ANALYSIS OF ESSENTIAL OIL COMPOSITION OF SOME SELECTED SPICES OF PAPUA NEW GUINEA

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ABSTRACT

Some selected spices from Papua New Guinea were analyzed to determine the chemical compositions of their respective essential oil contents. These included black and white pepper (Piper nigrum: *Piperaceae*); *cardamom* (Ellataria cardamomum: *Zingiberaceae*)*; ginger* (Zingiber officinale: *Zingiberaceae*)*; patchouli* (Pogostimon cablin: *Lamiaceae*)*; nutmeg* (Myristica fragrans: *Myristicaceae*)*; and the leaf and stalk of lemon grass* (Cymbopogon citratus: *Poaceae*). *The essential oils were obtained by exhaustive hydro-distillation and analyzed by a combined gas chromatography-mass spectrometry (GC-MS) method. The results indicated that d-3-carene (34.0 %), limonene (18.3 %) and b-caryophyllene (15.7 %) were the major components of black pepper (Piper nigrum); d-3-carene (23.7 %), limonene (23.7 %), b-caryophyllene (17.6 %) and b-pinene (16.9 %) were the major components of white pepper* (Piper nigrum); *1,8-cineole (44.4 %) and á-terpinyl acetate (39.7 %) were the major components of cardamom* (Ellataria cardamomum)*; citral (18.4 %), a-zingiberene (16.8 %) and camphene (11.2 %) were the main constituents of ginger* (Zingiber officinale)*; the patchouli alcohol (71.8 %) was the main constituent of patchouli* (Pogostimon cablin)*; a-pinene (22.6 %), sabinene (15.8 %) â-pinene (15.2 %) and myristicin (13.2 %) were the main components of nutmeg* (Myristica fragrans) *and* citral *was the main component in the leaf (91.0 %) and stalk (90.7 %) of lemon grass* (Cymbopogon citratus).

Keywords: *spices, essential oils, Piper, Ellataria, Zingiber, Pogostimon, Myristica, Cymbopogon, citral, floral diversity.*

INTRODUCTION

Essential oils are volatile organic compounds that are the major constituents in spice products and give rise to the perceived flavour and fragrance characters. These chemical compounds have a high vapour pressure and therefore are highly volatile, hence exist as vapour at ambient temperature and pressure. The analysis of these compounds in flavour and fragrance industries has served as the benchmark to ascertain the qualities of these products.

As an ongoing research program aimed at identifying the chemical compositions of the different volatile organic compounds in the floral diversity of Papua New Guinea (PNG) (Rali *et al.* 2003), we report here the chemical compositions within the matrix of the essential oil extracts obtained from some of the selected spice crops of PNG. The spices studied were black pepper (*Piper nigrum* L: Piperaceae), white pepper (*Piper nigrum* L: Piperaceae); Cardamom (*Ellataria cardamomum* White et Mason: Zingiberaceae); ginger (*Zingiber officinale* Roscoe: Zingiberaceae); patchouli (*Pogostimon cablin* Pellet: Lamiaceae); Nutmeg (*Myristica fragrans* Houtt: Myristicaceae); and the leaves and stalks of lemon grass (*Cymbopogon citratus* [DC] Stapf: Poaceae).

MATERIALS AND METHODS

The spice products were a donation from the New Guinea Spice Ltd of Rabaul, East New Britain Province, while patchouli and lemon grass samples were obtained from Tabubil in the Western Province. These samples were brought back to the laboratory while fresh and the essential oils extracted by exhaustive hydro-distillation, using an all-glass apparatus standard distillation setup. The oils obtained were dried over anhydrous sodium sulphate (Na_2SO_4) and analyzed using gas chromatography coupled to a mass spectrometer (GC-MS). The individual oil constituents were tentatively identified by their respective retention times and confirmed by comparison to the mass spectral data and that of the authentic reference compounds or with published data.

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RESULTS AND DISCUSSIONS

The results of the chemical analysis of essential oils extracted from some selected spices of PNG are presented in Table 1. The major constituents of both the black and white pepper (*Piper nigrum*) were d-3-carene, limonene, b-caryophyllene and b-pinene. However the composition of these essential oils were observed to vary in various proportions of composition in the individual types as presented in Table 1.0. Earlier reports by Martins *et al.* (1998) on the essential oil composition of black pepper reported a composition of limonene (18.8%), â-caryophyllene (15.4%), sabinene (16.5%) and â-pinene (10.7%). A further detailed phytochemical studies on the essential oil and other secondary metabolites in this species and others from the genus *Piper* were recently reviewed (Parmar *et al.* 1997). The oil extracts were previously reported to exhibit antimicrobial activity (Dorman & Deans, 2000; Hammer *et al.*, 1999). The oil obtained from *Piper nigrum* has been widely appreciated in culinary preparations.

Eleven components of cardamom oil were detected (Table 1). The major constituents were 1,8-cineole (44.4 %) and á-terpinyl acetate (39.7 %). In previous studies, Hussain *et al.* (1988) reported 74 % composition of 1,8-cineole contents while another study by Pieribattesti *et al.* (1986) reported 54.4 % 1,8-cineole and 24.0 % á-terpinyl acetate. Atta-ur-Rahman *et al.* (1999) further reported 1,8-cineole and á-terpinyl acetate compositions to be 30.7 % and 30.6 % respectively, and the oil was observed to inhibit the growth of the fungal species *Aspergillus flavus*. The cardamom seeds and oil have application in food flavouring in the forms such as whole, decorticated seeds, and grounded powder.

The chemical composition of essential oils from the rhizomes of ginger (*Zingiber officinale*) has been well documented including its health (Wilkinson 1999) and antimicrobial properties (Hammer *et al.* 1999; Hili *et al.* 1997; Habsab *et al.* 2000). The analysis of the rhizome oil extracts from this study indicated citral content (18.4 %) and á-zingiberene (16.8 %) to be the major components followed by camphene (11.2 %). In comparison with the oil yield and chemical compositions in the rhizomes from the published data (MacLeod and Pieris, 1984; Smith and Robinson, 1981; Kami *et al.* 1972), a variation in oil yield and chemical compositions in the extracts can be observed. Such difference can be attributed to earlier postulation that the oil yield and compositions are influenced by geographical locations, climate conditions and the age of the plant at harvest (Miyazaki and Taki 1955).

The major components identified for nutmeg (*Myristica fragrans*) seed oil were a-pinene (22.6 %), sabinene (15.8 %), b-pinene (15.2 %) and myristicin (13.2 %). A previous study, (Masada 1975) reported á-pinene (26.7%), â-pinene (20.7%), sabinene (14.5%), limonene (9.4%) and terpinen-4-ol (4.4%) as the main constituents. Recently, Atta-ur-Rahman *et al.* (1999) reported 37constituents representing 99.3% of the total nutmeg essential oils. The major component identified was terpinen-4-ol making up 31.3%.

Citral, the common indicator compound in lemon grasses (*Cymbopogon citratus*) is a mixture of two inseparable isomeric sesquiterpene aldehydes of geranial and neral (De Silva 1959). A previous study on citral content in lemon grass from Port Moresby was reported by Sino *et al.* (1992) to be 68 %. In this study, the samples from Tabubil had a higher citral contents in the leaf (91 %) and stalk (90.7 %). Such difference in citral yield can be attributed to geographical locations, weather patterns and conditions and age of grass as influential factors in the citral yield and quality of oil (Miyazaki and Taki 1955). This result is particularly encouraging because of the vast opportunity it has in the potential for cultivation and commercial production of this oil, which has applications in the synthesis of various flavour and fragrance substitutes, vitamin A and ionones (Kingston 1962).

The composition of various indicator chemical markers from this study are within established marketable values. For example, oil and citral contents in lemon grass (Table 1) suggests the possibility of cultivating this species for the commercial production. It is further recommended that detailed chemical study be pursued to establish the commercial potential for these and other spice products, hence establish adequate scientific basis on which to develop the untapped economic potential in the spice industry in PNG.

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Table 1. **Compositions (% area) of the essential oil extracts from the selected spices of Papua New Guinea**

Constituents	Black Pepper	White Pepper	Cardamom	Ginger	Patchouli	Nutmeg	Lemon grass leaf	Lemon grass stalk
a-thujene	-	-	-	-	-	4.0	-	-
a-pinene	7.4	8.9	1.5	3.5	-	**22.6**	-	-
b-pinene	**11.3**	**16.9**	-	-	-	**15.2**	-	-
sabinene	-	-	1.2	-	-	**15.8**	-	-
myrcene	2.7	2.6	1.4	2.2	-	2.1	8.9	2.7
r-cymene	2.6	1.9	0.8	-	-	2.6	-	-
limonene	**18.3**	**23.7**	3.4	1.7	-	3.4	-	-
1,8-cineole	-	-	**44.4**	4.8	-	-	-	-
linalool	-	-	2.1	-	-	-	-	-
terpinen-4-ol	-	-	2.0	-	-	9.6	-	-
a-terpineol	-	-	2.7	-	-	0.7	-	-
linalyl acetate	-	-	0.8	-	-	-	-	-
geranyl acetate	-	-	-	9.2	-	-	-	-
a-terpinyl acetate	-	-	**39.7**	-	-	-	-	-
camphene	-	-	-	**11.2**	-	-	-	-
a-phellandrene	3.5	2.3	-	-	-	-	-	-
b-phellandrene	-	-	-	3.9	-	1.6	-	-
g-terpinene	-	-	-	-	-	3.6	-	-
neral	-	-	-	6.4	-	-	**33.5**	**29.2**
geraniol	-	-	-	8.9	-	-	-	2.7
geranial	-	-	-	**12.0**	-	-	**57.5**	**61.5**
AR-curcumene	-	-	-	1.8	-	-	-	-
E,E-a-farnescene	-	-	-	6.4	-	-	-	-
a-zingiberene	-	-	-	**16.8**	-	-	-	-
b-bisabolene	-	-	-	2.9	-	-	-	-
germacrene-D	-	-	-	2.0	-	-	-	-
b-sesquiphellandrene	-	-	-	6.4	-	-	-	-
d-3-carene	34.0	23.7	-	-	-	-	-	-
a-terpinolene	0.6	-	-	-	-	0.8	-	-
d-elemene	1.9	-	-	-	-	-	-	-
b-caryophyllene	15.7	17.6	-	-	-	-	-	-
a-caryophylene	0.8	0.9	-	-	-	-	-	-
caryophyllene oxide	1.3	1.5	-	-	-	-	-	-
a-guaiene	-	-	-	-	7.5	-	-	-
seychellene	-	-	-	-	3.9	-	-	-
a-patchoulene	-	-	-	-	1.7	-	-	-
d-guaiene	-	-	-	-	9.9	-	-	-
pogostol	-	-	-	-	5.1	-	-	-
patchouli alcohol	-	-	-	-	**71.8**	-	-	-
a-terpinene	-	-	-	-	-	2.5	-	-
saffrole	-	-	-	-	-	2.3	-	-
myristicin	-	-	-	-	-	**13.2**	-	-

Note: - = not detected

REFERENCES

De SILVA, M.G. (1959) "Lemon Grass Oil from Ceylon". Manufacturing Chemist, 30, 415-416.

DORMAN, H.J.D. and DEANS, S.G. (2000), "Antimicrobial Agents From Plants: Antibacterial Activities of Plant Volatiles", J. Appl. Microbiology, 88, 308-316.

HABSAB, M., AMRAN, M., MACKEEN, M.M., LAJIS, N.H., KAKUJAKI, H., NAKATANI, N., RAHMAN, A.A and ALI, A.M., (2000) "Screening of Zingiberaceae Extracts for Antimicrobial and Antioxidant Activities", J. Ethnopharmacology, **73**, 403-410.

HAMMER, K.A., CARSON, C.F., and RILEY, T.V. (1999), "Antimicrobial Activity of Essential Oils and Other Plant Extracts", J. Appl. Microbiology, 86, 985-990.

HILI, P., EVANS, C.S. and VENESS, R.G. (1997), "Antimicrobial Actions of Essential Oils", Letters in Appl. Microbiol., 24, 269-275.

HUSSAIN, A., VIRMANI, O.P., SHARMA A., KUMAR A. and MISRA, L.N., (1988) "Major Essential Oil-Bearing Plants of India", Central Institute of Medicinal and Aromatic Plants, Lucknow, India.

KAMI, T., NAKYAMA, M., and HAYASHI, S. (1972) "Volatile Constituents of the Zingiber officinale", Phytochemistry, 11, 3377-3381.

KINGSTON, B.H. (1962) "A fresh look at Citral", Manufacturing Chemist, 33, 510-512 & 514-518.

MacLEOD, A.J. and PIERIS, N.M., (1984) "Volatile Aroma Constituents of Sri Lankan Ginger", Phytochemistry, 23, 353-359.

MARTINS, A. P., SALGUEIRO L., VILA, R., TOMI, F., Cañigueral, S., CASANOVA, J., PROENCA da CUNHA A. and ADZET, T. (1998) "Essential Oils from four Piper species" Phytochemistry, 49, 2019-2023.

MASADA, Y., (1975) "Analysis of Essential Oils by Gas Chromatography and Mass Spectrometry", Hirokawa Publishing Comp., Tokyo, p. 214-218.

MIYAZAKI, Y and TAKI, M. (1955) "Seasonal Variations in the Growth and the Oil Content of Lemon Grass", Bulletin of National Hygiene Laboratory, 73, 289-303.

ATTA-ur-RAHMAN, CHOUDHARY M.I., FAROOQ A., AHMED A, IQBAL M. Z., DEMIRCI B., DEMIRCI F and CAN BASER K. H (1999), "Antifungal Activities and Essential Oil Constituents of Some Spices from Pakistan", Third International Electronic Conference on Synthetic Organic Chemistry (ECSOC-3), September 1-30, 1999.

PARMAR, V.S., JAIN, S.C., BISHT, K.S., JAIN, R., TANEJA, P., AMITA, J., TYAGI, O.D., PRASAD, A.K., WENGELA, J., OLSENA, C.E., and BOLLA, P.M. (1997) "The Phytochemistry of the Genus Piper", Phytochemistry, 46, 597-673.

PIERIBATTESTI, J.C., SMADJA, J. and MONDON, J.M. (1986) "Flavors and Fragrances – A World Perspective", In Lawrence, B. M., Mookherjee, B.D. and Willis, B.J. (Eds.), Elsevier, Amsterdam, Netherlands, p.697-706.

RALI, T., LEACH, D.N. and WOSSA, S.W. (2003) "Preliminary Analysis of the Essential Oil Compositions in Some Aromatic Plants Species of Papua New Guinea", Proceedings of the 5th New Guinea Biological Conference, University of Goroka, Eastern Highlands, Papua New Guinea, September 2003.

SINO, D., ALAM, K., TAMATE, J., and RALI, T. (1992) "A Preliminary Study on the Lemongrass Oil From Papua New Guinea", Science in New Guinea, 18(3), 133-134.

SMITH, R.M. and ROBINSON, J.M. (1981) "The Essential Oils of Ginger from Fiji", Phytochemistry, 20, 203-206.

WILKINSON, J.M. (1999) "Ginger-A Review of its Medicinal Use", Biomedical Research, 1:23-32.