Nutmeg with yellow arils-potential tree spice with high essential oil content

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Abstract

The mace derived from the arils of *Myristica fragrans*, is red in colour, whereas the present study reports the evaluation of *Myristica fragrans* with yellow arils. Morphological features of fruits of *M. fragrans* with yellow arils and that of common red aril variant from cultivated locations in Kerala, South India, were similar. The average mace yield was higher for the yellow arils (2.5 kg plant⁻¹) than the red arils (2.0 kg plant⁻¹). The volatile chemical profiling revealed that the essential oil yield of the yellow mace (19.3% v/w) was more than twofold higher than standard red mace (9.2% v/w). GC-MS analysis showed that the distribution of monoterpenoids, sesquiterpenoids, and phenylpropanoids were similar in both the varieties, except for the percentage distribution. The major constituents of mace of both the red and yellow types were α -pinene, and sabinene followed by elemicin and safrole. Though an exotic species, *M. fragrans* has been naturalized in south India, and several high-yielding varieties have been reported from the region. The present study highlights nutmeg with yellow arils as a potential spice crop with high oil yield.

Keywords: elemicin, essential oil, sabinene, yellow mace, α -pinene

The spices, nutmeg and mace are derived from the seeds and arils of *Myristica fragrans* Houtt. (Myristicaceae), a small tree cultivated throughout the tropical regions (Pursglove *et al.* 1981; Choo *et al.* 1999; Asgarpanah & Kazemivash 2012). *M. fragrans* is native to the Eastern Moluccas Island of Indonesia, and the Arabs introduced this spice to Europe during the 11th century as a flavouring agent. Nutmeg was one of the major driving factors for the European expeditions to Asia, and the Portuguese reached the Moluccas Island in the year 1512. After the successful invasion by the Dutch and the French, the British occupied the Moluccas Island during the 1800s, and the species was introduced to India from Moluccas Islands by the British in the early 1800s. *M. fragrans* is now widely grown throughout the tropical regions, especially in Guatemala, Indonesia, Taiwan, and Malaysia. India produces around 15,000 MT of nutmeg and mace annually, and around 3000 tonnes were exported in 2019-20. In India, Kerala dominates in the production of nutmeg and mace, with around 14,340 tonnes cultivated in 22,510 ha (Spices Board 2020).

Nutmeg and mace are also popular as traditional medicinal ingredients and have been used as a folklore medicine for treating diarrhea, mouth sores, and insomnia (Somani & Singhai 2008, Chauhan et al. 2014). Nutmeg is also an essential ingredient in aphrodisiac formulations in Ayurveda (Chauhan et al. 2014). Various biological activities have been reported for nutmeg and mace, such as antioxidant, antiinflammation, anti-cytotoxicity, antibacterial, and antitumor properties (Jukić et al. 2006, Akinboro et al. 2014). The spices nutmeg and mace, have a characteristic pleasant fragrance and a slightly warm taste and are widely used as food flavours, medical formulations, and perfumes (Asgarpanah & Kazemivash 2012). Chromatography-Mass Gas Spectroscopy (GC-MS) studies on aril essential oils showed considerable variation in the volatile constituents among different accessions across different parts of the world (Ogunwande et al. 2003; Maya et al. 2004; Singh et al. 2005; Subarnas et al. 2010, Zhao et al. 2019). Generally, the major constituents were sabinene, terpinen-4ol, safrole, α -pinene, myristicin, and elemicin (Singh et al. 2005). Kerala state is one of the major producers of nutmeg and mace in India (Kumar et al. 2010). Recently M. fragrans with yellow arils instead of commonly found red (Fig. 1) was reported as a premium quality product among the farmers in Kerala, and the cultivation of the yellow variant is restricted to specific locations in Central Kerala (Fig. 2). Though colour variations within the red hue were reported for the arils, and there is no report of the yellow coloured variants from

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the region (Kumar *et al.* 2010). The vegetative characteristics like size, shape, and texture of the leaves and fruits, branching pattern, and bark texture of the yellow mace nutmeg variant were found to be similar to those of common red mace variants. The average crop yield of common red mace tree-¹ annually, while the average crop yield of the yellow mace variants is around 2.5 kg dried mace tree⁻¹ year⁻¹.

The present study reports the volatile chemical composition of M. fragrans with yellow arils and its comparison with the red aril variant, both collected from cultivated sources from Kerala, south India. Mature fruits of M. fragrans, three accessions of yellow and red arils, were collected from different localities across Kerala State, India. The plants were on an average 20 years of age, and a detailed morphological examination of the plant in its habitat and that of specimens collected were carried out. Fresh and dried mace samples were used for volatile chemical studies. Fresh and dried arils (100 g each) of the collected samples were hydrodistilled using a Clevenger-type apparatus for 4 h. The essential oils were dried over anhydrous sodium sulfate and kept at 4°C till further analysis. GC-MS analysis of the essential oil from fresh arils was performed by split mode (1:50) injection of 1 μ l of the diluted oil in diethyl ether, on a Shimadzu TQ triple quadrupole gas chromatograph fitted with cross bond 1,4-bis (dimethylsiloxy) phenylene dimethyl polysiloxane Rxi-5 Sil MS capillary column (30 m x 0.32 mm, film thickness 0.25 mm) coupled with 8030 series mass selective detector. The GC-MS operation conditions were: injector temperature, 220°C; transfer line temperature, 240°C; oven temperature programme, 60-250°C (3°C min⁻¹) and carrier gas He at 1.4 ml min⁻¹. The mass spectra conditions were Electron Impact (EI⁺) mode, 70 eV with a mass range of 40 to 450 m/z, and ion source temperature, 240°C. The volatile constituents were identified by MS library search (Wiley 275. L and NIST 14), relative retention indices



Fig. 1. Myristica fragrans fruits and seeds with arils. A- red aril variant; B- yellow aril variant.

(RRI) calculated using standard series of C_{30} hydrocarbons (Aldrich Chemical Co. USA), and by literature reference (Adams 2017).

The oil content and volatile organic composition have a detrimental role in the flavouring property of the spice. In the present study, the yellow mace recorded high oil yield compared to the red mace, indicating enhanced aroma and flavour value for the yellow variety (Table 1). Literature reports showed that the essential oil content in mace oil from South India varied from 6.0 to 26.1% (Maya *et al.* 2004).



Fig. 2. Collection sites of *Myristica fragrans*.

 Table 1. Essential oil yield of yellow and red mace

Spice	Essential oil yield (fresh) (% v/w)	Essential oil yield (dried) (% v/w)	
Mace (Yellow)	5.4 ± 0.36	19.3 ± 1.15	
Mace (Red)	3.3 ± 0.61	9.2 ± 1.04	

In the present study, thirty-two volatile compounds were identified from the mace essential oil samples (Table 2). The distribution patterns of volatile chemicals of the red and yellow varieties were similar, except for quantitative differences. The major volatile organic constituents in the mace of both the red and yellow varieties were monoterpenoids, followed by phenylpropanoids, and the results are in corroboration with previous reports (Mallavarapu & Ramesh 1998). Sesquiterpenoids could not be detected in the aril essential oils of red and yellow variants. The distribution of the monoterpenoids α -pinene and sabinene were found to be similar for red and yellow mace oils. Among the monoterpenoids, sabinene was the primary compound ranging from 13.9% to 30.6% in red and 6.6% to 21.7% in yellow mace oils. The phenylpropanoid, elemicin was the major constituent of the yellow mace oil (22.3% to 31.0%), while for the red variant, the content of elemicin varied from 9.0% to 25.2%. The phenylpropanoids, myristicin, elemicin, and safrole are hallucinogenic compounds, and also have a detrimental role in the flavour and medicinal properties of nutmeg, while

Compound	RRI _{Lit}	RRI _{Cal}	Red aril (%)	Yellow aril (%)
α-Thujene	924	924	0.4±0.2	0.8±0.4
α-Pinene	932	932	11.9±0.1	9.0±0.2
Sabinene	969	970	16.2±0.7	16.2±0.1
β-Pinene	974	976	0.8±0	9.8±0.1
Myrcene	988	988	1.9±0.3	2.0±0
α -Phellandrene	1002	1006	1.6±0.1	1.5±0.8
δ-3-Carene	1008	1008	3.0±0	2.8±0
α -Terpinene	1014	1015	0.6±0.1	1.0±0.2
Limonene	1024	1027	4.9±1	3.3±0.5
β-Phellandrene	1025	1028	2.1±0.4	2.1±1.2
1-8-Cineole	1026	1030	0.1±0	0
γ-Terpinene	1054	1055	0.8±0.2	1.5±0.8
cis-Sabinene hydrate	1065	1068	0.5±0	0.4±0.2
Terpinolene	1086	1084	3.3±0.7	2.9±0.4
Linalool	1095	1100	0	0.2±2.0
trans-Sabinene hydrate	1098	1100	0.5±0	0
cis-p-Menth-2-en-1-ol	1121	1122	0	0.2±0.1
trans-p-Menth-2-en-1-ol	1136	1142	0.1	0
Terpinene-4-ol	1174	1178	2.2±0.5	3.0±1.2
α -Terpineol	1186	1196	0.8±0.2	0.5±0
Safrole	1285	1286	2.4±0.3	1.1±0
Eugenol	1356	1349	0.7±0	0
Methyl eugenol	1403	1398	13.0±0.1	7.8±0.4
E-Methyl isoeugenol	1491	1492	0	1.7±0.7
Myristicin	1517	1516	4.5±0.3	3.9±0.1
Elemicin	1555	1545	23.2±0.1	22.3±0.2
Total			94.2±2.1	94.0±1.5
Monoterpene hydrocarbons			47.8±1.8	51.8±3.0
Monoterpene oxygenated			4.6±1.2	1.9±2.1
Total Monoterpenoids			51.1±2.3	55.2±3.1
Phenyl propanoids			42.1±1.8	33.8±2.5

Table 2. Essential oil constituents of different accessions of *M. fragrans* with red and yellow arils

Compounds listed in order of their elution on Rxi-5 Sil MS capillary column.

Linear retention index (RRI_{Cal}) on Rxi-5 Sil MS column, using homologous series of C_8 - C_{30} alkanes. Linear retention index (RRI_{Lit}) taken from Adams (2017) and/or NIST 14. sabinene imparts a sweet smell to *Myristica* products (Zhao *et al.* 2019). Compared to the red variants, the high oil yield with a similar distribution pattern projects the yellow variety as a superior grade with respect to flavour profile.

Literature review revealed that the oil yield and percentage of constituents differ considerably depending on geographical origin, quality, and duration of storage (Leela 2008). In the international spice market, the aroma has a detrimental role in the spice value of mace. The mace oils from the West Indies, derived from the arils of the plants cultivated in Granada, have α -pinene, β -pinene, and sabinene as the major constituents, while the East Indian oils, derived from Indonesian islands, have higher amounts of myristicin (Purgeslove 1981, Ehlers et al. 1998). Quality-wise, the oil of Indian origin is considered an intermediate between the West Indian and East Indian oils. In mace oil originated from India, α -pinene, β -pinene, and sabinene were the major constituents, and the content of myristicin and elemicin were also high (Goplalakrishnan et al. 1992). The main volatile constituents of M. fragrans arils identified in the present study were similar to those in previous reports, except for minor differences in relative quantities of a few components.

Kerala state is leading in mace and nutmeg production in India, and the state hosts a variety of *M. fragrans* with red arils. Though recently mace with yellow colour is available in the spice market of Kerala as a first grade and highly priced spice commodity compared to the red mace, the product was least investigated for its essential oil chemistry. The present study revealed the superior quality of the yellow mace compared to the red variety with respect to oil yield.

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