Bull. Chem. Soc. Ethiop. **1996**, 10(1), 57-71.
Printed in Ethiopia

ISSN 1011-3294

REVIEW ARTICLE

PROGRESS IN THE CHEMISTRY OF MARKETED TRADITIONALLY USED PLANTS OF ETHIOPIA

Berhanu M. Abegaz[1]

Department of Chemistry, University of Botswana, Private Bag 0022, Gaborone, Botswana

CONTENTS

1. INTRODUCTION

The first paper in chemistry to be published in an international journal from the Department of Chemistry of Addis Ababa University, AAU, is almost certainly that of L.R. Pittwell [1]. It is entitled **Color of Fusion of the Elements** and it appeared in *Chemist Analyst* in 1962. Pittwell, in subsequent years, published papers dealing with emission spectroscopy and the analysis of natural waters from Ethiopia [2,3,4]. Pittwell was an expatriate analytical chemist and he may also have been the first in the Department to acquire a research grade instrument - an emission spectrograph. W.J. Horton, J. Gay and R.M. Baxter would probably feature very

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[1] Permanent address: Department of Chemistry, Addis Ababa University, P. O. Box 1176, Addis Ababa, Ethiopia.

prominently in terms of training the first set of Ethiopian chemists in the early to mid sixties. Baxter, who was the last non-Ethiopian to hold the position of head of department until 1973 (now retired and living in Burlington, Ontario, Canada) has written an interesting account on *Some Thoughts on Traditional Ethiopian Chemistry* which has appeared in this Journal [5].

The mid seventies, witnessed the emergence of Ethiopian chemists who, since then, have made contributions to world science. The training of chemists continued over the years, and despite the unfavourable political climate and the economic difficulties of the seventies and the eighties, the Department developed and thrived. Several items of research grade instrumentation were acquired and funding agencies, particularly SAREC (now SIDA) supported research activities in the chemistry of natural products. In the early eighties we saw the creation of the Chemical Society of Ethiopia and the birth of the *Bulletin of the Chemical Society of Ethiopia*. Today, the Society is one of the most active chemical societies in Africa and the journal, one of the best scientific media serving chemists not only in Ethiopia but throughout Africa. The Department was one of the first in Addis Ababa University to launch postgraduate level of education leading to M.Sc. (1978) and Ph.D. (1985) degrees. Many graduates of the Department are today working in institutions within and outside the country. There are now Ethiopian chemists and pharmacognosists publishing papers on secondary metabolites from plants. Indeed, Ethiopian chemists have every reason to be proud of their achievements and contributions to the building of science in Africa. Just recently, two chemists from the Department of Chemistry (AAU) have won two of the three awards given to date for outstanding scientific achievement of the North America-based Ethiopian Scientific Society.

I have been very fortunate to have been one of the young chemists who came and started working in the Department of Chemistry, at the then Haile Selassie I University in 1973. The first paper in the area of natural products to be published in an international journal (*Phytochemistry)* from the Department of Chemistry is, based on work myself and Berhane Tecle (now Ph.D. working for American Cyanamid Company in New Jersey, USA) conducted in Room 128 of the old quadrangle on *Glinus lotoides* (*Mettere*). It is my singular privilege to be invited by the current Editor-in-Chief of the Bulletin to present a review of the research on natural products done by me and my co-workers and collaborators.

Our effort in the phytochemical studies of marketed plants could have not proceeded very well without the assistance of our colleagues in the Biology Department who provided us with the taxonomic identities of the plants. Several collection trips were jointly undertaken. A large number of markets were visited in various parts of the country. In turn, some of the chemical findings on the plants were also of some benefit to the botanists in their efforts to classify certain genera. Indeed some of the work was done jointly and this has led to joint publications [6,7]. The writing of the Ethiopian flora has resulted in re-examination of the identity of these plant species and as a result the names of some of these plants have now changed. Mostly this has been due to revision of some genera while in one case, it was due to incorrect identification (See Appendix 1).

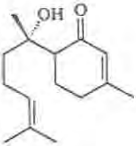
For the purpose of this review, the plants we investigated are grouped into three. The first group are the fragrance and flavor plants, including spices as well as food and beverage additives. The second are the poisonous plants and the third group are the medicinals. To keep this review within reasonable length, it has been necessary to present only a summary of our findings and to leave out the discussion on some plants completely. However a complete list of the plants we studied, together with references to the literature, are given in Appendix 1

2. FRAGRANCE AND FLAVOR PLANTS

2.1 Fragrance Plants

A typical setting on a holiday morning in most Ethiopian homes is the spreading on the floor of freshly cut green grass, just before starting the familiar coffee ceremony. A closer look among the green grass may reveal a few branches of *Ariti (Artemisia rehan)* or a few leaves of *Tej sar (Cymbopogon citratus)*, or of *Addes (Myrtus communis)* which impart a pleasant fragrance to the room. Our studies on the first two essential oil-rich indigenous plants began almost concurrent with the launching of postgraduate studies in the Department in 1978. This led to two publications [8,9] which arose in part from the M.Sc. theses of Dr. Paulos G. Yohannes, now a lecturer in a college in Atlanta, Georgia, USA. A total of 18 constituents were identified from the oil of *Tej sar* which accounted for 80% of the oil. Geraniol was found to be the major constituent (40%) of *Tej sar* oil. A new sesquiterpene, (-)-α-oxobisabolene (**1**) was also identified. The composition of the oil was found to be distinctly different from all other reported geographical varieties of *C. citratus* (where the major constituent is usually citral) and we indicated the close similarity of *Tej sar* oil to that reported in the literature for the oil of *C. nardus*. Our report has prompted a re-examination of the specimen we studied and we now believe that the Ethiopian *Tej sar* that we studied is, *C. nardus*.

Ariti, Artemisia rehan, upon hydro-distillation yielded a characteristic blue oil. The blue color is caused by chamazulene (**2**) which is an artefact produced during steam distillation. Twenty two components comprising 87% of the oil, were identified and the major constituents were found to be davanone (**3**) (44%) and camphor (23%) [10].



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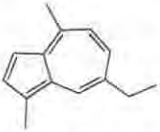

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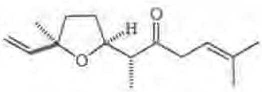

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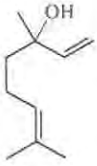
2.2 Flavor Plants

Ethiopia has a rich culture in terms of varieties of foods which are often prepared by blending a number of spices. Many of these spices are intended to impart flavor while in addition they may also act as preservatives, minimizing, if not inhibiting, the growth of bacteria and fungi. One of such spices *is Trachyspermum copticum* (Amharic name - *Nech Azmud*) which inhibits the growth of molds (the kind that grows on bread), giving credence to the Ethiopian empirical wisdom of making this an ingredient during baking [11]. The oil is predominantly made up of carvacrol (**3**) and such phenols have antiseptic properties and have been used to embalm bodies.

One of the interesting studies on essential oils has been on the plant that typifies the most popular dish, namely - *Kitfo*, of the Gurage ethnic group in Ethiopia. *Kitfo* is a type of food prepared from minced beef richly laced with warm spiced butter. A typical flavor that characterizes this dish results from the employment of the plant locally known as *Kosseret* in the preparation of the butter. *Kosseret* is a domesticated plant and is cultivated in home gardens and is also sold in traditional markets. A study of this culturally important plant was undertaken and one of the difficulties encountered in the early stages was taxonomic. The taxonomic name given to this plant is *Lippia adoensis* (Verbenaceae), which is also the same name given to the wild plant locally known as *Kesse*. *Kesse* is used for washing utensils and to impart fresh and spicy fragrance. Our investigations on the chemical constituents of the oils derived from these wild and cultivated plants of *Lippia adoensis* revealed qualitative as well as quantitative differences in the composition of the oils. Qualitatively, the rather sweet fragrance can be used to differentiate the cultivated from the wild form, which has a lemon-like odor . Fourteen compounds representing *ca* 90% of the oil of *Kosseret* and sixteen components representing *ca* 80% of the oil of *Kesse* were identified [12]. Linalool (**4**) is the major component (*ca* 75%) of the oil of *Kosseret* which is totally absent in the oil of *Kesse*. Only trace amounts (0.7%) of the rare monoterpene 2-methyl-6-methylene-2,7-octadien-4-one (ipsdienone, **5**) was present in *Kosseret* while *Kesse* oil contains up to 15%of **5**. Ipsdienone is an important compound since its reduction product, ipsdienol, is the principal component of the sex attractant of the male *Ips confusus* [13]. Based on the chemical and morphological characters the wild and cultivated plants were classified to varietal levels, the wild plant being *L. adoensis* var.*adoensis* and the cultivated, *L. adoensis* var. *koseret* [64].

Several projects were undertaken in collaboration with the Ethiopian Spice Extraction Company. Two of the staff of the Company were trained to M.Sc. level. Projects involved analyses of essential oils and oleoresins derived from exotic black pepper (*Piper nigrum*, Piperaceae), cardamon (*Elettaria cardamomum*, Zingiberaceae), cinnamon (*Cinnamomum verum*, Lauraceae), exotic and indigenous ginger (*Zingiber officinale*, Zingiberaceae), turmeric (*Curcuma longa*, Zingiberaceae) and the indigenous *Korarima* (Ethiopian cardamon, *Aframomum corrorima*, Zingiberaceae). These studies have enabled the establishment of quality standards and have also assisted the Ethiopian Spice Extraction Company in its efforts to find international markets for locally produced essential oils and oleoresins. Two papers were published [14,15] and several reports submitted to the company. It is very gratifying to learn that these efforts have been very useful to the company.



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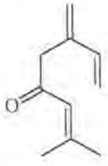

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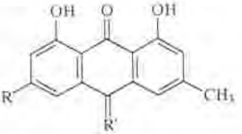
2.3 Studies on *Gesho* - *Rhamnus prinoides*

R. prinoides, (Rhamnaceae) is a plant which is also known to occur outside Ethiopia in Cameroon, Sudan, throughout East Africa to South Africa and Angola and also in Arabia [16]. It is cultivated in Ethiopia and is an important commodity which is sold in almost every traditional market in Ethiopia. Although it is quite common to find *Gesho* cultivations throughout the country, it is worth mentioning that regions in Tigrai, around Kara Kori in North Shoa and Sebeta, just west of Addis Ababa, are important centers of production of *Gesho*. Travelers passing through Kara Kori will undoubtedly notice villagers sitting by the street side chopping *Gesho* branches and allowing them to dry all along the length of the highway passing through the town. The leaves and stems of *Gesho* are ingredients in the making of the traditional fermented beverages *Tella* and *Tej*, respectively. Although it is generally known that *Gesho* imparts the characteristic bitterness of these beverages, more precise understanding of the scientific role of this plant in the brewing process is emerging only very slowly. Coady in 1965 [17] suggested that *Gesho* may have a role in the fermentation process. More concrete information on the microbiological role of *Gesho* came out in two publications [18, 19] where it is claimed that the plant regulates the microflora responsible for the fermentation process. It is also indicated that the bitterness of the brew is directly related to the amount of *Gesho* added. Recently efforts have been made to develop technologies that allow the utilization of extracts from *Gesho* to hop beer [20]. We have observed that care is always taken to remove the fruits of the plant from the leaves and stems during the making of *Tella*. The fruits are, however, used for the treatment of ring worm infections. Prinoidin (**6**), a novel emodinanthrone rhamnoside diacetate and related compounds have been reported from the fruits of *R. prinoides* [21,22]. Our recent investigations on the leaves led to the characterization of the previously unknown naphthalenic compound ß-sorigenin-8-*O*-ß-D-glucoside (**7**), which is responsible for the bitter taste of the leaves [23]. The name 'geshoidin' is proposed for this novel glucoside. The leaves were also found to contain ß-sorigenin (**8**), chrysophanol (**9**), physcion (**10**), emodin (**11**), musizin (**12**), rhamnocitrin (**13**), rhamnazin (**14**), quercetin (**15**) and 3-O-methylquercetin (**16**). The co-occurrence of musizin (**12**) and geshoidin (**7**) in the leaves of the same plant undoubtedly has biosynthetic significance, and most probably musizin is a precursor of geshoidin.



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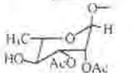
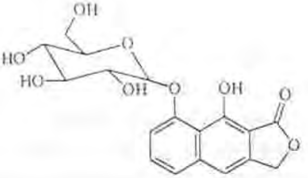
6		H,H
9	H	O
10	OCH_3	O
11	OH	O



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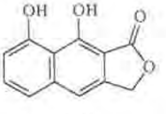

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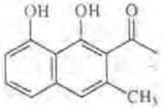

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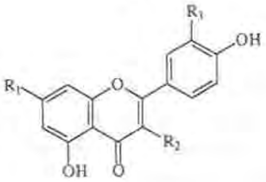


	R_1	R_2	R_3
13	OCH_3	OH	H
14	OCH_3	OH	OCH_3
15	OH	OH	OH
16	OH	OCH_3	OH

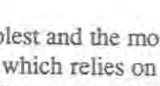
An organoleptic evaluation was made by locating five volunteers who independently confirmed that geshoidin possess bitter properties. It is interesting to note that geshoidin is bitter despite the presence of a glucose moiety in the structure. The toxicity of geshoidin to brine shrimp (*Artemia salina)* was evaluated at seven different concentrations over a range of 64 folds (15 to 1000 μg/ml)[2]. No lethality was observed. Preliminary assay for possible cytotoxicity of geshoidin has also been negative[3]. Although rigorous toxicity tests should be conducted on geshoidin, the results obtained so far suggest that this compound may have commercial potential.

3. POISONOUS PLANTS

3.1 *Lathyrus sativus (Guaya)*

One of the poisonous plants listed in Appendix 1 is the food legume, *Lathyrus sativus*, or grass pea, which is commonly known in Amharic as *Guaya*. In my childhood, I had been made aware of the debilitating effects of this plant. I recall being told not to lay down on the fresh plant, nor to consume any *Kita*[4] made from *Guaya* together with milk as this was thought to lead to enhanced toxicity manifestations. With this background, I was understandably keen to learn about the chemistry of this otherwise very tasty legume. Although the above claims are not scientifically substantiated, we now know a lot more about the chemical composition and toxicological properties of *Guaya*.

As early as 1913, *L. sativus* was reported as a cumulative poison causing paralysis of the lower limbs in man [24]. The human neurological disorder, Lathyrism, resulting from excess consumption of the seeds of grass pea, is caused by the non protein amino acid ß-N-oxalyl-2,3-diaminopropanoic acid (**17**) (ß-ODAP) [25, 26]. There are historical accounts on how several hundred Rumanian jews were intentionally fed with this legume, which resulted in many of them becoming lathyritic [27]. There have also been epidemic levels of lathyrism resulting from high level consumption of grass pea in communities where food shortages occur either due to flooding or drought. One such epidemic occurred in northwest Ethiopia in 1976 [28].



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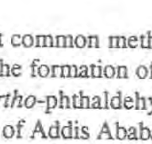

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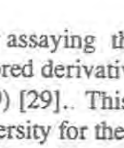

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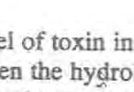

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The simplest and the most common method for assaying the level of toxin in *L. sativus* is that of Rao, which relies on the formation of a colored derivative when the hydrolysis product of ß-ODAP is reacted with *ortho*-phthaldehyde (**19**) [29].. This method has been employed in the Department of Chemistry of Addis Ababa University for the determination of the toxin level in seed samples and also in various food preparations (Table 1). While this method is quite simple and can be used to determine the amount of diaminopropanoic acid (DAP, which is the hydrolysis product of ß-ODAP) in seeds and other samples it also detects any other substance

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[2] We are grateful to Ato Mesfin Bogale of ESTC for making this determination.

[3] We are grateful to Dr. R. Becker of the University of the North, South Africa for this information.

[4] An unleavened bread prepared from the flour of cereals or legumes or a mixture of both.

(like α-ODAP, **18**) that would yield DAP under the hydrolysis conditions.

As early as 1966, Bell and O'Donovan reported that in ethanolic solution ß-ODAP slowly equilibrates with the α-isomer (**18**) and that these substances interconvert more readily when heated [31]. What is particularly interesting about this interconversion is the realization that α-ODAP is non toxic [32]. It is in this regard that our efforts have been geared towards understanding the mechanism of isomerization of ß-ODAP to the non-toxic isomer (α-ODAP).

Table 1. Levels of ß-ODAP in commonly consumed foods in Ethiopia [30] .

Sample type	ß-ODAP levels in mg/100 g		
	Highest	Lowest	Average
Whole seeds	810	282	455
Decorticated seeds	690	239	444
Shiro[*]	429	155	270
Wot[¶]	338	113	190
Nifro[§]	162	70	107

[*] The flour prepared from legumes and used for the preparation of *Wot*.

[¶] The traditional Ethiopian sauce prepared from *Shiro*, meat or other vegetables.

[§] Boiled cereals, legumes or a mixture of both and served as snack.

We found out that NMR spectroscopy can be used to study the isomerization of ß-ODAP to a 60/40 equilibrium mixture of the two isomers (ß and α, respectively) [33,34]. The isomerization of aqueous solutions of ß-ODAP (pH =2.3) to α-ODAP had initially been believed to proceed through the formation of a diketopiperazine (**20**) intermediate [31]. Later it was shown that such an intermediate was unlikely under acidic conditions. This was shown by actually synthesizing the alleged intermediate (**20**) and showing that it was in fact stable under acidic conditions, although at alkaline pH it underwent a reaction to give mixtures of α- and ß-ODAP [35]. Careful examination with particular care to detect intermediates failed. More recently Girma Moges and coworkers [36] have conducted kinetic experiments which provided further insight into the mechanism of this interesting rearrangement.

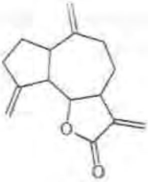
More scientific data have been generated during the last ten years and Ethiopian and other scientists elsewhere have made significant contributions to our current understanding of *Lathyrus* and lathyrism. There are many groups, neurologists, breeders, nutritionists, and chemists presently working on *Guaya* in Ethiopia. The interested reader is referred to a recent volume of the proceedings of an international conference on Lathyrus and lathyrism held in Addis Ababa, Ethiopia [37]. It seems very probable that further research will in the future result in the production of low toxin varieties of grass pea and the eradication of lathyrism.

3.2 *Echinops kebericho, E. amplexicaulis* and other *Echniops* spp.

Kebericho is a fumigant that is sold in traditional markets. Fumigating houses with the tubers of *Kebericho* is believed to get rid of typhoid and fever. Some communities use it as insect repellant. We began our studies by seeking taxonomic information from the National Herbarium (AAU). Our interest on this plant was met with enthusiasm by Dr. Mesfin Tadesse (AAU), who was interested in studying the genus. Although *Kebericho* has been widely used in Ethiopia to the extent of becoming an established article of trade in many regions, a taxonomic description of this plant was lacking. Thus the the name *Echinops kebericho* Mesfin was assigned to it by Dr. Mesfin. We collected several *Echinops* species and undertook a chemotaxonomic study of the genus which was subsequently published [7]. Four of the taxa (*E. kebericho, E. amplexicaulis, E. macrochaetus* and *E. pappii*) were investigated in more detail than the others and the following findings were made. *E. Kebericho* tubers are extremely rich in just one sesquiterpene, namely dehydrocostus lactone (**21**) amounting to *ca* 10% of the dry weight of the tubers (7). This sesquiterpene has been reported from a few other sources but was principally known as the major constituent of Japanese costus root oil. It is a biologically active compound having antitumor, antimutagenic and cytotoxic properties.

Echinops spp are characterized by heads which can range from 3 -5 (*E. pappii*) in some species to 10-15 cm in diameter in others (e.g. *E. longisetus*). *E. amplexicaulis* can be distinguished by a mid-size head (6 -10 cm) and is reddish brown. It is widely sought as a medicinal plant and is often violated on account of its brilliant color and solitary growth. It may very well be regarded as an endangered plant. The tubers are also rich in a sesquiterpene lactone known as costunolide (**22**). *E. kebericho* and *E. amplexicaulis* present themselves as commercially viable sources for the two sesquiterpene lactones **21** and **22** if successful efforts are made to develop suitable uses for these compounds. Modest efforts are presently underway, but they need to be intensified.

We were able to isolate and identify several thiophene derivatives from several *Echinops* species. Of particular significance may be the isolation of α-terthienyl (**23**) from *E. macrochaetus* and *E. pappii* [38]. A recent book [39] provides interesting insight into the properties of naturally occurring di- and tri-thiophenes. According to this source, α-terthienyl is a potent antiviral compound although its activity was shown to be virus dependent. It is phototoxic to gram-positive and gram-negative micro-organisms and is also fungicidal. It kills nematodes and its activity in exterminating mosquitoes has been impressive. The light activated terthienyl is more active than DDT and Malathion although less active than Dursban.



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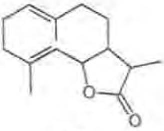

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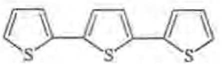

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4. MEDICINAL PLANTS

4.1 Anthelmintic plants - *Glinus lotoides* and *Salsola somalensis*

The most frequently encountered medicinal plants in traditional markets are, understandably, those that are used for ridding the body of parasitic worms and infections. Ethiopia has a rich plant lore in terms of plants that are used to deal with such problems. One of the most outstanding and very old remedy is *Hagenia abyssinica* (*Kosso*[5] in Amharic). This plant attracted attention ever since James Bruce gave a description of it in his book in 1793 and also

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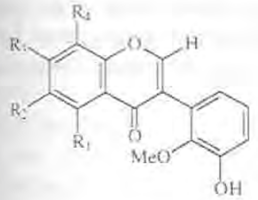
[5] *Kosso* is also the name used to refer to the beef tape-worm parasite, *Taenia saginata*.



	R_1	R_2	R_3	R_4
24	OH	H	OMe	OMe
25	OH	OCH_2O		H
26	OH	OMe	OMe	OMe
27	OH	OMe	OMe	H
28	OH	H	OMe	OH
29	OMe	H	OMe	OH
30	OH	OH	OMe	H
31	OMe	OH	OH	H
32	OMe	OMe	OMe	H
33	OMe	OMe	OH	H
34	OMe	OH	OMe	H

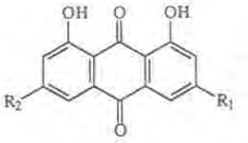
described the medicinal attributes of this plant as used in Ethiopia. Several scientists have worked on this plant for over one hundred years and there are also historical accounts of its use not only in Ethiopia but also in several European countries. Other popular medicines are *Mettere (Glinus lotoides)*, *Dima (Salsola somalensis)*, *Kechemo (Myrsine africana)*, *Enkoko (Embelia schimperi)*, etc. We have investigated two of these plants. The anthelmintic fraction of the aqueous ethanol extract of the seeds of *G. lotoides* yielded a novel triterpenoid glycoside [40]. One of the most impressive traditional medicine for the treatment of tape-worm infestation is *Dima (S. somalensis)*. The roots are used as tooth-stick and the juice is swallowed. The parasite is often expelled in the same day. However, we have observed that it is possible to develop mild allergic reactions to this crude drug, a phenomenon not unusual when dealing with plants belonging to the family Chenopodiaceae. Biological test on the various extracts of the roots indicated that the chloroform and ethyl acetate extract residues were very active against the test organism - *Taenia saginata*. The residue from these extracts yielded, in addition to known compounds, lupeol, β-sitosterol, β-amyrin, 12 novel isoflavonoids (**24-34**) [41,42]. This compounds were considered rather unusual because they lack oxygenation at 4'-position of ring B. Three of these compounds were tested for anthelmintic activity and were found to possess only moderate activity. One of the three that were tested in Addis was sent to Hoechst laboratories in Germany and after rigorous testing, it was decided that this compound is unworthy of further follow up as an anthelmintic. Paucity of material precluded conducting tests on pure samples of the remaining 9 compounds.

4.2 Anthranoids from Ethiopian *Senna*

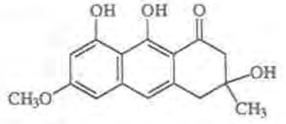
Senna is an important genus which has yielded important purgative drugs. The most famous is *Cassia senna (C. acutifolia)*, known as Alexandrian senna or *Cassia angustifolia*, also known as Tinnevelly senna, or a mixture of the two species. The biologically active constituents of *Senna* are the hydroxyanthracene glycosides known as sennosides. Many members of *Senna* have for a long time been considered in a broader classification together with the now separate genera of *Cassia* and *Chamaechrista* [16]. We have studied four taxa, namely: *S. didymobotrya*, *S. septemtrionalis*, *S. longiracemosa* and *S. multiglandulosa* and have identified several anthraquinones, anthrones, preanthraquinones, and novel bianthraquinones.

4.2.1 *Senna didymobotrya*

The leaves yielded the monoanthracene derivatives, chrysophanol (**9**), physcion (**10**), aloe-emodin (**35**), fallacinol (**36**), rhein (**37**), parietinic acid (**38**) and the preanthraquinone torosachrysone (**39**). The isolation from this taxon of fallacinol (**36**) and parietinic acid (**38**) constituted the first report on the occurrence of these two substances from higher plants [62]. Previously they had been reported from lichens and cultures of *Eurotium echinolatum* [43]. Chrysophanol, aloe-emodin and rhein had been reported earlier from this plant by Egyptian workers [44].



	R_1	R_2
35	CH_2OH	H
36	CH_2OH	OCH_3
37	COOH	H
38	COOH	OCH_3



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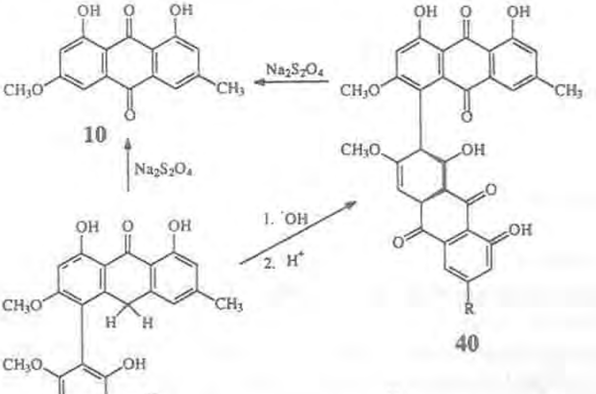
4.2.2 *Senna septemtrionalis* (synonyms: *Cassia laevigata, C. floribunda, S. floribunda)*

This legume has been shown to contain 8-mono- and digalactosides of physcion as well as chrysophanol and emodin [45,46]. Our studies on the leaves yielded the common anthraquinones, emodin, and physcion, in addition to two novel bianthraquinone pigments for which the names floribundone-1 (**40**) and floribundone-2 (**41**) were given [47]. These compounds represented the second set of examples of an anthraquinone dimer with a 5-7'-bianthracene linkage. The mixture of floribundone-1 and 2 was difficult to separate and it was observed that a solution of floribundone-2 (**41**) was easily oxidized to floribundone-1 (**40**). The



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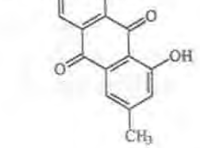

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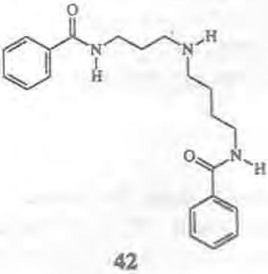

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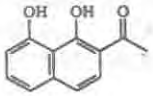

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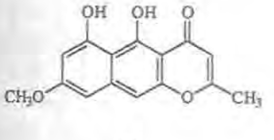
mixture of the two compounds was also cleaved to physcion by reaction with sodium dithionite. The reductive cleavage of floribundone-2 (**41**) presumably yields physcion anthrone which would be oxidized to physcion during work up. Alternatively and most probably, the oxidation of floribundone-2 (**41**) to floribundone-1 (**40**) may take place faster than the cleavage of reaction to yield physcion directly. Also isolated from the leaves was a N^1,N^8-dibenzoylspermidine (**42**) and other traces of pigments which at present are unidentified. Floribundone-1 (**40**) has since then reported from Mexican *Senna* species [48] and an atropi-isomer of it from *Cassia torosa* of Japanese origin [49].

4.2.3 *Senna longiracemosa* (synonym: *C. longiracemosa*)

The leaves yielded chrysophanol (**9**), physcion (**10**), torachrysone (**43**), rubrofusarin (**44**), nataloe-emodin (**45**) and the bianthraquinones 10,10'-bichrysophanol (**46**), 10,10'-chrysophanol-physcion (**47**), 10,10'-chrysophanol-isophyscion (**48**), 10,10'-biisophyscion (**49**) and 10-hydroxy-10,7'-(chrysophanol-anthrone)-chrysophanol (**50**) [50]. Compounds **45**, **47**, **48**, and **49** were reported for the first time as new natural products and **50** had not been reported from the genus *Senna* previously. The root bark of this plant also yielded 2-methoxystypandrone (**51**) in addition to **9**, **10**, **11**, **46**, **47**, **48**, and **49**.



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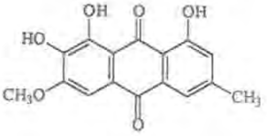

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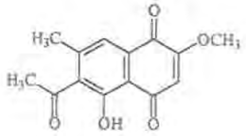

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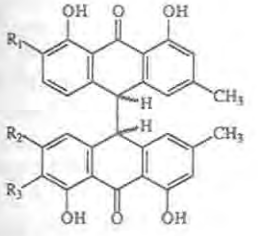
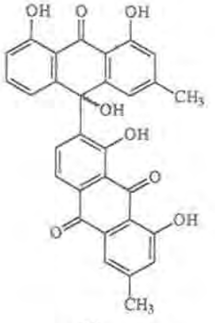


	R_1	R_2	R_3
46	H	H	H
47	H	OMe	H
48	H	H	OMe
49	OMe	H	OMe



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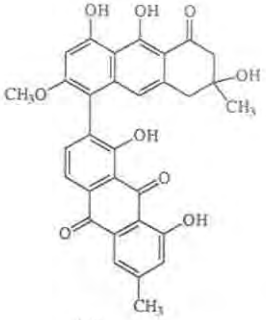

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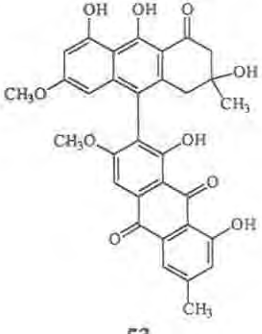
4.2.4 *Senna multiglandulosa*

Our earlier investigation of the leaves and stems had yielded chrysophanol, emodin, physcion and four bianthraquinones floribundone-1 (**40**), torosanin-9,10-quinone (**52**), anhydrophlegmacin-9,10-quinone (**53**) and the novel 1,4-quinone 9-(physcion-7-yl)-5,10-dihydroxy-2-methoxy-7-methyl-1,4-anthraquinone (**54**) [51]. Torosanin-9,10-quinone had been reported as an oxidation product of torosanin obtained from the unripe seeds of *Cassia torosa* [52]. The most interesting compound obtained from this plant is the 1,4-quinone **54** for which the name sengulone is proposed. This is the first example of an anthraquinone dimer containing a 1,4-quinone moiety. We were initially intrigued by the possibility of ano structure (55) which closely fitted the spectroscopic data for compound **54**. That the compo isolated from the leaves and stems was **54** and not **55** was unequivocally established by di experiments. Thus irradiation of the Me signal at 2.49 led to enhancement of the Ar-H sig at 7.70 and 7.11, and irradiating the other Me signal at 2.32 led to the enhancement of two H signals at 6.95 and 6.79. On the other hand, irradiation of the methoxy signals at 3.82 3.88 each led to a corresponding increase in the signals of only one proton at 6.20 and 7 respectively. These nOe data enabled us to reject structure **55** in favour of **54** for the struc of sengulone. Survey of the literature indicates that 1,4-anthraquinones have not been repo from higher plants. There are, however, a few 1,4-anthraquinones isolated from *Asperg cristatus* [53].

In a subsequent study we undertook to examine the chemical constituents of the seeds c *multiglandulosa* and we were able to identify an isomer of sengulone (named iso-sengul which fits the rejected alternative structure described above (55) in addition to other pigme namely physcion, torosachrysone (**39**), the bianthraquinones floribundone-1 (**40**) anhydrophlegmacin-9,10-quinone (**53**). Difnoe data were consistent with the structure assi for **55** as were homonuclear 2D-NMR measurements (COSY45) [54].



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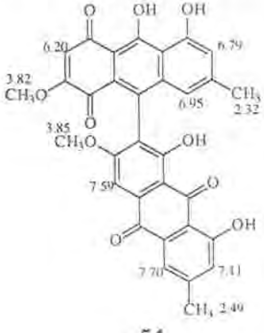

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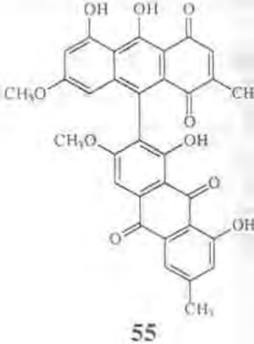

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5. ACKNOWLEDGMENTS

Most of the work reported in this review was conducted in the Department of Chemist Addis Ababa University (AAU). The Swedish Agency for Research Cooperation Developing Countries (SAREC, now SIDA) is gratefully acknowledged for a generous rese grant received through the Ethiopian Science and Technology Commission (ESTC) and A The author also acknowledges the contributions of several coworkers and internati collaborators whose names are mentioned in the references. Ato Merhatibeb Bezabih read manuscript and made valuable suggestions.

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