



主编 王海英 刘志明
主审 杨国亭

ZHIWU HUOXINGWU YINGYU DAODU

植物活性物英语导读



东北林业大学出版社

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前 言

我们的祖先与大自然亲密接触,并且深谙依赖安全的、永续利用的自然环境生存的道理,基于人与自然和谐的理念,环境友好的天然产品开发迫在眉睫,原料的消费和再生之间的平衡,废物的产生和它在环境中的生物学消除之间的平衡等都是产品开发中亟待解决的问题。本书编写的目的在于提供植物活性物质的提取、分离、分析和鉴定的新思路、新方法,提高本科生和研究生的专业外语阅读能力,培养天然产品开发的技术人才。

本书共分 20 个单元,每个单元由一篇课文和一篇阅读材料组成。阅读材料提供与课文相应的背景知识或是课文的续篇,以进一步拓展课文的内容。根据课文与阅读材料的内容,配有相应的练习题、注释和词汇表。课文与阅读材料共计 40 篇,均选自 2000 年以来出版的教科书、著作及专业期刊、科技报告等,并注有网址,方便查阅。

本书由王海英、刘志明主编。其中,第 1 单元到第 12 单元由东北林业大学林学院王海英副教授编写,第 13 单元到第 20 单元以及附录和词汇表由东北林业大学材料科学与工程学院刘志明副教授编写,全书由王海英负责统稿,杨国亭教授审稿。

本书可以作为植物资源学、林产化工、材料化学、食品科学与工程专业本科生的专业外语教材或参考教材,可供植物活性物提取以及植物提取物化学与精细化学品合成研究方向的研究生使用,也可从事天然产品开发的科技人员提供参考。

感谢韩国江原大学 김성문 博士、KOSEF (Korea Science & Engineering Foundation) with the 2006 Korea - China Young Scientist Exchange Program 和东北林业大学出版社对本书编写和出版的支持和帮助。感谢本书中所选阅读材料的作者们为植物天然产品开发的研究者们提供一个广阔的研究平台!

限于水平,疏漏、不妥之处在所难免,恳请读者批评指正。

编者

2007 年 9 月

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Unit 1 Chemistry of Essential Oils (I)

What Is an Essential Oil?

Essential oil, also defined as essence, volatile oil, etheric oil or aetheroleum, is a complex mixture of volatile constituents biosynthesized by living organisms. Essential oils can be liberated from their matrix by water, steam and dry distillation, or expression in the case of citrus fruits. Their occurrence and function in nature is still a question and the subject of ongoing research. However, there is evidence that organisms produce essential oils for defence, signalling or as part of their secondary metabolism. As a consequence essential oils comprise an important bioresource for renewable natural products.

Extracts of aromatic plant or animal materials obtained using organic solvents or fluidized gasses are not considered as essential oils. Concretes, absolutes, spice oleoresins, *etc.* which can be classified as aromatic extracts are not covered in this chapter.

Essential oils, their fractions and their isolates are utilized in flavour and fragrance, food, perfumery, cosmetics and toiletries, fine chemicals, pharmaceutical industries and therapy. They are used as such or in diluted forms in the budding aromatherapy sector.

Essential oils may comprise volatile compounds of terpenoid or non-terpenoid origin. All of them are hydrocarbons and their oxygenated derivatives. Some may also contain nitrogen or sulphur derivatives. They may exist in the form of alcohols, acids, esters, epoxides, aldehydes, ketones, amines, sulphides, *etc.* Monoterpenes, sesquiterpenes and even diterpenes constitute the composition of many essential oils. In addition, phenylpropanoids, fatty acids and their esters, or their decomposition products are also encountered as volatiles.

Owing to their liquid nature at room temperature, essential oils are called as such. They should not be confused with fixed oils or fatty oils, which are composed of a naturally occurring mixture of lipids which may not necessarily be volatile. Therefore, essential oils differ entirely both in chemical and in physical properties

from fatty oils. Essential oil evaporates completely when dropped on filter paper; however, fixed oil leaves a permanent stain which does not evaporate even when heated.

Essential oils occur mainly in aromatic plants. A few of them are found in animal sources, e. g. musk, civet and sperm whale, or are produced by microorganisms. The Council of Europe describes "essential oil" as a product obtained from "vegetable raw material". Owing to a ban on animal-based flavour and fragrance materials, essential oils of trade are entirely of plant origin.

Among many others, well-known families rich in essential oil bearing species are Apiaceae, Asteraceae, Cupressaceae, Hypericaceae, Lamiaceae, Lauraceae, Myrtaceae, Pinaceae, Piperaceae, Rutaceae, Santalaceae, Zingiberaceae and Zygophyllaceae.

In plants, essential oils occur in oil cells, secretory ducts or cavities, or in glandular hairs. In some cases, they are bound with carbohydrates in the form of glycosides. In such cases, they must be liberated by hydrolysis of the glycosidic bond. This is done by allowing enzymatic reactions to take place during wilting prior to distillation of fresh plant materials. Mosses, liverworts, seaweeds, sponges and fungi have also been shown to contain essential oils. Besides higher plants, some terrestrial and marine animals, insects, fungi and microorganisms are also known to biosynthesise volatile compounds.

Essential oils are frequently associated with gums and/or resins. They are freed from such products by distillation.

Essential oil constituents can be classified as terpenoids and non-terpenoid hydrocarbons.

Non-terpenoid Hydrocarbons

Non-terpenoid hydrocarbons found in essential oils such as short chain alcohols and aldehydes are formed by metabolic conversion or degradation of phospholipids and fatty acids.

Hydrocarbons consist of carbon and hydrogen. They may also contain oxygen, nitrogen or sulphur. The simplest hydrocarbon is methane (CH_4), which is a colourless and odourless highly flammable gas. The carbons are connected by single, double or triple bonds to form higher molecular weight hydrocarbons. Saturated homologous straight-chain structures are called alkanes, while their

unsaturated forms are called alkenes. Alkenes show isomerism owing to the position of the hydrogen atoms attached to the double bond as in the examples *trans*-but-2-ene and *cis*-but-2-ene.

Molecules with three carbon atoms can only form a straight chain; however, with four carbons or more they can form both straight and branched chains. Then, their naming also changes accordingly. Isoprene is one such molecule; it is chemically 2-methyl-1,3-butadiene.

Terpenoids

Terpenes, also called isoprenoids, are one of the largest classes of natural chemicals formed by head-to-tail rearrangement of two or more isoprene molecules. More than 30 000 terpenoids have been isolated from plants, microorganisms and animals. They are important constituents of essential oils. Molecules formed from two isoprene molecules are called monoterpenes ($C_{10}H_{16}$). C_5H_8 compounds are hemiterpenes. Sesquiterpenes contain three isoprene units; hence they have the formula $C_{15}H_{24}$. $C_{20}H_{32}$ compounds formed from four isoprene units are diterpenes. Heavier terpenes like diterpenes are generally not found in essential oils. Isoprene itself is considered the only hemiterpene, but oxygen-containing derivatives such as prenol and isovaleric acid are hemiterpenoids, too.

Kekulé, in 1880, was the first scientist to name $C_{10}H_{16}$ compounds as “terpenes”, because of their existence in turpentine. His assistant Wallach (1910 recipient of the Nobel Prize in Chemistry) hypothetically proposed in 1887 that terpenes were constructed via two or more isoprene units. Three decades later, Robinson (1947 recipient of the Nobel Prize in Chemistry) perfected Wallach’s “isoprene rule” by suggesting that the isoprene units should be connected in a head-to-tail fashion. A few years later, Ruzicka (1939 recipient of the Nobel Prize in Chemistry) proposed in 1950 the “biogenetic isoprene rule”, further developing Wallach’s hypothesis. The rule reiterated the formation of terpenes by head-to-tail rearrangement of two or more isoprene units. This rule stipulates that the terpenoids are derived from aliphatic precursors such as geraniol for the formation of monoterpenes, farnesol for the sesquiterpenes, geranylgeraniol for the diterpenes and squalene for triterpenes. It is interesting to note that three terpene scientists received the Nobel Prize in Chemistry within a span of 37 years.

Biosynthesis of Terpenes

Terpenes, biogenetically, arise from two simple five-carbon moieties. Isoprenyldiphosphate (IPP) and dimethylallyldiphosphate (DMAPP) serve as universal precursors for the biosynthesis of terpenes. They are biosynthesized from three acetylcoenzyme A moieties through mevalonic acid (MVA) *via* the so-called *mevalonate pathway*. About 10 years ago, the existence of a second pathway leading to IPP and DMAPP was discovered involving 1-deoxy-*D*-xylulose-5-phosphate (DXP) and 2*C*-methyl-*D*-erythritol-4-phosphate (MEP). This so-called *non-mevalonate* or *deoxyxylulose phosphate pathway* starts off with the condensation of glyceraldehyde phosphate and pyruvate affording DXP. Through a series of reactions, IPP and DMAPP are formed, respectively.

IPP and DMAPP lead to geranylpyrophosphate (GPP), which is an immediate precursor of monoterpenes. The formation of nerylpyrophosphate (NPP) from GPP gives rise to a wide range of acyclic, cyclic, bicyclic or tricyclic skeletons. Reactions like rearrangement, oxidation, reduction and hydration *via* various terpene cyclases result in the formation of numerous terpene derivatives. Condensation of GPP and IPP leads to farnesylpyrophosphate (FPP), the immediate precursor of sesquiterpenoids. Likewise, FPP and IPP are conducive to diterpenoids.

The mevalonate-independent pathway is present in most bacteria and all phototropic organisms. In higher plants and most algae both pathways run independently. The mevalonate pathway is located in the cytoplasm and is responsible for the biosynthesis of most sesquiterpenoids. The mevalonate-independent pathway, in contrast, is restricted to the chloroplasts where plastid-related isoprenoids such as monoterpenes and diterpenes are biosynthesized *via* this pathway.

Monoterpenes

Monoterpenes are formed from two attached isoprene units: 2, 6-dimethyloctane as the simplest skeleton. Thus, they can be acyclic or linear like β -myrcene, (*E*)- β -ocimene, (*Z*)- β -ocimene, and *allo*-ocimene. Or they can be cyclic, meaning ring-forming, such as in the simplest form like *p*-menthane or *p*-cymene. Monocyclic *p*-menthane, *p*-cymene, bicyclic δ -3-carene and tricyclic

tricyclene type monoterpenes are found in essential oils.

Aromatic monoterpenes which contain a benzene ring like *p*-cymene, carvacrol, thymol and phenylethyl alcohol are common constituents of many essential oils, e. g. oregano (*Origanum* sp.), thyme (*Thymus* sp.), savory (*Satureja* sp.) and rose (*Rosa* sp.) oils. Another important constituent class of essential oils is phenylpropanoids. They are not considered as terpenoids owing to their different biogenetic origins, which will be mentioned later.

According to the *Dictionary of Natural Products* (DNP), there are 25 different classes of monoterpenes.

The biosynthesis of different classes of monoterpenes formed from α -terpinyl cation.

Acyclic Monoterpenes

These regular monoterpenes constitute a small class which includes the trienes myrcene and ocimenes and the alcohols geraniol, nerol, citronellol, linalool, etc.

Citral is the naturally occurring mixture of the aldehydes geranial and neral. Citronellal is another acyclic aldehyde within this grouping. Variation of the 2,6-dimethyloctane skeleton is easily noticeable.

Cyclic Monoterpenes

Cyclic monoterpenes can be classified in three subgroups according their ring size such as;

- (1) Monocyclic monoterpenes
- (2) Bicyclic monoterpenes
- (3) Tricyclic monoterpenes

Monocyclic Monoterpenes

p-Menthane monoterpenes which possess the 1-methyl-4-isopropyl-cyclohexane skeleton comprise the largest group of naturally occurring monoterpenes.

p-Menthadienes are limonene, α -terpinene, β -terpinene, γ -terpinene, terpinolene, α -phellandrene and β -phellandrene resulting from different rearrangements of the α -terpinyl cation. This group is also classified among the monoterpene hydrocarbons.

Aromatic monoterpenes such as *p*-cymene and its hydroxylated derivatives thymol and its isomer carvacrol always occur along with α -terpinene, γ -terpinene and terpinen-4-ol. Metabolites, like *p*-cymene-8-ol and cuminyl alcohol may also be derived from *p*-cymene.

Other important members of this class include oxygenated derivatives such as α -terpineol, menthol, isopulegol and *cis*-hexahydrocuminyl alcohol, also classified as monoterpene alcohols.

Aldehydes in this group are as follows: carvone, dihydrocarvone, isomenthone, piperitone, pulegone (piperitenone), isopulegone.

Bicyclic Monoterpenes

1,8-cineole as well as 1,4-cineole are cyclic ethers. All including ascaridol are bicyclic oxygenated monoterpenes.

Pinane monoterpenes are bicyclic monoterpenes resulting from intramolecular rearrangement of the α -terpinyl cation yielding the [3.1.1] bicyclic system. α -Pinene and β -pinene are the main constituents of turpentine oil from pines. They occur widely in essential oils.

The bornane-, camphane- and fenchane-type monoterpenes possess the [2.1.1] bicyclic skeleton formed by different cyclization of the terpinyl cation. Important members include borneol, isobornyl acetate, camphene, camphor, fenchone.

Thujane-type monoterpenes, unusual monoterpenes with a cyclopropane ring in a bicyclo[3.1.0] skeleton, are formed from the terpinen-4-yl cation directly or via the sabinyl cation. Important members include α -thujene, sabinene, the *cis* isomer of sabinene hydrate, sabinol, sabinylacetate, α -thujone, β -thujone and isothujanol.

Carane-type monoterpenes possess a cyclopropane ring in a bicyclo[4.1.0] skeleton. δ -3-Carene is a common constituent in various essential oils.

Tricyclic Monoterpenes

Tricyclene or 1,7,7-trimethyltricyclo[2.2.1.0^{2,6}]heptane, is a good example which frequently occurs in various essential oils.

Irregular Monoterpenes

There are two major types of irregular monoterpenes:

1. The substituted cycloheptane monoterpenes, also called tropones. Eucarvone, nezukone (4-isopropyl-2,4,6-cycloheptatrienone) and γ -thujaplicin most probably arise by an unknown ring expansion of the cyclohexane skeleton.

2. The other major group of irregular monoterpenes is formed by non-head-to-tail fusion of isoprene units. Important members include Artemisia ketone, santolinatriene, chrysanthemol, yomogi alcohol and lavandulol. Lavandulane-type compounds occur in the families Lamiaceae (Labiatae) and Apiaceae (Umbelliferae), while chrysanthemane, artemisane and santolinane types occur in the family Asteraceae (Compositae).

Sesquiterpenes

Sesquiterpenes are formed by the addition of one more isoprene units to a monoterpene molecule, and thus have the molecular formula $C_{15}H_{24}$. There are linear, branched or cyclic sesquiterpenes. Sesquiterpenes are unsaturated compounds. Cyclic sesquiterpenes may be monocyclic, bicyclic or tricyclic. They are the most diverse group among the volatile terpenoids. The DNP treats sesquiterpenoids in 147 different structural types.

Acyclic Sesquiterpenes

β -farnesene is a constituent of hops oil and many other oils. α -farnesene is the structural isomer.

Farnesol is widely distributed in flower oils such as rose, acacia and cyclamen.

Nerolidol is isomeric with farnesol and is found in neroli oil and many other oils. Its *E* isomer is more frequently found in nature than its *Z* isomer.

Monocyclic Sesquiterpenes

Bisabolene-type sesquiterpenes, e. g. α -bisabolene, are widely distributed in nature. This sesquiterpene hydrocarbon is a constituent of bergamot, myrrh and a wide variety of essential oils. Its oxygenated derivatives α -bisabolol [6-methyl-2-(4-methyl-3-cyclohexane-1-yl)-5-hepten-2-ol] and β -bisabolol [4-methyl-1-(6-

methylhept-5-en-2-yl) cyclohex-3-enol] are found abundantly in chamomile.

Zingiberene [5-(1,5-dimethyl-4-hexenyl)-2-methyl-1,3-cyclohexadiene], is a constituent of ginger oil.

Lanceol or 2,7(14),10-bisabolatrien-12-ol is a primary alcohol found in the oil of sandalwood (*Santalum lanceolatum*). *Z* and *E* isomers exist.

Bicyclic Sesquiterpenes

Cadinene is a trivial name of a number of isomers which occur in a wide variety of essential oils e. g. cubeb oil. Actually, it is derived from the Cade juniper (*Juniperus oxycedrus* L.). The cadalene (4-isopropyl-1,6-dimethyldecahydronaphthalene) carbon skeleton is the base. Prominent stereochemical isomers are α -cadinene, γ -cadinene and δ -cadinene. This group is also known as naphthalene-type sesquiterpenes.

α -selinene, β -selinene, γ -selinene and δ -selinenes are found in celery oil and many other oils.

α -eudesmol, β -eudesmol and γ -eudesmol are tertiary alcohols found in many oils. They are practically the oxygenated forms of selinenes.

α -cyperone is a sesquiterpene ketone found in the essential oil of the tubers of *Cyperus rotundus*. Further hydroxylated derivatives such as α -cyperol and isocyperol can be found along with another ketone with a tricyclic unusual skeleton, namely cyprenone.

The bicyclic ketone eremophilone was first isolated from wood oil of *Eremophila mitchellii*. It is also found in many other oils. *allo*-eremophilone is also structurally related.

The azulenes are a group of bicyclic sesquiterpenes which are responsible for the blue colour of essential oils. They contain highly conjugated five- and sixmembered aromatic carbon rings fused together. Chamazulene, the bluecolouring principle of chamomile oil, is actually formed from matricin during distillation, through the carboxylic acid intermediate, whereas guaiazulene or 1,4-dimethyl-7-isopropylazulene is found in geranium oil.

Vetivanes are sesquiterpene ketones occurring in vetiver oil. Vetivane is basically a spiro decane. Although structurally different as in the case of α -vetivone and β -vetivone, they are characteristic compounds present in vetiver oil. Analogues such as α -vetispiroene and β -vetispiroene may occur as well.

The tertiary alcohol guaiol, also called 2-(1,2,3,4,5,6,7,8-octahydro-1,4-dimethylazulen-7-yl) propan-2-ol, is found in guaiacum wood oil.

Miscellaneous Sesquiterpenes

Caryophyllene, a common constituent of essential oils, was first isolated from clove oil. β -caryophyllene [(E) -caryophyllene] is the most widely encountered form of caryophyllenes. Caryophyllene derivatives are characteristic constituents of most birch oils.

Humulene is isomeric with caryophyllene. First isolated from hops oil (*Humulus lupulus*), it is a common constituent of essential oils.

Diterpenes

Head-to-tail rearrangement of four isoprene units results in the formation of diterpenes ($C_{20}H_{32}$). Diterpenes are generally found in resins, e. g. pimaric acid and abietic acid. Some diterpenoids are also constituents of essential oils, e. g. phytol. Like sesquiterpenes, diterpenes are heavier than monoterpenes; therefore, they require more energy to go to the vapour phase. For this reason, longer distillation times are necessary for their recovery. The DNP lists 118 different structural types for diterpenoids. Important diterpenes found in essential oils will be detailed.

Acyclic Diterpenes

Phytol, a diterpene alcohol (3, 7, 11, 15-tetramethyl-2-hexadecen-1-ol), occurs in two isomeric forms: *trans*-phytol and *cis*-phytol. Phytol was first isolated at the beginning of the nineteenth century during esterification of the chlorophyll molecule. It is a constituent of nettle and many essential oils. Another acyclic diterpene, geranylcitronellol, also occurs in essential oils.

Cyclic Diterpene

Camphorene, which is a constituent of camphor oil, is identical to dimyrcene. Several dimyrcene derivatives are constituents of pistachio oils. The gum resin of *Commiphora mukul* furnishes essential oil (0.4%) consisting chiefly of myrcene 4 and "dimyrcene".

Bicyclic Diterpenes

Sclareol, a ditertiary glycol, is a constituent of clarysage (*Salvia sclarea*) oil. The diterpene ketone sclareolide and the lactone ambrosia are important (bio) synthetic derivatives found in clarysage extract.

Manool and manoyl oxide are found in pine oils. They are common diterpenes encountered in many essential oils.

Labdane, abienol and labdanediol are representatives of volatile labdane derivatives.

Tricyclic Diterpenes

Kaur-15-ene, kaur-16-ene and phyllocladene are encountered in essential oils.

The diterpene pimaradiene and sandaracopimaradiene (or isopimaradiene) are found in some essential oils.

(Selected “essential oil research” from <http://www.springerlink.com/home/main.mpx>, K. Hüsnü Can Başer, Fatih Demirci, Department of Pharmacognosy, Faculty of Pharmacy, Anadolu University, 26470 Eskişehir, Turkey, to be continued)

New Words and Expressions

essential	adj. 本质的, 实质的, 基本的, 提炼的, 精华的; n. 本质, 实质, 要素, 要点
essential oil	n. (香)精油
essence	n. 基本, [哲]本质, 香精
etheric	adj. 醚的
organism	n. 生物体, 有机体
distillation	n. 蒸馏, 蒸馏法, 蒸馏物, 精华, 精髓
metabolism	n. 新陈代谢, 变形
comprise	v. 包含, 由……组成
concrete	adj. 具体的, 有形的; n. 混凝土; v. 用混凝土修筑, 浇混凝土, 凝结
oleoresin	n. 油性树脂, 树脂油
aromatic	adj. 芬芳的