

Chemical constituents of essential oil from the leaf of *Alpinia nigra* of Bangladesh

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Abstract

Essential oil obtained by hydro-distillation from fresh leave of *Alpinia nigra* was analyzed by Gas Chromatography Mass Spectrometry (GC-MS). Fifty three compounds were identified in the leaf oil. The main essential oil compositions were 1,5,9,9-Tetramethyl-1,4,7-cycloundecatriene (24.92%), 6,6-Dimethyl-2-methylenebicyclo[3.1.1]heptanes (12.90%), 5-Amino-6-(2-fluoroanilino)furazano[3,4-b]pyrazine (12.18%), 4,11,11-Trimethyl-8-ethylenebicyclo[7.2.0]undec-4-ene (10.76%), 6-(3-Fluorobenzyl)-1,2,3,4,5,6-hexamethyl-2,4-cyclohexadien-1-ol (5.77%), 1,6,10-Dodecatrien-3-ol, 3,7,11-trimethyl-, (E)- (5.41%), Caryophyllene oxide (2.61%), 5,5-Dimethyl-4-[(1E)-3-methyl-1,3-butadienyl]-1-oxaspiro[2.5]octane (2.44%), 1,5,5,8-Tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene (1.96%), Sabinen (1.79%), D-Limonene (1.39%), α -Cadinol(1.20%).

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Introduction

Alpinia nigra (Bengali name: Jangli Ada, Family: Zinziberaceae) is widely grown in Bangladesh, India and Srilanka. It is an aromatic medicinal plant found in China, Bhutan, India, Sri Lanka and Thailand at an altitude of 900–1,100 m (Guo and Jiang, 1977). It is an herbaceous medicinal plant. The medicinal applications of *Alpinia nigra* have also been reported in stomach problems related to gastric diseases, gout and colic. The shoot of this plant has traditional usage among the native tribes of Tripura, Northeast India who consume the raw juice of the green shoot for its presumed anthelmintic, antioxidant properties (Roy and Tandon, 1999).

Most of the species of *Alpinia* are economically important, since they are being used in the treatment of various ailments (Jitoe *et al.*, 1992) and as ornamental plants (Criley, 1988). *Alpinia* species are characterized by a wide range of volatile compounds and have been the subject of numerous phytochemical studies (Fujita *et al.*, 1994; Pooter *et al.*, 1995; Kuster *et al.*, 1999). Indigenous system of herbal therapy is becoming an increasingly attractive approach to control parasitic infection, particularly in developing countries. They are the most common infectious agents of human beings that contribute in the wide spread occurrence of undernourishment, anaemia, eosinophilia and pneumonia (Bundy, 1994). They are also responsible for considerable economic losses to the livestock industry of marginal farmers, particularly of developing countries (Singh *et al.*,

2008; Ortega *et al.*, 2010). People living in tropical and sub-tropical countries with low per capita income, poor hygienic conditions suffers because of the presence of favorable conditions for the proliferation of the parasite (Hotez *et al.*, 2007) and, also for the propagation of intermediate hosts that are an essential link in the life cycle of the parasite (Roy and Tandon, 1992).

However, despite of having developed health care facilities, sophisticated instrumentations and advancement in chemotherapy, there is still lacking of proper and effective tools to deal with helminthic infections. Therefore, there is always been a need to find new anthelmintic drugs because current drugs do not control all parasitic infections well. Moreover, high treatment frequency, single-drug regiment or frequent use of the same anthelmintic has led to the development of resistance among helminth population (Geerts and Gryseels, 2000; 2001). Similarly, the undesired effect of limited availability to the rural areas and further restricted the effective control of helminthiasis (Martin *et al.*, 1997; Waller, 1997; Suleiman *et al.*, 2005) causing new threat to human society.

Though the advancement of synthetic medicines, to certain extend, has lifted the health care and livelihood of people, yet the use and importance of plants and its botanicals for the same has never been neglected and a large number of plants are screened for their efficacy against various helminthic infections (Diehl *et al.*, 2004; Athanasiadou *et al.*, 2007; Adama *et al.*, 2009; Roy *et al.*, 2008; 2010).

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Several such studies based on traditional medicinal knowledge were done in Indian sub-continent to test the putative anthelmintic activity of different plants. *Alpinia nigra* is one such plant, shoot of the plant along with a part of rhizome is used by the indigenous tribal people of Tripura, India, as vegetable, whereas aqueous juice of shoot of the plant is consumed to get rid of intestinal helminth infection (Roy and Tandon, 1999; Roy *et al.*, 2009; Roy and Swargiary, 2009).

Materials and Methods

Plant material

The leaves of *Alpinia nigra* were collected from the plants grown in the campus of Bangladesh Council of Scientific and Industrial Research (BCSIR) Laboratory, Chittagong during April 2013. The voucher specimen was deposited in the herbarium of BCSIR Laboratory, Chittagong.

Extraction of essential oil

Leaves were harvested from healthy, well-grown plants. Freshly harvested leaves (400 g) were grounded in a blender separately. The grounded leaves were subjected to hydro-distillation using Clevenger apparatus for 4 h for isolation of oils separately. The oil samples were stored at 0°C in air-tight containers after drying them over anhydrous sodium sulfate and filtered before going to GC-MS analysis.

GC-MS analysis

The essential oil from leaves of *Alpinia nigra* were analyzed by GC-MS electron impact ionization (EI) method on GC-17A gas chromatograph (Shimadzu) coupled to a GC-MS QP 5050A Mass Spectrometer (Shimadzu); fused silica capillary column (30 m x 0.25 mm; 0.25 mm film thickness), coated with DB-5 ms (JandW); column temperature 100°C (2 min) to 250°C at the rate of 3°C/min; carrier gas, helium at constant pressure of 90 Kpa. Acquisition parameters full scan; scan range 40 - 350 amu. Samples were injected by splitting and the split ratio 1:20.

Identification of the compounds

Compound identification was done by comparing the National Institute of Standards and Technology (NIST) library data of the peaks with those reported in literature, mass spectra of the peaks with literature data. Percentage composition was computed from GC peak areas on DB-5 column without applying correction factors.

Results and Discussion

The chemical compositions of volatile oil of leaf of *Alpinia nigra* are shown in Table-1. The oil yield varied from 0.02% to 0.15% for the leaves on a fresh weight basis (W/W). There were similarities on the library of such major compounds 1,5,9,9-Tetramethyl-1,4,7-cycloundecatriene (24.92%), 6,6-Dimethyl-2-methylenebicyclo[3.1.1]heptanes (12.90%), 5-Amino-6-(2 fluoroanilino)fuzazano[3,4-b]pyrazine (12.18%), 4,11,11-Trimethyl-8-ethylenebicyclo[7.2.0]undec-4-ene (10.76%), 6-(3-Fluorobenzyl)-1,2,3,4,5,6-hexamethyl-2,4-cyclohexadien-1-ol (5.77%), 1,6,10-Dodecatrien-3-ol, 3,7,11-trimethyl-, (E)- (5.41%), Caryophyllene oxide (2.61%), 5,5-Dimethyl-4-[(1E)-3-methyl-1,3-butadienyl]-1-oxaspiro[2.5]octane (2.44%), 1,5,5,8-Tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene (1.96%), Sabinen (1.79%), D-Limonene (1.39%), α -Cadinol (1.20%) and 1,8-Cineol (1.04%).

1,8-cineole is the common compound of all the reported oils in the world including ours (Rath *et al.*, 1994; Tewari *et al.*, 1999; Kong *et al.*, 2009). The value of 1,8-cineole is very lower in Bangladesh than others country. I think, it's depending on soil, weather, climate and species. α -Fenchyl acetate was found to be one of the major constituents in the root oil of these two species and rhizome oil of *Alpinia calcarata* (Tewari *et al.*, 1999).

A large number of chemicals have been isolated and studied from the genus *Alpinia*; however, limited literature is available on *Alpinia nigra* and its phytochemicals. The seed clusters of *Alpinia nigra* and isolated two bioactive flavone glycosides, astragalins and kaempferol-3-O-glucuronide from the plant (Qiao *et al.*, 2007). Out of the two chemicals, kaempferol-3-O-glucuronide was found to be a dominant compound in the seed clusters distributed primarily in the pulp. In addition to this, two major volatile oils, β -Pinene and α -pinene have also been isolated from the fruits and rhizomes, of *Alpinia nigra* (Qiao *et al.*, 2000).

Conclusion

It may be concluded that *Alpinia nigra*, growing widely in Bangladesh, may be utilized as a source for the isolation of natural 1,5,9,9-Tetramethyl-1,4,7-cycloundecatriene; 6,6-Dimethyl-2-methylenebicyclo[3.1.1]heptanes respectively for medicinal and commercial use.

Table 1. Chemical constituents of the leaf oil of *Alpinia nigra*

S/N	Compound Name	Percentage
1	1,5,9,9-Tetramethyl-1,4,7-cycloundecatriene	24.92
2	6,6-Dimethyl-2-methylenebicyclo[3.1.1]heptane	12.90
3	5-Amino-6-(2-fluoroanilino)furanazo[3,4-b]pyrazine	12.18
4	4,11,11-Trimethyl-8-methylenebicyclo[7.2.0]undec-4-ene	10.76
5	6-(3-Fluorobenzyl)-1,2,3,4,5,6-hexamethyl-2,4-cyclohexadien-1-ol	5.77
6	1,6,10-Dodecatrien-3-ol, 3,7,11-trimethyl-, (E)-	5.41
7	Caryophyllene oxide	2.61
8	5,5-Dimethyl-4-[(1E)-3-methyl-1,3-butadienyl]-1-oxaspiro[2.5]octane	2.44
9	1,5,5,8-Tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene	1.96
10	Sabinen	1.79
11	D-Limonene	1.39
12	α -Cadinol	1.20
13	2-Isopropyl-1H-benzimidazole 3-oxide	1.13
14	1,8-Cineol	1.04
15	Oxirane, dodecyl-	0.87
16	Dodecahydro-as-indacene	0.86
17	Juniper camphor	0.82
18	Germacrene D-4-ol	0.77
19	1-Isopropyl-4,8-dimethylspiro[4.5]dec-8-en-7-one	0.76
20	4-Terpineol	0.73
21	Alloaromadendrene oxide-(1)	0.69
22	4-Isopropyl-1,6-dimethylenedecahydronaphthalene	0.60
23	7-Isopropenyl-1-methyl-4-methylenedecahydroazulene	0.45
24	1,2,3,6-Tetramethylbicyclo[2.2.2]oct-2-ene	0.40
25	11-Azabicyclo[4.4.2]dodec-11-ene, 12-ethoxy-	0.24
26	3-Carene	0.16
27	1-Methyl-cis-1,2-epoxycyclooctane	0.24
28	5-Isopropyl-2-methylbicyclo[3.1.0]hexan-2-ol	0.17
29	1,5-Heptadiene, 3,3-dimethyl-, (E)-	0.15
30	α -Terpineol acetate	0.31
31	6-Hydroxy-3-(hydroxymethyl)bicyclo[2.2.1]heptane-2-carboxylic acid	0.39
32	Eugenol	0.30
33	7-Isopropyl-4a,8a-dimethyl-4a,5,6,7,8,8a-hexahydro-2(1H)-naphthalenone	0.29
34	Methyl iso-eugenol 1	0.37
35	1H-Cycloprop[e]azulene, decahydro-1,1,7-trimethyl-4-methylene-	0.19
36	Chamigren	0.23
37	α -Panasinensin	0.38
38	2,4-Diisopropenyl-1-methyl-1-vinylcyclohexane	0.17
39	(E,E,E)-3,7,11,15-Tetramethylhexadeca-1,3,6,10,14-pentaene	0.36
40	β -Elemenone	0.30
41	Caryophyllene oxide	0.30
42	Patchoulene	0.13
43	Cubanol	0.23
44	(-)-Globulol	0.16
45	Longifolenaldehyde	0.25
46	9-cis-Retinal	0.28
47	5-Isopropyl-2-methylbicyclo[3.1.0]hexan-2-ol	0.10
48	(-)-Camphor	0.10
49	L-borneol	0.10
50	7-Methylene-9-oxabicyclo[6.1.0]non-2-ene	0.12
51	Germacrene D	0.12
52	γ -Murolene	0.13
53	1-Methyl-4-methylene-2-(2-methyl-1-propenyl)-1-vinylcycloheptane	0.17

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