

Influence of Harvesting Time on the Composition of Volatile Components in Different Anatomical Parts of Lovage (*Levisticum officinale* Koch.)

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The essential oils from different botanical parts (leaves, stems, flowers, and seeds) of lovage (*Levisticum officinale* Koch.) were analyzed at various phases of plant growth. The seasonal changes in leaves were less considerable than in the stems. Seeds and flowers possessed the highest yield of oil. α -Terpinyl acetate was found to be the dominating compound in leaves and stems (up to 70%), β -phellandrene in seeds and flowers (61.50% and 40.80%, respectively); Z-ligustilide was confirmed as a major lovage phthalide constituting from 4.40% to 11.70% in leaves and from 4.80% to 13.80% in the stem's essential oils depending on the harvesting time.

Keywords: *Levisticum officinale* Koch.; *Apiaceae*; lovage; volatile constituents; essential oil; leaves; stems; roots; blossoms; harvesting time

INTRODUCTION

Lovage (*Levisticum officinale* Koch.) is a tall perennial aromatic plant, cultivated in numerous European countries. The essential oils from leaves, seeds, and roots of lovage are used in the food, beverage, perfumery, and tobacco industry (Cu et al., 1993). The root of lovage has also been known for centuries as a medicine possessing carminative and spasmolytic activity (Segebrecht and Scilcher, 1989).

The composition of volatile compounds of lovage has been the subject of numerous investigations (Gijbels et al., 1982; De Pooter et al., 1985; Toulemonde et al., 1987; Toulemonde and Noleau, 1988; Segebrecht and Scilcher, 1989; Cu et al., 1990; Stahl-Biskup and Wichtmann, 1991; Blank and Schieberle, 1993; Venskutonis, 1995; Bylaite et al., 1996). Different botanical parts of lovage have been analyzed; however, most of the studies were focused on the root. Their specific flavor is characterized as medium aromatic (Heath and Reineccius, 1986) or as strong impact warmly aromatic with sweet, yeasty, musky, lemon-like, celery-like notes (Heath, 1981). The phthalides are believed to play a major role in the aroma of lovage. Seven phthalides have been identified in lovage roots constituting from 22% to 71% (Toulemonde and Noleau, 1988; Cu et al., 1990) in the total essential oil. Different isolation procedures (dynamic headspace, distillation, and extraction with different solvents) were used to obtain lovage aroma concentrates, and the content of phthalides in them was highly dependent on the method applied. However, only a few studies on the assessment of odor-active components of lovage were performed in recent years (Blank and Schieberle, 1993; Bylaite et al., 1996), and none of them demonstrated the significant contribution of phthalides to the aroma of lovage. On the contrary, the investigation of celery

aroma compounds revealed a significant correlation between sensory evaluation of flavor scores with the individual and total phthalide contents (Uhlig et al., 1987; MacLeod and Ames, 1989).

Toulemonde and Noleau (1988) performed a detailed study of lovage leaf, seed, and root essential oils as well as root and leaf solvent extracts in 1988, and identified 191 compounds. Quantitative analysis of seed and root essential oil showed that β -phellandrene was a major component in the seed essential oil (63%) while *cis*-3-*n*-butylidene-4,5-dihydrophthalide was a dominating constituent in the root oil (67%). It should be noted that the number of detected compounds significantly depended on the method of isolation; e.g., 116 compounds were detected in the leaves by using simultaneous distillation–extraction in the Likens–Nickerson apparatus, whereas only 31 constituents were found in dichloromethane extract. De Pooter et al. (1985) also performed comparison of different isolation procedures and their effects on the aroma composition of lovage. They found that the headspace of fresh lovage was constituted almost exclusively of mono- and sesquiterpenes, while phthalides appeared among volatiles only when hydrodistillation was used.

The variation of aroma composition depends on different factors, such as the isolation procedure, the botanical part of the plant, harvesting time, and state of maturity. A survey of the literature reveals that detailed investigations of the variations of yield and changes of aroma compounds in different botanical parts of lovage during the growth period have not been the subject of any detailed study. An exception was the study by Stahl-Biskup and Wichtmann (1991), in which the composition of the essential oil from the lovage roots in relation to the development of their oil duct systems was analyzed and phthalides were found in considerable amounts only after many secondary oil ducts had been developed.

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Table 1. Yields (% w/w of Dry Weight) of Essential Oils from Different Botanical Parts of Lovage at Different Harvesting Periods

| growth phase and date of harvesting | botanical part harvesting | | | | |
|--|---------------------------|-------------|-------------|------------|-------------|
| | leaves | stems | flowers | seeds | roots |
| 1. shoot up, May 15 | 1.14 ± 0.04 | 0.53 ± 0.04 | | | |
| 2. growing, May 25 | 1.21 ± 0.05 | 0.68 ± 0.04 | | | |
| 3. growing, June 9 | 1.35 ± 0.04 | 0.97 ± 0.04 | | | |
| 4. formation of buds, June 16 | 1.12 ± 0.06 | 1.16 ± 0.06 | | | |
| 5. beginning of flowering, June 28 | 1.01 ± 0.04 | 0.65 ± 0.04 | | | |
| 6. end of flowering, July 7 | 1.10 ± 0.05 | 0.53 ± 0.03 | 1.53 ± 0.06 | | |
| 7. formation of seeds, July 19 | 1.09 ± 0.04 | 0.73 ± 0.03 | | 2.7 ± 0.05 | |
| 8. 2nd shoot up (newly grown plant), Sept 21 | 1.10 ± 0.03 | 1.16 ± 0.03 | | | 0.05 ± 0.04 |

Thus, it was considered that the studies of the seasonal variations in the composition of volatiles in different botanical parts of lovage cultivated in Lithuania would be of some interest.

MATERIALS AND METHODS

Plant Material and Isolation of Volatile Compounds.

Plant material was collected in the experimental garden of the Lithuanian Institute of Horticulture in 1995. The whole herb was harvested every second week, a total of 8 times from the middle of May until the end of September. The plants were sorted by botanical part (leaves, stems, flowers, seeds, and roots) depending on the growth period and air-dried at 30 °C in the laboratory oven with active ventilation. Volatiles were isolated by hydrodistillation in a Clevengertype apparatus for 2 h. Triplicate analysis was carried out for every sample. Simultaneously, the content of moisture in air-dried herb was determined by distilling 5 g of the sample with toluene for 1.5 h (AOAC, 1995). The yields of oil are presented as percent (w/w) of dry weight.

Identification and Quantification of Volatile Compounds. The essential oils were diluted with Et₂O (1% v/v) and analyzed on a dual-column Carlo Erba Vega 6000 gas chromatograph with FID heated at 260 °C. Four GC analyses were carried out for each essential oil. Two different polarity-fused silica capillary columns were used: DB-1 (dimethylpolysiloxane) and DB-Wax [poly(ethylene glycol)], both 60 m length, 0.25 mm i.d., 0.25 μm film thickness. The oven temperature was programmed from 50 °C (5 min hold) to 250 °C at a rate of 3 °C min⁻¹ with a final hold of 10 min. Helium was used as a carrier gas with a flow rate of 2 mL min⁻¹. The injector temperature was set up at 230 °C.

Gas chromatography–mass spectrometry analyses were performed on a combined GC–MS (Varian 3400, Finnigan MAT 95) instrument with a mass selective detector using a BP-5 (5% phenylmethylpolysiloxane) capillary column (25 m length, 0.25 mm i.d., 0.25 μm film thickness); the GC oven temperature was programmed from 60 to 260 °C at an increase of 3 °C min⁻¹. Mass spectra were obtained with 70 eV electron impact ionization and scanned from *m/z* = 24 to 300 at 0.5 s decade⁻¹.

Identification was based mainly on comparison of retention indices (RI) and mass spectra (Adams, 1995). Also the results obtained were thoroughly compared with the published ones and found to be in good agreement. When available, pure chemicals were used as reference compounds. The content of the individual constituents was expressed as a peak area percent computed by a GC system integrator, and also in arbitrary units, which were calculated by the formula

$$C = \frac{Y \times I \times 1000}{100}$$

where *C* is the amount of individual component in arbitrary units (a.u.), *Y* is the yield of essential oil (% of dry wt), and *I* is the percentage content of individual component in the essential oil.

An arbitrary unit represents the absolute concentration of a particular compound in 1 kg of dry herb. An arbitrary unit

is approximately equal to μg kg⁻¹, and it could be converted into μg kg⁻¹ by determining the GC response factors for every identified constituent.

RESULTS AND DISCUSSION

Total Essential Oil Content. The yields of lovage essential oil at different harvesting periods are given in Table 1. The highest amount of essential oil (2.70%) was determined in the seeds, which were formed in the middle of July. The concentration of volatile oil in leaves and stems varied from 1.01 to 1.35% and from 0.53 to 1.16%, respectively, whereas the flowers (July 7) contained 1.53% of oil. It should be emphasized that lovage having a high concentration of oil in its stems is an exceptional plant as compared with many other aromatic herbs usually possessing only a negligible amount of volatiles in their stems (Venskutonis, 1995). However, an extremely low amount of oil was obtained by hydrodistillation from the roots (approximately 0.05%), and this finding is in disagreement with the results obtained by Maghami (1979), who reported the content of oil in lovage roots to range from 0.6 to 1%, in the seeds from 0.8 to 1.10%, and in the leaves from 0.05 to 0.15%. A much higher content of oil in lovage roots was also determined by Cu et al. (1990). The content of essential oil in the leaves was almost constant during the whole period of plant vegetation with a peak (1.30%) just before the formation of buds (the phase of intensive growing, beginning of June). From a previous harvest in 1992, the yield of essential oil from the leaves of lovage on June 17 (beginning of blossom) and July 8 (full bloom) was significantly higher, 1.47 and 1.98%, respectively, than the yield in 1995 most likely due to different climatic conditions, because the strains of the plant and the growing and drying conditions were the same (Venskutonis, 1995).

The concentration of the essential oil in the stems was steadily increasing from 0.53% in May 15 (initial growing) to 1.16% in June 16 (buds formation); however, afterward it dropped again. In general, it is accepted that most of the herbs accumulate the highest amount of volatile oil during the anthesis period; however, numerous studies show that every particular aromatic plant can possess its own peculiarities. For instance, the study of aroma compounds in dill by Huopalahti and Linko (1983) showed their highest value at the initial stage of growth, while thyme was richest in essential oil during or immediately after the full bloom (Senatore, 1996).

Composition of Essential Oils. The yield of root essential oil (0.05%) was very low which makes it difficult to separate the oil from the distillate. Therefore, for further analyses, only the oils from leaves, stems, seeds, and flowers were used.

Table 2. Identification Data of Volatile Compounds in Different Botanical Parts of Lovage

| no. ^a | constituent ^b | leaves ^c | stems ^c | seeds ^c | flowers ^c | content, % (min-max) |
|------------------|---|---------------------|--------------------|--------------------|----------------------|----------------------|
| 1 | tricyclene | a,b | a,b | — | — | tr |
| 2 | α -thujene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.11 |
| 3 | α -pinene | a,b,c | a,b,c | a,b,c | a,b,c | 0.37–1.47 |
| 4 | camphene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.43 |
| 5 | sabinene | a,b,c | a,b,c | a,b,c | a,b,c | 0.45–1.21 |
| 6 | β -pinene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.77 |
| 7 | myrcene | a,b,c | a,b,c | a,b,c | a,b,c | 1.30–7.12 |
| 8 | 2,3-dehydro-8-cineole | a,b | a,b | — | a,b | tr, 0.17 |
| 9 | octanal | a,b | b | b | b | tr, 0.21 |
| 10 | α -phellandrene | a,b,c | a,b,c | a,b,c | a,b,c | 0.15–1.96 |
| 11 | Δ -3-carene | — | — | a,b | — | tr |
| 12 | α -terpinene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.48 |
| 13 | <i>p</i> -cymene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.92 |
| 14 | β -phellandrene | a,b,c | a,b,c | a,b,c | a,b,c | 10.85–61.50 |
| 15 | limonene | a,b,c | a,b,c | a,b,c | a,b,c | 1.90–5.38 |
| 16 | β -ocimene (<i>Z</i>) | a,b,c | a,b,c | a,b,c | a,b,c | 0.13–1.64 |
| 17 | phenyl acetaldehyde | a,b | — | — | — | tr |
| 18 | β -ocimene (<i>E</i>) | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.39 |
| 19 | γ -terpinene | a,b,c | a,b,c | a,b,c | a,b,c | 0.16–1.87 |
| 20 | terpinolene | a,b,c | a,b,c | a,b,c | a,b,c | tr, 1.72 |
| 21 | linalool | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.23 |
| 22 | 2-methylbutyl-2-methylbutanoate | a | a | — | — | tr |
| 23 | isomer of compound 22 | a | a | — | — | tr |
| 24 | <i>cis-p</i> -menth-2-en-1-ol | a | — | — | — | tr |
| 25 | pentylcyclohexadiene | a,b,c | a,b,c | a,b,c | a,b,c | 0.21–0.95 |
| 26 | terpinen-4-ol | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.23 |
| 27 | 4-isopropylcyclohex-2-en-1-one | a | a | a | a | tr, 0.80 |
| 28 | α -terpineol | a,b,c | a,b,c | a,b,c | a,b,c | tr, 1.50 |
| 29 | methyl salicylate | — | — | a,b | — | 0.11 |
| 30 | carvone | a,b | — | — | a,b | tr, 0.27 |
| 31 | geraniol | a,b,c | b,c | — | — | tr, 0.22 |
| 32 | linalool acetate | b | a,b | — | — | tr |
| 33 | phellandral | — | — | — | a | 0.10–0.13 |
| 34 | bornyl acetate | a,b,c | a,b,c | a,b,c | a,b,c | 0.11–0.61 |
| 35 | <i>cis</i> -dihydro- α -terpinyl acetate | b | b | b | b | tr, 1.35 |
| 36 | α -terpinyl acetate | a,b,c | a,b,c | a,b,c | a,b,c | 4.56–70 |
| 37 | bornyl propanoate | — | a | — | a | tr, 0.81 |
| 38 | α -copaene | — | — | a | a | tr |
| 39 | geranyl acetate | a,b,c | a,b,c | — | a,b,c | tr, 1.52 |
| 40 | methyleugenol | — | — | a,b | a,b | 0.10–0.20 |
| 41 | bornyl 2-methylpropanoate | — | a | — | — | tr |
| 42 | β -farnesene | — | — | a,b | a,b | 0.13–0.78 |
| 43 | germacrene D | a,b,c | a,b,c | a,b,c | a,b,c | tr, 1 |
| 44 | β -selinene | a,b,c | a,b,c | a,b,c | a,b,c | tr |
| 45 | germacrene A | a,b | a,b | a,b | a,b | tr, 0.25 |
| 46 | bornyl 2-methylbutanoate | — | a | — | — | tr |
| 47 | bornyl 3-methylbutanoate | a | a | — | — | tr |
| 48 | δ -cadinene | a | — | a | a | tr |
| 49 | germacrene B | a,b | — | a,b | a,b | tr |
| 50 | α -cadinol | — | — | — | a | tr |
| 51 | 3-butylphthalide | a | — | — | a | tr |
| 52 | 3-butylidene phthalide (<i>Z</i>) | a,b,c | a,b,c | a,b,c | a,b,c | tr, 0.69 |
| 53 | 3-butylidene phthalide (<i>E</i>) | a,b,c | a,b,c | a,b,c | a,b,c | 0.10–0.38 |
| 54 | sedanolide | a | a | — | — | tr |
| 55 | ligustilide (<i>Z</i>) | a,b,c | a,b,c | a,b,c | a,b,c | 4.44–16.01 |
| 56 | ligustilide (<i>E</i>) | a,b,c | a,b,c | a,b,c | a,b,c | 0.13–0.47 |
| 57 | validene-4,5-dihydrophthalide | — | — | — | a | tr |
| 58 | neophytadiene | — | a | — | — | tr |

^a Peak numbers refer to Figure 1. ^b Constituents are listed in order of elution from the DB-1 column. ^c a, identification confirmed by mass spectra; b, identification confirmed on the DB-1 column; c, identification confirmed on the DB-Wax column.

Fifty-eight compounds were identified in different parts of lovage and are listed in Table 2 in order of their elution from the DB-1 column. Most of them have been already reported in previous studies of lovage (Toulemoude et al., 1987; Toulemonde and Noleau, 1988; Cu et al., 1993) except for some minor tentatively identified constituents (no. 23, 24, 29, 30, 35, 37, 41, 45, 46, 47). The quantitative composition of the oils was almost similar in all samples except for some minor components, which were detected only in particular botanical parts. Although most of the identified constituents were present in the essential oils during all investigation seasons, a few minor ones were not detected at some

harvesting periods. For further discussion, trace compounds of lovage (<0.10%) have not been considered. The percentage composition of the compounds exceeding 0.10% in the total essential oil is presented in Table 3.

All identified constituents were divided into the three groups: major constituents (the mean content >9%), compounds of low–medium concentration (constituting ~1–5%), and minor constituents (<1%). Three major components were common for all analyzed lovage parts, namely, β -phellandrene, α -terpinyl acetate, and *cis*-ligustilide. This finding is in agreement with the results published in the literature (Toulemonde and Noleau, 1988; Venskutonis, 1995). However the differences in

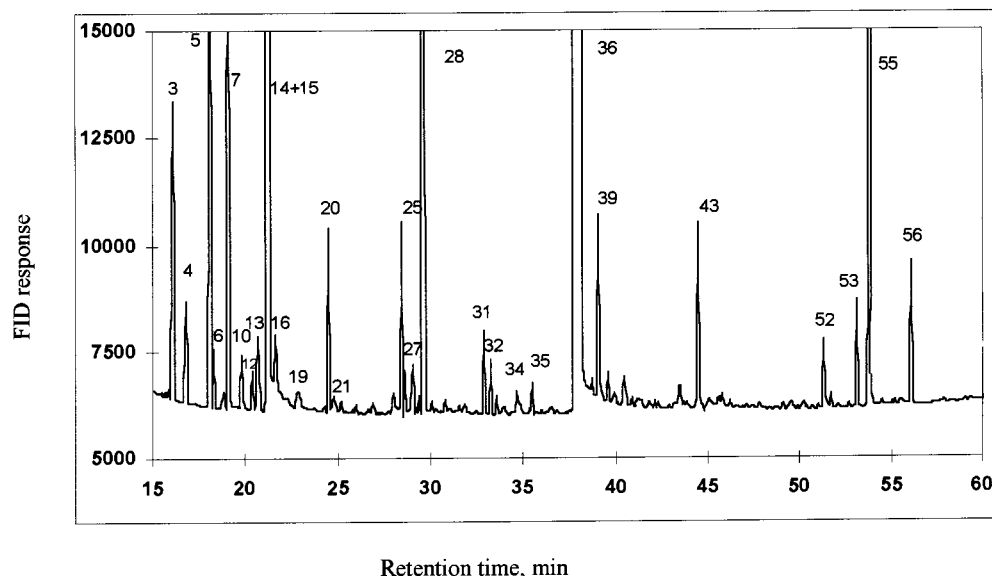


Figure 1. Gas chromatogram of the essential oil of lovage leaves harvested on May 15. See Materials and Methods for the GC conditions and Table 2 for the peak identifications

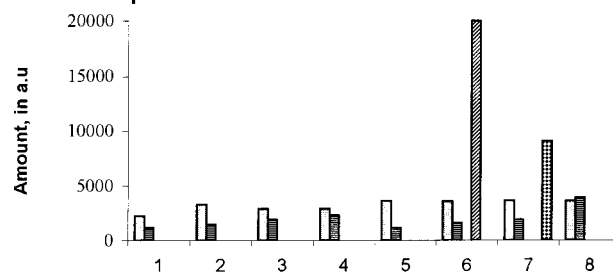
essential oil of leaves and from 10.85 to 19.21% in stems. It is interesting to notice that it increased during flowering both in leaves (5th harvesting, June 28) and in stems (6th harvesting, July 7). During this phase, the percentage content of α -terpinyl acetate decreased. β -Phellandrene was also the major constituent in seeds (61.50%) and flowers (40.82%). It was described as an odor-active compound having a grassy, chemical odor in our previous study of lovage (Bylaite et al., 1996).

Z-Ligustilide is known as a major lovage phthalide (Gijbels et al., 1982; Toulemonde and Noleau, 1988; Cu et al., 1990; Stahl-Biskup and Wichtmann, 1991; Venskutonis, 1995), and it was found in this study as well. The percentage concentration of Z-ligustilide varied from 4.44% to 11.70% in the leaves and from 4.80 to 13.85% in the stem essential oil depending on the time of harvesting. Its content was slightly higher in the seeds (14.18%) and flowers (16.01%). Corresponding contents of Z-ligustilide were found in the leaves and stems of lovage from Lithuania harvested in 1992 (Venskutonis, 1995). It can be noticed that the content of Z-ligustilide (Table 3) was lower both in leaves and in stems at the initial phase of growing.

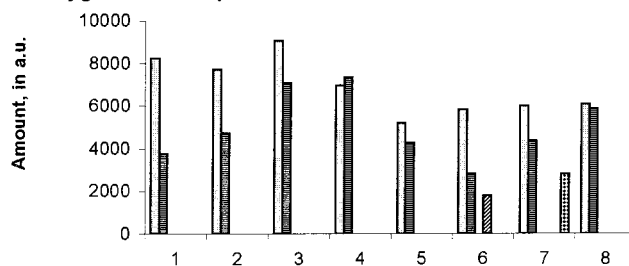
Low-Medium Concentration Constituents of Lovage. The following compounds can be included in this group: α -pinene, sabinene, myrcene, limonene, *cis*- β -ocimene, γ -terpinene, α -terpineol, and geranyl acetate, of which limonene and myrcene were the most abundant components. Limonene ranged from 1.90 to 4.65% in the leaves and from 2.04 to 5.64% in the stems, myrcene from 1.60 to 4.44% in the leaves and from 1.30 to 3.34% in the stems. The highest content of myrcene was determined in the flowers, 7.12%. Although the changes in monoterpene hydrocarbon content at different harvesting period are of a somewhat "jumping" character (e.g., exceptional increase of limonene and decrease of *cis*- β -ocimene in July 7), a clear tendency in the accumulation of these compounds including β -phellandrene during the flowering period can be observed (Table 3, Figure 2).

Two oxygenated monoterpene compounds, α -terpineol and geranyl acetate, were also present in appreciable amounts both in leaves (0.69–1.50% and 0.39–0.83%, respectively) and in stems (0.86–1.48% and 1.01–

A. Monoterpenes



B. Oxygenated compounds



C. Phthalides

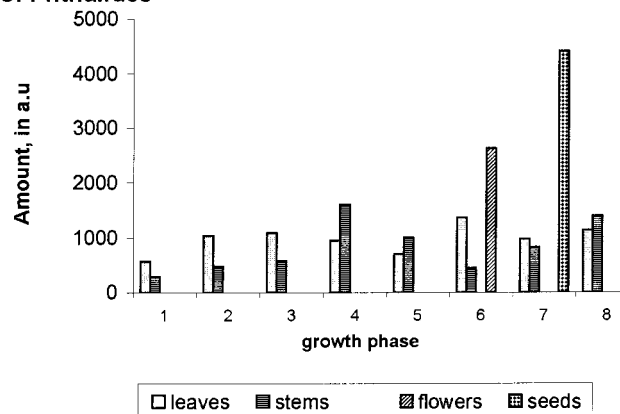


Figure 2. Amounts of monoterpenes (A), oxygenated compounds (B), and phthalides (C) in different botanical parts of lovage at different growth phases. a.u. = [essential oil yield (%) \times percentage content of individual component \times 1000]/100.

1.52%, respectively). These compounds were minor constituents or not detected at all in the seeds or flowers. On the contrary to monoterpene hydrocarbons, the content of oxygenated compounds, in general, was slightly higher at the initial phases of growth as compared with the flowering period.

Minor Constituents of Lovage. Other constituents identified in different botanical parts of lovage were in minor (<1%) and trace concentrations. A few compounds were detected only in certain parts; e.g., β -farnesene and methyleugenol were detected only in the oils from seeds and flowers, and germacrene D was not found in the stems, whereas geraniol was present only in leaves and stems. Three other phthalides were identified in all botanical parts at a concentration of up to 0.50%. A very specific compound of lovage essential oil, namely, pentylcyclohexadiene, present in appreciable amounts in the roots (Venskutonis, 1995), was found at all harvesting periods in all botanical parts; however, its concentration did not exceed 1% in the total oil.

CONCLUSIONS

The seasonal changes in the yield of essential oil in lovage leaves was less significant than in the stems, where it increased more than 2-fold from the initial phase of vegetation until the period of bud formation and subsequent decrease again. The highest yield of oil was found in the seeds followed by the flowers.

The major constituents of essential oils from lovage leaves, stems, flowers, and seeds are α -terpinyl acetate, β -phellandrene, and Z-ligustilide, constituting from 73 to 88% of the total oil. α -Terpinyl acetate was the dominating compound in leaves and stems (up to 70%), and β -phellandrene in seeds and flowers (61.50% and 40.80%, respectively).

Seasonal changes in the concentration of individual compounds in lovage leaves and stems were found to be rather complex: in general, the content of monoterpene hydrocarbons and Z-ligustilide was higher during the flowering period, whereas the percentage concentration of oxygenated monoterpene esters and alcohols at this phase was lower as compared to the initial stages of growing.

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