Production of lavender essential oil - evaluation of functional changes using chemometric methods

THESES OF DOCTORAL (PhD) DISSERTATION

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1. Background. objectives

Essential oils constitute an important group of plant-derived products that are widely used in the food, cosmetics and pharmaceutical industries, as well as in aromatherapy. Their quality is a key determinant of their applicability and is closely related to their chemical composition. Lavender (*Lavandula angustifolia*) essential oil is one of the best-known and most widely used essential oil. Its chemical profile consists mainly of monoterpenes and their oxygenated derivatives; however, the proportions of the individual components, and therefore product quality, are influenced by numerous factors, including plant variety, cultivation conditions, climatic characteristics, harvesting, storage and the extraction technology applied.

Steam distillation is the traditional and still widely used method for the industrial recovery of essential oils. Alongside this technology, alternative extraction processes are receiving increasing attention, including supercritical fluid extraction, which uses carbon dioxide to extract volatile components at relatively low temperatures and with adjustable solvent power. The effectiveness of supercritical extraction is influenced not only by operating parameters - pressure, temperature, solvent volumetric flow rate and co-solvent content - but also by factors related to the raw material. Owing to the combined effects of cultivation site, weather conditions and the time elapsed since harvesting, the quantity and composition of the extracted essential oil can be regarded as a complex system.

The aim of my doctoral research was to comprehensively investigate the functional changes occurring during the production of lavender essential oil using supercritical fluid extraction and steam distillation. Chemometric methods were applied to identify and rank the effects of multiple interrelated variables. Correlation analyses, regression models and variable-importance methods enabled me to evaluate not only the relationships between individual factors and the response variables examined, but also the relative contribution of the parameters. I determined and ranked the effects of the main operating parameters of supercritical extraction on yield and composition, investigated the role of the origin of the raw material, the time elapsed since harvesting and aggregated meteorological indicators, and compared the importance of these factors for the two extraction technologies. In addition, steam distillation generates a large amount of lavender hydrosol, for which rapid quality control is important for the further utilization of this by-product. Therefore, I developed a rapid estimation method based on UV-Vis spectroscopy and chemometric modeling to evaluate the volatile components of lavender hydrosol.

2. Experimentals and evaluation methods

2.1. Lavender raw material

For the experiments, *Lavandula angustifolia* lavender was used, harvested in Tihany and Tamási every July between 2021 and 2025 in the morning hours. After harvesting, the plants were tied into bunches and dried to constant mass in a shaded, well-ventilated area. To investigate environmental factors associated with the raw material, mean temperature and total precipitation during the plant maturation period from March to July were used as aggregated meteorological indicators.

2.2. Steam distillation

Lavender essential oil was extracted by steam distillation at both laboratory and industrial scale. In the laboratory apparatus, 100-200 g of plant material was processed per batch. The dried lavender was cut into 1-2 cm pieces and placed in the distillation vessel. Steam at 2 bar was introduced at the bottom of the vessel. As the steam passed through the plant bed, it carried the volatile components into the condenser, where they condensed. The condensate was collected in a phase separator, where the essential oil and lavender hydrosol were separated. The laboratory distillation time was 30 min. During the industrial experiments performed in Tihany, 250-350 kg of fresh lavender was processed per batch, allowing the laboratory results to be compared with samples obtained under industrial conditions.

2.3. Supercritical fluid extraction

Supercritical fluid extraction experiments were conducted in a laboratory-scale apparatus using carbon dioxide as the solvent and ethanol as the co-solvent. The extraction column was packed with ground lavender with a particle size of 400-2000 μm. During the experiments, the temperature was varied between 40 and 60 °C, the pressure between 100 and 200 bar, the solvent volumetric flow rate between 4 and 8 mL/min, and the co-solvent content between 1 and 3% (V/V). The extraction time was 60 min in all measurements. Essential oil yield was calculated from the masses of the raw material charged and the product recovered.

2.4. Analytical methods

The chemical composition of lavender essential oils was analyzed by gas chromatography-mass spectrometry (GC-MS) using an Agilent 5975C instrument equipped with an HP5 column. Pharmacopoeial-grade lavender essential oil and standards of linalool, linalyl acetate and 4-terpineol were used as references. Based on the mass spectra and the standards, 19 components were identified. The effect of parameter changes on composition was evaluated

from changes in the areas under the identified chromatographic peaks. Thin-layer chromatography was also used for supplementary rapid qualitative analysis.

2.5. Chemometric and statistical methods

During data analysis, I examined the relationships between technological, environmental and raw-material characteristics affecting lavender essential oil yield and composition, and ranked the relative importance of the predictor variables. Pearson correlation, Spearman rank correlation and p-value analysis were used to explore preliminary relationships. The effects of input variables were evaluated using linear regression; the LMG index was applied to rank the relative importance of variables, while LASSO regression was used for simultaneous variable selection and regression. Calculations were performed in MathWorks MATLAB R2021a.

2.6. UV-Vis/PLS analysis of lavender hydrosol

UV-Vis spectrophotometry was used to analyze the lavender hydrosol generated as a by-product of steam distillation over the wavelength range of 200-600 nm after a 100-fold dilution. The volatile components were recovered by liquid-liquid extraction and their composition was determined by GC-MS. A PLS regression model was constructed from chromatographic concentration data and UV-Vis spectra to estimate the amounts of oil components in the lavender hydrosol.

3. Theses

I. I investigated the effects of the operating parameters applied during the supercritical extraction of *Lavandula angustifolia* - pressure, extraction temperature, solvent volumetric flow rate and co-solvent content - on the yield and composition of the extracted essential oil. Using Pearson and Spearman correlation, p-value analysis, the LMG index and LASSO regression, I established that, within the experimental range investigated, pressure and solvent volumetric flow rate were the most important variables affecting yield. Increasing either parameter had a positive effect on yield. With respect to essential oil composition, pressure and extraction temperature showed the strongest relationships with the amounts of individual components. Increasing pressure showed a positive relationship with the amount of linalool and a negative relationship with the amount of linalyl acetate, while extraction temperature showed a weaker negative trend with the amount of linalool. Solvent volumetric flow rate had a smaller effect on composition, whereas co-solvent content was not found to be a determining variable within the investigated range. Based on my results, the operating parameters contribute to the quantitative and compositional characteristics of lavender essential oil to different extents; therefore, the variable-importance ranking can be used to support the targeted selection of technological parameters.

II. I investigated the relationships between factors affecting the characteristics of the *Lavandula angustifolia* raw material - weather conditions, cultivation site and time elapsed since harvesting - and the yield and composition of the essential oil obtained by supercritical extraction. Based on the applied correlation, regression and variable-importance methods, I established that, in the dataset investigated, factors describing raw-material variability often showed greater variable importance than the operating parameters. Pressure was positively related to yield, whereas extraction temperature was not found to be a determining variable in this dataset. Cultivation site showed relationships with yield and with the amounts of α-pinene and linalool, while the time elapsed between harvesting and processing showed a negative relationship with the amount of linalyl acetate. Among the aggregated meteorological indicators, precipitation correlated with the proportions of linalool and 4-terpineol, while mean temperature correlated with the amounts of β-cis- and β-trans-ocimene. I established that, in the investigated dataset, the yield and chemical profile of lavender essential oil obtained by supercritical extraction cannot be explained solely by operating parameters. The origin of

the raw material, the time elapsed since harvesting and the meteorological characteristics of the cultivation period also play important roles.

III. During the extraction of *Lavandula angustifolia* essential oil by steam distillation, I investigated the relationships between factors affecting the raw material - particularly the time elapsed since harvesting and meteorological indicators - and the yield and composition of the extracted essential oil. I established that, in the investigated dataset, the time elapsed since harvesting showed a negative relationship with essential oil yield and with the amounts of certain components, particularly linalool and 4-terpineol. For linalyl acetate, a non-linear change was observed, suggesting the possible existence of an optimal storage-time range. Among the aggregated meteorological indicators, mean temperature showed a positive relationship with the amount of linalool, while precipitation showed a positive correlation with both yield and linalool content. By comparing the results of steam distillation and supercritical extraction, I established that the trends associated with time elapsed since harvesting and meteorological indicators do not always appear in the same direction or to the same extent for the two extraction technologies. Based on my results, the effect of the raw material's prior history and environmental characteristics on the quantity and composition of the extracted essential oil is technology-dependent.

IV. I developed a rapid, simple and cost-effective estimation method based on UV-Vis spectrophotometry and PLS regression for investigating the volatile components of lavender hydrosol generated as a by-product during steam distillation of lavender essential oil. Lavender hydrosol samples of different origins, produced under industrial and laboratory conditions, were used for method development. Following liquid-liquid extraction, the composition of the extracts was determined by GC-MS, after which a calibration database was created from the UV-Vis spectra of the extracts and raffinates together with the chromatographic data. I established that, within the sample types, concentration ranges and measurement conditions covered by the calibration database, the developed UV-Vis/PLS-based model is suitable for rapid estimation of the amounts of selected valuable components in lavender hydrosol. The method can be applied as a preliminary screening and quality-control procedure, supporting rapid comparison of samples and assessment of whether further detailed chromatographic analysis is required.

4. Publications

Articles

1. Preiner, S.; Tarcsay, B. L.; Juhász, C. J.; Pethő, D.; Miskolczi, N. Identifying key parameters to maximize lavender essential oil components during supercritical extraction using machine learning models. Chemical Engineering and Processing - Process Intensification, 2025, 110523. DOI: 10.1016/j.cep.2025.110523
2. Preiner, S.; Tarcsay, B. L.; Pethő, D.; Miskolczi, N. Lavender hydrosol analysis using UV spectroscopy data and partial least squares regression. MethodsX, 2025, 14, 103304. DOI: 10.1016/j.mex.2025.103304
3. Preiner, S.; Petho, D.; Miskolczi, N. Effect of the Supercritical Extraction Parameters on Lavender Essential Oil Yield and Composition. Chemical Engineering Transactions, 2025, 117, 937-942. DOI: 10.3303/CET25117157

Conference Proceedings

1. Preiner S., Pethő D., Miskolczi N.: Levendula illóolaj előállítás szuperkritikus extrakcióval, Műszaki Kémiai Napok, Veszprém, 2022. április 26-28.
2. Preiner S., Pethő D., Miskolczi N.: Paraméterváltoztatás hatása a szuperkritikus extrakcióval kinyert levendula illóolaj mennyiségére és minőségére, PhD hallgatók anyagtudományi napja XXII., Veszprém, 2022. november 14.
3. Preiner S., Pethő D., Miskolczi N.: A levendula illóolaj-tartalmának és minőségének változása a betakarítást követően, Műszaki Kémiai Napok, Veszprém, 2023. április 18-20.
4. Preiner S., Pethő D., Miskolczi N.: Effect of the environmental phenomena and elapsed time since harvesting on the quality and quantity of extracted lavender oil, SSCHE, Tatranské Matliare, Szlovákia, 2024. május 20-24.
5. Preiner S., Pethő D., Miskolczi N.: Levendula illóolaj kinyerése különböző módszerekkel, PhD hallgatók anyagtudományi napja XXIV., Veszprém, 2024. november 18.
6. Preiner S., Pethő D., Miskolczi N.: A kinyerés módjának és a környezeti körülményeknek a hatása a levendula illóolajra, Műszaki Kémiai Napok, Veszprém, 2025. április 15-17.
7. Preiner S., Pethő D., Miskolczi N.: A vízgőzdesztilláció és a szuperkritikus extrakció hatékonyságának vizsgálata levendula illóolaj kinyerésére, XVIII. Szent-Györgyi Albert konferencia, Budapest, 2025. május 9-10.

8. Preiner S., Pethő D., Miskolczi N.: Optimization of lavender essential oil extraction: Method comparison and identification of key parameters, 51st International Conference of SSCHE, Hotel DRUŽBA, Jasná, Demänovská Dolina, Szlovákia, 2025. május 27-30.
9. Preiner S., Pethő D., Miskolczi N.: Effect of the supercritical extraction parameters on lavender essential oil yield and composition, iCheaP 17, Firenze, Olaszország, 2025. június 29. - július 2.