NATURE'S GIFTS: ESSENCES, OILS AND PLANTS

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by

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Mr. Vice Chancellor, Deputy Vice Chancellor (Administration), Deputy Vice Chancellor (Academic), Provost, College of Medicine, Registrar, Librarian, Dean of the Faculty of Science, Deans of other Faculties and Postgraduate School, Distinguished ladies and gentle men.

Introduction

I count myself fortunate and priviledged to be asked to deliver this particular inaugural lecture on behalf of the Faculty of Science. It is very significant because the University, the Faculty of Science and the Department of Chemistry are celebrating their 60th Anniversary and this lecture is taking place on the 20th Anniversary of my appointment to the Chair of Organic Chemistry!!!.

The Faculty of Science, one of the three foundation faculties of the then University College, Ibadan, commenced academic activities at the old 56th Army General Hospital, Eleyele, in January 1948 with 37 students out of 104 fresh men and women. The Faculty has contributed immensely to the development of science and science related disciplines in Nigeria. The Faculty continues to maintain world acclaimed highest standards of scholarship in all its academic endeavours inspite of the financial and institutional crises of recent years. We must pay tribute to the thirty-one deans who have provided leadership in the Faculty since 1948. A number of these deans have been appointed Vice-Chancellors in the UK, West Indies and Nigeria. More than 12 retired and serving professors in the Faculty have been elected to the Fellowship of the Nigerian Academy of Science—a 103-member apex honour body in science and science based disciplines in Nigeria.

Chemistry, a foundation department in the Faculty was at a time at "par with the best in the world" and it is still associated with excellent and distinguished achievements over the years. This inaugural lecture is the eighth from the Department and is titled: "Nature's Gifts: Essences, Oils and Plants". It is my intention to offer a *tour d'horizon* of my research in the area of the science and chemistry of essential oils from Nigerian medicinal and economic plants. The lecture is the fifth from the Natural Products Chemistry group which has been engaged in the study of secondary metabolites from Nigerian medicinal plants since 1952. Professor R.J.M. McIloroy (the first Head of Department) and Professor C.W.L. Bevan (HOD: 1953-1966) pioneered extensive research into Nigerian mahogany trees, gums and mucilages.

Medicinal Plants

The strong bond between mankind and plants predates history. Plants are being used as sources of life sustenance in form of food, energy, medicine and shelter. It is well known that there is generally a plant for every human need, ranging from special diets to religious and sometimes magical ceremonies. The most striking and important use of plants is in the treatment of diseases. There is a great wealth of remedies from nature which the early men recognized in their primary preoccupation while gathering their daily food. Medication by plants or herbs was recognized as the only path against diseases by the Egyptians (3000BC), Greeks (400BC) and Romans (37BC). The Arab physicians in the middle ages relied mainly on plants for therapy.

Prior to the advent of systematic and scientific studies of medicinal plants along with their curative properties, the utilization of the plants was entirely empirical as illustrated by the so called "Law of Signatures". It was believed that nature had assigned a plant for the cure of every ailment and also that an obvious sign existed for which disease a plant or part of it could be used. For example, the walnut which has the shape of the brain was taken to be a good cure for diseases of the brain!!!.

Early traditional medical practice in West Africa involved the use of both mysticism and plants or parts thereof. Sometimes, the medicines may also comprise of parts or whole animals and other unrelated ingredients. However, plants still remained the critical source of local traditional drugs.

The documentation of the medicinal use of West African plants was started in the 1930s by Dr. J.M. Daziel, a Botanist and Naturalist at Royal Kew Gardens, London. In 1937, he published the *magnus opus* on West African medicinal plants titled "The Useful Plants of West Tropical Africa". The scholarly work was an important contribution to our knowledge of traditional uses of plants in the Nigerian ethnomedical practice. Dalziel's book has since been revised, reorganized and updated into a five-volume edition by H.M. Burkill in collaboration with Royal Kew Gardens.

The upsurge in the interests in medicinal plants and herbal drugs has led to a phenomenal increase in the study of these plants and their associated curative properties. There is therefore the need for more basic scientific research into this vast field with the emergence of new disciplines such as ethnobotany, ethnomedicine and ethnopharmacology. Basically, the plant cell is an incredible manufacturing crucible of a diverse array of chemical compounds. These natural products are the physiologically active substances which exert the curative properties of the plants. Natural products are also being used as flavours in food, fragrances, natural pigments, sweetners, pharmaceuticals and pesticides.

Numerous Nigerian plants have now been investigated. Their natural products have been isolated and assayed for biological activities. The ethnomedical folklores were systematically investigated and facts were separated from myth and legends. Shellard (1979) reasoned that "the significance of research into medicinal plants is that it extends our knowledge of their botany, their constraints, their pharmacological activity and their therapeutic value, so that they can be used more effectively in the treatment and prevention of disease".

The main objective of our research has been to conduct the in-depth investigation of these medicinal plants as well as economic plants in the Nigerian bio-diversified flora, for their essential oils. There is no strict dividing line between medicinal plants and economic plants per se, because some medicinal plants may prove to be economically important e.g. *Artemisia annua* used for the production of the natural product artemisin—the drug of choice in the modern treatment of malaria. A few economic plants such as *Eucalyptus* species could be medicinally useful in the treatment of certain diseases.

Essential Oils

Essential oils are volatile components which are extracted or distilled from odorous plant materials. The process for the procurement of these oils must be such that the nature and composition are preserved as much as possible. These procedures include distillation and mechanical expressions. The technique of distillation has been used as early as 3000BC in Persia and 2000BC in China to produce "aromatic oils". Brunschwig, a Strasbourgh physician described the manufacture of essential oils in his 1500 Book titled *Distillation Book*. It was not until 1826 that steam distillation was formulated in Germany to manufacture 'pure oils' which were used in flavouring foods.

The oil is basically 'essential' because of its unique odour or scent and it is regarded as the 'essence' of the plant. Essential oils could be isolated from whole plants or parts thereof e.g. flowers, leaves, wood, root, seeds, fruits, rhizomes and peels. The methodologies of isolating essential oils from plants have undergone tremendous refinements over the years and these processes are listed below.

Methods of Essential Oil Preparation

1. *Distillation*
    (a) Steam distillation
        - atmospheric pressure
        - reduced pressure
    (b) Hydrodistillation
    (c) Hydrodiffusion
    (d) Flash distillation
        - atmospheric pressure
        - reduced pressure
        - combination
    (e) Vacuum distillation
        - vacuum degassing
        - fractional distillation
        - high vacuum distillation
        - molecular distillation
    (f) Carbon dioxide distillation

2. *Extraction*
    (f) Solvent extraction
    (g) Supercritical inert gas extraction

3. *Combined Methods*

- simultaneous distillation and extraction (SDE)
- simultaneous distillation adsorption (SDA)

4. *Head Space Extraction*

- dynamic e.g. closed loop stripping
- static

5. *Miscellaneous Methods*

- freeze concentration
- expression—mechanical cold press—citrus peels
- enfleurage—flowers
- zone melting

The most widely used method is steam distillation which simply entails passing steam through the crushed plant material in boiling water. The cooled emerging steam and oil are easily separated. A variant of the steam distillation procedure requires that the plant material be boiled in water but without the involvement of external steam. This procedure is called hydrodistillation. Hydrodiffusion is a relatively new form of distillation in which the steam spray is passed through the plant material from above. The steam percolates by gravity. The essential oil and water are collected as steam distillate and hydrodistillate. Generally, the steam releases the essential oils embedded in the special oil glands in plants.

These steam driven extractions are widely used in the industries—(steam distillation and hydrodiffusion) and research laboratories (hydrodistillation). In fact, the International Organization for Standardization in its ISO 9235 definition proclaimed that "Essential Oils are obtained from vegetable raw material by distillation with water or steam" (ISO 1997).

Extraction methods with solvents are utilized to a lesser extent due to the fact that numerous other substances usually co-extract with the essential oils. However, the advantage of this technique is the favourable distillation coefficient of volatiles in the solvents. Solvents could range from ether to pentane and liquid carbon dioxide. Chromatographic methods are principally used to clean up the extracts in order to improve the purity of the essential oils.

Combined steam distillation with extraction was developed in the Brewery industries by Linkens and Nickerson (1964) to improve on the efficiency of the production of essential oils in simultaneous distillation extraction (SDE). The advantages of this method are that the volatiles could be obtained in high concentrations and thermal degradation is mostly minimized.

The liquid-liquid extraction method is also suitable for cleaning processes where many water constituents are co-extracted with the volatile oil. A plethora of distillation methods which include flash distillation (at atmospheric and reduced pressure) have also been used to extract essential oils from edible fruits and plants. High vacuum sublimation entails a variant of the distillation process to isolate volatile oils. Vacuum distillation has been utilized to procure high purity oils for sensory studies. The volatile oils are recovered and concentrated without the intervention of heat which could affect the sensitive aromas from typical tropical fruits.

The head space (HS) technique is employed in the collection of plant volatiles in a closed vapour space above the plant material, particularly flowers. The vapour phase presumably contains the volatile compounds which are responsible for the flower odour. The head space vapour has the disadvantage of containing components of high volatility whereas the substances of low vapour pressure remain uncaptured. The volatiles are normally collected by absorption into liquid traps or adsorption by solid adsorbents such as alumina, charcoal, silica gel and porous polymers. The oils are recovered from the absorbents by solvent extraction or simultaneous distillation extraction (SDE). The advantage of the head space (HS) technique is the propensity to form artefacts and additional volatile impurities formation are reduced considerably.

The miscellaneous procedures for preparing essential oils are rather restrictive in their applications and these methods are used when others have proved unsatisfactory. Sometimes aqueous solutions remain a handy solvent in the extraction of essential oils from plants. The water content of the extracts are removed by freeze concentration or lyophilisation. The yields of nature identical volatile oils are often good.

Zone melting is anchored on the principle that a solvent freezes slowly in pure solid form and the equilibrium conditions tend to exclude impurities. Cyclohexane and other aromatic hydrocarbons are usually the solvents of choice. Expression method in which the essential oil is squeezed out of the oil glands in the plant materials, is reserved exclusively for citrus fruits peels due to large quantities of oils in them. The oil is mechanically expressed by a cold press. The nature of the expressed oils is nearly identical with that in the peels oil glands.

The process of enfleurage is useful in the extraction of volatile oils from delicate flowers. Glass plates coated with a layer of purified fat are used in this method. The flowers are placed between the glass plates for a day after which the flowers are removed and replaced with fresh ones. This process is repeated daily for over two months until the fat is saturated with the essential oils from the flowers.

The methods or procedures which are employed to isolate essential oils from plants determine the composition of the oils. It is unlikely that two different isolation methods would produce volatile oils of identical composition for a specific plant or its organs. The drawbacks and advantages of an isolation method over the others are dependent on the objectives of the study of essential oil composition. However changes leading to the formation of new compounds which did not exist in the original plant is not a desirable phenomenon.

Supercritical carbon dioxide is increasingly being used in research laboratories and industrial settings to extract essential oils from plant materials. The advantage of this procedure is that the carbon dioxide ultimately reverts to gas and the petrochemical residues which originate from normal organic solvents are avoided. The lower temperature of extraction prevents the decomposition or alteration of the essential oil constituents and the product is nature identical, as much as possible.

Industrial Uses of Essential Oils

Essential oils are unique and widely known for their scents, flavours and medicinal properties. Consequently, the uses are multifaceted. Two major basic applications could be recognized:

(a) industrial – as flavour and fragrance material in perfumery;

(b) medicinal – as expectorants, home remedies, conventional and alternative medicines.

Industrially, essential oils remain the basic raw material for perfumers and flavourists. The odourant attributes of their products are solely due to essential oils. Perfumery compositions for fine fragrances, toiletries and house products are of essential oil origins. The widespread industrial utilization has led to a growing field of research and rapid advances in the science of odour.

The human nose is the most sensitive organ to 'odour perception' which has developed into a science in its own right in the highly lucrative perfumery field. Fascination with the sense of smell is now the basis of numerous studies. Findings have shown that the description of sensory properties remain incomplete due to the fact that odour perception is an individual experience and language may not be exact in describing a particular odour. In the same way as "beauty is in the eye of the beholder", odour has become more a matter of choice and preference. A Wigan teacher in the UK was reported to have sent a boy home because of his smell. The boy's mother sent the teacher a note: "Dear Miss, our Johnny smells the same as his Dad and his Dad smells lovely. I should know, I've slept with him for 25 years. The trouble with you Miss is that you are an old maid and don't know what a proper man smells like" (Van Toller and Dodd 1988).

Essential oils are quite adept and skillful in masking malodours as well as imparting their own nice scents on man's surroundings and environment. Numerous cosmetics in the market today owe their odour to delicate amounts of essential oils and other ingredients which promote the sense of attractiveness.

A number of essential oils impact on food their typical and special flavour which have always been appreciated and sought after. In addition to food and food products, volatile oils are used in flavouring beverages, pharmaceuticals and domestic toiletries. Minor utilities of essential oils include, the use as stabilizers in bottled and canned food due to their preservative and antioxidant properties.

Cough and cold remedies often contain essential oils as key ingredients on account of their antiseptic, rubefacient and carminative activities. Some essential oils have shown promise as both chemopreventive and chemotherapeutic agents for certain solid tumors (Morse and Toburen 1996).

Besides conventional medicine, essential oils have found application in alternative medicine leading to the concept of 'Aromatherapy' and 'Aromachology'. Aromatherapy is the use of essential oils from plants for holistic and curative purposes. It is as much an art as it is a science because identified specific essential oils have healing properties on account of their appealing aromas. The phenomenon of aromatherapy has been known for ages as the healing effect of essential oils. Aromatherapy attempts to organize these curative properties and the particular essential oils into a systematic science. Aromatherapy is administered by inhalation or topical application on the surface of the body and is not usually by taking by mouth. Clinical studies have shown improvements in the symptoms of nausea or pain as well as lower blood pressure, and improved pulse and respiratory rates on administration of aromatherapy.

Aromachology on the other hand is a "science that is dedicated to the study of interrelationship between psychology and fragrant attributes to elicit a variety of specific feeling and emotions" (Anon 1992). There is growing evidence that essential oils, through their odour effects, have reduced patient stress during surgical operations, improved performance at the work place and promoted general feelings of well-being. Odour stimuli also affect mood and anxiety. There is no fundamental difference between the effects of single essential oils and single aroma chemicals (natural or synthetic) and blends of natural or synthetic materials.

World Trade in Essential Oils

Due to the extensive industrial uses of Essential oils, the size of its world-wide trade is quite enormous – about US14 billion, annually (Table 1).

Table 1: World Trade in Essential Oils, Perfumes and Flavours

	1986	1990	1994	1998	1986-1998 % pa
Exports*	2149	4122	5052	7439	10.9
Imports*	2008	4206	4802	6811	10.7

*Figures in US$millions.
Source: United Nations International Trade Year Book 1999 and 1992.

The major exporters of essential oils are the United States (US), European Union (EU) and a handful of developing countries such as China, India, Brazil and Indonesia, with EU countries being responsible for 52% of the total. The principal importers are the US and EU countries (55%). Nigeria is a very small player in the league of importers (Table 2).

At an annual growth rate of 6.1 percent the worldwide sales of essential oils would translate to a projected value of $20 billion in 2008. This is an enormous trade volume and there is still an increasing demand for essential oils, perfumes and flavourants. This demand is driven by food flavouring, cosmetic, and fragrance industries. Consumer preference for natural over synthetic substances is another factor which is responsible for the increasing worldwide demand.

Noteworthy is the fact that Nigeria does not export any essential oil whatsoever for the simple reason that there is no essential oil industry in the country. There is also no definite trend in the import of essential oils into Nigeria. It is particularly difficult to gauge the economic factors which drive these fluctuating importation trends. It would be interesting to commence research into the economics of essential oil importation in Nigeria.

Table 2: Import of Essential Oils, Resinoids, etc. into Nigeria

Year	1975	1978	1983	1984	1985	1986	1987	1988	1989	1990	1991	1993	1995
Amount*	2.7	2.8	4.07	2.66	0.8	4.67	0.8	34.2	7.7	59.3	21.2	18.3	7.21

*Figures in millions of Naira
Source: Nigerian National Bureau of Statistics (2008)

Natural Functions of Essential Oils

Essential oils do not seem to play any role in the metabolic processes of the plant growth and development. However, it is now obvious that essential oils are important participants in the chemical ecology of the producing plants, as defences against pathogens and herbivores, attractants for pollinators, feed deterrents and alleopathic agents. The variations in the composition of essential oils have been implicated in tritrophic interactions of the plants with the environment. Certain insects have developed the capacity to detoxify essential oils and in some cases formed allomones from them.

Chemical Composition of Essential Oils

The nature of the constituents of essential oils is a vital determinant of the use to which the oil is applied industrially and medicinally. Generally, essential oils are complex mixtures of organic compounds of all chemical classes. The dominant constituents are generally terpenoid in nature—mostly mono- and sesquiterpenoids which are typically hydrocarbons, alcohols, esters, aldehydes, ketones, lactones and carboxylic acids. A number of aromatic compounds have also been detected in essential oils. The constituents usually occur in different concentrations which make each oil unique in its composition and applications.

Analyses of Essential Oils

The centerpiece of the chemistry of essential oils is the capacity and ability to isolate and identify individual components. The method of choice for this is chromatography in which the components are separated on the basis of their volatilities and ability to move through a stationary phase at different speeds. Analyses have been excellently achieved nowadays with high resolution gas chromatography (GC) and more recently gas chromatography (GC) coupled to a mass spectrometer (GC-MS). The analyses are normally fast, sensitive, accurate and reliable. Extremely minor or trace components (10^{-9} g range) could be detected and identified. Oftentimes, a gas chromatograph coupled to a sniffing port would facilitate the identification of trace components with strong odour threshold in GC-O analyses.

Essential Oils of Nigerian Medicinal and Economic Plants

Over 350,000 species of plants are known to exist throughout the world and less than 5% of those plants could be found in West Africa. Dalziel (1936) collated botanical data, economic uses and vernacular names of 2,565 medicinal plants which belong to 1,016 genera spread over 151 plant families. In the recent revised edition of Burkill (1985, 1994, 1995), the number of plants increased threefold to 7,349 plants. Oliver (1960) identified 59 plants which could yield essential oils in the Nigerian flora. Another 52 species appeared in the second monogram on "Nigerian Medicinal Plants" (Oliver 1960b). However, only 36 plants were itemized in both publications. The implication is that about 75 Nigerian plants have the potential of producing essential oils. In view of our research, the list appears restrictive.

Prior to the commencement of our investigations, information on the chemical compositional data of Nigerian essential oils was scanty. The oils were analysed semi-quantitatively using methods which are not acceptable by today's standards. Talaj's pioneering work on West African essential oils was carried out on 18 Ghanaian plants which belong to 12 families (Ekundayo 1986). Our studies of Nigerian essential oil bearing plants have covered 101 plants distributed over 30 families (Table 3). The plants were collected from different areas of Nigeria ranging from Akwa Ibom to Kaduna States, Jos and Ibadan.

Table 3: List of Nigerian Essential Oil-bearing Plant Families in Current Studies

Plant family	Number of plants	Plant family	Number of plants
Alleaceae	1	Graminaceae	1
Anarcadiaceae	8	Labiatae	5
Apiaceae	1	Lythraceae	1
Apocynaceae	1	Meliaceae	2
Asclepiadeae	1	Menispermaceae	1
Asteraceae	5	Moraceae	4
Burseraceae	2	Myrtaceae	18
Caesalpiniaceae	1	Pinaceae	10
Casuarinaceae	1	Piperaceae	1
Chenopodiaceae	1	Poaceae	1
Combretaceae	1	Rubiaceae	2
Cupressaceae	2	Rutaceae	8
Cyperaceae	2	Solanaceae	3
Euphorbiaceae	2	Verbenaceae	5
Fabaceae	4	Zingiberaceae	4

The highest number of plants in our studies belong to the Myrtaceae family with the fourteen (14) *Eucalyptus* species dominating. An interesting Myrtaceae plant on our list is guava fruit (*Psidium guajava).* 10 *Pinus* species were also investigated in the current work. The table includes five *Citrus* species and twenty two of their cultivars. The other plants in our research were widespread in terms of families.

Essential oils were isolated from whole plants and parts thereof in our laboratories by hydrodistillation. The yields of oils were generally low – less than 0.5%. However, yields were comparatively higher in rhizomes of ginger (*Zingiber officianle*) – 1.8%; tumeric (*Curcuma longa*) – 1.33% and sedge nut (*Cyperus tuberosus*) – 0.8%. Majority of the oils possess pleasant organoleptic properties with essentially woody, herbaceous, spicy and aromatic notes.

The essential oils were then analysed comprehensively using gas chromatography (GC) and gas chromatography – mass spectrometry (GC-MS). An analysis was considered satisfactory if more than 80% of the constituents were identified. New compounds were routinely isolated from the oils by means of preparative GC and the structures of these compounds were characterized by spectroscopic methods namely mass spectrometry (MS), nuclear magnetic resonance (NMR), ultraviolet (UV) and infra red spectroscopy (IR).

The resulting analytical data showed that the Nigerian essential oil could be delineated into two groups:

1. Economically useful oils
2. Medicinally valuable oils

Essential Oils of Nigerian Economic Plants

The economically useful oils were collected from plants belonging to three families:

(a) *Pinus* species (Pinaceae)
(b) *Eucalyptus* species (Myrtaceae)
(c) *Citrus* species (Rutaceae)

***Pinus* Species**

The needle (leaf) essential oils from ten *Pinus* species growing in Forest Research Institute of Nigeria (FRIN) trial plantation in Ibadan were investigated. *Pinus caribaea, P. massoniana, P. merkussii* and *P. elliotti* volatile needle oils contained predominantly α- and β-pinene whilst in *P. pseudostrobus, P. cubensis, P. insularis, P. oocarpa* and *P. kesiya,* the major constituents were borneol, bornyl acetate and α-terpineol (Ekundayo 1978, Ekundayo and Akintewe 1983). The *Pinus* species are therefore potential sources of marketable turpentine-like oils whose dominant components are starting materials in the industrial synthesis of other useful chemicals (Fig. 1). The principal components in the Nigerian *Pinus* species essential oils are α and β-pinene which are widely used as feed storks in the industrial syntheses of insecticides, disinfectants flavour and fragrance materials. Chemically, α-pinene is easily converted into myrcene from which citral and vitamins A and E could be produced.

The Nigerian *Pinus* species were planted primarily for timber and pulp purposes even though the plants are not indigenous to Nigeria. In case of commercial exploitation, the needle oils would be secondary products because the timber qualities of the Pines constitute the primary objectives. In fact, the needles under the exploitative circumstances are waste products from which marketable essential oil or pine oil could be extracted.

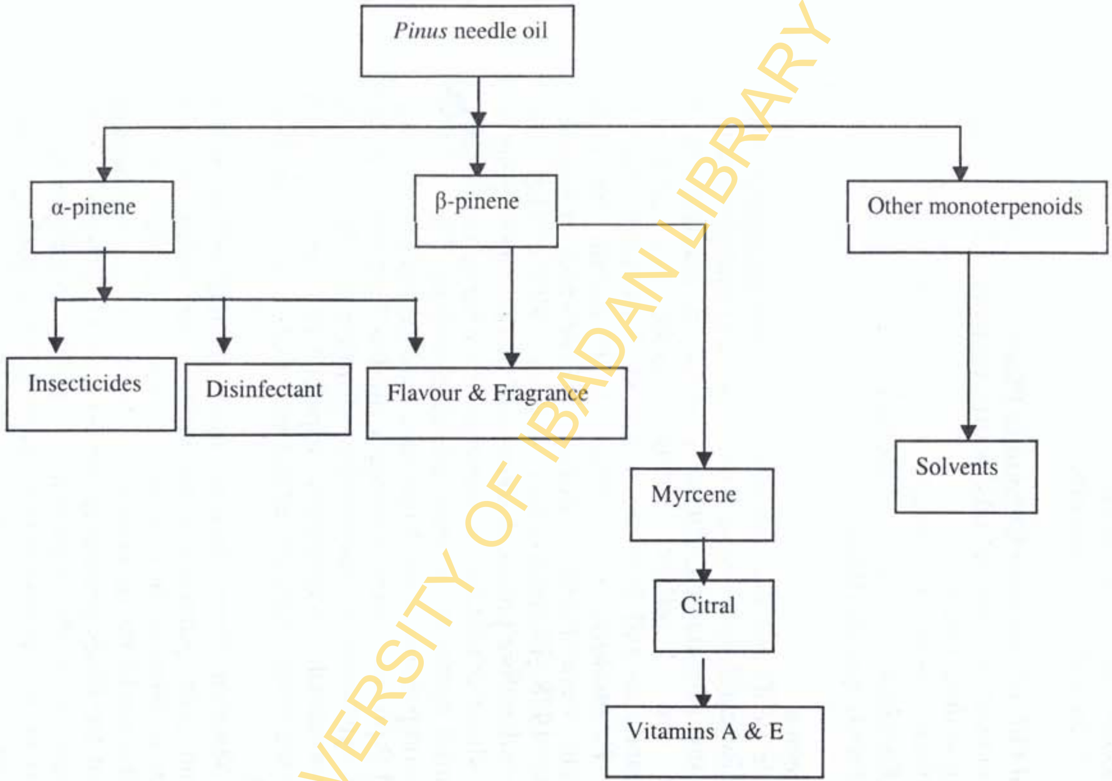


Fig. 1: Pontential industrial conversion of pine needle oils.

Eucalyptus Species

Leaf volatile oils were extracted from fourteen *Eucalyptus* species growing in FRIN trial plantations in Ataka, Kaduna State, Forest Station, Jos and Agodi Gardens, Ibadan.

In commerce, three types of *Eucalyptus* oils are recognized:

(a) Oils of high 1,8-cineole content;
(b) Oils of high citronellal content;
(c) Oils of high phellandrene or piperitone content.

Type 'a' is medicinal *Eucalyptus* oil, whilst the types 'b' and 'c' are sometimes called perfumery *Eucalyptus oil,* on account of their citrus-like odour.

The results of the analysis of the oils are displayed in table 4.

Table 4: Classification of the Essential Oils of Nigerian grown *Eucalyptus* Species

Type 'a' (Cineole Group)	Type 'b' (Citronellal Group)	Type 'c' (Phellandrene/ Piperitone Group)	Others
1. *E. propinqua* – 61.8% 2. *E. camadulensis* var *nasee* – 72.2% 3. *E. camadulensis* var *myrore* – 70.4% 4. *E. camadulensis* var *petford* – 45.6% 5. *E. globulos* – 45.9% 6. *E. camadulensis* var *catharina* – 32.8%	1. *E. tesselerasis* – 37.3%	Nil	1. *E. cloezina* α-pinene (67.2%) 2. *E. saligna* α-thujene (63.8%) – Ibadan 3. *E. saligna* p-cymene (20.3%) – Kaduna 4. *E. alba* α-thujene (32.9%) 5. *E. maculate* car-3-ene (10.5%) 6. *E. paniculata* p-cymene (24.4%) 7. *E. decaseana* limonene (28.8%) 8. *E. robusta* α-pinene (22.1%) 9. *E. deglupta* α-pinene (24.7%)

Sources: Oyedeji *et al* 1999; Oyedeji *et al* 2000; Ogunwande *et al* 2005; Olawore *et al* 1999; Ogunwande *et al*; 2003.

One *Eucalyptus* species could be classified as belonging to the citronellal group while no *Eucalyptus* fell within the phellandrene/piperitone group. The eight other plants contain a variety of monoterpenoids which did not belong to any of the three main types. Only three *Eucalyptus* volatile oils from *E. camadulensis* var *nanse*, *E. camadulensis* var *myrore* and *E. propinqua* are potentially useful medicinally. These plants could be commercially exploited in future essential oil development programmes in Nigeria, in addition to the particular interest in the timber capability of the *Eucalyptus* plants at the inception of FRINs trial plantations.

Citrus Species

Analysis of the leaf essential oil from Nigerian grown sweet orange (*Citrus sinensis*) shows sabinene, car-3-ene, (E)-β-ocimene and linalool to be the principal components. Eleven cultivars named Etinan, Hamlin, Meran, Umudike, Agege I, Bende, Person, Pineapple, Personbrown, Kinnow and Pope were investigated. Another eleven cultivars of *Citrus reticulata* of the tangerine, tangelos and Satsuma mandarin types displayed essentially similar compositional patterns to the sweet orange oil. However, lime fruits (*Citrus aurantifolia*) contained high amounts of limonene (34%) whilst lemon (*Citrus limon*) oil comprised of mainly citral (50.1%) and limonene (29.3%). Lime and lemon oils exhibited the characteristic lemon odour which would be ideal in flavouring soft drinks, baked goods and confectioneries. The orange may find prime use as a source of cheap fragrances for detergents, air freshners and beverages (Kasali and Ekundayo 2008).

All these *Citrus* plants were purposely cultivated for their fruits at Nigerian Horticultural Research Institute (NIHORT) orchards in Ibadan. Exploitation of their leaves for citrus oil is quite feasible and should be profitable.

Essential Oils of Nigerian Medicinal Plants

The bulk of our research of Nigerian essential oils was devoted to medicinal plants. Seventy nine (79) plants have been investigated up to date. The results are deep, revealing and extensive. However, the limitation of time would only allow me to review a very small fraction of our findings in this lecture. For the purpose of chemical profiling of essential oils of Nigerian medicinal plants, my intention is to offer a brief synopsis of the investigations of four plants.

Piper guineense (Ashanti pepper) (Piperaceae) is a herbaceous plant which is commonly used as a remedy for rheumatism and bronchitis. The dried fruits or berries which are usually on sale in local markets constitute the flavourant ingredient of the local "pepper soups". A total of 56 compounds were identified in the fruit essential oil which was very rich in phenylpropanoid derivatives such as myristicin, sarisan, safrole and dillapiole. The last three compounds are toxic and psychotropic. At the level of 0.15 μg/g of the fruits, there is an attendant potential toxicity on ingestion (Ekundayo *et al.* 1988). (Peppersoup aficionados beware !!!).

Hyptis suavolens (Bush Tea) (Labiateae) is a very popular remedy for fever and a number of diseases. The stem is reputed to possess anticancer properties and the leaves exert anti-fertility activities in women (Oliver-Bever 1986). The plant is characterized by a strong mint-like odour. Analysis of the leaf essential oil afforded the identification of thirty nine (39) components with a dominant amount of sabinene (Asekun and Ekundayo 2003). The sesquiterpenoid fraction comprised of β-caryophyllene and E-α-bergamotene. The latter imparted a pleasant spicy odour on the oil which displays significant antimicrobial activities (Asekun *et al.* 1999).

Lantana camara var *aculeata* (Verbenaceae) has a wide application in Nigerian ethnomedicine in the treatment of rheumatism, asthma, cough and cold (Oliver-Bever 1996). The volatile oils from the leaves and flowers contained forty-six compounds with sizeable amounts of sabinene, 1,8-cineole and β-caryophyllene (Kasali *et al.* 2004).

The case of the common onion (*Allium cepa*, Alleaceae) is very interesting. It was cultivated by the Egyptians 5000 years ago as a condiment and medicinal plant. Onions are used in the treatment of respiratory and infectious diseases in northern Nigeria (Oliver-Bever 1986). The essential oil from onion bulbs purchased in Ibadan was found to contain mainly diallyl disulphide (59%) with minor amounts of methylallyl trisulphide (19%) and diallyl tetrasulphide (3%) (Ehiabi *et al.* 2006).

However, all these dialkyl(en)yl sulphides, disulphides, trisulphides and tetrasulphides are formed from S-Alk(en) cysteine sulphoxide when onion is crushed. The allinase enzyme is released along with the substrate from the previous intact cellular structure. A side product of this enzyme mediated biochemical reaction is thiopropanal-S-oxide which induces tears. Uncut onion is absolutely odourless in contrast to the pungent and sulphurous odour of the released chemicals on crushing. These sulphur compounds exhibit wide ranging pharmacological activities such as fungicidal, antibacterial, hyperglycaemic, anti-tumor and anti-asthmatic activities.

Biological Activities of the Essential Oils of Nigerian Medicinal Plants

Just like the onion oil, numerous essential oils display a whole spectrum of biological activities. Normally, evaluation of biological activities of naturally occurring substances provides a direct avenue to the utilization of plant sources for the treatment of diseases. We conducted five bioassays on Nigerian essential oils. These assays were relatively rapid, reproducible and significant.

1. Brine Shrimp Lethality Assay

Brine Shrimp Lethality assay is a preliminary screening system for the toxicity of plant extracts using laboratory cultured brine shrimp (*Artemia nauplii*). The assay is quantitatively measured in terms of LC_{50} which is the lethal dose needed to kill 50% of the brine shrimp. An LC_{50} value of less than 1000 μg/ml is indicative of toxicity. The assay results are displayed in table 5.

Table 5: Brine Shrimp Lethality Assay of Nigerian Essential Oils

Plant (Family)	LC_{50} μg/ml
Hyptis suaveolens (Labiatae)	4.1
Cedrela odorata (Meliaceae)	10.0
Daniela ogea (Caesalipinaceae)	10.9
Costus after (Zingiberacae)	10.8
Xylopia aethiopica (Annonaceae)	TOXIC
Ocimum canum (Labiateae)	109.9
Ocimum basilicum (Labiateae)	95.5
Annona senegalanis (Annonaceae)	19.3
Tetrapleura tetraptera (Fabaceae)	111.45
Treculia africana (Moraceae)	18.7
Ficus capensis (Moraceae)	16.3
Artocarpus communis (Moraceae)	245.0

Sources: Asekun and Ekundayo (2008); Aboaba and Ekundayo (2008)

The assay results were obtained for the leaf essential oils of twelve medicinal plants belonging to different families. The plants are of widespread use in West African folkore medical practice. The LC_{50} assay values for the essential oils depict considerable levels of toxicity. These preliminary screening assay results could be the basis for potential bioactivities investigations of the oils.

2. *Antitumor Activities*

Essential oils from only seven Nigerian medicinal plants displayed cytotoxic activities against 3 different carcinoma cell lines in a survey conducted on eighteen plants. Volatile oils from the leaves of *Hyptis suaveolens*, *Cedrela odorata* and *Xylopia aethiopica* were toxic against human epidermal carcinoma cells (Hep 2-cell line). Essential oils from *Datura metel* (fruits and flowers), *Solanum erianthun* (leaf), *Cassia alata* (leaf), and *Ficus mucoso* (leaf) exhibited *in vitro* cytoxicity against human breast carcinoma ($H_2$5781 cell-line) whilst only the leaf volatile oil of *Solanum erianthum* showed significant inhibitory effects against human prostate carcinoma cells (UPC-3 cell line) (Asekun and Ekundayo 2008; Essien and

Ekundayo 2008). These demonstrable antitumor activities would certainly form the basis of future studies of the active essential oils.

3. *Antimicrobial Activities*

The antimicrobial effects of numerous essential oils have been extensively recorded in the literature. In our assays, the agar diffusion methods were utilized. Assays were conducted on forty-six plants and sixty-one essential oils; whole plants and their organs were investigated. Majority of these volatile oils exhibited a broad spectrum of activities against gram-positive, gram-negative bacteria, and fungi. *Eucalyptus*, *Ocimum* and other Labiateae plants were very active. Additional inhibitory essential oils were extracted from *Causarina equistifolia*, *Datura metel* and *Cassia alata*. However, *Solanum macranthum*, *Ficus mucoso*, and *Melaleuca leucadenron* did not inhibit microbial growth in test organisms (Essien and Ekundayo 2008; Asekun and Ekundayo 2008).

4. *Larvicidal activities*

Malaria is one of the world's leading causes of morbidity and mortality with about 2 million annual deaths. Natural products are increasingly been used against malaria vectors since synthetic insecticides are environmentally hazardous. There is no doubt that essential oils are capable of interfering with the reproductive stages of the *Anopheles* mosquito.

Eight essential oils from Nigerian plants were assayed for larvicidal activities against *Anopheles gambiae* and all the oils displayed significant activities (Table 6).

Table 6: Larvicidal Activities of the Essential Oils of Nigerian Plants

Plant	LC_{50} µg/mL
Acalypha segetalis	45.4
Tetrapleura tetraptera	187.5
Xylopia aethiopica	194.5
Ficus capensis	196.4
Treculia Africana	263.7
Artocarpus communis	852.0
Curcuma longa (leaves)	50.0
Curcuma longa (rhizome)	12.5

Sources: Aboaba and Ekundayo (2008); Ajaiyeoba *et al* (2008)

Acalypha segetalis exhibited a dose of LC_{50} of 45.4µg/ml. The other oils had values of 187.5, 194.5, 196.4, 263.7 and 852.0µg/ml for *Tetrapleura tetraptera, Xylopia aethiopica, Ficus capensis, Treculia africana* and *Artocapus communis* respectively (Aboaba and Ekundayo 2008). Leaf and rhizome essential oils of *Curcuma longa* (tumeric) were also toxic to mosquito larvae at concentrations of 50 µg/ml and 12.5 µg/ml respectively (Ajaiyeoba et al. 2008). These comparatively low LC_{50} values are indicative of the potential of essential oils from some Nigerian plants to serve as larvicides in the vector control of mosquitoes. There is considerable interest in developing plant derived insecticides to replace the synthetic analogues, and at lower costs.

5. Acetylcholinesterase (AChE) Inhibitory Assay

Acetylcholine plays a role in the central nervous system especially in the realm of information processing. Acetylcholine and its receptors have been implicated in numerous sensory related diseases such as Parkinson, Huntingtons chorea, myasathemia gravis and even leukemia. Acetylcholine is customarily hydrolysed by the enzyme, acetylcholinesterase (AChE), and an inhibition of the activity of the enzyme has led to human long-term improvement and enhanced performance in word recall tests.

AChE inhibition was measured for thirteen essential oils from eight plants. Most of the volatile oils demonstrated activities at all concentrations (Table 7).

However, five volatile oils from *Treculia africana* leaves, stembark and *Tetrapleura tetraptera* leaves did not inhibit acetylcholinesterase activities at low and medium concentrations.

Isolation and Chemical Characterization of Biologically active Compounds from Nigerian Essential Oils

In view of the array of biological activities displayed by numerous essential oils from Nigeria, the next logical phase of our research would be to identify the volatile oils possessing biological activities. Then activity-directed fractionation of the oils would follow with the aim of isolating the active constituents in the pure form. The compounds would be assayed. Spectroscopic methods would be employed to identify the constituents unambiguously. Chemical syntheses and structure activity relationships will constitute the final phase in our search for pharmacologically active compounds in essential oils.

Novel Chemical Aspects of Nigerian Essential Oils

The significance of our research into the essential oils of Nigerian plants is in the magnitude of the interesting multifaceted Chemistry thereof, as indicated in the following sections.

Isolation and Identification of New Naturally Occurring Organic Compounds

Isolation of new naturally occurring substances is always of academic interest, especially the intellectual rigour which goes into the elucidation of their structures.

A new thymol derivative—6-methoxythymol, was detected in *Ocimum viride* (Effinrin) hydrodistilled essential oil. The compound with a molecular formula of $C_{11}H_{16}O_2$ was assigned its chemical structure on the basis of its spectroscopic properties especially the mass spectral features. It is structurally related to thymol which is the principal antibiotic compound in the plant's essential oil.

Table 7: Acetylcholinesterase Assay of Thirteen Essential oils

Essential Oil	Conc of ChE over Control (Y) µg/ml	% age inhibition	% age decrease or increase compared to Huperzine A % age inhibition	Equivalent Huperzine A deduced from std. graph (X)	Remark on cholinesterase inhibition
XASB	5 10 20	-3.1 -12.7 -22.1	-41.1 -41.9 -45.2	6.5 6.8 8.5	Good inhibition at all concentrations
ASWP	5 10 20	-0.9 -9.7 -12.0	-43.3 -44.9 -55.3	7.5 8.3 13.3	Good inhibition at all concentrations
TTL	5 10 20	-16.4 -17.6 -18.2	-27.8 -37.0 -49.1	- 4.3 10.3	No inhibition at low concentration and good inhibition at high conc.
SJR	5 10 20	-7.3 -13.9 -24.2	-36.9 -40.7 -43.1	4.2 5.8 7.3	Good inhibition at all concentrations
TAL	5 10 20	-23.6 -24.8 -29.7	-20.6 -29.8 -37.6	- 0.5 4.5	No inhibition at low concentration, trace at medium & good inhibition at high concentration
TASB	5 10 20	-21.8 -29.1 -33.9	-22.4 -25.5 -33.4	- - 2.5	No inhibition at low & medium conc. But moderate

TARB	5	-35.2	-9.0	-	No inhibition at all concentrations
	10	-37.6	-17.0	-	
	20	-47.3	-20.0	-	
FCL	5	-5.5	-38.7	5.0	Good inhibition at all concentration
	10	-10.9	-43.7	7.6	
	20	-12.7	-54.6	13.0	
FCSB	5	-7.3	-36.9	4.2	Good inhibition at all concentration
	10	-12.7	-41.9	6.8	
	20	-27.3	-45.2	8.5	
FCRB	5	-3.0	-41.2	6.4	Good inhibition at all concentration
	10	-6.7	-47.9	9.5	
	20	-10.3	-57.0	14.2	
ACL	5	-2.8	-41.4	6.3	Good inhibition at all concentration
	10	-8.5	-46.1	8.8	
	20	-11.3	-56.0	13.8	
ACSB	5	-18.2	-16.0	-	No inhibition at all concentration
	10	-32.1	-22.5	-	
	20	-39.9	-24.3	-	
ACRB	5	-3.0	-41.2	6.4	Good inhibition at all concentration
	10	-12.1	-42.5	7.0	
	20	-21.2	-46.1	8.8	

Source: Aboaba and Ekundayo (2008)

Notes

XASB=*Xyplopia aethiopica stem bark*
TARB=*Treculia africana root bark*
SJR=*Sphenocentrum jollyanum roots*
FCRB=*Ficus capensis root bark*
TASB=*Treculia africana stem bark*
TTL=*Tetrapleura tetraptera leaves*
FCSB=*Ficus capensis stem bark*
ACL=*Artocarpus communis leaves*
ASWP=*Acalypha segetalis whole plant*
FCL=*Ficus capensis leaves*
TAL=*Treculia africana leaves*
ACRB=*Artocarpus communis root bark*
ACSB=*Artocarpus communis stem bark*

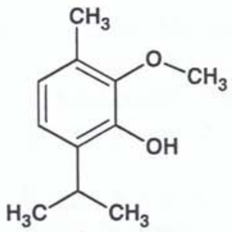
In the examination of the leaf essential oil of *Ageratum conyzoides*, two new dihydro-derivatives of chromenes were isolated. These chromenes—dihydrodimethylencecalin and dihydroencecalin have not been previously encountered in nature.

Comprehensive GC and GC-MS analyses of the essential oil of *Calotropis indica* from Ogbomoso afforded the isolation of a new natural product which was identified as (-)-eudesman-1,4(15),11-triene.

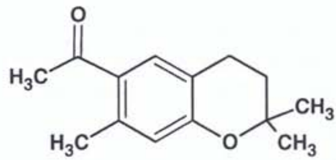
Rare p-menthane derivatives – 1,2:3,4-diepoxy-p-menthane and 1,4-diepoxy-p-menth-2-ene were recently found in leaf volatile oils of *Chenopodium ambrosiodes*—a highly aromatic herb. These compounds are closely related to the customary principal component of *Chenopodium* volatile oils—ascaridole—a powerful antihelmintic compound.

Chemical Synthesis of Fragrant Compounds

Synthetic aroma and flavour chemicals have been known since 1859 when "synthetic wintergreen oil" was first utilized as a substitute for the natural oil from vernal grass: *Meliatus officinalis* (Johnston 1859). α-and β-Ionones are naturally occurring substances which possess sweet and floral flavour organoleptic properties. In fact, α-Ionone is the flavour ingredient in raspberries, carrots and black tea. These compounds are chemically synthesized from citral and propanone because of their low levels in essential oils (Fig. 2).

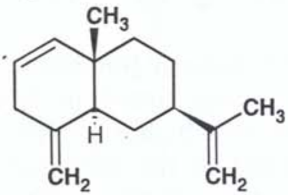


I



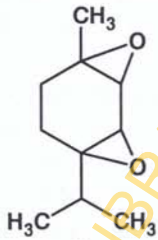
II R = H dihydrodemethylencecalin

III R = OCH_3 dihydroencecalin



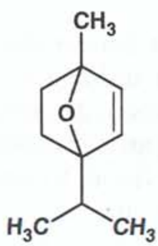
IV

(-)-eudesman-1, 4(15), 11-triene



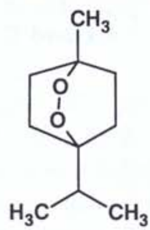
V

1, 2: 3, 4- diepoxy-p-menthane



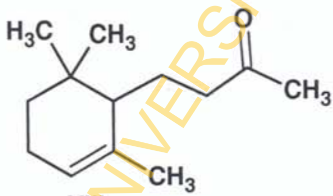
VI

1, 4-epoxy-p-menth-2-ene



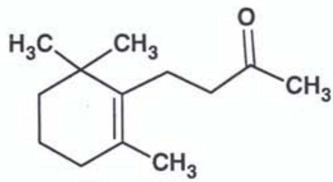
VII

ascaridole



VIII

alpha- ionone



IX

beta- ionone

Fig. 2: Synthesis of α- and β-Ionones

Eight ketones were used in the chemical synthesis of various types of ionones which were evaluated for their odour properties in our laboratories (Table 8).

Table 8: Characteristics of Organoleptic Ionones derived from Ketones

Ketone	Odour of ionone	Application
1. 2-octanone	Slightly fruity	Deodorants
2. propanone	Flowery	Perfumes and lotion
3. phenylethanone	Sweet aromatic	Paints and air freshners
4. 3-butanone	Peppery and fruity	Paints and air freshners
5. butanone	Flowery	Perfumes and toiletries
6. 3-pentanone	Slightly chocking and flora	Paints
7. mesityl oxide	Mildly flowery	Perfumes and fabrics
8. 2-methyl cyclohexanone	Aromatic and faintly fruity	Paints

Source: (Moronkola, Aiyelaagbe and Ekundayo 2005)

The artificiality of these synthetic chemicals has been the subject of a new concept called 'Green Chemistry' which seeks to address the interrelationship between chemical industries and environmental policy management issues in order to achieve sustainable development. 'Green Chemistry' also recommends that industrial synthetic processes should not be directed at the production of toxic substances whatsoever. Safety issues such as minimization of chemical accidents, explosion and fire are also important in 'Green Chemistry'.

These key aspects of chemical synthesis have been known for years but it was only recently that an integrated form of the relationship between industrial chemical synthesis and environment was formulated into 'Green Chemistry'. Renewable resources are encouraged for use in chemical syntheses of industrial chemicals. The syntheses and uses of the products should, as much as possible, not affect nature. A number of 'green reactions' are being conducted on a pilot scale in the laboratories and the resultant industrial production has taken cognizance of catalysis, reagents, solvents, mineral resources and biodegradable products.

Economic Prospects of Production of Essential Oils in Nigeria

Our analytical data on the essential oils of *Pinus*, *Eucalyptus* and *Citrus* species have established unequivocally their economic viability, more especially that their volatile oils sell for between 21-30 US dollars per kilogram in international markets. *Pinus* and *Eucalyptus* are usually forest managers' favourites on account of their high timber yield and ease of cultivation. Pines are increasingly important as sources of essential oils in the areas where they have been planted as commercial timber trees. Since the needles find no use when trees are felled, they represent rich, yet cheap sources of essential oils—the integral profit potential is quite high.

Simple steam distilling equipment could be fabricated locally and the only raw materials are the leaves or needles. An essential oil industry would definitely be viable and serve as a vehicle for economic development around the three major cities—Ibadan, Kaduna, and Jos where the plants are currently cultivated in trial plots.

The long-term objectives of an essential oil industry in Nigeria should be the selection of economically valuable plants for cultivation and breeding on a large scale. Conventional essential oil bearing plants such as lemon grass (*Cympogon citratus*), ginger (*Zingiber officinale*), basil (*Ocimum basilicum*), mint (*Mentha* sps.) and Rose (*Rosa damscema*) would be ideal for this purpose. Apart from these conventional essential oil plants, Nigeria is richly endowed with other numerous aromatic plants which grow in the wild and are cultivable. Examples are custard apple (*Anona reticulata*), Ashanti pepper (*Piper guineense*), and Pitanga sherry (*Eugenia uniflora*). All these are medicinal plants in their own rights and our investigations have covered them all.

There is evidence of a growing worldwide demand for essential oils and at the current prices, an essential oil industry in Nigeria would definitely thrive and have considerable merit. However, in order to underpin the strategic development of the industry, focused research must be conducted to understand the market mechanisms and opportunities, production systems, supply chains and sustainable generic plant sources. There is no doubt that essential oil production in Nigeria has a very sound commercial base with room for considerable expansion in future.

Conclusion

Mr. Vice-Chancellor, in concluding this lecture, I wish to avoid the customary cliché of inaugural lectures proffering solutions for the plethora of problems besetting this country and especially the university system. There is no shortage of prescriptions. I have no intention of re-inventing the wheel!!. But I could not end this lecture without drawing attention to a former Ph.D student of mine who is now a Senior Lecturer in the University of Zululand in South Africa. Her Essential Oil research is principally funded by the South African National Research Foundation (NRF) to the tune of 1 million rands (₦14 million). Her vibrant research group comprises of two Ph.D, three M.Sc. and five undergraduate students. Need I say more?

Finally, Mr. Vice-Chancellor, in order to give meaning to the title of this lecture, I wish to refer to the Holy Bible, and specifically to the story of the Wise Men from the east, who visited Baby Jesus at his birth. "Then they opened their treasure chests and gave him gifts of gold, frankincense and myrrh" (Mathew 2: 11). The Oxford Advanced English Learners' Dictionary defines "frankincense" as "a kind of resin from trees, giving a sweet smell when burnt" and "myrrh" as "sweet smelling bitter tasting kind of gum or resin obtained from shrubs used for making incense and perfumes". The specific plants for frankincense are *Boswellia* species, and myrrh—*Commiphora myrrha,* an Absyssian tree. Plants have been treasure troves of valuable gifts for ages.

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I must start by an expression of gratitude to my Heavenly Father, God Almighty, my Creator who has been especially good to me. He has always shown me in all aspects of my life that He has the last say no matter the situation. This is evidenced by all that I have achieved through His grace, and His making this day possible.

I owe a debt of gratitude to numerous persons who have touched my life in one way or the other. Time and space constraints would not allow me to name everyone. (Kindly forgive me if I do not mention you in this acknowledgement).

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My family members have played invaluable roles in my life. This lecture is dedicated to all of them. First, my mother—Mrs. Aina Olukoya—deserves unrivalled appreciation for her sacrifice and support even unto this moment. I acknowledge with gratitude my siblings—Mrs. Funke Osotimehin, Mr. Kemi Olukoya, and Enitan Olukoya and their spouses. I wish to thank deeply my in-laws—the Salawu's—Mrs. Adebisi Salawu, Dr.

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I now thank you, Mr. Vice-Chancellor and this wonderful audience for your kind attention.

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