
# FEDERAL UNIVERSITY OF TECHNOLOGY MINNA

# NATURAL PRODUCT CHEMISTRY AND PROSPECTS FOR TRANSFORMING TRADITIONAL MEDICINES INTO MODERN PHARMACEUTICALS

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*B.Sc. (ABU), M.Sc. (Manchester), PhD (Manchester)*
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***Professor of Chemistry***

**INAUGURAL LECTURE SERIES 54**

**5TH OCTOBER, 2017**

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## INTRODUCTION

For centuries man has exploited plant extracts for therapeutic purposes, and there has therefore been an enduring interest in the nature of their constituents. It is only recently, however that science has enabled us to isolate and determine the chemical structures of these natural products with the development of new and more powerful analytical tools.

Natural product chemistry not only deals with characterization of these substances but also their preparation (synthesis). This is no longer used for traditional proof of structure, but is vital for making available actual and potential medicinally useful compounds.

'Natural products' today is quite commonly understood to refer to herbs, herbal concoctions, dietary supplements, traditional folk medicine, or alternative medicines.

## TRADITIONAL HEALERS: SCIENTIFIC FRIENDS OR FOES?

### HISTORY

The use of natural products (herbs, leaves, roots, stem bark, fruits, seeds) for therapeutic purposes has been described throughout history in the form of traditional medicines, remedies, potions and oils with many of these bioactive natural products still being unidentified. The dominant source of knowledge of uses of natural products from plants is a result of man experimenting by trial and error for hundreds of centuries through palatability trials or untimely deaths, or while searching for available foods for the treatment of diseases (Kinghorn, 2011; Hicks, 2011).

WHO defines traditional medicine as 'the sum total of the knowledge, skills and practices based on theories, beliefs and experiences indigenous to different cultures, whether explicable or not, used in the maintenance of health as in the prevention,

diagnosis, improvement or treatment of physical and mental health.' It is estimated that 80% of Africans especially rural communities rely on traditional medicines to meet their health-care needs (WHO, 2008). Their wide-spread popularity is largely based on their affordability and sustainability (Cordell, 2015).

Over the years, careful study of traditional medicines led to the establishment of the pharmacopoeia which is a catalogue of medicaments. Various societies *e.g.* the Chinese, Indians and indeed Western Europeans have exploited their rich history of traditional medicines and have developed them into modern medicines. The Chinese and Indians for instance, have given adequate recognition to their extensively developed traditional medical practices and have incorporated them into their national health programmes (Ravishankar and Shukla, 2007). Though Nigeria has a national policy on traditional medical practice (Federal Ministry of Health Traditional Medicine Policy For Nigeria, 2007), it is still in its infancy as there is little synergy between Western and our traditional medical practitioners today.

In contemporary Nigeria, most 'moderns' will not openly confess patronizing herbalists-(*babalawos bokas, dibias, etc*). But come to think of it, we owe a whole lot of gratitude to these 'uneducated' maybe 'uncivilized' and often underrated medical practitioners. For without them and their efforts over the years we will all be in dire straits health-wise. Since pre-history, this class of people identified and used various plants (and sometimes animal parts) to treat various ailments with varying degrees of success. Their skills earned them a pride of place in the society. In most rural communities, these are the medical doctors whom they consult when sick. Their practice is based on knowledge gathered over the years and passed down. Our traditional healers however have the habit of dabbling into the occult, preferring to rely more on spiritualism than solely herbal

remedies. A healthy dose of spiritualism /occultism is often therefore, an essential part of their practice (Borokini and Lawal, 2014). Many gullible Nigerians are taken in by this and would rather prefer spiritually-assisted remedies to plain medicaments (herbs). Most practitioners, however jealously guard their 'trade secrets' (form of intellectual property rights?)

## FROM HERBAL REMEDIES TO MEDICINAL DRUGS

Based on trado-medical claims, many plants have been investigated by several researchers in Nigeria. For instance, in a survey, 70 medicinal plants used in Edo state for the treatment of various ailments were listed by Okoli *et al.* (2007). These include the neem tree (*Azadirachta indica*), cashew (*Anacardium occidentale*), and paw-paw (*Carica papaya*), and in 2013, the Nigerian Society of Pharmacognosy published a list of 91 commonly used medicinal plants from all over Nigeria (NSP, 2013). Other authors include Sofowora (1982, 1993) Mann *et al* (2003), Edeoga (2006), Obiukwu (2010); Raphael, (2011) and Olutayo *et. al* (2013).

Natural products, containing inherently large-scale structural diversity than synthetic compounds, have been the major resources of bioactive agents and will continually play a vital role as protagonists for discovering new drugs (Shen *et al* 2003). Drug discovery from medicinal plants has mainly relied on biological activity guided isolation methods which have led to the discovery of important drugs. Thus natural products have been, and will be, important sources of new pharmaceutical compounds. Current renewed interest in natural product research is primarily due to the failure of alternative drug discovery methods to deliver many lead compounds in key therapeutic areas, such as immunosuppression, anti- infectives and metabolic diseases (Lahlou, 2007).

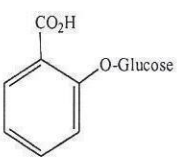
Some useful classes of drugs include:

## ANALGESICS

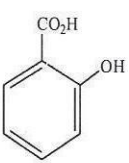
Analgesics are drugs that provide relief from pain an example of which is aspirin.

Aspirin, chemically known as acetyl salicylic acid, is one of the most widely used over-the-counter drug. It is used as a pain killer and an anti inflammatory. For years, extracts of the bark of the European willow plant was found to relieve pains. Users however had to contend with some serious side effects.

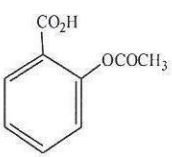
PLATE I: The willow tree




Salicin




Salicylic acid




Acetyl salicylic acid
**(Aspirin)**


(panadol)

Paracetamol

The active ingredient in the plant extract was found to be salicylic acid present as the glycoside salicin. When later the acid was acetylated, it was converted into acetyl salicylic acid (aspirin) which was more tolerable and caused far less discomfort. Today aspirin is widely employed as a pain killer and anti-inflammatory, though its usage has been reduced due to the introduction of acetaminophen (paracetamol). Recently, however aspirin is staging a comeback especially with doctors now prescribing low doses for those at risk of heart disease.

This is what Natural Product chemistry is all about. Search, isolate, synthesise (natural and synthetic analogues)

**ANTIBIOTICS**: - PENICILLIN

Undoubtedly one of the most famous natural product is penicillin. It is an antibiotic obtained from the fungus, *Penicillium notatum* by Alexander Fleming in 1929. This discovery and subsequent commercialization of synthetic penicillin ultimately revolutionized medicinal drug research and discovery.

The story is told of how Dr. Fleming returned from vacation to find his uncovered petri dishes containing *Staphylococcus aureus* colonies contaminated with mold from left-over bread. The amazing observation was that this mold called *Penicillium notatum* had inhibited the normal growth of *S. aureus* colonies. For this discovery, Dr. Fleming was awarded the Nobel Prize in medicine in 1945 (Mann, 1994).

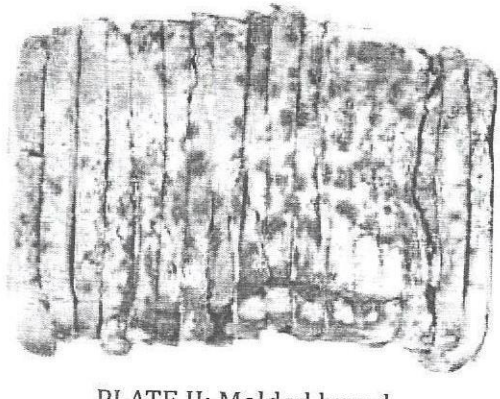
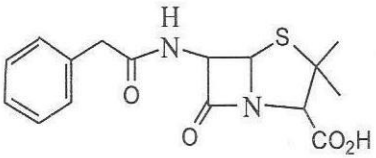
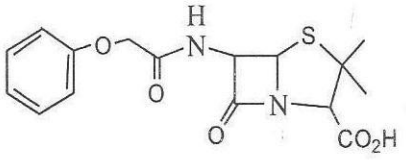


PLATE II: Molded bread

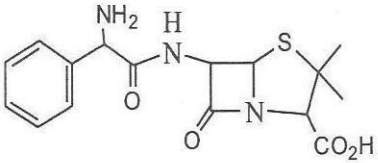
The active ingredient was isolated and found to be penicillin which has been employed to save millions of lives worldwide in the fight against many anti microbial infections. Further modifications led to the development of many presently used antibiotics such as ampicillin and amoxicillin



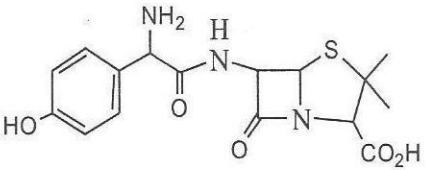
penicillin G



penicillin V



ampicillin



amoxicillin

Apart from the penicillins, many other classes of antibiotics have also been discovered, some natural others synthetic. These include tetracyclines, cephalosporins, quinolones (*e.g.* ciprofloxacin – Cipro), sulphonamides (Septrin) and macrolides (Erythromycin) (Anderson, 2016).

**ANTI-MALARIALS**: – QUININE

For centuries, South American Indians had used the Cinchona (*Cinchona ledgeriana*) bark to treat fevers and later, Spanish colonialists found that it was very effective against malaria (Taylor, 1945; Russell, 1955; Jarcho, 1993)

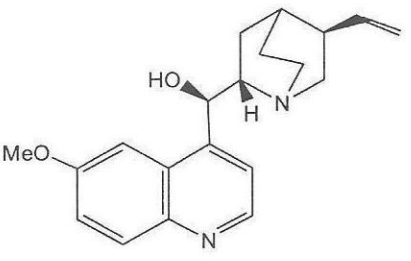
PLATE III: The *Cinchona* tree

Analysis of the bark extract showed that it contained quinine. Consequently, quinine was extensively used, as it was then the only effective anti-malarial by the early 20th century. It was however seen to have a number of unpleasant side-effects, if consumed in quantity, including; nausea and tinnitus (Roche, 1990).

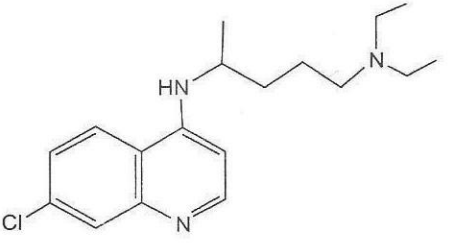
Because of the complex chemical nature of many natural products, their synthesis on an industrial scale may be impossible, hence the need for substitutes. During World War II, supplies of quinine, for example became difficult. Thus research into synthetic substitutes such as chloroquine became imperative.

Chloroquine has a close structural similarity to quinine and its synthesis is an example of how chemists use natural templates to

design and produce synthetic compounds which can be tested for medical potency and efficacy. If useful, modifications on a drug may be carried out to improve potency, tolerance and acceptability.



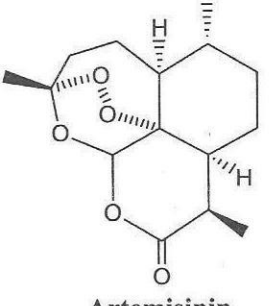

Quinine




Chloroquine

Chloroquine has been for a long time the drug of choice against malaria, but over time, the parasite developed resistance and alternatives are being developed, one of which is artemisinin.

Artemisinin **comes** from a common herb *Artemisia annua* which had been used by the Chinese to treat many diseases as well as malaria. It is fast and effective. Recently there have been signs that malarial parasites are developing resistance even to this drug. Artemisinin-based combination therapy (ACT) *i.e.* artemisinin in conjunction with some other drug(s) is currently the preferred treatment for malaria. This has been found to be both effective and well tolerated in patients.




**Artemisinin**

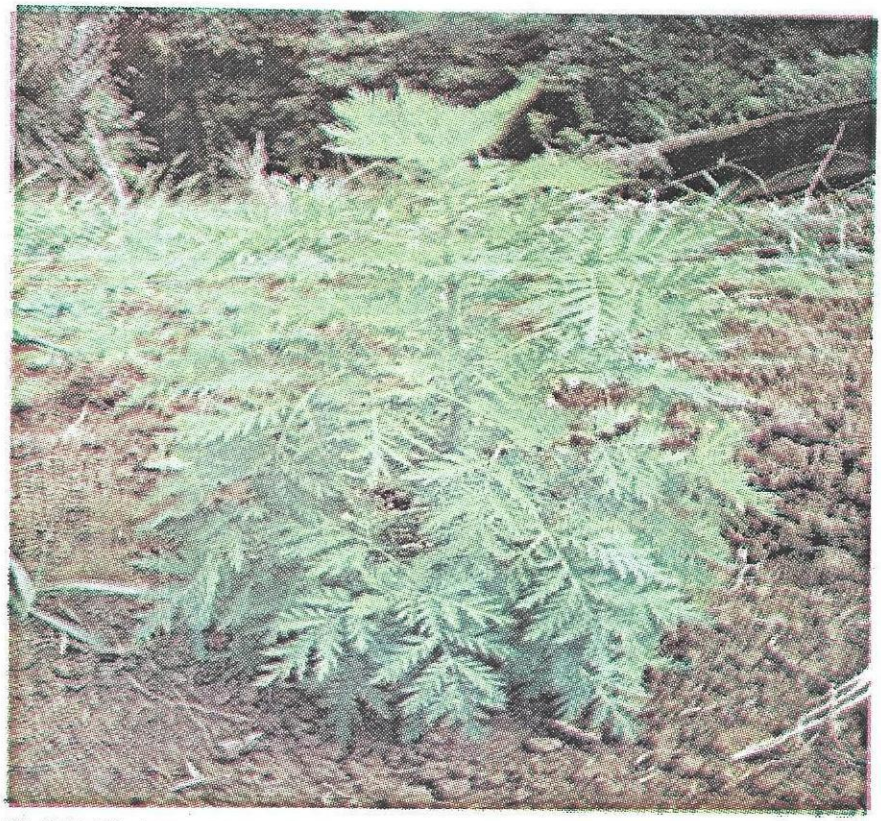
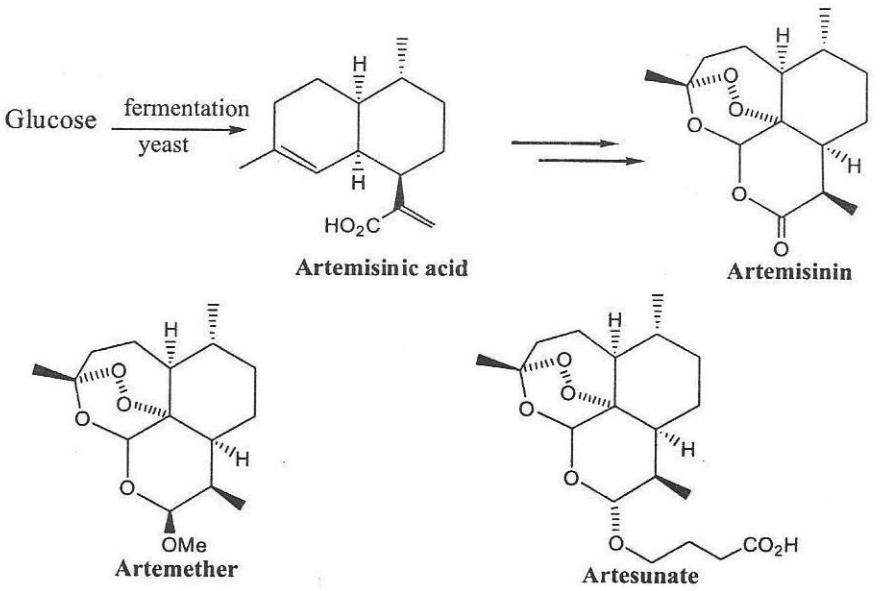


PLATE IV: *Artemisia annua*

Sourcing artemisinin from *Artemisia annua* has its own problems including instability of regular supply, and price fluctuation which can adversely affect the availability of ACTs, hence the need for a synthetic route to the compound. The chemical structure of artemisinin is quite complex and so also its synthesis, which necessarily involves several steps. Organic chemists are constantly seeking for simpler and shorter synthetic routes as this eases production and usually gives better overall yields. Thus, artemisinin was recently prepared in four steps from artemisinic acid obtained as a fermentation product of glucose by a strain of yeast (*Saccharomyces cerevisae*) vividly

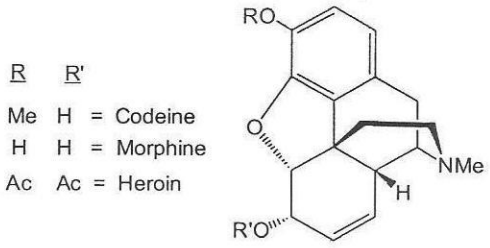
showcasing how natural product chemists exploit biotechnology in many of their reactions. Thus through this procedure, artemisinin, artemether and artesunate - frontline drugs in the fight against malaria can be now be obtained in commercial quantities (Paddon *et al.*, 2013).

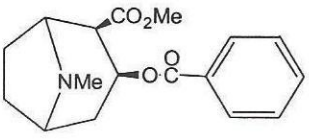



Natural products are used as medicinal drugs directly or more often in combination with others. Currently however, many of them have served as templates for the synthesis of many pharmaceutical medicines.

There is a bewildering array of bioactive secondary metabolites found in medicinal plants among whom are the following;

The opiates - morphine, codeine and heroin from the poppy plant (*Papaver somniferum*) and the alkaloid cocaine from coca plant (*Erythroxylum coca*) which are addictive and often lead to drug abuse.




Cocaine

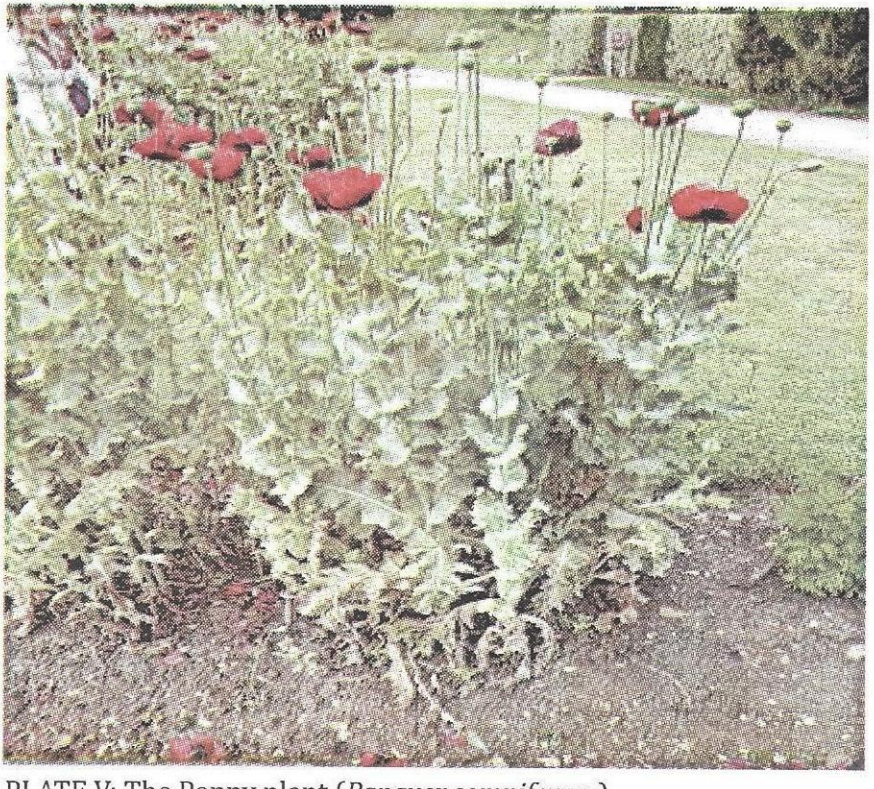
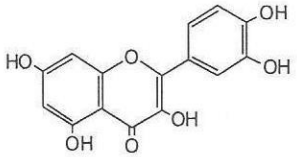
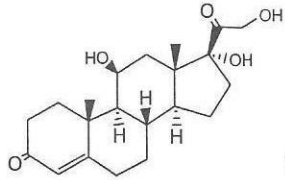


PLATE V: The Poppy plant (*Papaver somniferum*)

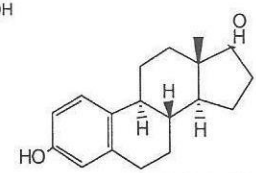
Others are: Terpenes *e.g.* terpineol (Dauda *et al.* 2011), Steroids, (*e.g.* Cholesterol, Oral contraceptives, Sex hormones, Corticosteroids), Phenolic compounds – Tannins *e.g.* Polygalloyl tannin (Jigam *et al'*, 2010), Phlobatannins, Flavonoids (in teas, flower colours), Anthraquinones, etc.



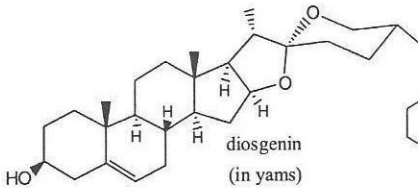
**quercetin** (flavonoid)
(in fruits, vegetables, leaves & grains)



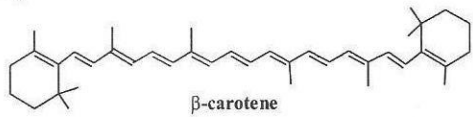
**hydrocortisone** (corticosteroid)



**estradiol** (steroid)
female sex hormone



diosgenin
(in yams)

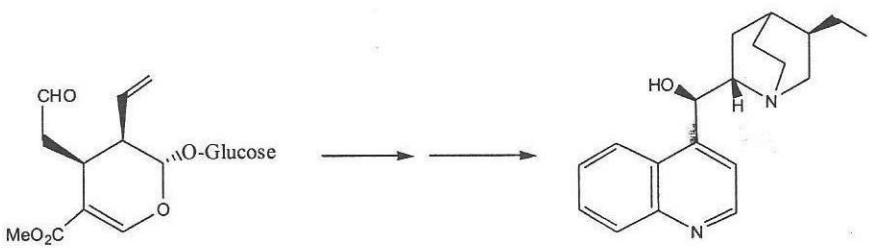


**β-carotene**
in carrots

## ALKALOIDS

Alkaloids are 'alkali like' compounds because of the presence of basic nitrogen, hence their name. They are important because many of them have pharmacological activity. Just as alkalis, alkaloids are bitter (due to the basic nitrogen). This is responsible for the bitter taste of many herbal medicines. Thus quinine, chloroquine, codeine and cocaine are all alkaloids.

i. The quest for the synthesis of quinine analogues took many forms. Some examples of synthesized alkaloidal derivatives include that of dihydrocinchonidine. One of the approaches was the route by Brown *et al.* (1986) starting from the naturally available biological precursor secologanin.

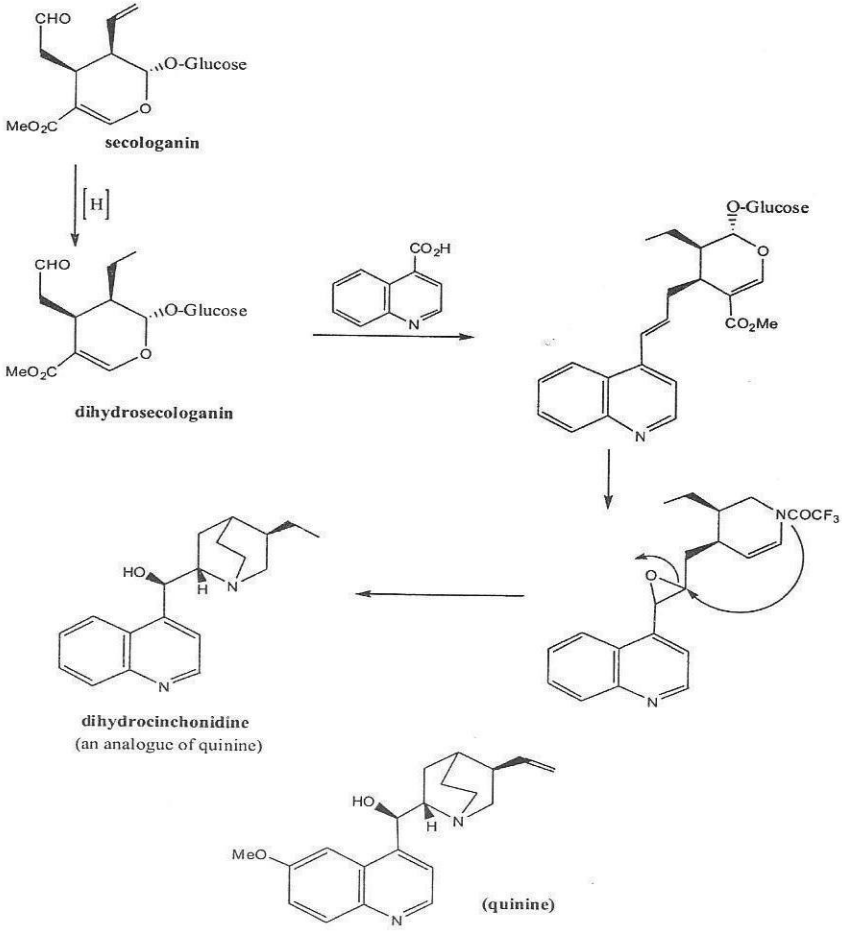


**secologanin**

**dihydrocinchonidine**

Tracer studies, has shown that isotopically labeled secologanin is incorporated into many indole alkaloids and it is therefore a biological precursor of a variety of complex alkaloids. Thus, this 10 carbon unit has often been employed in biomimetic syntheses.

Secologanin was reduced to dihydrosecologanin which was condensed with tryptamine and subsequently transformed in a series of steps to dihydrocinchonidine as follows:



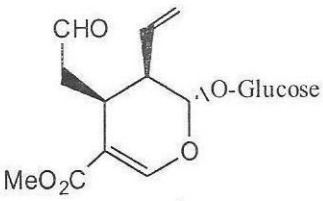
## MY CONTRIBUTIONS

### SECOLOGANIN

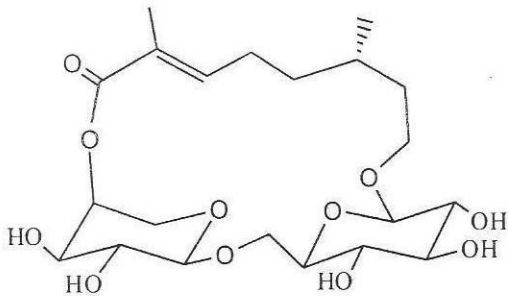
Secologanin is a biological precursor to many indole alkaloids. This 10-Carbon unit forms the monoterpenoid non-tryptamine portion of many classes of indole alkaloids.

It therefore is a favoured starting material of many alkaloid syntheses. Much of my synthetic work on indole alkaloids involved the use of secologanin as starting material

The diglycoside lonitoside, a monoterpenoid macrolide was also isolated along with secologanin from the plant *Lonicera nitida* (Brown *et al.*, 1991).




**secologanin**




**lonitoside**

Thus glycolysis of secologanin ethylene acetal was conducted at different pH resulting into various aglycones (Dauda, 2007) which were subsequently employed for the synthesis of the following:

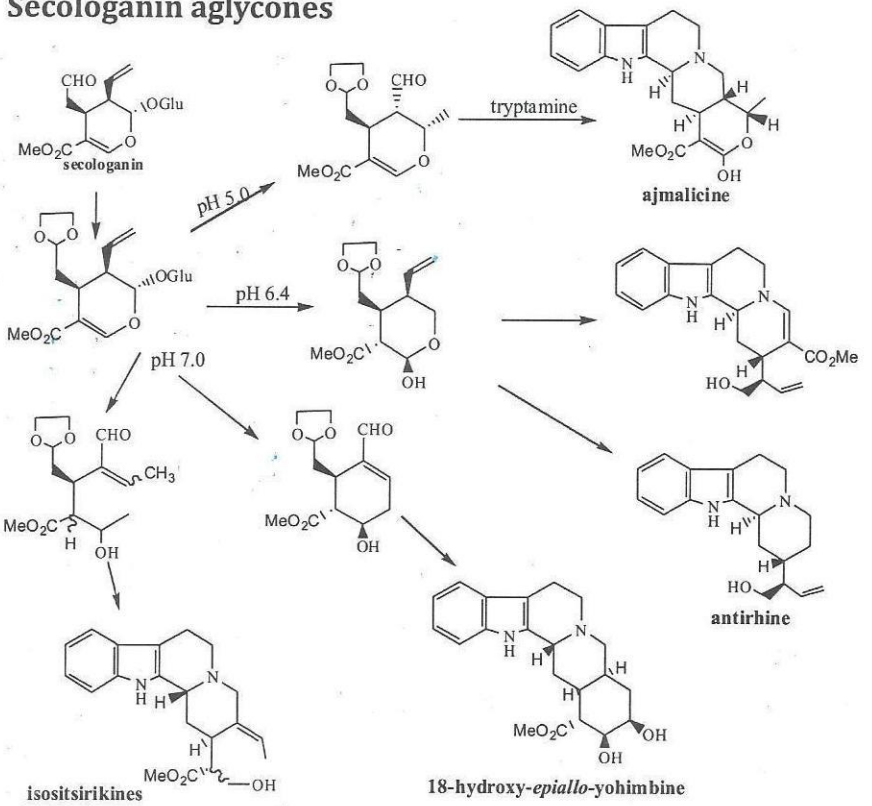
i. the aglycone obtained at pH 5.0 was treated with tryptamine to give ajmalicine (Brown, 2002)

ii. pH 6.4 aglycones afforded antirhine (Brown, 1991) and 16-methoxycarbonyl-16.17-dehydroantirhine (Brown, 2000)

iii. while the pH 7.0 aglycone gave 18-*R*-hydroxy-epiallo-

yohimbine (Brown 2000) as well as isositsirikines and dihydroakuamigine (Dauda, 2008).

**Secologanin aglycones**




*Source: Dauda, 2007, 2008*

**DESERPIDINE (Analogue of Reserpine)**

Reserpine is the main alkaloid from *Rauwolfia vormitoria* (Hausa - *wada,* Yoruba - *asofeyeje,* Igbo - *akanta*). It is an antipsychotic and antihypertensive drug that has been used for the control of high blood pressure and for the relief of psychotic symptoms. Traditionally, Rauwolfia has been used in Ghana and Nigeria as an emetic and is reportedly used for the treatment of jaundice, snake bites and fever (Onike, 2010).

PLATE VI: *Rauwolfia vormitoria*

The first total synthesis of reserpine was achieved by R.B. Woodward in over 18 steps (Woodward *et al.*, 1958). This brilliant feat contributed to an award of a Nobel prize in 1963.

In 2000 we published a paper in detailing the synthesis of a reserpine analogue 18-R-hydroxyohimbine starting from secologanin in just 7 steps (Brown *et al* 2000). A product which is convertible into deserpidine in two steps.

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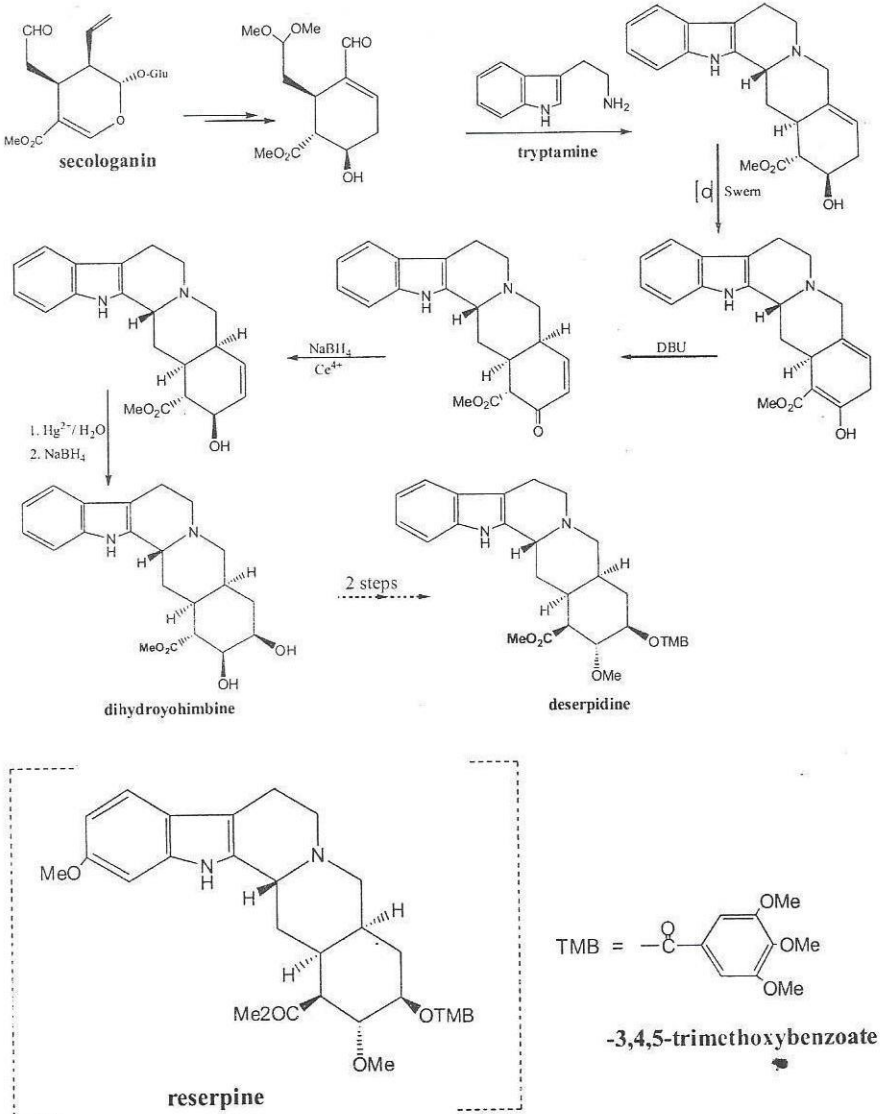
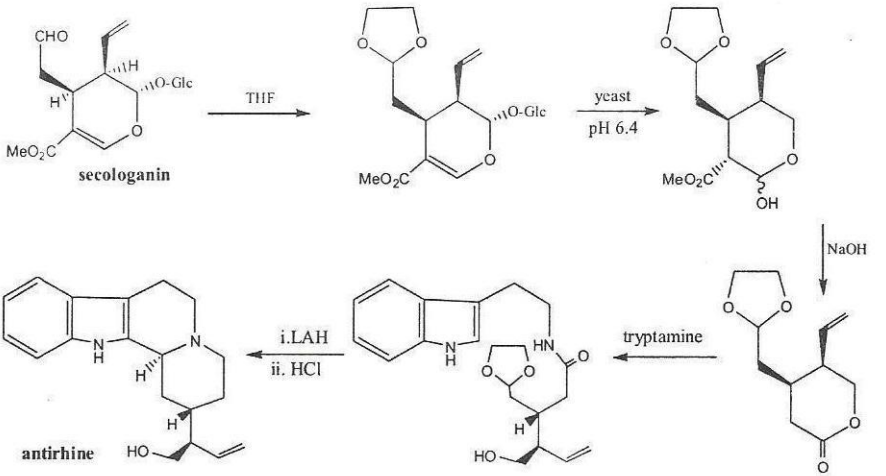



## ANTIRHINE

. Antirhine is an alkaloid from *Antirhea putaminosa* and *Rhazya stricta* it is reported to have anti-cancer properties (Obaid *et al.* 2017)

Its previous total synthesis had involved a minimum of ten steps with less than 15% yield. Starting from secologanin, however, we were able to achieve over 45% overall yield in five steps (Brown *et al.* 1991):




Thus trees and plants are, indeed, rich reservoirs of various types of drugs and chemicals and potential sources of revenue if properly exploited in a sustainable and environmentally friendly manner.

## PLANT-DERIVED DRUGS

The following is a list of drugs / chemicals obtained from some plants (Taylor, 2000):

| Drug/Chemical | Action/Medicinal Use | Plant Source |
|---|---|---|
| Acetyldigoxin | Cardiotonic | *Digitalis lanata* |
| Aesculetin | Anti-dysentery | *Frazinus rhychophylla* |
| Agrimophol | Anthelmintic | *Agrimonia supatoria* |
| Ajmalicine | Circulatory Disorders | *Rauvolfia sepentina* |
| Allantoin | Vulnerary | Several plants |
| Allyl isothiocyanate | Rubefacient | *Brassica nigra* |

| Drug/Chemical | Action/Medicinal Use | Plant Source |
|---|---|---|
| Anisodamine | Anticholinergic | *Anisodus tanguticus* |
| Anisodine | Anticholinergic | *Anisodus tanguticus* |
| Atropine | Anticholinergic | *Atropa belladonna* |
| Benzyl benzoate | Scabicide | Several plants |
| Benzyl benzoate | Scabicide | Several plants |
| Betulinic acid | Anticancerous | *Betula alba* |
| Caffeine | CNS stimulant | Several plants |
| Chymopapain | Proteolytic, mucolytic | *Carica papaya* |
| Cocaine | Local anaesthetic | *Erythroxylum coca* |
| Codeine | Analgesic, antitussive | *Papaver somniferum* |
| Curcumin | Choleretic | *Curcuma longa* |
| Deserpidine | Antihypertensive, tranquillizer | *Rauvolfia canescens* |
| Digitalin | Cardiotonic | *Digitalis purpurea* |
| Digitoxin | Cardiotonic | *Digitalis purpurea* |
| Emetine | Amoebicide, emetic | *Cephaelis ipecacuanha* |
| Ephedrine | Sympathomimetic, antihistamine | *Ephedra sinica* |
| Etoposide | Antitumor agent | *Podophyllum peltatum* |
| Gossypol | Male contraceptive | *Gossypium species* |
| Hesperidin | Capillary fragility | *Citrus species* |
| Hyoscyamine | Anticholinergic | *Hyoscyamus niger* |
| Irinotecan | Anticancer, antitumor agent | *Camptotheca acuminata* |
| Kawain | Tranquillizer | *Piper methysticum* |
| Methyl salicylate | Rubefacient | *Gaultheria procumbens* |
| Monocrotaline | Antitumor agent (topical) | *Crotalaria sessiliflora* |
| Morphine | Analgesic | *Papaver somniferum* |
| Nicotine | Insecticide | *Nicotiana tabacum* |
| Papain | Proteolytic, mucolytic | *Carica papaya* |
| Papavarine | Smooth muscle relaxant | *Papaver somniferum* |
| Pinitol | Expectorant | Several plants |
| Quinidine | Antiarrhythmic | *Cinchona ledgeriana* |

## PROSPECTS AND CHALLENGES

India and China have pursued a deliberate programme of research and transformation of their traditional medicines into modern pharmaceuticals. Thus they have been able to produce cheap alternatives to western brands. As such they have successfully gained a fair share of the international medicinal drug market.

Promoting a synergy between herbalists, scientists and entrepreneurs can help establish a vibrant pharmaceutical industry rivaling those Asian nations, in view of our diverse and untapped flora. This is a potential large-scale foreign exchange earner if pursued in a sustainable manner. However, despite the existence, abundance, universal awareness and use of herbal/traditional medicine over centuries in most West African countries, there is still a vacuum in their regulation, assessment of safety, efficacy and quality control of their use. Research into the chemistry and biological / pharmacological properties of medicinal plants has not received adequate attention, such that traditional medical practices continue in their undeveloped forms. This needs to be adequately addressed. Screening and development of our Natural Products will enhance national health-care programmes, expand the repertoire of our pharmacopeia and usher us into potential large scale commercial/industrial production of novel pharmaceuticals both natural and synthetic.

In addition Natural Product Chemistry has contributed immensely to the structural elucidation of many compounds used in traditional medicines. This has afforded scientific study of the nature and potential, of these versatile metabolites. Indeed because of their diversity and the fact that any natural product may have more than one pharmacological activity, their full potential is only limited by time, scope and adequate screening of these innumerable God-given medicines. It is hoped that with improved quality, and modernization the traditional medical

system will gradually be integrated into the mainstream of our medical system so as to improve accessibility to healthcare, conserve our scarce foreign exchange and indeed serve as a potential revenue earner as well as providing employment for our teeming masses.

Traditional medicines/herbal remedies have not been fully appreciated and needs to be re-appraised for their potential contribution to our economy and the health-care programme. Thus, It is time that Nigeria promotes the understudy of our traditional medicines as they are a much-needed pool of new pharmaceuticals and a veritable source of foreign exchange.

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Vice-Chancellor Sir, I will like to start by thanking the Lord Almighty for his benevolence, care, support and sustenance. He it is that has granted me favour, good health and protection throughout my life. May his name be praised forever and ever. Amen.

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## PROFILE OF THE INAUGURAL LECTURER

**Prof. Bukar Emmanuel Naroka Dauda** was born in Diko, Gurara L.G., Niger State on 21st September 1948. He attended Primary School Diko and Abuja Secondary School (Suleja). He went to Federal School of Science, Lagos from 1967-1969 for GCE (A-Levels). He was admitted into Ahmadu Bello University (A.B.U.), Zaria to read Agriculture, but was counseled by Prof. M. Olatunji to switch over to Chemistry Department where he obtained a Second Class Upper Degree (1969-1972). He attended the University of Manchester, UK from 1975-1977 for Masters in Organic Chemistry, and again at the University of Manchester, for a PhD. in Natural Product Chemistry (1986-1989) on Commonwealth Academic Fellowship. He took up appointment in the School of Basic Studies as a Lecturer upon graduation in 1972 till 1981. Between 1981-82, he was in Abuja as the Principal, FCT Secondary School, Kwali and later Secondary School, Bwari (1982-1984) before moving to the Federal University of Technology, Minna in 1984 as a Senior Lecturer in the Department of Chemistry. Between 1993-2001 he served as Provost College of Education, Minna. He was the Head of Department of Chemistry, Federal University of Technology, Minna (2004-2010), and at Ibrahim Badamasi Babangida University, Lapai (2010-2011) while on sabbatical leave. He was promoted to the rank of Professor in October 2012. He has been appointed to many boards both at Federal and State levels and has served as External Examiner to some Universities. He is a Member of the Chemical Society of Nigeria; Member the Royal Society of Chemistry, (U.K.); and a Chartered Chemist (U.K.)

He holds two traditional titles - the Magayakin Ija and the Mai Arewan Kwali (FCT).

He is married and blessed with two children.

# Note