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PRODUCTION, CHEMISTRY AND SENSORY PROPERTIES OF NATURAL ISOLATES

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1. INTRODUCTION

Natural isolates may for instance concern distilled essential oils, cold-pressed oils, extracted oleoresins, concretes and absolutes, plant - or animal exudes, and tinctures. The main aspects of the production of these isolates are the preparation of the plant material, the isolation methods, the yields and economics and the quality control.

The preparation of the plant material may concern e.g.: harvesting, threshing, chipping, drying, grinding, hydrolysis and fermentation. During these preparations various volatile compounds, such as unsaturated aliphatic alcohols and aldehydes, mono- and sesquiterpene oxides, coumarins, polyfunctional benzenoids and irones, can be formed.

Citrus oils can be produced by expression such as the Italian pellatrice and sfumatrice methods, the American Brown oil extractor, and the FMC apparatus. The biogenesis of limonene in citrus oils will be discussed. Leaf, seed and flower oils are manufactured by steamdistillation, hydrodistillation and hydrodiffusion. The production of mint oil, bitter orange leaf & flower oil and rose oil will be shown. The chemistry of the olfactively character-impact compounds in these oils will be demonstrated. Biogenesis of the following characteristic compounds in Mentha species will be discussed: menthone and menthol in peppermint and cornmint, carvone in spearmint, menthofuran in watermint, pulegone in pennyroyal.

Other natural isolates, such as oakmoss absolute, treemoss absolute and labdanum gum are manufactured by solvent extraction, and for instance rose and bitter orange flower oils can be produced by supercritical fluid carbondioxide extraction. The formation of volatile compounds, such as amberoxide etc., by acidic photochemical oxidation on the labdanum plants will be discussed. Modern continuous distillation and extraction processes are practiced today.

Yields and economics of volatile naturals will be commented. Ranges and anomalies in the yields are noticed. Scope and limitations of the economics, such as raw material, capital, energy and labour costs will be discussed.

The physico-chemical standards for many volatile natural isolates oils have been published. The analysis of rose oils and bitter orange flower oils by modern spectroscopic techniques will be treated. The odour intensities of the oils were determined.

1.1. Importance of natural isolates

Essential oils are mixtures of volatile compounds isolated from plant- and animal materials. It as been published that about 350,000 different plant species should exist (1), and from these plant species approximately 60,000 should be medicinal plants and about 17,500 (5%) should be aromatic plants (2). The practical use of medicinal plants is estimated at a number of 10,000. (see Tables 1-4).

About 300 different plantspecies are used for the production of essential oils for the food, flavour and fragrance industry. The annual world production of volatile oils is estimated at around 50,000 tons (3). This production, however, could be up to 100,000 tons with a value of about 1 billion $, based on the figures of the world consumption (2,3).

More than 50% of the quantity of all the essential oils are citrus and mint oils. Especially sweet orange oil is produced in thousands of tons. Apart from the production of volatile natural oils 250,000 - 300,000 tons of turpentine is producedl from which about 100,000 tons are used for the productions of terpenoids for the flavour and fragrance industry.

Excellent reviews about the production of essential oils have been written by Meyer-Warnod, (4), Arnaudo (5) and Lawrence (6). Flores and Segredo (7) published on the citrus oil recovery during juice extraction. Boucard and Serth (10) wrote about a continous steam stripping process for the distillation of essential oils. Boelens et al. (11) published about ten years of hydrodiffusion of oils. Moyler (14) reviewed thoroughly ten years of carbondioxide extracted oils. With respect to the produdion of essentiai oils the following subjects will be treated: the preparation of the plant material, the isolation methods, yield and economics, and quality control.

Table 1.
INCREASING PRODUCTION OF SOME ESSENTIAL OILS

OIL	*1984*	*1990*	*1996*
Mentha arvensis	4000	5100	7000
Litsea cubeba	900	1050	1500
Tea tree	10	100	300

Table 2.
SOME IMPORTANT MEDICINAL PLANTS (19,20)

Common name	*Botanical species*
Ginseng	*Panax ginseng*
Maidenhair tree	*Ginkgo biloba*
Grapple plant (Wood spider)	*Harpagophytum procumbus*
Bockbean (Buckbean)	*Menyanthes trifoliata*
Taxus species (I ndian yew)	*T. vallichina, T. brevifolia, T. baccata*
Goldenseal	*Hydrastis canadensis*
Witch hazel	*Hamamelis virginiana*
Costus	*Saussurea lappa*
Valerian	*Valeriana officinalis*
Chamomile (Roman)	*Chamaemelum nobile*
Tea tree	*Melaleuca alternifolia*

Table 3.
PRODUCTION OF SOME IMPORTANT MEDICINAL PLANTS

Common name	*Botanical species*	*Plant material in metric tons*	*Annual growth %*
Ginseng	*Panax ginseng*	8,000	16
Maidenhair tree	*Ginkgo biloba*	2,000	26
Grapple plant	*Harpagophytum procumbens*	700	20

Table 4.
TURNOVER OF MEDICINAL PLANTS

	Million of dollars
Woridwide	10,000
Germany	2,200
France	200
United Kingdom	150

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2. PREPARATION OF THE PLANTMATERIAL

2.1. General

Various pre-preparations of the plant materials are often necessary before the essential oils can be isolated. Sometimes the desired volatile products are even not present as such in the fresh material, therefore hydrol ysis (mosses) or fermentation (orris root) is necessary.

2.2. Harvesting

All plant material must be harvested before the volatiles can be isolated. The harvesting may simple concern picking of fruits or flowers, or cutting branches of the trees. In practice, however, modern apparatus have been developed, as for example for the cutting of the flowertops of lavandula species. Leaves of plants can, after damaging during harvesting, produce a series of volatile compounds, as for instance (Z)-3-hexenol (leaf alcohol) and (E)-2-hexenal (leaf aldehyde) in freshly mown grass by enzymatic lipoxidation of linolic acid (16).

2.3. Threshing

It will be clear that seeds and fruits must be threshed before the oil can be isolated. Special apparatus have been developed for the threshing of for instance umbelliferous fruits, such as aniseed (see grinding).

2.4. Drying

Often the plant material is dried before the volatiles are produced. Some herbs, as for example mint plants, are only partly dried before production of the oil. Others, as for instances spices, are more thoroughly dried as a type of conservation before the oils are isolated. During damaging and drying of plant material again chemical reactions nnay occur, as for instance with the enzymatic conversion of the glycoside melitoside into glucose and (Z)-coumaric acid, which easily cyclizes to coumarin with a characteristic hay-like odour. This reaction occurs during drying of grass and in some labiatae species, like spike lavender and rosemary (17).

2.5. Grinding

Some fruits and seeds are sometimes ground before isolation of the volatiles as for instance with some umbelliferous fruits. Often the yields of the oil increase after grinding of the fruits, especially with wet grin ding directly into the apparatus.

2.6. Chipping

Woody plant material, such as cedarwood, sandalwood etc., are chipped before steam-distillation. The chipping of cedarwood before steamdistillation has been described by Boucard and Serth (10).

2.7. Hydrolysis

In some plant material the volatiles are not present as such but are formed after hydrolysis of less - or non-vol atile compounds. An example of these materials are oak - and treemoss, which contain nonvolatile depsides. These depsides are polyfunctional dimeric benzene-derivatives, which hydrolyze into monomers after treatment with hot water or steam, or in an enzymatic process. Atranorin is an odourless depside. which occurs in oakmoss. This depside is by hydrolysis converted into methyl beta-orcinyl carboxylate, the olfactively character-impact compound of oakmoss extract, and odourless atranylic acid, which by decarboxylation afford the volatile compound atranol.

2.8. Fermentation

Some volatiles, as for instance the irones from orris root, are only obtained after fermentation of the dried roots. During this fermentation process, hydrolysis and oxidation may occur.

3. ISOLATION METHODS

3.1. General

The methods for the production of essential oils have been modernized during the last 15 years. Most modern continuous distillations and extractions have been introduced during the last decenium. The following isolation methods will be treated in more detail: expression of citrus fruits, steamdistillation of labiatae oils, hydrodistillation of flower oils, hydrodiffusion of leaf oils, solvent extraction of mosses, and supercitrical carbondioxide extraction of flower concretes.

3.2. Expression

For the expression of citrus fruits there are mainly four methods in use, the Italian pellatrice, sfumatrice method, the American Brown oil extraction and the FMC Corporation process. The methods will be discussed more in detail. One should notice that expressed oil always contain nonvolatile residue, which can vary in con centration in the oil from 2 to 7%.

The dominant monoterpene in all citrus oils is (+)-(6R)-limonene, which occurs for instance in cold-pressed sweet orange oil for over 95%. Limonene most probably is formed during growing and ripening of the fruit via mevalonate, geranyl- and (-)-(3R)-linalyl pyrophosphate.

3.2.1. Pellatrice method

The pellatrice expression of citrus fruits concerns the abrasion of the surface of entire fruits.The fruits are rotating against an abrasive surface of a moving Archimedes' screw, covered with abrasive parts. During this movement the oilcells burst and the oil is released with a water spray. An oil-water emulsion is obtained and the oil is isolated by centrifugal separators. The advantages of the method are: good yield and quality with more oxygen compounds. Some drawbacks may be a darker oil and slightly more residue.

3.2.2. Sfumatrice method

Before treatment the peel and pulp are separated from the fruits, and the peel hardened in a lime bath. With the sfumatrice method the oil is isolated from the peel by a ribbed roll er pressing and a water spray (8). The oil-water emulsion is centrifuged. Advantages of the method are: the separated pulp, lighter product and less residue. Drawbacks are the peeling and lime treatment, and no optimal yield.

3.2.3. Brown oil extractor

In the Brown method the whole fruit is used. The fruits move on a bed of rolls covered with needles, and a water spray removes the oil-water emulsion. There are drying rolls and a solid el iminator for the solid materials. The oil is centrifuged. Advantages are the low solid content and water recycle, a drawback is the relatively high capital costs (8).

3.2.4. FMC Corporation method

The most ingenious apparatus for the expression of citrus fruits is the FMC apparatus. More than 50% of the quantities of citrus oils are isolated by this method (see Table 5). The method is based on the whole fruit extraction principle. The recovery of the oil occurs during juice extraction (7). During the extraction cycle the components of the apparatus interact to separate the various parts of the fruit instantaneously. The citrus oil glands burst and release their oil when the peel is deflected by the pressure created between the cup fingers during the extraction cycle. Recycle water is introduced during extraction, through a special ring located at the upper cup, to capture the oil. The oil is finally isolated by centrifuging. Advantages of the method are: fully automatic, minimum labour costs, juice and oil production. Minor drawbacks are: grading of the fruits, high capital costs (leasing is possible), yields up to 85% of the oil content.

3.3. Distillation

Another important method for the isolation is the distillation of essential oils. One can distinguish batch or continous steamdistillation, hydrodistillation and hydrodiffusion. A modern approach to essential oils distillation from the herb has been described by Denny (9). The different distillation methods will be discussed below.

Table 5.

WORLD PRODUCTION OF CITRUS ISOLATES IN 1995 (18)

(in thousands of metric tons)

Type	*Fruits*	*Processed Fruits*	*Oils*	*Oil-percentage calculated on processed fruits*	*Value in million dollars*
ORANGE	53,410	d-limonene 23,040 essence oils	36 68 42.5	0.64	220 ($1.5/Kg)
TANGERINE	13,251	845	1.5	0.18	15 ($15/Kg)
LEMON/LIME	7,653	1,555	5.44	0.35	92.5 ($17/Kg)
GRAPEFRUIT	5,561	1,950	3.5	0.18	35 ($10/Kg)
TOTAL	79,875	27,390	157	0.573	362.5

3.3.1. Steamdistillation

Steamdistillation is featured by the fact that the plant material is extracted by direct steam (produced in the still) or by indirect steam. The still often has a grill at the bottom and the plant material sometimes is in a perforated basket. Steamdistillation is used for the production of labiate leaf and flower oils, laurel leaf oil, eucalyptus leaf, bitter orange leaf oil and umbelliferous fruit oils etc. For yields of the steamdistillation of umbelliferous fruit oils see Table 9. The bulk of the essential oils is, apart from the expression of citrus oils, still manufactured by steamdistillation.

3.3.2. Hydrodistillation

Hydrodistillation are mostly carried out with flowers, e.g. bitter orange flower, rose or jasmine.The flowers are in a perforated basket and they are heated in 2 - 3 times their weight of water with indirect steam ffrom outside the still). A volume of water equal to the weight of the flowers is distilled. Yield of the separated oils is in general below 0.1 % and the distillate water is saturated with more soluble oxygen-derivatives (see Analysis).

3.3.3. Hydrodiffusion

Hydrodiffusion is carried out with low pressure steam (<0.1 bar) replacing the volatiles from the intact (uncommited) plant material by osmotic action. In the hydrodiffusor the low-pressure steam flow goes, according to the law of gravity, from the top through the vegetable load down to the conden ser at the bottom. The isolation sequence of the volatile components is determined to a great extent by their water solubilities. As a consequence, the condensed water is more or less saturated with the polar constituents of the oil (11). Test results of a hydrodiffusor are shown in Table 6.

Table 6.
TEST RESULTS OF SCHMID HYDRODIFFUSOR LS 500 COMPARED WITH HYDRODISTILLATION
(according to Schmid Hydrodiffusion SA, Switzerland, May 1981)

Product (Origin)	*Hydrodiffusion*		*Hydrodistillation*	
	Time (hr)	*Yield (%)*	*Time (hr)*	*Yield (%)*
Cistus leaves (France)	8	0.13	16	0.04
Cistus leaves (Spain)	8	0.15	16	0.05
Lavender (France)	0.5	0.73	1	0.75
Lavandin (France)	0.5	1.7	1	1.4
Cumin fruits (Poland)	4	5.0	12	3.7
Caraway fruits (Poland)	4	3.6	10	4.5

3.4. Extraction

A third method for the isolation of essential oils is the extraction of plant material, which for in stance can be solvent extraction , subcritical liquid carbondioxide and supercritical fluid carbon dioxide extraction. One has to keep in mind that with every type of extraction a certain amount of nonvolatile compounds will be extracted.

3.4.1. Solvent extraction

Solvent extraction can be carried out in two types, namely by percolation and by immersion. By percolation runs the solvent through the raw material, and by immersion covers the solvent completely the plant material and moves the solvent from the bottom to the top. A wide range of solvents are in use, such as alkanes, haloalkanes, benzenoids, ethers, ketones etc. The most usual solvent is hexane.

Yields of the extraction of oakmoss lichen with various solvents and transesterification of the depsides is shown in Table 7. Oakmoss and treemoss (Pinetree) absolutes were produced and the odour intensities determined by a group of observers. Besides the absolutes were analyzed by gaschromatographic-mass spectrometric techniques. The concentrations of the main constituents are shown in Table 8.

Table 7.

YIELDS OF EXTRACTION AND TRANSESTERIFICATION OF OAKMOSS LICHEN

Solvent	*Method*	*Yield (%)*
hexane	extraction	2
dichloromethane	extraction	5
benzene	hydrolysis/extraction	7
acetone	hydrolysis/extraction	10
methanol	alcoholysis/extraction	15
benzene/methanol	transesterification	10

Table 8.

MAIN CONSTITUENTS IN OAKMOSS AND TREEMOSS ABSOLUTE

Compounds	*Oakmoss absolute* %	*Treemoss absolute* %
Methyl 2,4-dihydroxy-3,6-dimethylbenzoate (methyl beta-orcinolcarboxylate)	47	57
3-Chloro-3,6-dihydroxy-4-methylbenzaldehyde (chloro atranol)	10	10
2,6-Dihydroxy-4-methylbenzaldehyde (atranol)	5	6
Cembrene	2	1
Methyl 2,4-dihydroxy-6-methylbenzoate	0.5	0.7

3.4.2. Subcritical liquid carbondioxide extraction

The subcritical liquid carbondioxide extraction is carried out at 50 to 80 bar and a temperature of 0 to 10 °C. Moyler (14) has published in detail about this extraction. The most practical extraction of this type is with hop cones (fruit cones of *Humulus lupulus* L.). The yield of a steamdistilled oil of hop cones is about 0.5%, whereas the yield of the carbondioxide extraction is ca 12%, due to the fact that nonvolatile polyfunctional diterpenes (humulones) are soluble in carbondioxide. About fifty essential oils obtained by liquid carbondioxide extraction are commercially available.

3.4.3. Supercritical fluid carbondioxide extraction

The supercritical fluid carbondioxide extraction is carried out at pressures over 80 bar and in general with temperatures above room temperature (14). The supercritical fluid extraction (SFE) comes more and more into practice during the last five years. One can extract rather fresh plant material (labiataes) with this method. One may also make first a solvent extraction (e.g. hexane) to prepare a concrete, and subsequently carry out a SFE-extraction, as for instance with flower concretes. Some analysis of examples of bitter orange flower concrete and rose concrete are shown in Table 6 and 7.

4. YIELDS AND ECONOMICS

4.1. General

Yields and economics are imported for successful production of an essential oil.

4.2. Yields

One can sometimes find quite a range of yields for one and the same essential oil. This range may be due to several explainable reasons, as there are climate, soil, isolation method etc. More often, however, published yields are too optimistic and not reproducible.

Table 9 demonstrates the variation in the yields of umbelliferous fruit oils as collected by Moyler (14). In generai the yield of carbondioxide and of ethanol extractions are higherthan those of steamdistillation, these differences are mainly due to the fact that the extracts contain non-volatile residue.

Table 9.

YIELDS (%) OF UMBELLIFEROUS FRUIT OILS

	steamdistillation	*liquid CO_2-extraction*	*fluid*	*ethanol extraction*
Angelica	0.3-0.8	3	-	-
Anise	2.1-2.8	-	7	15
Caraway	3 - 6	3.7	-	20
Carrot	0.2-0.5	1.8	3.3	3.3
Celery	2.5-3.0	3	-	13
Coriander	0.5-1.0	1.5	-	-
Cumin	2.3-3.6	4.5	-	12
Fennel	2.5-3.5	5.8	-	15
Parsley	2.0-3.5	3.6	-	20

4.3. Economics

The production costs of an essential oil may concern: the raw material costs, capital costs, labour costs, and en ergy costs. The raw material costs comprise the plant material, solvents etc. The capital costs are the investmen ts (leasing), depreciation and interest. Labour costs concern working, maintenance and quality control costs.

Because many aromatic plants grow in developing countries and they are used as raw materials in industrialized countries there is often a controversy between financial (capital/profit) economy and social (work) economy (6).

Raw materials costs may be relatively high with flower oils (also working hours). Certain apparatus (CO_2-extraction, FMC-apparatus) have high capital costs. Steam - and hydrodistillation have increase energy costs. CO_2-extraction and hydrodiffusion need more labour costs.

5. QUALITY CONTROL

5.1. General

The quality control of essential oils may concern the physicochemical standards, the chromatographic and spectroscopic analysis, the sensory analysis.

5.2. Physicochemical Standards

The physicochemical standards can comprise acid -, alcohol -, carbonyl -, and ester number; the solubility in ethan ol/water of various concentrations, specific gravity, optical rotation, refractive index, freezing/concealing and flash point, moisture content and the evaporation residu. A lot is known about the physicochemical properties of essential oils. One can find physicochemical standards in publications of the International Organization for Stan dardization (ISO), Essential Oil Association of the United States (EOA), in the Food Chemicals Codex (1996-IV), Monographs of the Research Institute for Fragrance Materials (RIFM), the Pharmacopoeias (EP, BP, USP, DAB, etc.) and in the published country standards (AFNOR, DIN etc.)

5.3. Analysis

5.3.1. Rose and Bitter Orange Flower Oils

Analyses of essential oils are carried out using the most modern gaschromatography and up-to-date spectroscopic techniques. For high resolution gas-chromatography analysis, high precision fused silica capillary columns are in use. Some examples of these analysis are given in Tables 10-12 with the headspace and oil analysis of rose flowers and of bitter orange flowers.

Table 10.

HEADSPACE ANALYSIS OF ROSA DAMASCENA; VARIATION IN COMPOSITION AFTER PICKING

Compounds	*Living (%)*	*Picked (%)*
Monoterpene hydrocarbons	28	57
2-Phenylethanol	40	2
Citronellol	8	2
Geraniol	2.5	+
Nerol	2.5	+
Phenylethyl acetate	3	1
Eugenol	+	2
Methyleugenol	0.5	2
cis-Rose oxide	+	1
trans-Rose oxide	+	0.5

These analysis were carried out in the rose fields near Isparta in central Southern Turkey by Dr. Robin Clery, Quest International, Ashford, Kent, UK.

Table 11.

CHEMICAL COMPOSITION OF ROSE OILS

Compounds	*hydrodistilled (%)*	*carbondioxide extracted (%)*
Citronellol	30	8
Geraniol	8	4
Nerol	9	2
2-Phenylethanol	2	67
Rose oxides	0.5	0.15
Methyl eugenol	2	0.7
beta-Damascenone	0.015	<0.005

From these analysis it is clear that a significant variation exists in the concentration of the main constituents of rose headspaces and oils. Especially the concentration of 2-phenylethanol can vary from 2 - 67 %.

An odour intensity comparison test between hydrodistilled rose oil and fluid carbondioxide extracted rose oil was carried out. The oils were presented in 0.5, 0.75 and 1 % solution to 22 observers in all possible paired combinations. The intensity score plotted against the logarithm of the concentration is shown in Figure 1. The hydrodistilled rose oil is about 2 to 3 times more odour-intensive than the carbondioxide extract.

Table 12.

CHEMICAL COMPOSITION OF BITTER ORANGE FLOWER OILS

Compounds	*hydrodistilled (%)*	*carbondioxide extracted (%)*
Citronellol	30	8
Monoterpene hydrocarbons	38	28
Linalylacetate	4	24
Linalool	38	35
Nitrogen compounds	0.5	2
Sesquiterpene alcohols	4	2

The odour value - quality and intensity - of hydrodistilled and fluid carbondioxide extracted orange flower oils were determined by a group of five perfumers. The oils were evaluated as such and in 1% and in 0.1% solution. The carbondioxide extracted oil was preferred over the hydrodistilled oil. The odour intensity of the carbon dioxide extract of the orange blossom concrete was about twice of that of the hydrodistilled oil.

6. CONCLUDING REMARKS

It was discussed that during the production of volatile natural isolates from plant materials

- several chemical reactions occur during the preparation of the plant material;
- the isolates can be prepared by expression, distillation and extraction;
- the isolation methods can be improved by continuous processes;
- the yields of the isolates are depending on the method, which is used;
- the chemical composition and the sensory properties can vary by different isolation methods.

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