### Flavour characteristics of Piper betle L.

V R BALASUBRAHMANYAM AND A K S RAWAT National Botanical Research Institute, Lucknow, U.P., India

### ABSTRACT

The betel leaves are aromatic with varied taste ranging from sweet to pungent due to the presence of essential oil. Compounds that contribute to the pungency, flavour and stimulating properties of the leaf are of interest to the flavour industry. The results of investigations carried out so far on the chemical analysis of essential oil of betel leaves have been reviewed. The results of the gas chromatography and mass spectrometry (GCMS) of the oil of all the five recognized varieties of Piper betle are discussed. The yield and composition of essential oil of betel leaves have been influenced by age and position of the leaf on the stem, season of harvest, potassium nutrition etc. Promoting the use of this important essential oil in flavour industry and confectioneries so as to utilise the surplus leaves from the assembling and wholesale markets which will otherwise go waste is suggested.

Key words: betelvine, Piper betle, quality, flavour.

Betelvine (*Piper betle* L.) is cultivated for its leaf which is used as a masticatory. The basic 'Pan" preparation consists of betel leaf spread over with hydrated lime and catechu (*Acacia catechu* L.) and few scrappings or pieces of arecanut (*Areca catechu* L.) rolled in. 'Pan' chewing is an ancient habit, prevalent amongst all classes of people in South-East Asian countries. As a masticatory, the betel leaf is credited with being aromatic, digestive, stimulant and carminative. Its initial effect when chewed, lies in the excitation of salivary glands and production of a mild degree of stimulation, resulting in a sensation of warmth and well-being besides, imparting a pleasant odour to the oral cavity.

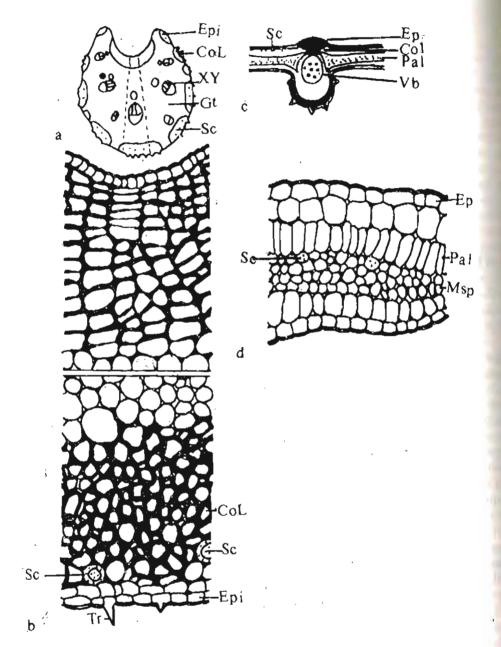
# Flavour characteristics of Piper belle

Betelvine is an important commercial crop of India, Bangla Desh, Sri Lanka and to a small extent of Malaysia, Myanmar. Singapore, Thailand, Philippines, Vietnam. Indonesia, Papua New Guinea, etc. In India, it is grown over an area of 40,000 ha, providing livelihood to hundreds of families engaged in its cultivation and trade. Next to India, Bangla Desh has a nourishing betel leaf industry with 12,700 ha under betelvine cultivation and an ennual production of 60,000 tones of betel leaves. According to the 'Susruta Samhita' (600 A.D.), a treatise on the indigenous Ayurvedic system of medicine. hetel leaves are beneficial to throat, help in digestion, act as deodorant. The volatile oil of betel leaves has been found to possess antiseptic properties. It is also good for the respiratory system and is used in treatment of bronchitis, cough, cold, and chills (Chopra et al. 1954, 1958).

Piper betle belongs to the family Pineraceae. The family is of considerable economic importance because of the presence of essential oil in fruits and leaves of many species, the most important one being Piper nigrum L. (black pepper). Piper betle is next to black pepper in its economic importance. The betel leaves are aromatic with varied taste, ranging from sweet to pungent due to the presonce of essential oil. Secretory cells containing essential oil occur throughout the plant. They always occur singly, larger than the surrounding parenchymatous cells. The essential oil is present in the form of globules in the mesophyll tissue of the leaf lamina. The oil also occurs in the superficial tissues of petiole (Fig. 1). The presence of oil help to reduce transpiration, mollify the heat and inhibit fungal infection (Chibber 1912).

A knowledge about the chemical composition of betle leaf oil which gives taste to the leaf is of interest mainly because of the medicinal uses as well as its use in flavour industry. Of obvious importance are the compounds that contribute to the pungency, flavour and stimulating properties of the leaf. Besides, the betel leaf contains the other compounds in all plant tissues.

The aroma of betel leaf intrigued researchers for many years. Consequently, the composition of betel leaf oil has been the subject of many studies, the first being that of Mann and Sahasrabuddhe (1913) who identified phenols and terpene-like constituents in the essential oil of betel leaf and among the phenols, eugenol was the chief one. Ueda and Sasaki (1951) studied the chemistry of betel leaves collected from different South Asian countries and identified phenolseugenol, cadinene, chavicol, sesquiterpenes and allylpyrocatechol as the chief constituent in betel leaf oil, besides cineole, cadinene, caryophyllene, chavicol, methyl eugenol and allylpyrocatechol which are the other constituents identified. Sethi and Agarwal (1956) have isolated hydroxy chavicol (4-allylpyrocatechol) from betel leaf oil. Deshpandey et al (1970) observed sitosterol. B-sitosterol, stearic acid and chavicol in the 'Banarasi' variety betel leaves. Atal et al (1975) reviewed the work of Mafuku and Kato (1935), Dutt (1956), Ally (1960), Nigam and Purohit, (1962) amongst others and reported chavicol, allyl-pyrocatechol, chavibetol, methyl chavicol, eugenol, estrogol, methyl-eugenol, 1,8-cineole, B-caryophyllene and cadinene as the major components in the oil of betel leaves. The oil of cv. Bangla is found to contain sitosterol and stigmasterol (Ganguly and Chaudhury 1975). Das and Chattopadhyay (1984) observed that phenols of 17 West Bengal varieties studied, ranged between 4.10 and 10.20 per cent on dry



- Fig. 1: Transverse section of the petiole and leaf lamina (diagrammatic) a) T.S. of petiole
  - b) Portion of the petiole showing details
  - c) A portion of transection passing through lamina showing details

1. Col. - collenchyma 2. epi - epidermis 3. Gt. ground tissue 4. Msp - spongy parencyma 5. Sc - secretory cell 6. Pal - palisade layer 7. Vb - vacsular bundles

### Flavour characteristics of Piper betle

weight basis. Li Yun Lin (1989) has analysed the volatile constituents of betel quid and has observed eugenol and its isomers as major ones in betel leaves.

An accurate knowledge of differences in the composition of essential oil of betelvine varieties being of fundamental importance, extensive investigations were carried out at the National Botanical Research Institute, Lucknow using modern techniques like gas chromatography and mass spectrometry (GCMS) (Rawat et al 1989, Balasubrahmanyam and Rawat 1990). They observed that the chemical constituents and their proportion in the essential oil of different cultivars provide a consistent and reliable tool for the identification of varieties and concluded that on the basis of the chemical composition, the existing betelvine types in the country could be grouped under one or the other of the following five cultivar groups viz, Bangla, Desawari, Kapoori, Meetha and Sanchi (Table 1).

Bangla is one of the most vigorous high yielding cultivars, tolerant to some of the key diseases and is cultivated on a commercial scale in north and east India. The oil of cv. Bangla is heavier than water, specific gravity being 1.0001 to 1.1090, possess clove-like spicy odour and a sharp pungent taste, while the yield varied from 0.15 to 0.2% on fresh weight basis amongst the three types studied (Sharma et al 1983). As part of the gas chromatographic study (GLC) on the basis of retention times on two columns and peak areas, they tentatively identified pcymene. a-terpineol, terpenyl acetate, eugenol, methyl eugenol and caryophyllene in three types of Bangla cultivars. Rawat et al (1989) observed the following constituents in their analysis usingGCMS: eugenol, 63.56%, isoeugenol 5.2%, chavicol 1.07%, safrole 2.33% and eugenol acetate 18.68%.

|                          | Sanchi | Kapoori | Desawari | Bangla | Meetha |
|--------------------------|--------|---------|----------|--------|--------|
| Per cent oil (Fresh wt.) | 0.19   | 0.10    | 0.12     | 0.16   | 0.85   |
| (A) Monoterpenes         |        |         |          |        |        |
| α-Thujene                | -      | 1.90    | -        | -      | -      |
| Camphene                 | -      | 0.35    | -        | -      | 0.50   |
| Sabinene                 | 3.68   | 6.13    | -        | -      | -      |
| β-Myrcene                | 0.53   | 2.16    | -        | -      | -      |
| β-Ocimene                | -      | 0.40    | 0.36     | -      | -      |
| Bornylene                | 1.18   | -       | -        | -      | -      |
| β-Pinene                 | 3.38   | -       | -        | -      | -      |
| Trans β-ocimene          | -      | 0.70    | -        | -      | -      |
| Y-Terpinene              | 0.46   | 1.60    | 0.51     | -      | -      |
| Terpinolene              | -      | 0.50    | -        | •      | -      |
| Allo ocimene             | -      | 0.63    | -        | -      | -      |
| a-Terpinene              | 0.23   |         | -        |        | 0.94   |
| β-Phellandrene           | 1.28   | -       | 0.39     | -      | -      |
| Limonene                 |        | -       | 0.37     | -      |        |
| p-Cymene                 | 0.31   | -       | 0.35     | -      | -      |

 Table 1. Comparative chemical composition of the oils of betelvine cultivars

| 2,6,6 Trimethyl<br>1-Methylene cyclo<br>Hex 2-eno<br>α-Pinene | -<br>0.41 | -     | 0.42<br>0.42 |       | 0.37      |
|---|-----------|-------|--------------|-------|-----------|
| (B) Sesqui terpenes   |           |       |              |       |           |
| γ-Cadinene  | -         | -     | 1.10         | •     | 9.44      |
| Δ-Cadinene  | -         | 1.18  | -            | -     | -<br>2.90 |
| α-Cadinene  | -         | -     | -            | -     |           |
| β-Salinene<br>β-Elemene                                       | 6.36      | -     | 2.57<br>2.60 | -     | -         |
| •   | 1,18      | -     | 2.80<br>0.86 |       | -<br>0.56 |
| γ-Elemene<br>Cia companya hullon a                            | 1.78      | 0.89  | 3,30         | -     | 10.64     |
| Cis-caryophyllene   | -<br>7.78 | -     |              | -     | 0.40      |
| Trans-caryophyllene<br>Aroma dendrene                         | 1.10      | -     | -            |       | 1.66      |
| a-Cubébene  | 4.43      | -     | 0.46         | •     | 4.71      |
| β-Cubebene  | 4.40      | -     | 0.40         | -     | 1.12      |
| p-Cubebene  | -         | -     | 0.00         | •     | 1.12      |
| Oxygenated compounds  |           |       |              |       |           |
| (C) Alcohols  |           |       |              |       |           |
| Linalool  | 1.2       | 1.46  | 0.23         | -     | 0.32      |
| a-Terpineol   | 0.69      | 2.30  | 0.32         | •     | -         |
| Terpinen-l-ol   |           | 1.45  | -            | -     | -         |
| a-Costol  | -         | 0.36  | -            | _     | -         |
| δ-Cadinol   | -         | 1.19  | -            | ·     | -         |
| 3,7,11,15 Tetra   | -         | -     | -            | -     | -         |
| Methyl-2-hexadecan-l-ol                                       |           | 0.76  | -            | -     | -         |
| Geraniol  | -         | 2.46  | -            | -     | -         |
|   |           |       |              |       |           |
| (D) Aldehydes   |           |       |              |       |           |
| Decanal (Capric aldehyde)                                     |           | 2.87  | -            | -     | -         |
| Decanal (Laural aldehyde)                                     |           | 4.23  | <u>.</u> .   | -     | -         |
| Stearaldehyde   | 2.69      | -     | -            | -     | -         |
| (E) Acids   |           |       |              |       |           |
| Hexadecanoic acid   |           | 0.41  |              |       | _         |
|   |           | 0.41  | -,           |       |           |
| (F) Oxides  |           |       |              |       |           |
| 1,8 Cineole   | -         | -     | -            | -     | 2.33      |
| Caryophyllene oxide   | -         | 0.66  | 1.63         | •     | -         |
|   |           |       |              |       |           |
| (G) Phenols   | 12.00     | 22.00 | 20.47        | 60 56 | 18.92     |
| Eugenol   | 13.90     | 33.22 | 20.47        | 63.56 | 10.92     |
| Isoeugenol  | -         | 10.59 | -            | 5.2Q. | -         |
| Chavicol/Chavi betol  |           | -     | -            | 1.07  | -         |
| (H) Phenolic ethers   | 0.00      |       | 1 00         |       |           |
| Methyl eugenol  | 0.90      | -     | 1.90         | -     |           |
| Methyl chavicol   | -         | •     | 5.81         | -     | 7.62      |

| Flacom   |            |           |       |            |            |
|--|------------|-----------|-------|------------|------------|
| Anethole<br>Safrole                              | -<br>22.75 | 6.45      | 45.34 | -<br>2.33  | 19.31<br>- |
| (I) Esters<br>Eugenol acetate<br>Methyl benzoate | -          | -<br>2,23 | -     | 18.68<br>- | -          |

Terpenic constituents and hydrocarbons were totally absent in Bangla. In Vietnam, a cultivar resembling Bangla in its composition of oil, contained eugenol 56.5% and eugenol acetate 18.46% (Dung, unpublished).

manur characteristics of Piper belle

Cv. Desawari is cultivated in parts of Uttar Pradesh and Madhya Pradesh. Its leaves yield 0.12-0.14% yellow coloured oil with a specific gravity of 1.0002, pungent followed by sweet taste. Sharma et al (1987) used thin layer chromatography to confirm the constituents identified by GLC in the oil of two types of Desawari 'Desi' and 'Mahoba'. They identified a-pinene, 1,8-cineole, psymene, methyl chavicol,  $\alpha$ -terpeneol, terpenylacetate, eugenol, methyleugenol, anothole and caryophyllene tentatively on the basis of retention time and Rf values. Pursuant to this study Rawat and coworkers (1989) identified the main constituents as safrole 45.34%, eugenol 20.47%, methyl chavicol 5.81% and methyl eugenol 1.90%. The hydrocarbons were low in Desawari - 14.24%. The ratio of safrole to eugenol was 10:3 in Desawari

Kapoori, the most important commercial cultivar in peninsular India, yield 0.1% oil-yellow with greenish tinge and has an aromatic odour, lighter, specific gravity being 0.9650. Sharma *et al* (1982) used a combination of thin layer and gas chromatography to isolate and confirm the presence of  $\alpha$ -pinene,  $\beta$ -pinene, 1,8-cineole, camphene, p-cymene,  $\alpha$ -terpineol, methyl chavicol, terpenyl acetate, eugenol and methyl eugenol in Kapoori leaves of *Piper betle* while the GCMS studies of Rawat *et al* (1989) recorded the presence of 26 constituents in Kapoori oil, the maximum number of constituents in any of the five cultivars analysed. The oxygen containing components constituted 70.64% of which eugenol 33.22% and isoeugenol 10.59% are the major ones (Table 2).

35

Cv. Meetha, which is commercially cultivated in Midnapore and Howrah districts of West Bengal only, but marketed throughout the country because of its excellent flavour, yields the maximum oil 0.85% although per hectare yield of Meetha leaves may be the minimum amongstall betelvine cultivars. The oil is vellowish brown with fennel-like odour and sweet taste, its specific gravity being 0.9991. Rawatet al (1987) used retention time data to characterize the presence of α-pinene, β-pinene, p-cymene, α-terpineol, methyl chavicol, terpenyl acetate, eugenol, methyl eugenol, anethole and caryophyllene in Meetha. Two years later Rawat et al (1989) used a combination of GC and MS to examine the terpene hydrocarbons and oxygenated compounds present in the oil. In Meetha the oxygenated compounds were comparatively low 48.50% of which anethole (19.31%) and eugenol (18.92%) are the major ones. Ciscaryophyllene has been observed to be maximum (10.64%) in this cultivar; the hydrocarbons constituted 33,24% of the oil.

| Cultivars | Monoterpene<br>hydrocarbons<br>% | Sesquiterpene<br>hydrocarbons<br>% | Oxygenated<br>compounds<br>% |
|-----------|----------------------------------|------------------------------------|------------------------------|
| Bangala   | Nil                              | Nil                                | 90.84                        |
| Desawari  | 2.82                             | 11.42                              | 75.70                        |
| Kapoori   | 14.37                            | 2.07                               | 70.64                        |
| Meetha .  | 1.81                             | 31.34                              | 48.50                        |
| Sanchi    | 11,46                            | 21.53                              | 44.03                        |

Table 2. Hydrocarbons and oxygenated compounds in the oil of betelvine cultivars

Cv. Sanchi, which is commonly cultivated in almost all betelvine growing states, yield 0.19% oil. The oil is dark brown with specific gravity 1.0001 and sharp pungent taste. The hydrocarbons in Sanchi were 32.99% while the oxygenated compounds were 44.03%, safrole 22.75% was conspicuous among the oxygenated compounds. Cv. Sanchi has been marked by the presence of a unique compound stearaldehyde (2.69%) which is absent in other cultivars.

From a chemical compositional stand point, the studies have shown that eugenol is the major common molecular species amongst the constituents so far identified in the five cultivars and eugenol is also quantitatively the most important component. The pleasant sweet taste and fennel-like odour of Meetha is due to the presence of anethole in its essential oil. Desawari and Kapoori leaves are comparatively less pungent due to low concentration of eugenol. Some of • these compounds identified were not previously known to be present in the essential oil of betel leaves.

# Factors influencing the yield of essential oil in leaves

The older the leaves, the higher the yield of essential oil. Leaves which are well matured from the lower portion of the vine

stem are richer in oil than the tender ones from the apical parts. Matured leaves harvested during winter recorded maximum yield than the ones harvested during rainy season or summer. Of all the nutrient elements considered essential for the vine. potassium has been shown to exert a profound influence on increasing the essential oil content of leaves (Misra et al unpublished). They observed change in the composition of the oil due to differential application of potassium, however, the variation was not significant. The oil content in the leaves increases with the storage and bleaching due to loss of moisture in the stored leaves. Interesting possibilities are opened up by the report that application of triacontanol increase the vield of oil in betel leaves (Unpublished).

The oil from betel leaves is known to have antifungal properties (Baby et al 1989). Kapoori oil exhibits minimum inhibitory concentration (MIC) of 250 ppm against the pathogens Collectrichum capsici, Sclerotium rolfsii and Alternaria alternata (Tripathi et al 1985). Five propenyl phenols isolated from the chloroform extract of leaves of Piper betle showed significant nematocidal activity, These compounds are chavicol, chavibetol, allylpyrocatechol, chavibetol acetate and allylpyrocatechol diacetate (Evans et al. 1984).

# Flavour characteristics of Piper belle

## Conclusion

Much progress has been made towards the identification of components of betel leaf essential oil. Out of more than 50 compounds identified so far, eugenol, anethole, terpenyl acetate and isoeugenol are found to be of importance to flavour industry. The process of essential oil synthesis appears to be influenced by a host of environmental factors which influence through their effects on the growth and developmental process of the vine. A close relationship between potassium nutrition and essential oil content has been indicated; potassium is obviously of importance in betelvine nutrition.

Almost the entire production of betel loaves is consumed fresh as a masticatory. In parts of Sikkim, liquor is made with betel leaf flavour. In recent years, the oil is increasingly used as an adjunct in confectionery preparations like chewing gum, lozenges, in blending perfumes, in tooth-paste, mouth-wash, etc. Large mantities of leaves 10-25% depending upon the variety, distance transported, packaging, etc. are damaged while repacking at Pandaribhas and in wholesale markets (Anon 1983). A cheap method of distillation for the recovery of oil from the surplus leaves from the wholesale and assembling markets will go a long way in exploiting these leaves otherwise going waste, for remunerative returns. Steps to promote the use of oil in flavour industry will also ensure better economic returns to the traditional cultivators who are sticking to the profession betelvine cultivation in spite of heavy odds.

#### References

\* Ally M M 1960 Preliminary observations on the pharmacology of betel leaf. Proc. 4th Pan Indian Ocean Sci. Congr. Karachi, sect. G. 31 p.

- Anon 1983 Marketing of betel leaves in India. Directorate of marketing and Inspection, Ministry of Rural Development, Faridabad.
- Atal C K, Dhar K L & Jagdev Singh 1975 The chemistry of Indian Piper species. Lloydia 38(3): 256-264.
- Baby P, Rao G S J G & Prasad Y R 1989. Leafoil of *Piper betle* L. The *in vitro* antimicrobial studies. Proc. XI International conference of essential oils, fragrances and flavours 12-16 Nov. 1989, New Delhi Vol. 1 p.3.
- Balasubrahmanyain V R & Rawat A K S 1990 Studies on morphology and chemistry of *Piper betle* L. J. of Plantation crops 18(2): 78-87.
- Chibber H M 1912 The morphology and histology of *Piper betle* L. Linn. J. of Botany, 41: 357-382.
- Chopra I C, Jamal K S & Khajuria B N 1954 Pharmacological action of some common essential oil bearing plants used in indigenous medicine including *P. betle.* Indian J.Med. Res. 42: 385.
- Chopra R N, Chopra I C, Handa K L & Kapur L D 1958 Chopra's Indigenous Drugs of India.
- Das A K & Chattopadhyay N C 1984 Studies on the standardization of betelvine cultivars of Midnapore District. Proc Natl. Workshop on Betelvine. Eds. S K Roy et al. Calcutta Univ. Calcutta. The Agril. society of India, Calcutta. 23-32.
- Deshpandey SN, Upadhyay R & Singh R P 1970 Chemical study of *Piper betle* L. leaves. Curr. Sci. 30: 372.
- Dutt S K 1956 The Indian Pan and its essential oils. Indian Soap J. 21: 275.
- Evans P H, Bowers W S & Funk E I 1984 Identification of fungicidal and nematocidal components in the

#### Balasubrahmanyam and Rawat

leaves of Piper betle L. (Piperacease) J. Agric. Food Chem. 32: 1254-56.

- Ganguly P & Chaudhury M 1975 Phytochemical studies on Bangla variety of Betel leaf (*Piper betle* L.). Indian Agric. 19: 199-200.
- Guenther E 1960 The Essential Oils. Vol. 1 and 5, 160-61. D Van Nostrand Co. Inc. Princeton New Jersey.
- Li Yun Lin 1989 The volatile constituents of *Piper betle* L. Proc. 11th International conference of essential oils, fragrances and flavours 12-16, Nov. 1989. New Delhi, Vol. 1 p. 40.
- Mafuku K & Kato R 1935 Volatile oil of Piperaceae(*Piper betle* L.)J. Chem. soc. Japan 56: 1150.
- \*Mann H, Sahasrabudhe D S & Patwardhan 1913 J. Soc. Chem. Ind. 32: 922.
- \*Nigam SS & R M Purohit 1962 Chemicalexamination of the essential oil of the leaves of *Piper belle*. Reichsfoffe Aromen 12: 185.
- Rawat A K S, Sharma M L & Balasubrahmanyam V R 1987 Essential oil of betel leaf (*Piper betle* L.) cv. Meetha. Indian Perfumer 31(2): 146-149.
- Rawat A K S, Usha Shome & Balasubrahmanyam V R 1987 Analysis of the volatile constituents of *Piper betle* L. cultivars. A chemo-systematic approach. Poc. XIV International Botanical congress. 24 July-1 Aug. 1987 pp. 104-108.
- Rawat A K S, Banerjee R & Balasubrahmanyam V R 1989 Chemical polymorphism of essential oil of *Piper betle* L. grown in India. Feddes Repertorium 100, (7-8): 331-334.
- Rawat A K S, Tripathi R D, Khan A J & Balasubrahmanyam V R 1989 Essential oil components as markers

for identification of *Piper belle* L. cultivars. Biochemical Systemat. ics and Ecology 17(1): 35-38.

- Sethi S C & Agarwal J S 1956 Stabiliza. tion of edible fats by spices II. A new anti-oxidant from betel leaf. J. of Scientific Ind. Res. 15B: 34-36.
- Sharma M L, Rawat A K S, Balasubra. hmanyam V R & Singh A 1982 Essential oil of Betel leaf ( *Piper* betle L.) var. Kapoori. Indian Per. fumer 26(2-4): 134-137.
- Sharma M L, Rawat A K S, Balasubrahmanyam V R & Singh A 1983 Studies on the essential oil of Betel leaf (*Piper belle* L.) Part II. Vars. Desi Bangla, Ramtek Bangla and Calcutta Bangla. Indian Perfumer 27(2): 91-93.
- Sharma M L, Rawat A K S, Balasubrahmanyam V R & Singh A 1984 Essential oil of Betel leaf ( Piper betle L.) cv. Kapoori. Proc, Gr. Discussion on Improvement of Betelvine cultivation Feb. 1981, 161-164, National Botanical Research Institute, Lucknow.
- Sharma ML, Rawat AKS & Balasubrahmanyam V R 1987 Studies on the essential oil of Betel leaf (*Piper betle* L.) Part III Vars. Desi Desawari and Mahoba Desawari. Indian Perfumer 31(4): 319-324.
- Tripathi R D, Rawat A K S, Johri J K, Chaurasia R S, Nainon M O & Balasubrahmanyam V R 1985 Tolerant factors of *Piper betle* L. cv. Kapoori to some fungal pathogens. Indian J. Plant Pathol. 3(1): 128-133.
- Ueda E & Sasaki T 1951 Chemical studies of Formosan Plants I Chemical constituents of the leaves of *Piper betle* L. Pharm. Soc. Japan, 71(6): 559-560.
- \* Original not seen.