated three known coumarins from ***C. hystrix***. Lawrence *et al*[85] examined leaf oil of Thai origin by combination of column chromatography, preparative GC and IR spectroscopy and found citronellal (65.4%) as main component. Moreuil and Huet[86] reported that Combava petitgrain oil of Madagascan origin contained citronellal 58.92%. In 1990 Sato *et al*[87] examined this oil of Thai origin using combination of GC and GC-MS techniques; it was found to contains 81.49% citronellal. These studies revealed that ***C. hystrix*** leaf oil contains citronellal as main component (Table 8).


Citrus hystrix


Citrus hystrix

Citrus unshiu Marcovitch (Satsuma mandarin)

It is also known as Unshiu, Mikan or Satsuma mandarin and produces an essential oil from its leaves. The chemistry of its oil is quite meagre (Table 8). Ogihara *et al*[88] examined the composition of leaf extract of a tetraploid form and a diploid form of ***C. unshiu***; it was found to contain γ-terpinene as major components. In 1984, Kekelidze *et al*[89] compared the chemical composition of the leaf oil of ***C. unshiu*** graft on different root stocks and found same component (γ-terpinene) as major component but in greater extent. Kim *et al*[90] isolated three antiallergic polymethoxy flavones namely 3',4',5,6,7,8-hexamethoxy flavone, 5-hydroxy 3',4',6,7,8-pentamethoxy flavone and 3',4',5,7,8-pentamethoxy flavone from immature peels of ***C.unshiu***.

Citrus medica Linn. (Citron, Hindi-*Bara nimbu*)

Recently, we have investigated[91] the chemical composition of leaf and peel oil of this plant by using HPLC, GC and GC-MS techniques (Table 9). The result showed that leaf oil contained citronellal (63.3%), citronellol (15.1%) and limonene (8.0%) while peel oil contained limonene (32.0%), citronellal (27.5%) and citronellol (13.0%) as major components. Shiota[92] studied the volatile component in peel oil from fingered citron (***C. medica***)

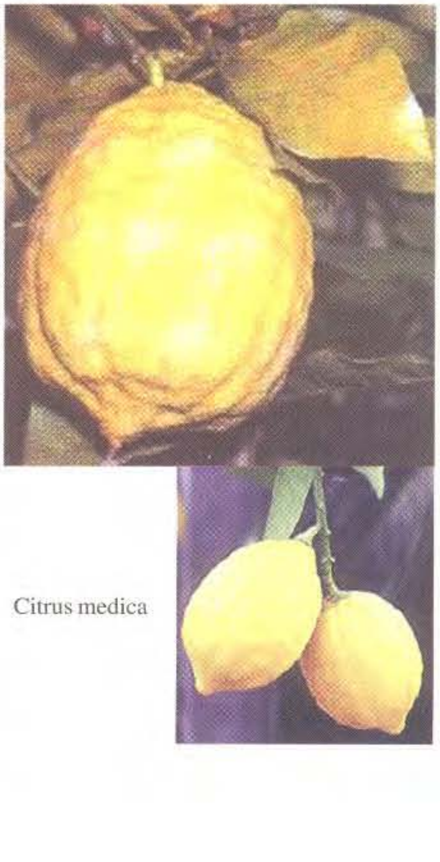

Citrus medica

Table 8: Major components of essential oils of *Citrus bergamia*, *C. hystrix* and *C. unshiu*

C. bergamia

α-Pinene	α-Terpineol	Limonene	Myrcene	Linalool	Terpinen-4-ol	Geranial	Nerol	Linalyl acetate	Reference
0.37	5.17	0.63	1.04	55.16	1.60	2.49	4.05	22.30	16
0.09	4.21	1.21	1.63	22.39	2.53	3.15	1.21	51.64	40

C. hystrix

Citronellal	Linalool	Sabinene	Terpinen-4-ol	Myrcene	Limonene	Citronellol	α-Pinene	Citronellyl acetate	Reference
65.4	2.9	4.9	-	0.6	0.6	76.4	0.2	5.1	85
58.92	4.67	4.81	0.85	1.44	0.48	-	0.37	-	86
81.49	-	1.57	0.20	0.43	0.03	8.22	0.06	0.90	87

C. unshiu

Limonene	p-Cymene	Elemene	β-Caryophyllene	Terpinene	Linalool	Reference
3.2	9.1	6.1	6.4	19.6	2.1	88
2.6	7.4	8.4	6.4	21.8	2.3	88
3.9	15.2	5.3	0.8	35.7	6.2	89
3.7	15.9	8.1	3.8	36.4	3.2	89

and found limonene (47.79%) as main component. Dung *et al*[93] reported more than 40 components in the oil of two different chemotypes, one contained limonene (48.4%) and p-cymene (33.7%) while the other contained limonene (55.5%) and γ-terpinene (22.5%). Govindachari *et al*[94] extracted three tetranortriterpenoids namely limonin, limonol and nomilinic acid from *C. medica* and tested their antifungal activity. Lota *et al*[95] examined five peel oils and six leaf oils of different varieties of citron (*C. medica* and *C. limonimedica*) by capillary GC, GC-MS and C^{13} NMR. They found three chemotypes: limonene, limonene/ γ-terpinene and limonene/ geranial/ neral for peel oil while leaf oil exhibited limonene /geranial/ neral composition.

Citrus junos Sieb. ex Tanaka (Yuzu)

It is a lemon-like sour *Citrus* fruit and has been cultivated mostly in Japan, China and Korea. It is used in cooking, beverages, and as seasoning for salads, sauces, sea food, pickled vegetables and vinegar. Chemistry of essential oil obtained from *C. junos* is little known (Table 9). Song *et al*[96] examined cold pressed peel oils of Japanese (Kochi, Takushima, Ehime, Oita, Wakayama) and Korean (Cheju and Chindo) origin, by GC and GC-MS and found significant differences in concentration of limonene, β-phellandrene linalool, β-farnesene,

Citrus junos

bicyclogermacrene and nerolidol which are the major constituents. They also analysed[97] quantitative and characteristic flavour of yuzu peel oil from Japan using GC and GC-MS and found limonene, γ-terpinene, α-pinene and β-phellandrene, as main components. These studies showed high content of limonene in peel oil of this species.

C. ichangenesis Swingle (Ichang lemon)

It belongs to subgenus *Papeda*. Swingle[98] classified yuzu and its relatives as hybrid of *Papeda* subgroup. Sawamura[2] investigated Ichang lemon which was rich in limonene (68.8%), γ-terpinene (16%), α-pinene (3.25%), α-terpinene (2.35%) and β-phellandrene (3.7%).

C. tachibana (Makino) Tanaka (Tachibana)

It is another species of ***Citrus*** whose chemical composition is mostly similar to other varieties and was found to be rich[1] in limonene (80.12%), γ-terpinene (8.07%), linalool (2.03%), myrcene (2.66%), β-pinene (1.87%) and α-pinene (1.17%).

C. indica Tanaka (Shiikuwasha)

The oil of this species is reported[2] to contain limonene (51.84%), γ-terpinene (28.3%), linalool (2.18%), β-pinene (2.08%), α-pinene (2.9%), p-cymene (2.28%) and β-caryophyllene (2.02%). It is interesting that this oil contains comparatively high amount of γ-terpinene among most of ***Citrus*** species.

Table 9: Major constituents of *C. medica* and *C. junos* oils

C. medica

Citronellal	Citronellol	Limonene	p-Cymene	γ-Terpinene	Citronellyl acetate	Geranial	Reference
63.3	15.1	8.0	-	-	5.2	-	91
27.5	13.0	32.0	6.5	2.0	0.4	2.3	91
-	-	47.79	-	-	-	-	92
-	-	48.4	33.7	-	-	-	93
-	-	55.5	22.5	-	-	-	93

C. junos

α-Pinene	Myrcene	α-Phellandrene	Limonene	β-Phellandrene	Linalool	Bicyclogermacrene	Reference
1.7	1.1	tr	77.5	1.6	2.7	1.5	96 (Kochi)
2.2	2.7	tr	72.8	31	2.5	0.9	96 (Tokushima)
2.1	2.7	tr	75.2	2.8	2.1	0.9	96 (Ehime)
2.1	2.7	tr	73.4	3.1	1.9	1.1	96 (Iota)
2.1	2.6	tr	72.9	2.8	2.7	1.2	96 (Wakayam)
2.0	1.1	1.3	75.3	2.1	3.3	1.0	96 (Cheju)
1.9	1.1	1.3	72.2	2.2	4.7	1.3	96 (Chindo)
2.0	0.4	0.9	77.44	2.16	1.6	0.7	97

Rare species

Some other species are also obtained by hybridization between two well known species. The chemistry of their volatile oils is quite meagre. Some such species whose chemical compositon is little known, are: ***C. elementina*** **Monreal,** ***C. tamurana*** **Hort. et Tanaka,** ***C. flaviculpus*** **Hort. ex Tanaka,** ***C. junos*** **Sieb. ex Tanaka,** ***C. sudachi*** **Hort. ex Tanaka,** ***C. inflata*** **Hort. ex Tanaka,** etc. The essential oil of Uruguayan ***C. elementina*** from fruit of Nules and Comune cultivars about 69 components were identified[99] among which α-pinene, sabinene, limonene, linalool and α-terpineol were the major components. Ruberto *et al*[100] examined the chemical composition of a new ***Citrus*** hybrid obtained by cross breeding the diploid elementine (***C. elementina***) and a tetraploid lemon (***C. limon***). The essential oil of such hybrid plant was found to be similar to that of lemon and contained limonene, γ-terpinene, β-pinene as major components. Choi and Sawamura[101] examined volatile oil of ***C. tamurana*** (Hyuganatsu) obtained by cold pressing method using capillary GC and GC-MS and reported 126 volatile constituents, among which limonene (80.35-83.39%), γ-terpinene (7.71-9.03%), myrcene (2.11-2.28%), linalool (1.37-2.0%), α-pinene (1.17-1.43%) and a sesquiterpene (E)–β-farnesene (0.6-0.9%) as major components. Choi and Sawamura[102] examined cold pressed ripe and over ripe Ki-mikan (***C. flaviculpus***) peel oil samples by GC and GC-MS , and compared with the Hyuganatsu (***C. tamurana***), and found limonene (ripe fruit, 82.44%, over ripe fruit, 73.1%) as the most abundant components in Ki-mikan oil, this being followed by γ-terpinene (8.83 and 13.74%), trans-β-farnesene (1.76 and 3.12%) and myrcene (1.54 and 1.13%) and limonene was 84.78% in Hyuganatsu oil. Njoroge *et al*[42] examined ***C. sudachi*** (sudachi) peel oil using GC and GC-MS technique and identified 83 components out of which limonene (69.0%) was the major component. They also investigated the chemical composition of ***C. inflata*** (mochiyuzu) peel oil by same technique and found limonene (77.2%) as major component. Recently, Sawamura *et al*[103] examined ***C.grandis*** **(Linn.) Osbeck** (tosa- buntan) cold pressed peel oil from Japan by GC and GC-MS and reported limonene (87.07%), myrcene (1.81%), α-pinene (1.13%) and γ-terpinene (6.64%) as major components.

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