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Changes in the Qualitative and Quantitative Composition of Essential Oils of Clary Sage and Roman Chamomile During Steam Distillation in Pilot Plant Scale

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1. Introduction

Clary sage (*Salvia sclarea* L.*)* from the genus Salvia is a biennial or short-living herbaceous perennial plant. Its leaves are united to a basal rosette in the first year, 12 to 25 cm long and 7 to 15 cm wide, ovate, cordate at the base, obtuse and long-stemmed. All the leaves are reticulate-rugose and hairy on both sides. The flowers seem loose to fairly dense, often branched paniculate. They reach approximately 2 cm and the large heart-shaped bracts are long, tapering and purple in early stages, later greenish white. Blossom: June or July to August. The seeds are 2 to 3 mm long nuts. Its native regions are the northern Mediterranean

Fig. 1. Clary sage

along with some other areas in Central Europe, North Africa and Central Asia. It is drought resistant and grows in dry, rocky places at an optimal soil pH of 4.5. Clary sage has a long history as medicinal herb and is currently grown mainly for the production of its essential oil reaching oil yields of 0.1 to 0.3%. (Board, 2003) (Dachler & Pelzmann, 1999) The clear, colourless oil is used in perfumes with a light, warm and sweet fragrance and as muscatel flavouring in food industry for vermouths, wines, and liqueurs. It is also used in aromatherapy for relieving anxiety, stress and fear, menstruation problems such as PMS (premenstrual syndrome) and cramping, and helping with insomnia, amenorrhea, dysmenorrhoea, pre-menopause, depression, fatigue, nervousness, migraine, varicose veins, haemorrhoids, oily skin and hair, spasmodic cough. It shows strong estrogenic, aphrodisiac and regenerative activity. (Lis-Balchin, 2006), (Kintzios, 2000) Sclareol, the essential oil main compound, is used in the perfume industry as a fixative and in the tobacco industry for flavouring. It is part of a large number of amber fragrances with woody note (Heinrich, et al., 2004), (Ferichs & Rimbach, 1992).

Fig. 2. Roman chamomile (Copyright Bernhard Bergmann)

Roman chamomile (*Chamaemelum nobile* L.) is a small (up to 80 cm), perennial plant found in dry fields and around gardens and cultivated grounds. It has daisy-like white flowers and is native to Western Europe, North America, and Argentina. Cultivation areas are mainly found in France (main producer), Belgium, Italy, Czech Republic, Slovakia, India, North America, Brazil and Argentina. Its stem is procumbent with ascending basic axis, the leaves are alternate, twice pinnate, finely dissected and fluffy to almost glabrous. The solitary, terminal flower heads rise eight to twelve inches above the ground and consist of prominent yellow disk flowers and silver-white ray flowers. Seeds are almost triangular, bald and shiny. (Dachler & Pelzmann, 1999) According to (Barnes, et al., 2007) Roman chamomile flower heads contain 0.4 – 1.75%. Blossom time is June and July, and its fragrance is sweet, crisp, fruity and herbaceous. The plant is used in tisanes, perfumes and cosmetics and to flavour foods. It is popular in aromatherapy, whose practitioners believe it to be a calming agent. The pharmacological profile of the Roman chamomile flower heads is similar to German chamomile (*Matricaria recutita* L.). It is used as mild sedative, anti-emetic and antispasmodic (Heinrich , et al., 2004), (Barnes, et al., 2007).

Essential oils are in general a complex mixture of natural compounds (terpene hydrocarbons like mono-, sesqui- and diterpenes - cyclic or non-cyclic – and their corresponding

oxygenated isoprenoid compounds alcohols, ketones, esters and aldehydes) with various applications in medicine, pharmacology, perfumery, cosmetics, food preservation or as insect repellent. Essential oils also contain phenolic compounds and alkanes. They are usually produced by distillation, but also by mechanical processes from different plant parts, such as seeds, fruits, fruit peel, roots, leaves, needles and balsams. Unlike fatty oils essential oils are volatile. Several data published in literature show that the quality of essential oils and the oil yield depend on many different parameters e.g. plant development stage, genetic determination, plant organ, drying and storage conditions, soil structure, climate, insolation and weathering, habitat, harvesting methods, date of collection of plant material, analysis conditions used for identification of the compounds, etc. Relative amounts of the compounds and the chemical composition of essential oils may also vary due to the applied distillation process itself (lab or pilot plant scale) and the duration of the distillation.

The essential oil of clary sage is a complex chemical mixture, consisting of up to 100 mostly terpenoid compounds. These are mainly (oxygenated) monoterpenes with small amounts of (oxygenated) sesquiterpenes. The main compounds are linalool (17.2%), linalyl acetate (14.3%), geraniol (6.5%), geranyl acetate (7.5%), terpineol (15.1%), nerol (5.5%), neryl acetate (5.2%) and sclareol (5.2%). Furthermore, α-pinene, β-pinene, camphene, myrcene, limonene, cis-and trans-ocimene, p-cymene, terpinolene, cis-3-hexen-1-ol, caryophyllene, terpinen-4-ol, citronellol, β-gurjunene, caryophyllene oxide, germacrene D, (2R, 5E)-2.12-epoxycaryophyll-5-ene, (2R, 5E)-caryophyllene-5-en-12-al, (2S, 5E)-caryophyllene-5-en-12-al, isospathulenol, (1R, 5R)-1,5-epoxysalvial-4 (14)-ene, salvial-4(14)-en-1-one (Kintzios, 2000).

Unlike chamomile from the Roman chamomile only few pharmacological studies exist. The composition of Roman chamomile oil is very complex and so far app. 100 compounds were identified. Main compounds are the sesquiterpenes and sesquiterpene lactones from the germacranolide type (e.g. nobiline). It is characterized by the presence of terpenoids and saturated and unsaturated fatty acids with four or five carbon atoms, such as butyric, valeric, crotonic, angelic, tiglin and methacrylic acid. These are esterified with C3 to C6-alcohols such as n-butanol, isobutanol, isoamyl alcohol and 3 - methylpentane-1-ol. The relative proportion of total esters in the essential oil of Roman chamomile is known as the highest of all essential oil-producing plants. Furthermore, Roman chamomile flowers contain hydroperoxides (e.g. 1-β-hydroperoxyisonobiline, a sesquiterpene peroxide from germacran**e**-type and allylic hydroperoxides as well). The content of hydroperoxides in the dried drug varies and is decreased during prolonged storage (Barnes, et al., 2007), (Bajaj, 1996), (Ferichs & Rimbach , 1992).

2. Methods

2.1 Cultivation and harvest

Both herbs were cultivated in the years 2002 and 2003 and Roman chamomile additionally in 2004 and 2010 by organic farming in three habitats (in 2010 only one habitat) of different altitude (Bad Blumau 285 m, Oberlungitz 400 m, St. Jakob 1000 m above sea level) in the East of Styria (Austria). Fields of the individual sites are divided into parcels a and b to ensure repeatability at each site.

The location of Bad Blumau (I) was located in the thermal spa area Blumau (Austria). Parcels were located within the spa recreation in order to attract interested visitors. The

herbal plantation had been converted from a meadow and spa gardener took care of herbal test fields.

Agricultural fields of the family "Ocherbauer" in St. Jakob im Walde were site (II) of the project. As in Blumau a meadow was transformed into a field. The use of machineries for cultivation of herbal test areas was found difficult, because plantation was located on a steep slope.

Agricultural site (III) of the family "Oswald" is located in Oberlungitz. Agricultural areas are laid out in rows for simplifying organic farming cultivation.

Used eight parcels had 10 per 20 m each at the cultivation sites Bad Blumau and St. Jakob, thus covered a total area of 1600 m². In Oberlungitz plants were grown in rows with 4.5 m x 35 m and 4.5 m x 59 m respectively, yielding a total area of 1692 m².

Harvesting techniques differed at the three sites, because available machines and devices were different.

Plant material was harvested in different stages of development. A code for development stages based on the catalogue of codes for grain plants ("BBCH – scale") was established (Meier, 2001). This catalogue was developed by the BBA (Federal Biological Research Centre for Agriculture and Forestry, Germany), the BSA (Protection and National Listing of new plant varieties, Germany), the IVA (Germany) and the IGZ (Institute of Vegetable and Ornamental Crops, Germany) and represents a code for the phenological development stages of mono- and dicotyledonous plants. The used herbs were characterised according to this code.

Clary sage is a biennial plant, which is harvested in the second year of cultivation, when the flowers are blooming. The optimum time for harvesting is June, since at that time the plant reaches the highest oil content. Moreover, in the period of 9 p.m. to 3 a.m., the highest oil yield can be achieved. Roman chamomile is harvested, when the florets wreath in the second third of the domed receptacle is already open. With increasing flowering process, the content of essential oil and chamazulene decrease. The characteristic blue colour is due to the formation of chamazulene in traces, which is formed from matricine by heating the oil. Delayed harvest may also lead to the disintegration of the flower heads. Harvest product should be the blooming chamomile heads (Dachler & Pelzmann, 1999).

For this project the entire herb of both plants was harvested in full bloom using bar- or power mower. For investigational reasons plant parts of clary sage were separated after harvest manually in order to compare the oil yield and composition of the essential oils gained by distillation of different plant parts. The vegetation progressing had been measured and documented regularly by taking several biomass yield samples at various stages; additionally sampling was done in order to document the total content of essential oil of the two herbs and their oil compositions. These data provided important information to determine the optimal harvest time, because the oil content depends on the plant development.

Plant height and inflorescence status of the herbs were noted and the samples were recorded photographically. The timing of sampling was based on the time of harvest.

According to the developed index ("BBCH scale") clary sage was harvested at growth stage 6 (blossom) and the subsidiary stage 66 (beginning of the flowering of the side shoots).

Roman chamomile was harvested at growth stage 6 (blossom) and the subsidiary development stages 65 (full flowering: 50% of flowers open, first ray florets may fall) and 49 (permanent development of young shoots, broadening of the main shoot).

After determining the moisture content in the samples with a moisture analyzer, plants were distilled immediately or dried at 35 ° C in a well ventilated area.

2.2 Distillation

Steam distillation was carried out using a hundred litres batch volume distillation plant of the type TWE 250-2000 Herba-Tec produced by the Innotec-Tetkov GmbH in Germany, which in average processes about 10 to 15 kilograms of fresh plant material per batch. For distillation in pilot plant scale plant material was weighed and used fresh or partly dried. Prior to distillation, the plants were cut using a slicer to a size of about 4 to 8 cm.

Main parts of the distillation plant:

1. steam generator
2. steam inlet area
3. pivoted distillation tanks
4. tube cooler
5. cooling water inlet
6. cooling water outlet
7. separation funnel of essential oil
8. control glass
9. lifting device

Fig. 3. Distillation plant in pilot plant scale TWE 250-2000 Herba-Tec

Disintegration of plant material had three main reasons:

1. Smaller parts of plants are generally easier to handle than the whole herb.
2. For distillation a homogeneous plant material filling in the column should be achieved avoiding cavities, which lead to shortcut steam flows.
3. The capacity of the distillation column is for cut plant material in general higher than for uncut material, if stuffing is avoided. A special designed cutter was used to prevent any squeezing and pressing of the herbs during cutting to create a smooth, sharp cutting surface.

A gentle disintegration of plant material is of importance to avoid losses of essential oil.

The steam distillation unit consists of two cylindrical distillation tanks (inner diameter 25 cm, height 2 m), with a 100 litres volumetric capacity. The tanks are rotatable with a swinging sieve to be used separately to allow an alternating filling and distillation process. Thus a continuous batch distillation process can be realized.

The steam production is carried out in a separate, electrically powered steam generator outside the tanks. A constant steam flow of approximately 200 g steam per minute or 12 kg of steam per hour is supplied. This specific amount of steam was claimed by the producer as the optimum amount of operation in connection with the geometry of the tanks.

Fig. 4. Distillation processing steps

After the steam passed through the distillation column the condensation of the water/oil mixture takes place in a heat exchanger (cooler), which is traversed in counter current flow by the cooling water. Separation of the essential oil is carried out in a separation funnel device. By the geometry of the funnel flow conditions are created that support the floating

of the oil on the hydrosol (condensed water vapour).The entire distillation process is run under normal atmospheric pressure conditions. All wetted parts are made of stainless steel with an electro-polished surface.

After the cutting process the herbs were weighed and filled into the distillation column manually. A homogeneous bulk body was produced to create comparable conditions and a plugging of plant material was avoided. All distillations were performed using a specific vapour flow of 12 kg of steam per hour. The amount of water content was also measured with a moisture analyzer. The temperature differences of cooling water and steam temperature and any condensate accumulating within the tank were documented. After distillation the used plant material was again weighed and the amount of accumulated flavour water calculated. The yield of essential oil was measured and calculated of 3 to 4 distillations to receive an average result. Prior to chemical analysis the essential oil was stored in a refrigerator.

In some cases every 10 minutes of the distillation process samples were taken in order to investigate the changes of essential oil yield and composition within a steam distillation process. Taken samples represent therefore a mixture of the gained essential oil during the period of 10 minutes. Samples were subjected to capillary gas chromatographic analysis coupled to a mass selective detector (GC/MSD) and coupled to a flame ionization detector (GC/FID).

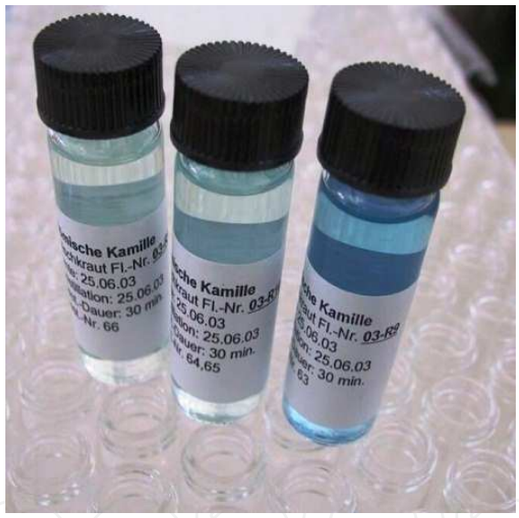

Fig. 5. Essential oils of Roman chamomile produced in different time frames during distillation

2.3 Analysis

The essential oils were analysed to state on the essential oil composition and its variation during the steam distillation process.

For quality analysis of essential oils such as the comparison of different samples taken in the distillation process it is necessary, to characterize the high number of compounds qualitatively and quantitatively. This is done by the use of capillary gas chromatographic techniques (GC/MSD and GC/FID). Main principles of these techniques are the separation of a mixture of compounds by transporting the sample through a capillary gas chromatography column. As mobile phase a gas, mainly helium is used. The separated

components leave the column due to different exchange mechanisms in the column at different times and are analyzed using special detectors. A frequently used detector is the flame ionization detector (FID), which is characterized by a high dynamic range for the universal detection of a large number of organic compounds. Because of this characteristic, this detector is often used in quantitative determinations. However, only limited information on the structure of the compounds is received. More specific information about the detected substances is provided by mass-selective detectors (MSD). The resulting mass spectra allow identification of chemical structures. By using the values of retention times, mass spectra and comparing results in literature and data bases, the chemical structure of a compound is detectable in most cases unambiguously.

Gas chromatographic method:

GC: Hewlett Packard 6890 GC system with integrated auto sampler
MSD: Hewlett Packard 5973 Mass Selective Detector
Column: J&W DB-5MS Capillary Column; Length: 30m; Diameter: 0.25 mm; Film thickness: 0.25 µm (non polar, coated with a phenyl arylene polymer – comparable to a 5% phenyl methyl polysiloxane column)
Mobile phase gas: Helium
Flow rate: 1 ml/min
Split ratio: 20:1
Temperature program: 10 min at 50 °C, with 2 °C/min heated to 220 °C, 10 min hold at 220 °C
Injection temperature: 220 °C
Injection volume: 1 µl
Temperature in Ion source: 250 °C
Quadrupol temperature: 200 °C
MS-Scan area: 20 - 350 m/z
Temperature of FID detector: 250 °C

In this project compositions of the essential oils were determined by comparing the relative retention times of standards and mass spectra from data bases of oil compounds (Adams, NIST, WILEY). Progression and development of the qualitative composition and the relative amounts of the most predominant compounds (>2%) were examined. These are important factors for the pharmaceutical application of essential oils. The results provide the possibility to determine the composition of the oils or the optimised distillation time with the highest relative amount of the main compounds. In addition trends and correlations among these compounds will be shown.

3. Results

3.1 Clary sage (*Salvia sclarea* L.)

3.1.1 Oil yield

Clary sage could only be harvested in the year 2003, because it's a biennial shrub. It shows big differences in essential oil yields in different parts of the plant - highest oil yields were obtained by distillation of the flowers. The oil yield of 7.9 ml/kg dry matter of the distillation of the flowers is 2.5 times higher than by distillation of the whole plant and 4.6 times higher than the distillation of stems (see figure 6).

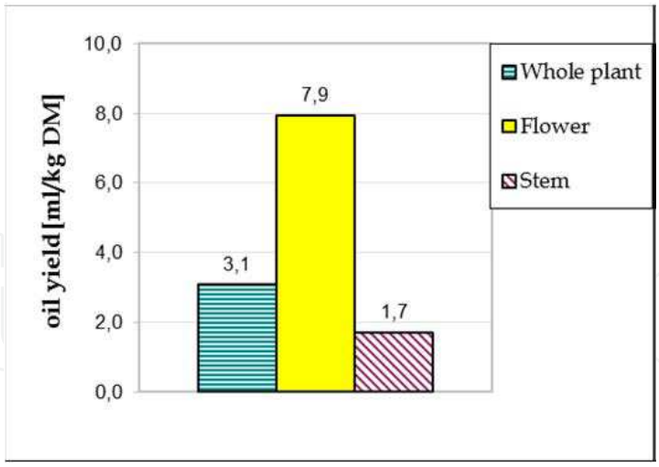

Fig. 6. Oil yield of different plant parts of clary sage (DM…dry matter)

Distillations of the fresh whole plants indicated an oil yield of app. 60% (app. 6.0–7.0 ml/kg dry matter) after 20 minutes distillation. After 30 minutes app. 80% (8.5-9.5 ml/kg dry matter) were obtained. In the last 30 minutes only small oil yields were achieved. Figure 7 indicates the oil yield measured in between specified time frames (9.3 to 20 minutes, 30 to 40 minutes, 50 to 60 minutes).

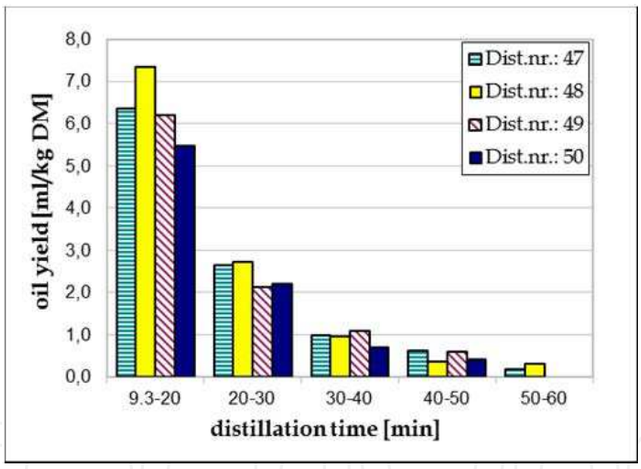

Fig. 7. Oil yields for specific time frames during distillation of the whole plant of clary sage (DM…dry matter)

According to literature data 90% of oil yield was reached after 2 hours distillation time (Kintzios, 2000). The average oil yield of all three agricultural sites reached 7.9 to 9.8 ml/kg dry matter by distillation of the flowers (app. 0.8 to 1.0% oil yield) and by distillation of the whole plant app. 3 ml/kg dry matter (app. 0.3%). These results correspond to the range of the literature values of the essential oil yield of 0.1 to 0.3%.

For economic reasons and according to the results, a distillation time of the fresh plant of 40 minutes and for drug distillations of 30 minutes is recommended by the authors. After that time the oil content increases only slightly and a yield of 90 - 95% is satisfactory. One reason for the extremely high oil content of the distillations of the fresh plant in 2003 could

probably have been the weather of spring and summer 2003 with many sunny days (and record heat in Austria).

3.1.2 Chemical investigations

75.9 - 91.8% of the essential oil of clary sage, harvested and distilled in pilot plant scale in 2003, was identified. The qualitative and semi-quantitative compositions of the distilled oils differ from each other depending on plant part, location and duration of distillation. The oil consisted mainly of linalyl acetate (16.7 – 69.7%), germacrene D (2.2 – 31.2%), β-caryophyllene (1.2 – 10.7%), bicyclogermacrene (0.6 – 9.1%), linalool (0.6 – 8.9%), α-copaene (0.5 – 6.1%), trans-A/B-sclareol oxide (0.5 – 5.7%) and sclareol (1.6 – 6.9%). Some samples exhibited a relatively high content of the pharmacological important diterpene sclareol. Sclareol is used as starting material for a number of amber fragrances. As the most important compound linalyl acetate was identified (approx. 70%). In most cases, the value of linalyl acetate corresponded to the value of germacrene D - if the relative amount of linalyl acetate decreased the value of germacrene D rose. Based on the data presented (see figures 10 – 12) it can be postulated, that the higher the amount of the flowers in the distilled material, the higher was the amount for linalyl acetate; the higher the amount of stems and leaves the higher was the content of germacrene D (see figures 9 to 11). The highest relative amount of sclareol was detected by distillation of the flowers after a distillation time of 50 minutes.

For economic reasons, only compounds with a higher relative amount than 2% are included in the following graphics.

Figure 10 indicates that linalyl acetate can be seen as the most important compound in the essential oil of the whole clary sage plant, the content does not considerably decrease with

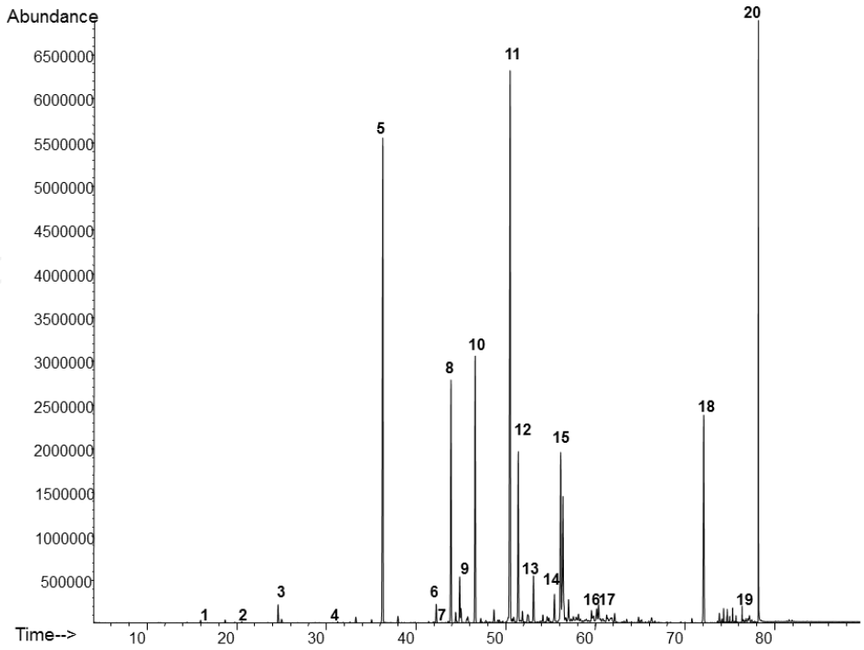

Fig. 8. Chromatogram of the essential oil of clary sage (Peak numbers correspond to compounds in table 1)

peak number	compound	retention time t_R [min]
1	myrcene	15.99
2	trans – β - ocimene	20.57
3	linalool	24.46
4	α - terpineol	31.22
5	linalyl acetate	36.39
6	neryl acetate	42.24
7	α - cubebene	43.54
8	α - copaene	43.87
9	β - cubebene	44.84
10	β - caryophyllene	46.57
11	germacrene D	50.48
12	bicyclogermacrene	51.38
13	δ - cadinene	53.08
14	1,5 - epoxysalvial-4(14)-ene	55.41
15	caryophyllene oxide	56.36
16	spathulenol	59.54
17	β - eudesmol	60.11
18	trans – A/B – sclareol oxide	72.03
19	manool	76.34
20	sclareol	78.16

Table 1. Identified compounds from the essential oil of clary sage from the harvest 2003

increasing distillation time. The other compounds are below 10%, linalool and germacrene D decrease with increasing distillation time and α-cubebene and sclareol rise.

In comparison to the whole plant distillation flower distillation shows an increase in the relative amount of linalyl acetate in the final minutes of the distillation. The same trend

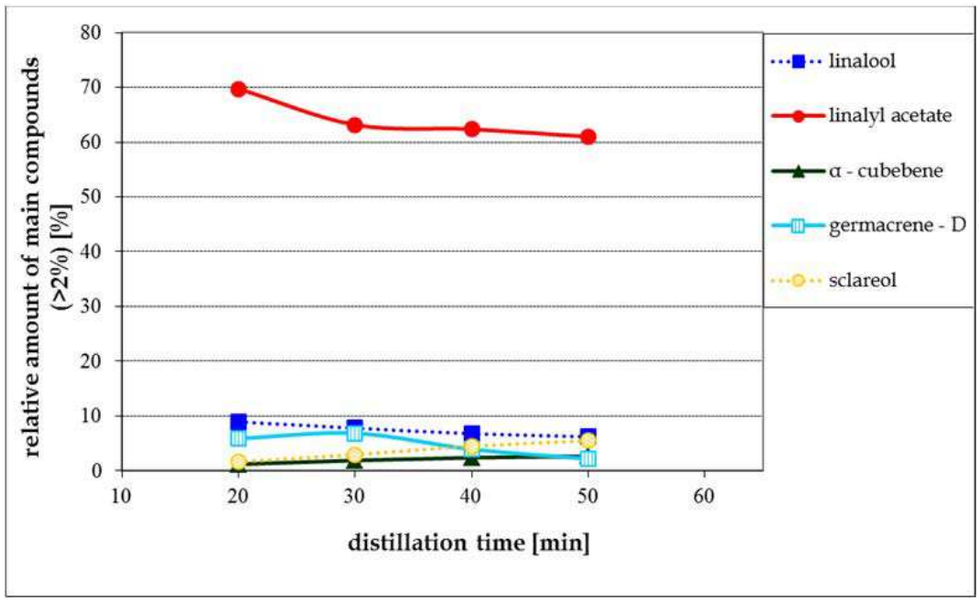

Fig. 9. Change of essential oil composition of main compounds during steam distillation of clary sage (habitat Bad Blumau, first cut, whole plant, beginning of flowering of the side shoots)

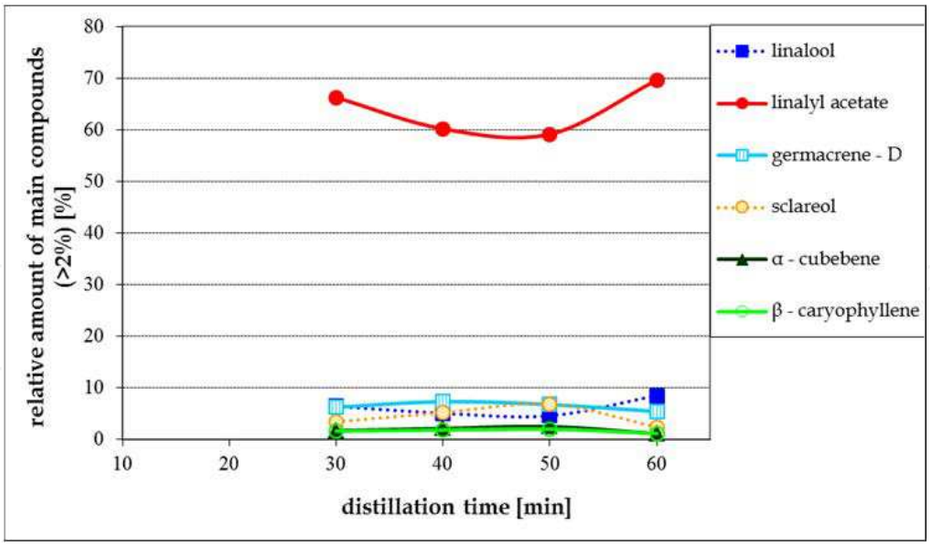

Fig. 10. Change of essential oil composition of main compounds during steam distillation of clary sage (habitat Bad Blumau, first cut, flowers only, beginning of flowering of the side shoots)

(increasing or decreasing with changes of direction in the final minutes respectively) show the graphs for linalool, sclareol and germacrene D.

The content of linalyl acetate in the essential oil produced by distillation of the stems and leaves is lower. It can be concluded that linalyl acetate is predominantly present in the flowers. The content of linalyl acetate decreases in the oil of the stems and leaves material with increasing distillation time, the levels of germacrene D, α-copaene, β-caryophyllene and bicyclogermacrene increase; sclareol content remains the same.

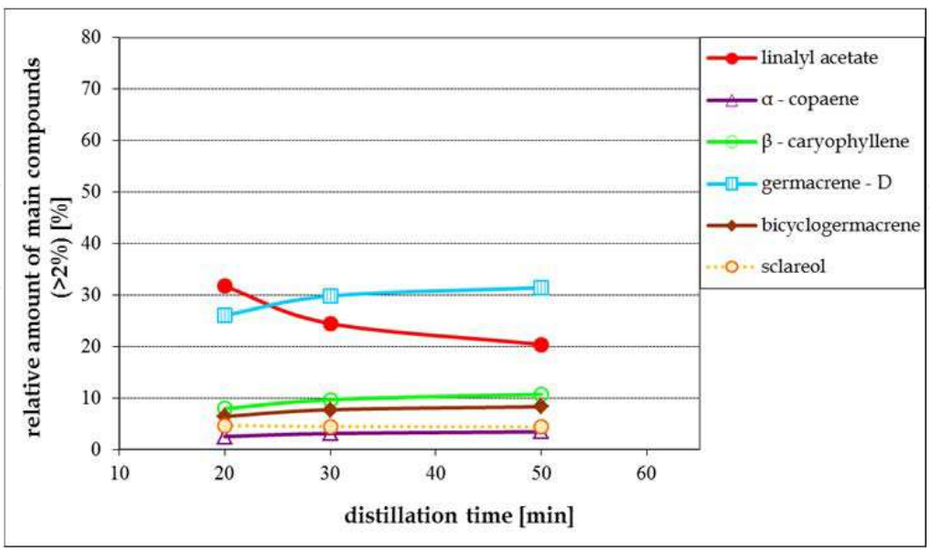

Fig. 11. Change of essential oil composition of main compounds during steam distillation of clary sage (habitat Bad Blumau, first cut, stems and leaves, beginning of flowering of the side shoots)

3.2 Roman chamomile

3.2.1 Oil yield

In 2003 by distillation of fresh Roman chamomile plants an average of 4.6 ml oil/kg dry matter is obtained after 20 minutes distillation time (equivalent to about 80% of oil yield - 3.5 ml oil/kg dry matter) and after another 10 minutes, 0.8 ml oil/kg dry matter. A distillation time of 30 minutes, thus proves to be useful in order to achieve an oil yield of about 95%.

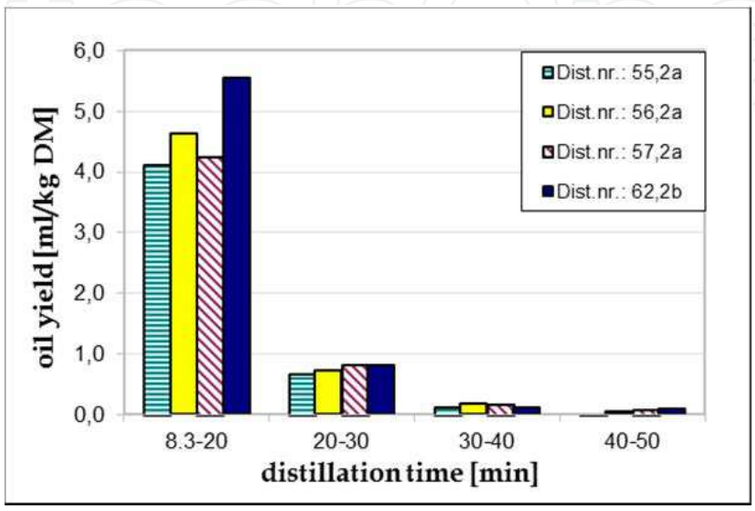

Fig. 12. Oil yields for specific time frames during distillation of the whole plant of Roman chamomile in 2003 (DM…dry matter)

Basically, Roman chamomile can be distilled easily. The colour of the essential oil is light to medium blue (see Figure 5), clear and low viscous. From an economic point of view a distillation time of 30 minutes of the fresh, and distillations of 20 minutes for dried plant material makes sense. The average yield of essential oil by distillation of the fresh Roman chamomile plant material reached values of 3.5 ml/kg dry matter (0.4%) and the dried 2.4 ml/kg dry matter (0.2%). The achieved oil content of Roman chamomile distillations in literature refers to the distillation of the flower heads and is not comparable with the results of whole plant distillations.

Harvest at the full flowering stage of the plants and the high number of sunshine hours in 2003 provided very good conditions for the increase of the content of the essential oil in the plants. By distillation of fresh Roman chamomile plants higher oil content with less effort (drying process) was achieved compared to distillation of drugs. A distillation of the drug is therefore not useful, if the capacity of steam distillation is capable for fresh processing.

Apart from a lower oil yield in 2004 compared to 2003, the curves of the distillations were not related. While in 2003, already after 25 minutes 90% of the oil yield was achieved, in 2004 this required a period of about 40 minutes. The oil yield was approaching its maximum more slowly. The reason for this fact could possibly have been the weather conditions, since, as already mentioned, the summer was exceptionally hot and dry in 2003. The oil cells of the plants in 2003 were better developed and thus distillation has been accelerated.

Due to the variability of results from different years, a distillation time of 50 minutes is advised.

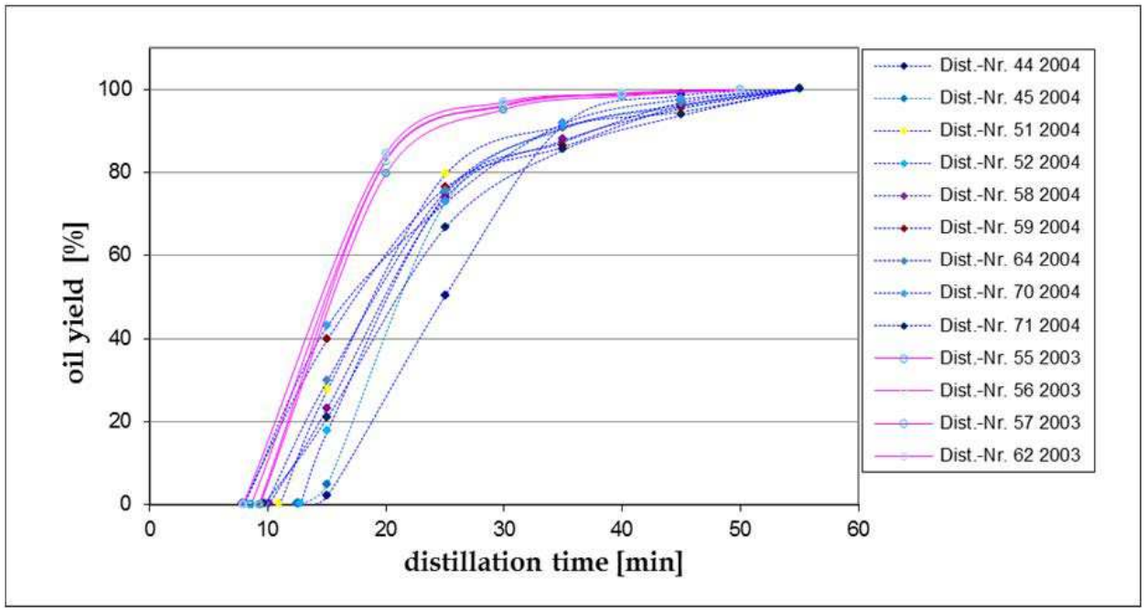

Fig. 13. Comparison of approximated oil recovery curves for the whole plant distillation of Roman chamomile in 2003 and 2004

By comparing the relative oil yields for the last two years the recovery curve is significantly different. One reason for the inhomogeneous curves from 2004 may have been the defective water dosing pump. The water supply had to be operated manually in this year, which might have led to fluctuations in the amount of steam. Apart from the already described different distillation maxima of 2004 and 2003, different heights of the curves can be recognized.

In 2003 Roman chamomile plants were harvested in full bloom and without any weeds, while in the cultivation year 2004 weeds proportion was high and the plants were harvested partially wet and prostrate. In summary, not only the weather, but also inflorescence, moisture and weeds are involved in the formation of the oil.

The distillation in 2010 differs from the distillation of the years 2003 and 2004. In the first cut 90% of the total oil yield was achieved after 20 minutes. The same distillation behaviour can be described for the second cut.

3.2.2 Chemical investigations

As the number of literature surveys on the Roman chamomile is scarce, few comparisons could be drawn to literature regarding the composition of the essential oil. In addition identification of compounds proved to be rather difficult, since the spectra exhibited many very similar esters, which are difficult to distinguish. As the composition also differed in the investigated years the following table is an example for the results of one investigation year (2003).

In both test series (fresh plant and drug material), the relative amounts of angelic acid 3-methylpentyl ester, 3-methyl-2-butenoic acid 3-methylbutyl ester, myrtenal and trans-pinocarveol increase, while the values for 3-methyl-2-butenoic acid pentadecyl ester, 3-methyl-2-butenoic acid cyclobutyl ester, 3-methyl 2-butenoic acid pentyl ester, α-pinene,

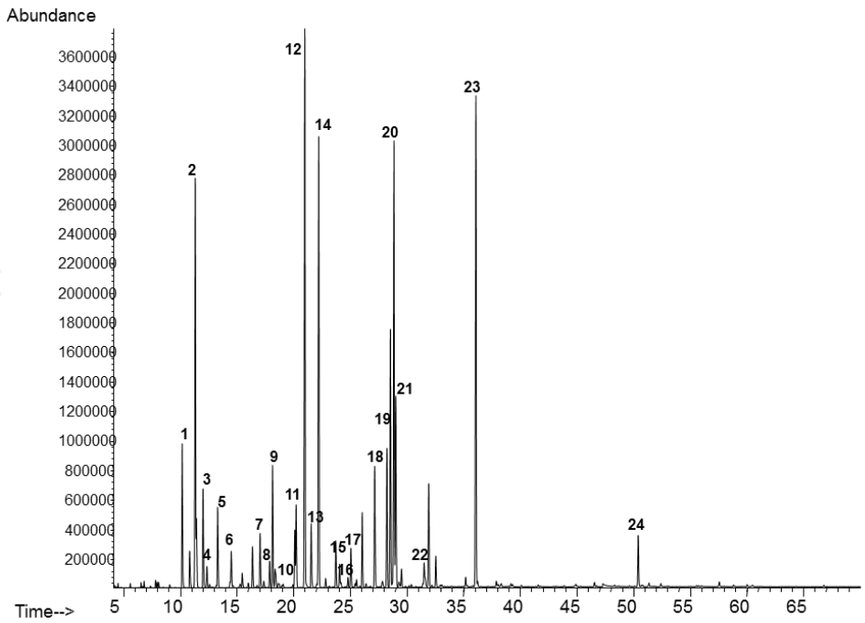

Fig. 14. Chromatogram of the essential oil of Roman chamomile (Peak numbers correspond to compounds in table 2)

peak number	compound	retention time t_R [min]
1	isobutyl isobutyrate	10.13
2	α - pinene	11.29
3	(E)-2-butenoic acid 2-methylpropyl ester	11.98
4	camphene	12.32
5	3-methacrylic acid-1-butene-4-yl-ester	13.27
6	β - pinene	14.47
7	2 - methyl butanoic acid 2-isobutyl ester	17.02
8	isoamyl butyrate	17.85
9	isobutyric acid 3-methylbutyl ester	18.12
10	p - cymene	18.37
11	cyclopropane carboxylic acid 3-methylbutyl ester	20.08
12	3-methyl-2-butenoic acid pentadecyl ester	20.97
13	isobutyric acid 3-methyl-2-butenyl ester	21.54
14	3-methyl-2-butenoic acid cyclobutyl ester	22.28
15	butyl angelate	24.00
16	isoamyl 2-methylbutyrate	24.75
17	2-methyl-butanoic acid 2-methylbutyl ester	25.04
18	trans - pinocarveol	27.15
19	3-methyl-2-butenoic acid pentyl ester	28.54
20	3-methyl-2-butenoic acid 3-methylbutyl ester	28.85
21	6,6-dimethyl-2-methylene bicyclo[2,2,1]heptane-3-one	29.00
22	myrtenal	31.47
23	angelic acid 3-methylpentyl ester	36,07
24	germacrene-D	50,39

Table 2. Identified compounds from the essential oil of roman chamomile harvested in 2003

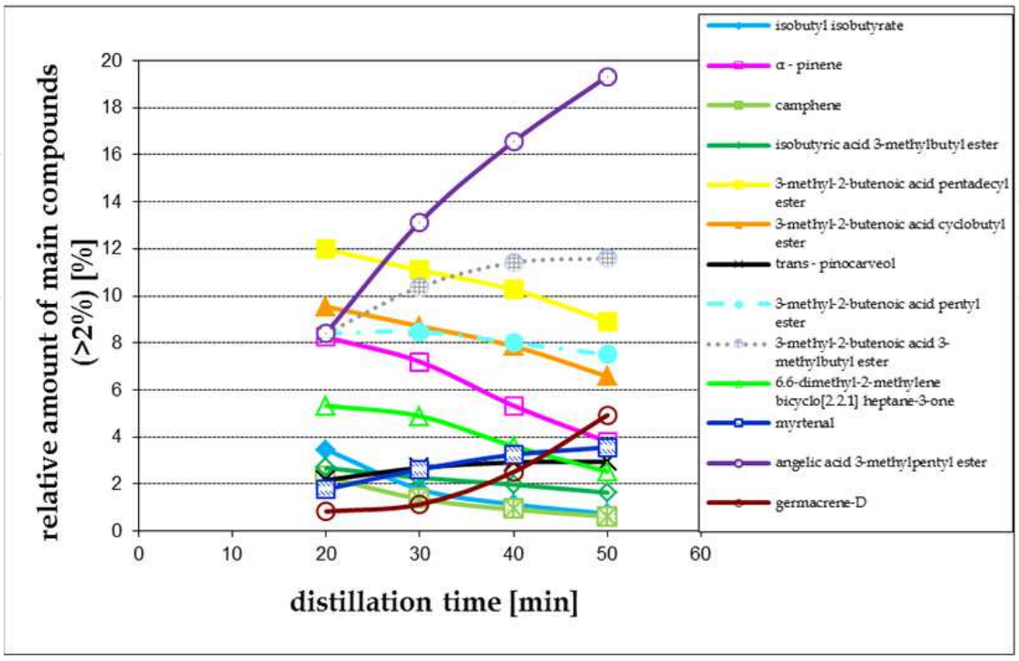

Fig. 15. Change of essential oil composition of main compounds during steam distillation of Roman chamomile (habitat Oberlungitz, first cut, fresh plant)

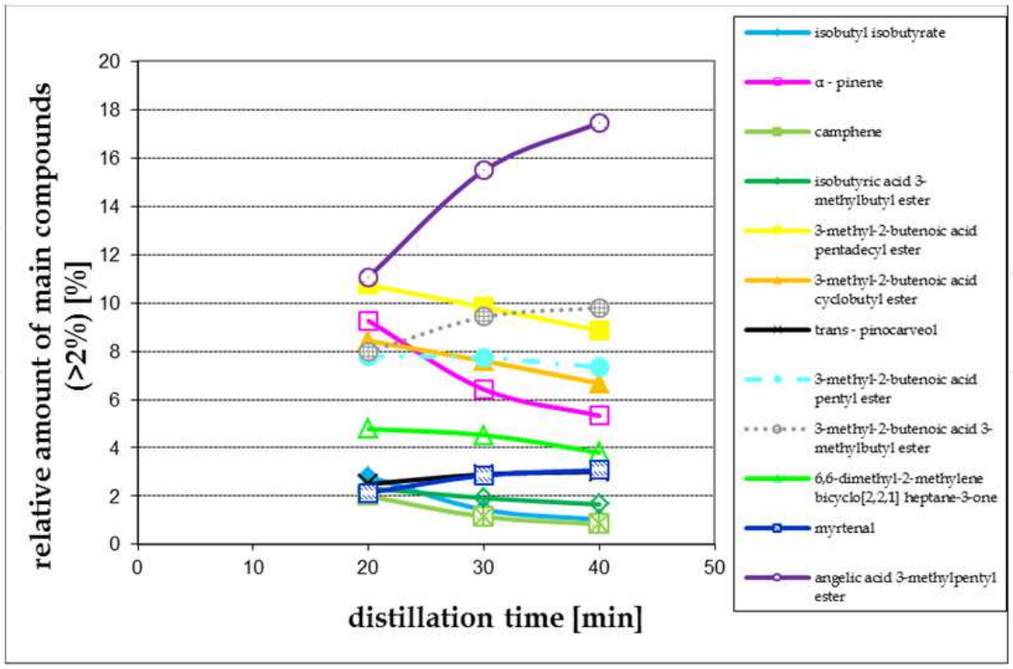

Fig. 16. Change of essential oil composition of main compounds during steam distillation of Roman chamomile (habitat Oberlungitz, first cut, drug)

6,6-dimethyl-2-methylene-bicyclo[2.2.1]heptane-3-one, isobutyl butyrate, isobutyric acid 3-methylbutyl ester and camphene decrease. 78.0 – 87.6% of the total Roman chamomile oil was identified and angelic acid 3-methylpentyl ester (8.4 - 19.3%), 3-methyl-2-butenoic acid pentadecyl ester (8.0 - 13.2%), α-pinene (3.8 - 12.2%), 3-methyl-2-butenoic acid 3-methylbutyl ester (7.0 - 11.6%), 3-methyl-2-butenoic acid cyclobutyl ester (6.6 - 10.2%), 3-methyl 2-butenoic acid pentyl ester (7.3 - 9.0%) and 6.6-dimethyl-2-methylene-bicyclo[2.2.1]heptane-3-one (2.5 - 6.1%) were identified as main compounds.

4. Conclusions

Based on the sampling during the steam distillation process of the two essential oils changes in the composition of the oils were observed during distillation. Thus, the achievable relative content of individual compounds of the essential oil depending on the plant (plant part) can be optimized. Additionally other plants and their essential oils were investigated. This might be of importance for future product developments, if specific essential oil compositions are required. In most cases the pilot plant scale distillation must not exceed 60 minutes distillation time, because after this period only very small amounts of essential oils are generated. That fact would not justify a longer distillation process for economic reasons.

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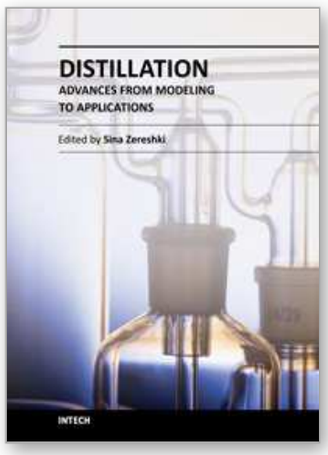

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Distillation modeling and several applications mostly in food processing field are discussed under three sections in the present book. The provided modeling chapters aimed both the thermodynamic mathematical fundamentals and the simulation of distillation process. The practical experiences and case studies involve mainly the food and beverage industry and odor and aroma extraction. This book could certainly give the interested researchers in distillation field a useful insight.

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