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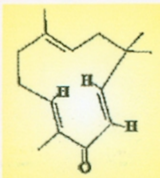
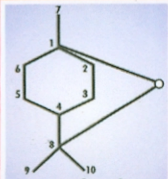
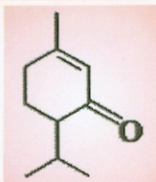


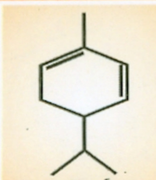
Zerumbone



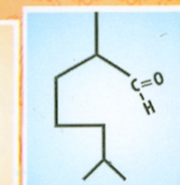
Eucalyptol



Piperitone



α-phellandrene



Citronella

17TH INAUGURAL LECTURE

titled: *Essential*

OILS FROM NIGERIAN MEDICINAL PLANTS AND THEIR BENEFITS

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Professor of Organic Chemistry,
Faculty of Pure and Applied Sciences

Thursday, 7th December, 2017

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17TH INAUGURAL LECTURE

By

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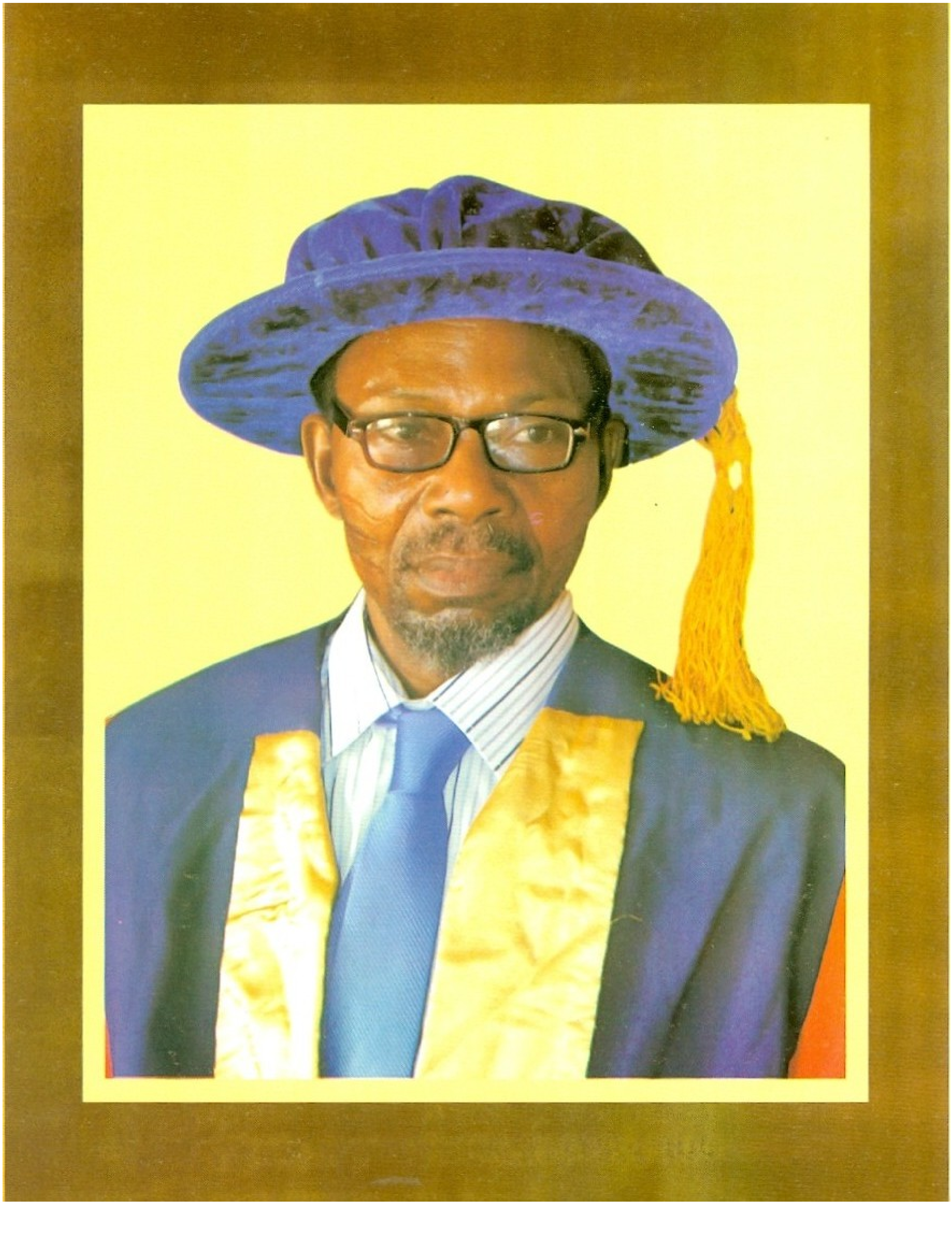


The Vice Chancellor,
Deputy Vice Chancellor,
Members of the Governing Council present,
Principal Officers of the University,
Dean of the Faculty of Pure and Applied Sciences,
Dean of other Faculties and The Post-graduate School
Gentlemen of the Press,
Distinguished Ladies and Gentlemen

Introduction

I am particularly grateful to the Vice Chancellor for giving me this opportunity to deliver the 17th inaugural lecture of this University and the fifth in the Faculty of Pure and Applied Sciences. I joined the University in January 1991 as one of the foundation staff of the Department of Pure and Applied Chemistry. Four of us who are foundation staff are still in the Department, while others have left or transfer their services to other departments in the University. Notably, our dear Chemistry Department has grown over the years, and today, there are twenty- three (23) academic and ten (10) laboratory staff members in the Department.

Medicinal Plants

From the title of this lecture, there is the need to discuss medicinal plants in relation to essential oils.

A medicinal plant is a plant, flowering or non-flowering that is used throughout history to either cure or lessen

symptoms from an illness. Its properties are similar to that of conventional pharmaceutical drugs. Medicinal plants are also called medicinal herbs.

Plants have been used for medicinal purposes long before prehistoric period (Sofowora, 1984), and today, traditional systems of medicine continue to be widely practiced. There is increased emphasis on the use of plants as a source of treatment for a wide variety of human ailments because of rise in population, inadequate supply of drugs, prohibitive cost of treatment, side effects of several synthetic drugs and development of resistance to currently used drugs. Recently, the World Health Organization (WHO) estimated that 80% of people worldwide rely on herbal medicine for primary health care needs, most especially in the rural areas. This, of course, mirrors the Nigeria situation and in fact, it has been observed that in developed countries, the use of herbal health products is on the increase. In China for example, herbal medicine is provided along with orthodox medicine in public hospitals for patients to state their preference for treatment. Also, it has been estimated that in developing countries, plant drugs contribute as much as 80% of the total drug needs.

Of note is the fact that the economic importance of medicinal plants in Nigeria is high. These herbs synthesize hundreds of chemical compounds, some of which were produced by pathways of primary metabolism. These are lipids, carbohydrates amino acids and nucleic acids. Primary metabolites are essential to the survival of the plant. There are also compounds which are produced by pathways of secondary metabolism and are referred to secondary metabolites.

In medicinal chemistry, secondary metabolites are also called natural products. Examples of secondary metabolites are terpenoids, alkaloids, phenols, polyphenols, tannins, steroids, phenylproparoids and polyketides (Hanson, 2003).Secondary metabolites are not essential for survival but nevertheless provide organisms that produce them an evolutionary advantage (Demain et al., 2000). Some of the natural products have therapeutic benefit as traditional medicines for treating diseases. An example of natural products is the essential oil obtained from aromatic plants.

Essential Oils

Essential oil is a concentrated hydrophobic liquid containing volatile aroma compounds in plants. Essential oils are also known as volatile oils, ethereal oils or aetherolea, or simply as the "oil of" the plant from which they were extracted, such as oil of clove. Oil is "essential" in the sense that it carries a distinctive scent, or essence, of the plant. Essential oils are natural aromatic compounds found in seed, bark, stems, roots, flowers, and other parts of plants. They can be both beautifully and powerfully fragrant. In addition to giving plants their distinctive smells, essential oils provide plants with

protection against predators and disease and play a role in plant pollinations. Essential oils are non water – based photochemical made up of volatile aromatic compounds. Although they are fat soluble they don't include fatty lipid or acids found in vegetable and animals oils. Essential oils are very clean, almost crisp to touch and are immediately absorbed by the skin. Pure and unadulterated essential oils are translucent and range in color from crystal clear to deep blue. They are obtained from plant in various ways, depending on the nature of the part in which they occur.

Methods of extraction of essential Oils

There are various methods used in the extraction of essential oil

(1) Hydrodistillation

This is the most common method used in the extraction. It involves the use of an all glass Clevenger-type apparatus. The pulverized plant material is immersed in water, which is contained in a round bottom flask. It is then heated in a regulated heating mantle at a temperature of 60oC for 3 hours. During heating, the steam and oil vapour are then separated after condensation in the condenser connected to the apparatus. The resulting oil is then collected into a tightly closed glass vial. Anhydrous magnesium sulphate could then be added to remove any traces of water left in the oil

(Okoh et al., 2010).

(2) Steam distillation

This method is also similar to hydrodistillation. In this technique, the steam is produced in a steam generator and passed through the pulverized sample contained in a vessel fitted with water-cooled condenser. The steam vaporizes the volatile compounds in the plants and carries volatile substances through the condenser. The essential oil is allowed to stand for minutes and being lighter than the water, will stay at the top. The essential oil can then the easily separated from the aqueous layer and collected into a glass vial (Setzer, 2017).

(3) Expression

This method is suitable for extracting essential oils embedded in citrus fruits, like orange, lemon, grape and lime. This technique involves the cleaning of the outer layer of the plant that contains the oil. The peels are separated from the fruits and then pulverized and pressed. The oil is separated from the mixture by centrifugation. The oil produced is pure, has short shelf life and cheap (Hamid et al., 2011).

(4) Effleurage

This method is highly suitable for the extraction of essential oil from flowers, that yield very low amount of oil when hydrodistillation or steam distillation methods are used, or which are too delicate to withstand the boiling

point of water. Fresh flowers are added at intervals so that the vegetable oil can absorb more of the flower essential oil. When the vegetable oil is saturated with essential oil, ethanol is added to separate fatty substances from the flower oil. The resulting mixture is filtered and the filtrate is made to undergo vacuum distillation whereby only the essential oil remains (Hamid et al., 2011).

(5) Super critical carbon dioxide

This technique involves placing the plant sample in a stainless steel tank and carbon dioxide is injected into the tank, pressure inside the tank builds. The carbon dioxide turns into liquid under high pressure and act as solvent to extract the essential oils from the sample. At low pressure, liquid carbon dioxide turns back to gaseous state leaving no residue behind. The resulting essential oil obtained through this process contain no organic solvent, it is safe, non-toxic and non-combustible. Moreover, many compounds show an appreciable solubility in super critical carbon dioxide at easily accessible conditions. A comparison of oil extracted with carbon dioxide in the supercritical state and with other techniques, seems to indicate that super critical carbon dioxide afford quality essential oils since the oil is obtained in its most natural form (Kubatova, 2001).

Analysis of Essential Oils

Essential oils are complex mixtures of volatile secondary metabolites in plants. Each of the constituent contributes to the beneficial or adverse effects of the oils. Therefore, the intimate knowledge of the oil composition is important for better and specially directed applications. Such applications include; insecticides, herbicides and as therapeutic agents. However, oil composition can only be obtained using standard analytical techniques (Buchbauer, 2000). The techniques in use are gas chromatography and gas chromatography-mass spectrometry.

Gas Chromatography

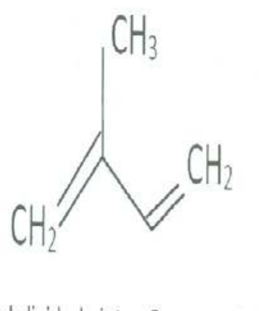
This technique involves the introduction of the extracted oil into the injection port with hypodermic needle. On introduction, the oil vaporizes immediately and it is propelled by the carrier gas, such as helium, hydrogen or nitrogen down the column. The column allows various constituents of the oil to partition themselves. Ideally, each component separates before eluting from the column. On elution, detector measures different compounds as they emerge from the column. They usually emerge as chromatogram that can be stored electronically and displayed on a computer monitor. The identity of each constituent is established by comparing the retention time of the oil constituents with that of n-alkanes run with the oil. Relative percentage of the oil constituents can be determined from the peak area of each component in the chromatogram. Surprisingly,

constituents with the same molecular mass could co-elute and be detected as one compound based on its retention index. It is on the basis of this, that the detector in the gas chromatograph is replaced with mass spectrometer which led to the emergence of gas chromatography-mass spectrometry.,

Gas chromatography – Mass spectrometry (GC/MS)

In this technique, the oil is introduced into the GC inlet where it vapourizes and then swept onto the column by the carrier gas. The sample flows through the column experiencing the normal separation processes. As the various sample components emerge from the column openings they flow into the capillary column interface. The function of the interface is to separate and concentrate the eluant via the removal of the carrier gas. From the interface, the sample enters the ionization chamber of the mass spectrometer where the oil constituent molecules are ionized by electron ionization techniques to form molecular ions. Molecular ions subsequently fragment into fragment ions. The latter ions are passed to the mass analyzer, where they are separated according to their mass to charge ratios. The ions enter a detector and then on to an amplifier to boost the signal. The detector sends information to the computer, which act as a "clearing house". It records all the data produced, converts the electrical impulses into visual displays and hard copy displays to produce mass spectrum of each of the constituents of the essential oil.

(Baser and Buchbauer, 2010). They are made up of various isoprene units: C_5H_8



Terpene is subdivided into C_{10} monoterpenes, C_{15} sesquiterprenes, C_{20} diterpenes, C_{25} sesterterpenes, C_{30} triterpenes, C_{40} tetraterpenes and C_{5n} polyterpenes, where n in $C_{5n} > 8$.

Applications of Essential Oils

(a) Pharmacological Purposes

Some of the essential oils are very important because they are bioactive. Plants that have phytochemicals that inhibit or destroy the activities of microorganisms are called antimicrobial plants.

The microbial pathogens could be bacteria, fungi, virus while some antimicrobial agents are usually in the form of natural products such as ampicillin and gentamin. Some essential oils which are used in the treatment of these pathogens are: oil of thyme, (Thymus vulgaris) and oil of bitter-kola (Garcinia kola) which are used as antibacterial agents while, oil of camphor (Cinnamomum camphora) and oil of chamomile (Matricaria chamomilla) are both used as anti-inflammatory agents.

(b) Industrial Purposes

Essential oils have a variety of uses in the industry in form of flavoring, perfumery and cosmetics. Some essential oils used in the industry are: Oil of Lavender (Lavandula officinalis) used as insecticides, oil of angelica (Archangelica officinalis) used as flavouring, oil of birch (Betula lenta) used as insect repellent and oil of cajeput (Melaleuca leucadendron).

(c) Air Purifier

Essential oils can also be inhaled or applied topically to certain areas of the body to increase energy, relaxation or focus and decrease stress or deepen meditation. Diffusing oils like lemon and grape fruit essential oils can be used to purify the air thereby generating a refreshing and uplifting energy to the space.

(d) Path way to an emotional State of Mind

Diffused or directly inhaled, oils such as lavender essential oil or frankincense essential oils are used for a deepened more relaxed experience.

(e) Breathing

The pure aroma of essential oils such as frankincense, peppermint and raven essential oils assist in opening pathway to a more emotional state of mind via the pineal gland. Directly inhaling peppermint can help bring more oxygen to the brain. It may also help to alleviate muscle soreness and tension. Pepper mint oil can also aid in increasing focus and mental clarity and when added to a cool glass of water, can help sooth minor stomach discomfort. Lavender essential oil can also be used to decrease post practice muscle irritation.

Uses of Essential Oils in the Industry

1. FRAGRANCES: The main reason for the expansion of the essential oil industry and the growing demand for products was the development of the food, soap, and cosmetics industries. These multinational companies are the main users of fragrances and flavors from essential oils.

2. FLAVORS: The development of the soft drinks industry is of great importance because it is a major consumer of essential oils, especially those of citrus origin. It is enough to say that nowadays, only Coca-Cola-produced beverages are consumed worldwide in a quantity exceeding 1 billion drinks per day. If we consider that the average content of the appropriate essential oil in the final drink is about 0.001 – 0.002%, and the standard drink is ca. 0.31 (300 g), we approach a daily consumption of essential oils by this company alone at the level of 3-6 tons per day, which gives an annual usage well over 2000 tons. Although all other brands of the food industry use substantial quantities of essential oils in ice creams, confectionery, bakery, and a variety of fast foods (where

spice oils are used), these together use less essential oils than the beverage manufacturers.

3. TOILETRIES: There is one special range of products that can be situated between the food and cosmetic, i.e. toiletries industry. It is a big consumer of essential oils, especially of all kinds of mint and eucalyptus oils. These are oral care products, chewing gums, and all kinds of mouth refreshing confectioneries.

4. FOOD AND PACKAGING: The dairy, coffee and tea, vegan, and meat/fish/sea food industries can all use essential oils in their applications as natural preservatives.

Present study on essential oils

We have been able to investigate the chemical composition of fifty three (53) Nigerian plants belonging to eighteen (18) different families. We also carried out biological activity experiments on some of these plants and we were able to confirm the traditional uses of these plants.

In the course of our studies, we carried out isolation and characterization of pure components either from the essential oils or extracts of some of the plants.

Table 1: Families of essential oil bearing-plants studied

Family	Number of plants
Amaranthaceae (chenopodiaceae)	1
Annonaceae	3
Araucariaceae	1
Asteraceae	5
Burseraceae	1
Clausiaceae	2
Cupressaceae	5
Cyperaceae	2
Euphorbiaceae	1
Fabaceae	1
Fabaceae (caesalpiniaceae)	1
Lamiaceae	3
Moraceae	1
Myristicaceae	1
Myrtaceae	19
Poaceae	1
Rutaceae	3
Zingiberaceae	2

Eucalyptus species (Myrtaceae)

Thirteen leaf essential oils and seven seed essential oils of *Eucalyptus* species have been studied here. The yields varied from 0.2% in *E. deglupta* to 4.7% in *E. grandis*. The colours also varied from colourless in *E. camaldulensis* variety nansee to light brown in *E. torelliana*.

Monoterpenoids constituted the bulk of essential oils of most of the *Eucalyptus* species studied, with the exception of *E. deglupta* in which sesquiterpenoids were the major (48%) components of the leaf oil (Oyedeji et al.,

1999).1, 8 – cineole was the component present in almost all Eucalyptus species we studied. The few exceptions were in leaf oils of *E. tereticornis*, *E. deglupta* and *E. grandis* as well as the seed essential oil of *E. torelliana*. The observation in the case of the leaf oil of E. tereticornis is unusual and it could be due to differences in environmental conditions.

Eucalyptus oils in trade matters are categorized into three broad types (Copper and Hone, 1992).

(i) Oils that are rich in 1, 8-cineole. These are medicinal and are used in the pharmaceutical industry.

(ii) Oil that is rich in citronella. They are used as perfumes.

(iii) Oils that have piperitone or □-phellandrene as their main constituents. These are for industrial uses.

Our results on the analyses of oils of Eucalyptus species from Nigeria are shown below:

Table 2: Classification of Eucalyptus plants studied.

1,8-cineole group	Citronella group	Piperitone/phellandrene group	Others
E. camaldulensis var. catharine. Leaf oil 70.4%	•	•	*E. alba* leaf oil α-thujene 32.7%
E. camaldulensis var. mysore Leaf oil 32.8%			*E. cloeziana* leaf oil α-pinene 46.6%
E. camaldulensis var. nansee Leaf oil 72.2%			*E. degluptaleaf* oil E-β-nerolidol 34.8%
E. camaldulensis var. nansee seed essential oil 40.8%			*E. grandisleaf* oil α-pinene 30.4%
E. camaldulensis var. petford Leaf oil 85.6%			*E. maculataseed* essential oil α-pinene 8%
E. camaldulensis var. petford seed essential oil 52.0%			*E. salignaleaf* oil α–thujene 63.8%
E. cloeziana seed essential oil 23.8%			*E. tereticornisleaf* oil β- pinene 39.4%
E. microtheca Leaf oil 35.18%			*E. tereticornisseed* essential oil α-selinene 15.5%
E. microtheca seed essential oil 53.8%			*E. torellianafruit* oil α-pinene 55.8%
E. propinqua Leaf oil 61.8%			
E. torelliana Leaf oil 33.8%			

Sources: Oyedeji et al., Jimoh et al., Ogunwande et al., Olawore & Ololade, Olawore et al., Ololade & Olawore.

Eight of the *Eucalyptus* oils are in the 1, 8-cineole group. The varieties of *E. camaldulensis*, petford, catharine and nansee are particularly suited for medicinal use because of their high 1,8-cineole contents (>70%). *E. camaldulensis* var. petford has the best quality (Jimoh et al., 2005) among Nigerian grown *Eucalyptus* species with the highest 1, 8-cineole content of 85.6%. The oil is

therefore good for commercial exploitation in Nigeria.

Structure of the major constituents of Eucalyptus species

Eucalyptol

1,8-cineole

Analyses of Essential Oils from other Medicinal Plants investigated

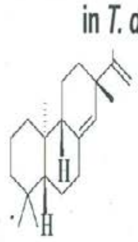
The chemical compositions of a few of the aromatic plants we studied will be discussed in this lecture because of time constraints. In all cases, the part of the plant investigated was pulverized and then subjected to hydrodistillation. The essential oil obtained was analyzed by combined gas chromatography – mass spectrometry (GC-MS).

***Taxodium distichum* (L.) L.C. Rich *Cupressaceae* (*Taxodiaceae*)**

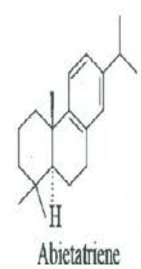
T. distichum, commonly called bald cypress, was selected because the plant has been reported to possess antimicrobial, antiplasmodic and anti-inflammatory activities (El Tantawy *et al.*, 1999). Hydrodistillation of leaves and fruits of this plant afforded yellowish pink oils with pleasant odours. The fruit oil was majorly composed of monoterpenoids (67.7%) with α-pinene, the most abundant component (60.5%). Thujopsene (17.6%) is the other significant component.

The main components of the leaf oil were thujopsene (27.7%), widdrol (12.8%) and β-caryophyllene (11.4%). The diterpene present in both oils was pimara-8(14),15-diene (leaf oil 13.1%, fruit oil 0.5 %) while diterpenes, abietatriene (2.7%) and abietadiene (1.7%) were present only in the leaf oil (Ogunwande *et al.*, 2007). Bioactivities of the oils will be discussed shortly.

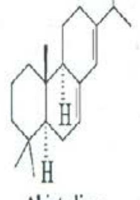
Structures of Diterpenes detected in *T. distichum* oils.



Pimara-8(14),15-diene



Abietatriene



Abietadiene

Hyparrhenia rufa (Nees) Stapf (*Poaceae*).

This plant is used as a natural perfume among the rural community. There are no references in the literature on the chemical composition of its volatile oil. The aerial parts of the plant gave yellow oil with sweet-smelling odour. The main components were τ–cadinol (17.43%) and β–selinene (11.64%). The most interesting characteristics of the oil was isolation and characterization of four unusual and rarely occurring terpenoid esters: (*E*,*E*)-methyl-10,11-epoxyfarnesoate, (*E*,*E*)- methylfarnesoate, (2*E*,6*E*)-methyl-3,7,11-trimethyl-10-oxododecadienoate and (2*E*,6*E*)-methyl-10-hydroxy-3, 7,11-trimethyl-2,6,11-dodecatrienoate. These esters and other ester components are believed to be responsible for the characteristic sweet-smelling odour of the oil. The oil of the plant will therefore be a good raw material in the perfume industry. Isolation and characterization of these esters will be discussed shortly.

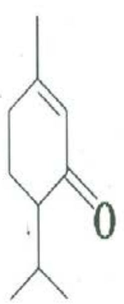
Tagetes erecta L. *Asteraceae*

The essential oils of this plant have been reported to possess antimicrobial and insecticidal properties (Singh *et al.*, 2003). This plant was therefore investigated on account of its aromatic and medicinal properties.

The leaf and flower essential oils were light yellow in colour. The leaf oil was dominated by piperitone (50.7%) and piperitenone (13.2%). Other monoterpenoid constituents included (*E*) -β-ocimene (6.7%) camphor (4.8%) and limonene (4.1%). The flower oil on the other hand, contained mainly 1,8-cineole (23.1%), α-pinene (11.7%), α-terpineol (10.7%) and piperitone (8.0%), with sizeable amounts of sabinene (5.6%) and piperitenone (4.0%).

There is relatively high proportion of piperitone and its derivative, piperitenone in the oil. Both are flavouring agents, therefore, the Nigerian leaf oil of the plant has potential as important raw materials in the flavouring industry (Ogunwande *et al.*, 2006).

Structures of the main components of *T. erecta*



Piperitone

Piperitenone

***Cyperus articulatus* L. (*Cyperaceae*)**

The rhizomes of the red and black varieties of the plant are known to possess repellant, anticonvulsant, antimalaria and antimicrobial properties (Etkin, 1997, Abubakar, *et al.*, 2000 and Bum *et al.*, 2001). Literature information is scant on the chemical composition of the essential oil of *C. articulatus*.

Our studies on this plant (Olawore *et al.*, 2006), showed

that the rhizome oils of both red and black varieties of the plant were rich in sesquiterpenoids, 71.7% and 60.6% respectively. The red rhizome oil was characterized by the abundance of cyperotundone (42.3%), piperitone (10.1%), β-maaliene (8.4%) and germacrone (5.3%). Cyperene epoxide is present as a minor component (2.2%). The main constituents of rhizome oil of the black variety were cedrol (19.0%), guaia-5-en-11-ol (14.9%) and cyperotundone (9.6%). Bioactivity of the plant will be discussed shortly.

***Plectranthus aegyptiacus* (Forsk) (*Lamiaceae*).**

The aerial parts, especially the leaf, was used as a spice in Southwestern part of Nigeria and in Saudi Arabia, for the treatment of laryngitis and sore throats (Lukhoba *et al.*, 2006).

The leaf oil was dominated by sesquiterpenoids (53.02%) and monoterpenes (18.46%). The constituents detected in significant amounts in the leaf oil were α-gurjunene (9.29%), 8-oxatetracyclo [5.2.1.1(2,6).1(4,10)] dodecane (7.99%), α-pinene (6.80%), humulene (6.55%), 2-isopropylidene-3-methylhexa-3,5-dienal (6.31%), farnesene epoxide (6.24%) and linalool (5.07%). The stem oil was majorly composed of hydrocarbons (64.66%), sesquiterpenes (23.72%) and monoterpenes (5.16%). Compounds present in appreciable amounts in the stem oil were *n*-octane (7.22%), 1,2-dimethylcyclohexane (7.14%), germacrene D (6.68%), *n*-

decane (6.08%), limonene (5.16%) and 1,2-dimethylbenzene (5.07%).

There are qualitative and quantitative differences in compositional profiles of the two volatile oils. The oils would be useful as spices (Aliyu *et al.*, 2017).

Syzygium guineense (Willd) DE. (*Myrtaceae*)

Traditionally, the fruit of the plant was used as treatment for dysentery (Burkill, 1987). It has been established that the triterpenoids isolated from the crude extract of the leaf of *S. guineense* have antibacterial activity (Djoukeng *et al.*, 2005), while the plant extract has been reported to be active against malaria (Segawa and Kasenene, 2007). Moreover, the ethanolic leaf extract of the plant possessed both analgesic and anti-inflammatory activities (Ior *et al.*, 2012).

Thirteen components (99.5% of the oil), were identified from the volatile oils of the leaves of *S. guineense*. Cyperene (23.0%), β-pinene (14.2%), D-limonene (13.0%), α-pinene (11.9%), caryophyllene (8.9%), cyclohexane (7.4%) and epiglobulol (6.4%) were the principal constituents identified in the oil. Eighteen components representing 99.2% of the oil were identified from the seed essential oil. The seed oil was dominated by octadec-9-enoic acid (14.3%), guaiol (10.5%), humulane-1,6-dien-3-ol (10.3%), epiglobulol (8.8%), selina-6-en-4-ol (8.5%) and palmitic acid (6.5%). Further work is expected to be carried out to confirm the use of this plant in ethnomedicine (Odewande *et al.*, 2017)

Bioactivites of the essential oils of some of the plants studied

We carried out the biological activities on some of the plants we studied. These types of investigations will determine the veracity of the ethno-medicinal uses of these plants.

(i) *Eucalyptus* species

Table 3: Antimicrobial activities of essential oils from some *Eucalyptus* spp. growing in Nigeria.

Microorganisms	Inhibition zone (mm)[a]							
	E_1	E_2	E_3	E_4	E_5	Amp	Gen	TiO
Gram positive								
Staphylococcus aureus UCH 560	-	10	10	10	12	R	14	n.t
S. aureus UCH 681	-	11	11	-	11	-	10	n.t
S. aureus UCH 511	-	10	12	12	11	R	12	n.t
B. cereus UCH	-	11	10	12	12	-	14	n.t
Gram negative								
Escherichia coli NCTC 7001	10	10	-	10	10	-	14	n.t
E. coli UCH 307	18	16	18	24	22	12	14	n.t
E. coli UCH 270	-	11	14	-	18	-	-	n.t
Pseudomonas aeruginosa NCTC675	-	9	-	10	-	9	10	n.t
P. aeruginosa UCH 655	10	12	-	12	11	-	-	n.t
Fungus								
Candida albicans	-	12	12	14	13	n.t	n.t	16

a= values are the mean of three replicates. E1 to E5= essential oil of *E. alba, deglupta, saligna, camaldulensis var.* catharine, *camaldulensis var.* mysore respectively (in 10% DMSO dilution)

5 mg/mL; Amp= ampicillin 2.5 µg/mL; Gen= gentamicin 1

Table 5: Zones of inhibition (mm) showing the antimicrobial properties of the seed essential oil of two varieties of *E. camaldulensis* var. nansee and petford.

Concentration (µg/mL)	Seed essential oil						ERY	CFM
	nancee			petford				
Organisms	1000	100	10	1000	100	10	10 µg	5 µg
Escherichia coli	18	18	14	10	10	-	22	-
Klebsiella pneumonia	19	17	17	24	23	19	21	07
Pseudomonas aeruginosa	20	18	18	13	13	13	20	10
Streptococcus agalactiae	-	-	-	15	10	-	-	-
Staphylococcus aureus	20	13	12	20	20	20	-	-
Salmonella typhimurium	-	-	-	-	-	-	21	-

Key note: - = Resistant, 6-9mm = low inhibition, 10-14mm = moderate inhibition,

≥ 15mm = high inhibition, ERY= erythtomycin, CFM= cefixime

The seed essential oils of *E. camaldulensis* var. nansee (Olawore and Ololade, 2017) displayed strong activity against *P. aeruginosa, S. aureus, K. pneumonia* and *E. coli* but *S. typhimurium* and *S. agalactiae* were resistant to the oil. The seed essential oils of *E. camaldulensis* var. petford exhibited high inhibition against *K, pneumonia, S. aureus* and *S. agalactiae* while it showed no activity against *S. typhimurium*. These results confirm the use of *E. camaldulensis* var. nansee and petford in traditional medicine.

Table 6: Antibacterial activities of the essential oils of *Eucalyptus torelliana* F (MIC, µg/mL)

Sample	Bc	Sa	Ec	Pa	Ca	As
Fruit oil	2500	156	156	625	312	156
Leaf oil	625	78	156	625	312	156
α-pinene	625	312	312	625	n.t	n.t
1,8-cineole	156	625	625	625	n.t	n.t
Gentamicin sulfate	1.22	0.61	2.44	1.22	n.t	n.t
Amphotericin B	n.t	n.t	n.t	n.t	0.61	0.61

Bc= Bacillus cereus (ATCC no. 14579); Sa= *Staphylococcus aureus* (ATCC no. 29213); *Escherichia coli* (ATCC no.25922), Pa=*Pseudomonas aeruginosa* (ATCC no. 27853), Ca= *Candida albicans* (ATCC no. 10231), As= *Aspergillus niger* (ATCC no. 16401); n.t=Not tested.

The fruit and leaf oils of *Eucalyptus torelliana* (Jimoh *et al.*, 2005) showed relatively weak antimicrobial properties.

Table 7: Antimicrobial activities of rhizome essential oils of red and black *Cyperus articulatus*.

	Diameter zones of inhibition at different concentration (mL of oil/mL of Tween 80)				
	1(r,b)	0.5(r,b)	0.25(r,b)	+Control	MIC (µg/mL) (r,b)
Gram positive bacteria					
Bacillus megaterium	6,10	4,9	1,8	15	0.2, 0.1
Bacillus cereus	-,9	-,5	-,4	13	12, 0.2
Streptococcus pyogenes	8,7	-,5	-,4	12	1.0, 0.2
Staphylococcus epidermidis	-,7	-,5	-,4	10	14, 0.2
Staphylococcus aureus	-,-	-,-	-,-	14	14,12
Gram negative bacteria					
Escherichia coli	-,15	-,14	-,12	30	8, 0.1
Proteus mirabilis	5,5	3,3	1,2	25	1.0,0.5
Klebsiella pneumonia	9,9	7,6	-,-	15	2.0, 2.0
Serratia marcescens	4,6	2,4	-,3	12	2.0,2.0
Pseudomonas aeruginosa	-,-	-,-	-,-	10	10, 12

r= red type, b= black type, + positive control, -No detectable activity

The comparison of the inhibitory actions of the two oils revealed that the organisms are more susceptible to the oil of black type than the oil of the red type. Four of the Gram-negative bacteria tested were sensitive to the oil of the black type as against three of the organisms that were sensitive to the oil of the red type. However, the results (Oladosu *et al.*, 2011) suggest that the essential oils of the red and black types of *C. articulatus* can serve as good cut for the formation of some antimicrobial drugs and also serve to confirm the use in traditional medicine.

Table 8: Determination of Minimum Inhibitory Concentration (MIC) of *Psorospermum corymbiferum* root bark extracts

Test Organism	Methanol	Chloroform	n-Hexane	Ethyl-acetate
	Concentration (µg/mL)			
P. aeruginosa	1.0×10^3	1.0×10^2	1.0×10^2	1.0×10^2
S. aureus	1.0×10^3	1.0×10^2	1.0×10^2	1.0×10^2
P. mirabilis	1.0×10^3	1.0×10^2	1.0×10^2	1.0×10^2
B. subtilis	1.0×10^3	1.0×10^2	1.0×10^2	1.0×10^2
C. albican	1.0×10^3	1.0×10^2	1.0×10^2	1.0×10^2
S. tyhpi	1.0×10^3	1.0×10^3	1.0×10^3	1.0×10^2
B. aureus	1.0×10^3	1.0×10^3	1.0×10^3	1.0×10^2

Pseudomona aeruginosa, Staphylococcus aureus, Proteus mirabilis, Bacillus subtilis, Candida albican, Salmonella typhi, Bacillus aereus.

The root bark extracts of *Psorospermum corymbiferum* showed good antimicrobial activities over the concentration range of 1.0 x 104 µg/mL to 1.0 x 101 µg/mL. These results (Zubair *et al.*, 2009) verified the usefulness of the plant for medicinal purposes as an antimicrobial agent. Its traditional use should therefore be upheld while scientific evaluation of its active principles should be given serious consideration.

Table 9: Anticandidal activities (MIC) results on the Root Bark of Psorospermum corymbiferum

Sample	MIC (µg/mL)
Candida albicans	20
C. stelloroides	15
Amphotericin B	0.39

The isolated compound (diosgenin) from the root bark of *Psorospermum corymbiferum* exhibited strong anticandidal activities, which supports its use in folk medicine (Zubair, 2011).

Table 10: Antimicrobial activities of the Fruit and Leaf oils of Taxodium distichum (MIC, µg/mL)

Sample	Bc	Sa	Ec	Pa	Ca	An
Fruit oil	156	156	312	625	312	19.5
Leaf oil	156	156	312	625	312	156
Gentamicin sulfate	1.22	0.61	2.44	1.22	Nt	Nt
Amphotericin	Nt	Nt	Nt	Nt	0.61	0.61

Bc= *Bacillus cereus* (ATCC no. 14579); Sa= *Staphylococcus aureus* (ATCC no. 29213); *Escherichia coli* (ATCC no.25922), Pa=*Pseudomonas aeruginosa* (ATCC no. 27853), Ca= *Candida albican* (ATCC no. 10231), An= *Aspergillus niger* (ATCC no. 16401); N=Not tested.

The essential oils of the fruit and leaf of *Taxodium distichum* exhibited weak activities against the tested microorganisms. The fruit oil showed a promising activity against *A. niger*, with a minimum inhibitory concentration

of 19.5 µg/mL (Ogunwande *et al.*, 2007). This is contrary to literature report of El Tantawy *et al.*, 1999, in which the fruit volatile oil of Egyptians grown *T. distichum* exhibited pronounced antibacterial activities against *P. aeruginosa* and *E. coli* and anticandidal effect against *C. albican*. This may be due to differences in the chemical composition of the Nigerian and Egyptian *T. distichum* oils.

Toxicity activities and Antitumor

Table 11: Cytotoxic potentials of *Taxodium distichum* oils

Sample	PC-3[b]	Hep G2[C]	Hs 578T[d]
Fruit oil	99.77 (0.23)	100	100
Leaf oil	97.58 (0.15)	95.19 (1.47)	0

a% kill at tested concentrations (standard deviations in parentheses).
bHuman prostate tumor cells (concentration tested 100µg/mL)
cHuman liver tumor cells (concentration tested 250µg/mL)
dHuman breast (ductal) tumor cells (concentration tested 250µg/mL)

The essential oils of the fruit and leaf of *Taxodium distichum* showed good cytotoxic effects towards the growth of human tumor cell lines PC-3, Hep G2, and Hs 578T, except for the leaf oil which showed no inhibitory effect toward Hs 578T cells (Ogunwande *et al.*, 2007). The results suggest that the oils offer new possibilities for cytotoxicity chemotherapy and also provide an insight into the medicinal importance of the plant growing in Nigeria.

Table 12: Cytotoxic potentials of the essential oils of *Eucalyptus torreliana* F

Sample	PC-3[b]	Hep G2[C]	Hs 578T[d]	MDA-MB-231[d]
Fruit oil	99.4 (0.1)	99.5 (0.5)	100	98.9 (1.4)
Leaf oil	98.5 (0.6)	87.9 (3.7)	100	94.6 (2.6)
α-pinene	0	95.0 (1.0)	5.5 (8.3)	24.1 (7.3)
1.8-cineole	0	7.6 (4.6)	0	0

* % kill at tested concentrations (standard deviations in parentheses).

a Human prostate tumor cells (concentration tested 100μg/mL)

b Human liver tumor cells (concentration tested 250μg/mL)

cHuman breast (ductal) tumor cells (concentration tested 250μg/mL)

dHuman breast (adenocarcinoma) tumor cells (concentration tested 100μg/mL)

The essential oils of *Eucalyptus torelliana* displayed strong activity against the four human tumor cell lines (Ogunwande *et al.*, 2005). It is suggested that synergy between several components of the oil or relatively potent minor components are responsible for the broad cytotoxicity observed. These results indicate the potential cytotoxic properties of *E. torelliana* essential oils and hence a hope in the near future to identify new cytotoxic compounds from these natural sources. The findings also provide support for some of the traditionally accepted medicinal uses of the plant in Southwest Nigeria.

Table 13: Brine Shrimp lethality results for the Rhizome Essential Oils of the Red and Black varieties of Cyperus articulatus

C. articulatus	Dosage (μg/mL)	Survivals 1	2	3	No of mortality	% mortality	LC_{50} (μg/mL)
Red	10	3	2	1	24	80	
	100	0	0	0	30	100	2.59
	1000	0	0	0	30	100	
Black	10	0	9	0	21	70	
	100	0	0	0	30	100	3.38
	1000	0	0	0	30	100	

For the red *C. articulatus*, none of the larvae survived at 1000 and 100 μg/mL, but six larvae survived at 10 μg/mL which account for 80% mortality rate. For the black *C. articulatus*, the cytotoxicity effects also follow the same manner, while nine of the larvae survived at 10 μg/mL of which amount to 70% mortality rate. Hence, the oil of the red variety has higher lethality than the oil of black variety has reflected in their LC50. This result (Ameen *et al.*, 2011) corroborates the folk medicinal uses of *C. articulatus*. The oils may possess potent antitumor compounds which are of importance in pharmaceutical industries.

Anti-inflammatory activities

Table 14: Anti-inflammatory activities of Psorospermum tenuifolium

Group	dose (mg)	no of animals in the group	change in ear weight (Mean ± SEM) mg	% edema reduction
Control	-	8	7.8±0.3	-
Indomethacin	0.25	8	3.3±0.1	57.7
PT1	5.0	8	0.6±0.3	92.3
PT2	2.5	8	1.8±0.5	76.9
PT3	1.25	8	4.0±0.5	48.7
PT4	0.075	8	6.6±0.3	15.4

P<0.05, student's t-test, PT= *Psorospermum tenuifolium*

The leaf essential oil of *Psorospermum tenuifolium* at 5.0 and 2.5 mg dose levels displayed a significant anti-inflammatory activity with a percentage oedema reduction of 92.3 and 76.9, respectively. The oil at these concentrations showed very potent activity in reducing oedema. Hence, the results confirm the folk medicinal uses of the plant (Zubair *et al.*, 2009).

Antioxidant activities

Table 15: IC50 of the free radical scavenging activities (DPPH) Antioxidant Properties of the Seed Essential Oils of *Eucalyptus* species and Reference Compound.

Essential Oils and Reference compound	DPPH IC_{50} (μgmL^{-1})
Eucalyptus tereticornis	2.00
Eucalyptus cloeziana	2.80
Eucalyptus maculata	5.00
Eucalyptus torelliana	4.00
Eucalyptus camaldulensis var. petford	3.50
Eucalyptus camaldulensis var. nansee	3.50
Asccorbic acid	9.00

Data are presented as the mean value ± S.D. of triplicate IC50 value is the concentration at which 50% of the DPPH is reduced. The lower the IC50 value the higher the antioxidant potential. The *Eucalyptus* oils displayed stronger free radical scavenging activities compared to the reference compound, ascorbic acid used, with *E. tereticonis* being the most antioxidant Ololade *et al.*, 2013; Ololade and Olawore, 2013, 2017; Olawore and Ololade, 2017).

Table 16: EC_{50} of the FRAP Antioxidant Properties of the Seed Essential Oils of *Eucalyptus* species and Reference drug

Essential Oils and Reference compound	FRAP EC_{50} (μgmL^{-1})
Eucalyptus tereticornis	5.00
Eucalyptus cloeziana	1.50
Eucalyptus maculata	10.00
Eucalyptus torelliana	7.50
Eucalyptus camaldulensis var. petford	5.00
Eucalyptus camaldulensis var. nansee	5.00
Asccorbic acid	11.00

Data are presented as the mean value ± S.D. of triplicate

The seed essential oils of *Eucalyptus* species significantly have higher antioxidant activities compared to the ascorbic acid. Therefore, the oils could be used as sources of antioxidant agents (Ololade and Olawore, 2017).

Isolation and characterization of phytochemicals

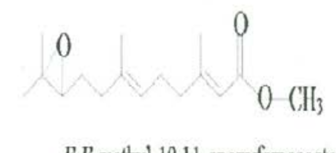
We carried out isolation and characterization of some compounds from the essential oils or extracts of some of the plants studied.

Hyparrhenia rufa

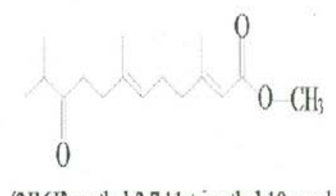
The essential oil obtained by hydrodistillation was fractionated by preparative gas chromatography. This yielded two fractions. Single compounds were obtained from the fractions by using column GC. The compounds

separated out as oils.

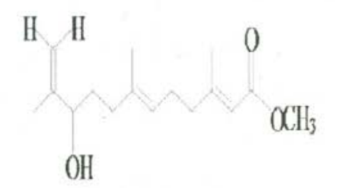
The structures of the four isolated compounds were established by comparison of their mass spectra and NMR data with those reported previously in the literature (Liedtke and Djerrasi, 1972; Anderson *et al.*, 1972).



E,E-methyl-10,11-epoxyfarnesoate



(2*E*,6*E*)-methyl-3,7,11-trimethyl-10-oxododecadienoate



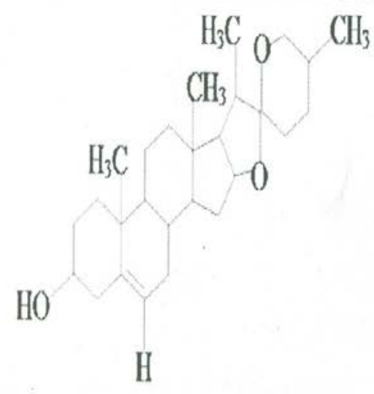
(2*E*,6*E*)-methyl-10-hydroxy-3,7,11-trimethyl-2,6,11-dodecatrienoate

These esters were isolated from *H. rufa* for the first time. They are homologues of juvenile hormones of the giant silkworm moth *Hyalophora cecropia*.

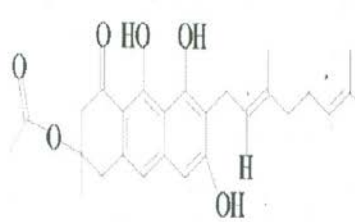
Psorospermum corymbiferum (Hochr) Clusiaceae (Guttiferae)

Our previous bioassay on this plant showed that it is an antibacterial agent (Zubair *et al.*, 2009). The ethanol extract of the plant was macerated with n-hexane, chloroform, ethyl acetate and methanol successively to give respective solvent extracts. Open column chromatography of the ethyl acetate extract on silica gel with cyclohexane/ethylacetate (1:1) as eluents gave acetylvismione F, prenylated bianthrone and 1,8-dihydroxyanthraquinone and a fraction, which was further rechromatographed to give a single spot (Zubair *et al.*, 2011).

Infrared (IR), ^{1}H and ^{13}C NMR data were identical to values reported in the literature (Han *et al.*, 1999) for the known aglycone, diosgenin. The steroidal compound was characterized for the first time from *P. corymbiferum*.



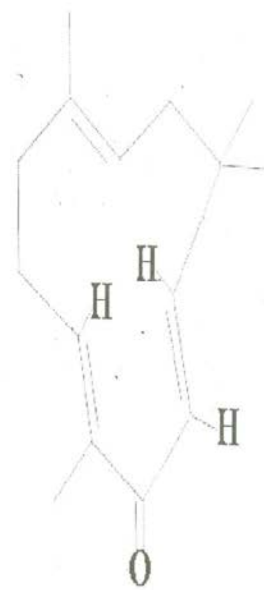
Diosgenin



Acetylvismione F

Xylopia aethiopica

The method of isolation of the single pure component was similar to that used for *H. rufa* i.e fractionation by preparative GC followed by column GC to obtain the single component. ^{1}H and ^{13}C NMR were used to determine the structure (Ogunwande *et al.*, 2005).



Zerumbone

$C_{15}H_{22}O$

Zerumbone was being reported as a constituent of *X. aethiopica* for the first time. The compound has been shown to possess considerable anti-HIV and cytotoxic properties (Dai *et al.*, 1997).

The plant therefore has potential to be used for treating HIV.

CONCLUSION

We have been able to detect an array of phytochemicals, in the plants investigated. Many of these are being reported as constituents of the plants for the first time.

Our results have shown that a considerable number of aromatic plants in Nigeria have very good prospects for use in the perfumery, pharmaceutical and flavouring industries. *Eucalyptus camaldulensis* var. petford is particularly good for use as a medicinal plant. *Hyparrhenia rufa* is recommended as a very cheap raw material in the perfumery industry.

Our biological activity studies have confirmed the uses of these aromatic plants in folk medicine and some of these can prove useful as antioxidants against degenerative diseases such as hypertension, cancer, skin diseases and diabetes.

We have been able to show that the isolated compound, diosgenin possess anticandidal activity and could be used as an antifungal drug.

RECOMMENDATIONS

There is need to provide researchers, the necessary tools to work with here in Nigeria. The traffic used to be both ways when I was an undergraduate at Ibadan. Lecturers and students came from Europe and America on sabbatical leave or to study at the University of Ibadan because there was substantial provision of

tools to work with, and this was one of the reasons why the University of Ibadan was one of the best ten (10) universities in the world at the time. Today, the traffic is only one way, that is, from Nigeria to the rest of the world. No doubt there is need for cross fertilization of ideas among universities in the world. Research should be properly funded since it is the basis for development.

There is need for proper preservation of these plants. Most of the *Eucalyptus* plants species are gone, even in Forestry Research Institute of Nigeria (FRIN), and I hope attempts are being made to plant new ones to replace those that have disappeared.

Callistris columellaris plant species at the secretariat, Ibadan, should be properly taken care of. Some of the *Eucalyptus* species at Agodi gardens have also disappeared. Now that Oyo State has renovated Agodi gardens, effort should be made to replace those that are no longer there and also plant new ones. The story is the same in the North at Afaka near Kaduna and Jos. The botanical garden in this university should continuously plant new species for research purposes.

GRATITUDE

Mr. Vice Chancellor Sir, I was working on the C-13 Nuclear Magnetic Resonance of fluoro organic compounds for the Ph.D degree at the University of Manchester. My desire, when I returned to Nigeria, was to continue in that area. However, there was no functional

Fourier transformed (NMR) spectrometer in this country for me to use. This was still the situation when I resumed work in LAUTECH in 1991. My first natural option at that time was to seek for collaborative research work with my colleagues at the University of Ibadan. I met separately with Profs. Ekundayo and Adeleke, the latter, a former Vice Chancellor of this university, who also offered to discuss with Prof. Ekundayo. He did so and Ladies and gentlemen, this was the beginning of the research efforts I have been narrating to you today. Moreover, these studies culminated in my being awarded the best researcher of the year, by the National Universities Commission, in 2005. For these I am grateful to God, Prof. Ekundayo, Prof. Adeleke and my other collaborators.

I am first and foremost grateful to Almighty Allah, the most merciful and the most beneficent. I give all praises, thanks and adoration to Allah who is the architect of all my achievements since childhood.

I must also mention my late parents- Mr and Alhaja Shittu Olawore. They took very good care of us and emphasized the importance of western and Koranic education to us right from childhood. I will always cherish their memories. May Allah make Aljanah their everlasting abode, Amin.

I am grateful to the University for giving me the opportunity to serve and contribute my humble quota to the growth of this University. I also thank the Head of Department of Pure and Applied Chemistry, Prof. (Mrs).

Onawumi and my other colleagues in the Department for making the environment conducive for work. Please sir, allow me at this juncture, to mention the names of Profs. Olajire, Ayodele and Adeoye, as the remaining three foundation academic staff members (apart from myself), who are still in the Department. I thank the Dean of the Faculty of Pure and Applied Sciences, Prof Oni and all other academic, administrative and laboratory staff of the faculty for their assistance and comradeship at all times. I thank the Vice Chancellor and other Principal Officers of the University. The Vice-Chancellor, Prof. Gbadegesin, was introduced to me shortly before assuming office and since then, I have been particularly struck by his humility. I pray God to continue to give him more blessings. May I also remark that I have worked closely with most of the other Vice-Chancellors of this University, even before they were appointed into office. They are Profs. Salawu, Raji, Adeleke and Nassir. I appreciate them all. I pray that God let the souls of late Profs. Salawu and Raji rest in peace, Amen.

My two younger sisters are here Alhajas Olaitan and Kareem. They have always been my pillar of support. I thank both of them for everything. Allah will continue to support you and your families too.

I also must mention my benefactor, Late Dr Adetunji, former Registrar of the then University of Ife and my God father, Dr. Saka Balogun. They played very vital roles in my life at one time or the other. My supervisors at the University of Ibadan, Profs Adesogan, Okogun and late Prof. Ekong and at Manchester, Dr Fields, are all worthy of my gratitude.

My research collaborators are worthy of mention. They are Prof. Ekundayo, Drs Ogunwande, Kasali, Oladosu, Lamidi, Ololade and Dr. (Mrs) Zubair. Also included are Prof (Mrs) Bello, Mrs. Ogunjinmi, and a host of my former M.Sc and B.Sc students, particularly Dr. (Mrs) Ibrahim, Prof. Amuda, Odewande and my colleague and son, Mr. Aliyu Akeem.

Now my immediate family - my children- Engrs. Jaleel and Sulaiman, Dr. (Mrs) Alatishe, Engrs. Taofeek and Kabir and my baby, Rukayat. I must acknowledge their spouses too, Dr Alatishe, Nafisat and Salama. Then my wife, Alhaja Adijat Olawore, I appreciate your love and concern for me at all times. I thank you for your support especially during difficult times. You are really a wife in a million. I can't stop loving you.

I must also appreciate the presence of my friends and colleagues, Profs. Fakunle, Ibiyemi, Adediran, Adesomoju, Oderinde, Odebunmi and Adekola. Mr. Ajuwon, Mr. Sola James and many others that space does not allow me to mention. I also want to thank my colleagues in other Universities like Fountain, LASU, Unilag, KWASU, Unilorin and those at Oyo State Ministries of Education and Industry, Science and Technology.

Thank you ladies and gentlemen for your presence here today. I wish you safe journey to your destinations.

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