

**Quality, energy requirement and costs of drying tarragon
(*Artemisia dracunculus* L.)**

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Dit onderzoek is uitgevoerd binnen de onderzoekschool MGS

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Akbar Arab Mohammad Hosseini

Proefschrift

ter verkrijging van de graad van doctor
op gezag van de rector magnificus
van Wageningen Universiteit,
Prof. Dr. M.J. Kropff
in het openbaar te verdedigen
op maandag 7 november 2005
des namiddags te half twee in de Aula

Akbar Arab Mohammad Hosseini (2005)

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Thesis Wageningen University

- with references
- with summary in English, Dutch and Farsi

ISBN: 90-8504-297-6

Abstract

Tarragon (*Artemisia dracunculus* L.) is a favorite herbal and medicinal plant. Drying is necessary to achieve longer shelf life with high quality, preserving the original flavor. Essential oil content and color are the most important parameters that define the quality of herbal and medicinal plants. Hot air batch drying is the most common drying method for these plants but affects the essential oil content and color. The drying conditions affect essential oil content and color as well as the energy consumption and costs. Process engineers and farmers need to know how they have to dry to obtain the best quality. The objective of this work is to investigate the conditions for optimal drying in terms of quality, energy consumption and costs.

Adsorption and desorption experiments were done to find the equilibrium moisture content and water exchange between the material and surrounding air during drying and storage at temperatures of 25°C to 70°C and relative humidities of 5% to 90%. Drying of tarragon leaves and chopped plants was investigated separately and the best model was selected from the drying equations in literature. The effect of drying temperature and relative humidity on the essential oil content and color change was studied. Experiments were done at temperatures of 40°C to 90°C and the optimal conditions were. Long-term effects of the drying conditions were also investigated during the storage time. Material dried at 45, 60 and 90°C was stored and the essential oil content and color of the material was measured after 15, 30, 60 and 120 days of storage. Drying at 45°C was found as the best condition based on the changes of essential oil and color during drying and storage.

Optimization of drying of tarragon was studied based on the results of the sorption isotherms, drying equations and the changes of essential oil content and color during drying and storage. Models were made for the drying process, energy consumption and cost calculation. The current conditions in The Netherlands and Iran were applied for various drying scenarios. The simulation model was run at selected ranges of temperature, humidity ratio, air speed and bed heights, with and without recirculation of the drying air. Considering the quality of dried material and costs, a temperature of 50°C was found as the optimal temperature for drying tarragon. The costs per kg dried product was about three times higher for The Netherlands compared to Iran. The recirculation is less important in Iran because the ambient air has a higher drying potential and energy prices are lower.

Acknowledgment

I am very grateful, deep down in my hearth, what God has done in my life.

This work reflects exactly what I have been up to in the last four years. It started by browsing the Wageningen University website to apply for a PhD program. After some communication and correspondence with Dr. Ir. Willem Huisman, I got admission to start my PhD education in the Farm Technology Group. Now I am almost at the end of this period and it is a pleasure to express my gratitude to all whom have been involved in my work. Since it is impossible to thank them all by name, I will therefore only mention those who were essential to accomplish this study.

First of all I would like to express my special gratitude to my wife who was always alongside me and also for her patience. Many thanks to my daughter *Fatemeh* who supported me mentally and to my son *Matin*, who was recently attended to our family and made our life much sweeter. I am deeply grateful to my father and my mother, who gave me the courage to get my education and supported me in all achievements. I appreciate my father-in-law and my mother-in-law for their support and taking care of my family for some time. I also thank my brothers and sisters for their help in whole my life.

I am grateful to all my supervisors; *Prof. Dr. Ing. Joachim Müller*, *Dr. Ir. Willem Huisman* and *Dr. Ir. Ton van Boxtel*; without your guidance it would have been impossible to do my research. Thank you very much for keeping up the spirit, I really enjoyed working with you.

Prof. Dr. Ing. Joachim Müller, you offered me your experience in the field of drying of medicinal plants. You were strongly guiding me, even after you left to Hoheinheim University.

Dr. Ir. Willem Huisman, when I think about Wageningen University, you are the first person who comes to my mind because my first contact was with you. You arranged my admission, and you were the person who was always in touch for all miscellaneous matters (educational and social) and you learned me a lot.

Dr. Ir. Ton van Boxtel, in the second half of my research period the discussion with you about my work increased. Now I can see how effective our discussion to improve my work was. I learned a lot from you about modelling and programming. Once again thank you all very much.

I am very grateful to all my colleagues of the Farm Technology Chair, especially *Sam Blaauw*, *Albert Boers*, *Michel Govers*, *Corrie Seves*, *Miranda Tap*, my room mate *Ana de Lima* and other colleagues for all their help, support, interest and valuable hints.

Albert Boers, I never forget your friendly assistance to prepare the requirements for my work, translation of the Dutch letters and all you have done for my private requests.

I really appreciate the help of the people of the Laboratory of Organic Chemistry, *Teris van Beek*, *Maarten Posthumus* and *Elbert van der Klift* concerning the GC and GC-MS experiments.

Many thanks to Agrotechnology and Food Innovation for the use of the drying equipments. Special thank goes to *Sudhakar Padhye*, MSc student in Wageningen University, who assisted me with experiments.

I would appreciate *Hans van der Mheen* and the people of Unifarm (Wageningen University) for their advices and the tarragon crops used for my experiments, *Adrie Reimerink* and *Roel Hooghordel* from Euroma Company and also *Bauke van der Veen* from VNK Company for their help and discussions

I would like to thank the Iranian Ministry of Science, Research and Technology (MSRT), which supported and funded this research for four years. Hereby I also want to thank my former colleagues from Tehran University – Aboureihan Campus for their help.

Finally, I extend my sincere thanks to the Iranian students in Wageningen University and their family members. We had a great time and nice gatherings together during this time and it helped us to keep ourselves fresh and relaxed.

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1

General introduction

1 Introduction

Agricultural crops stored at high moisture content will deteriorate because of microbiological growth. One of the most common preservation methods for herbs and spices is drying [1]. Transport and storage costs are lower after drying since weight and volume decreases. Drying is a process of simultaneous heat and moisture transfer, and can be done by various systems. Crops have different structures, shapes and characteristics and therefore they have a different drying behavior.

The use of herbal plants has increased significantly over the past few years, due to renewed interest in dishes that use a wide variety of spices [2]. The focus of this research is tarragon, one of such herbs and also a medicinal plant. During the drying process the condition, or quality, of tarragon changes. It can result in loss of essential oil, changes in color and texture and decrease in nutritional value. Consumer prefer processed products that keep more of their original characteristics [3]. Correct drying is needed to save the quality during the drying process, longer shelf life and support marketing. The drying conditions also define energy and capital costs. In brief, quality of the products and income are strongly related to each other and optimum drying causes higher income.

Nowadays, food supply networks are globally active with complex relationships. Currently, even fresh products can be shipped far away around the world. There is always competition between the food companies on the world market. Good quality and low price are more important than ever before, because consumers have a choice out of a number of products offered by competing chains. Optimization of individual links in the chain usually results in sub-optimal overall chain performance. Optimization of the production chain consisting of crop harvest, processing and storage helps to have a good performance and higher income.

The main goal of this research is to find the optimum scenario for the drying process of tarragon as defined by quality, energy consumption and costs.

2 Tarragon

2.1 Varieties and cultivation

Tarragon (*Artemisia dracunculus* L.) is a plant of the Asteraceae family, which is called the "King of Herbs" by the French. Within the genus *Artemisia*, it is one of the most polymorphic species with a very wide distribution in Eurasia and North America [4]. Two kinds of tarragon are cultivated in herb gardens. The French Tarragon, which is a native of the South of Europe and the Mediterranean area, has very smooth, dark green leaves and has the true tarragon flavor, [5]. The other variety is Russian Tarragon, native in Siberia and has an erect stem up to 1.2 m tall while the leaves are less smooth, large (about 5 cm long), narrow and dark green.

French Tarragon rarely, if ever, sets seed, so it must be propagated asexually by cuttings while the Russian Tarragon produces seeds [6].

Distinction between these two varieties is usually based on external morphology, anatomy, fertility, cytogenetics and essential oil content [6, 7].

There is another variety of tarragon, which is actually a member of the marigold family (*Tagetes lucida*), commonly called Mexican Marigold or Winter Tarragon.

2.2 Essential oil

The compounds of the essential oil in tarragon varies widely, according to geological location, climate, light conditions, soil type, and variety [8]. A number of compounds have been reported for French Tarragon and the main compounds are estragole (70% of essential oil), limonene, terpinene and elemicine [7, 9-11]. The main constituents of Russian Tarragon are, sabinene (40% of essential oil), methyl eugenol, β -ocimene and elemicin [7, 10].

2.3 Usage

Tarragon can be used fresh, dry and frozen as a flavor of foods, cream sauces and herbal vinegars, oils and mustards. French Tarragon has a long history of use as culinary and medicinal plant [8, 12]. Russian Tarragon is more used as medicinal plant. It has a traditional Persian history of use as a natural cleanser of the blood and for the treatment of headaches and dizziness [6].

3 Drying

Most common equipment for drying tarragon are batch dryers on farm level and belt dryers in industrial drying. For the experiments a laboratory thin bed dryer has been chosen, to achieve short drying times and to avoid over-drying of the bottom layer. Furthermore, in thin bed drying no differences in quality between bottom and top layer are to be expected. Variables during the drying process are temperature, relative humidity, airflow and bed height. Tarragon leaves and chopped plants are investigated in this research but the tarragon leaves are more in the focus as the main part of crop. Drying air is recirculated to minimize the energy consumption and costs. In the simulation also bed height was varied in order to investigate the consequences.

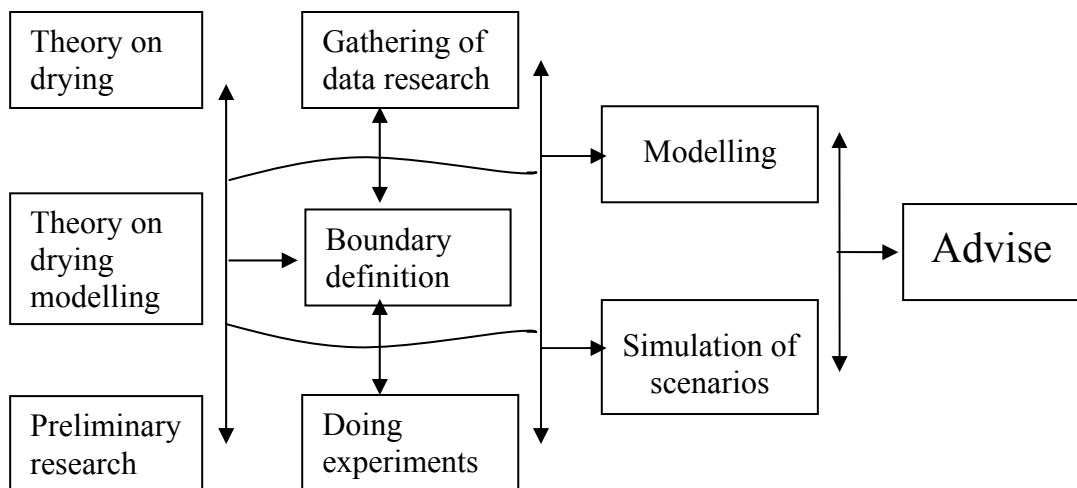


Fig. 1 Research framework of modeling and simulation for selection of relevant drying scenarios. The boxes from left to right are in time order.

4 Research framework

Fig.1 presents the structure and sequence of the stages of the research project.

In the framework diagram the horizontal arrows present the sequence of the research steps. The vertical arrows show the research steps that must be confronted with each other to draw conclusions for the next column.

5 Problem definition and objectives

Quality of the crop, energy consumption and costs in a drying process are related to each other and the crop properties and process parameters affect the level of these items. Higher quality will be more accepted by consumers and results in more income for the producer. The knowledge of the optimal adjustment of the drying process parameters is required for overall optimization of the processing chain.

The main goal of this research is to reduce energy consumption and drying costs and to optimize quality of dried tarragon in terms of essential oil and color.). Own experimental data and available information in literature are used for modeling and simulation. The research is done based on the conditions of The Netherlands and Iran.

6 Research items

The following items are studied in this research:

- The adsorption and desorption isotherms of tarragon, which determine the drying behavior
- The drying speed as influence by the relevant drying conditions
- Color and essential oil content changes during drying under these conditions
- Color and essential oil content changes during storage
- Energy consumption and costs of the drying process of tarragon by modeling

7 Boundary

In this research boundaries are defined to focus on an appropriate area of research. The boundaries are chosen based on the goal of the research, duration of the research project, available information in literature and technical facilities. The boundaries are mostly defined by a minimum and a maximum value of numerical parameters like temperature, relative humidity, air velocity and bed height. Climate conditions and economic factors are selected on both, Dutch and Iran conditions.

8 Thesis outline

Chapter 2 deals with the sorption isotherms of tarragon (stems and leaves separately). The desorption curve is relevant for drying and adsorption curve for storage conditions. In **chapter 3** the drying speed of tarragon leaves and chopped whole plants at temperatures of 40°C to 90°C and certain relative air humidities have been investigated and the best fitting

equation for modeling of the drying behavior has been selected among the available drying equations in literature. The effect of drying on the essential oil content is evaluated in **chapter 4** and the absolute values of oil in the dried samples are given. In this chapter the compounds are also analyzed individually and compared to find interactions between the compounds. In **chapter 5**, the effect of drying on the color is evaluated based on the color parameters L^* , C^* and h^o . As the essential oil and the color of the dried material can change during storage, in **chapter 6** the long term effect of drying on these parameters has been investigated during 120 days of storage time. As the main goal of this research is optimization of the drying of tarragon, in **chapter 7** all information of chapters 2 to 6 is used to find the optimal operation mode of dryers in terms of energy consumption, quality and costs.

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2

Modeling of the equilibrium moisture content (EMC) of tarragon (*Artemisia dracunculus* L.)

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Accepted in “International Journal of Food Engineering”

Abstract

The equilibrium moisture content of tarragon, *Artemisia dracunculus* L. (stem and leaf separately) was determined by using the saturated salt solutions method at three temperatures (25, 50 and 70°C) within a range of 5 to 90% relative humidity. Both adsorption and desorption methods were used for stem and leaf of two varieties: French and Russian Tarragon. Experimental curves of moisture sorption isotherms were fitted by modified Henderson, modified Halsey, modified Oswin, modified Chung-Pfost and GAB equation and evaluated by residual sum squares, standard error of estimate and mean relative deviation. The modified Halsey and GAB equations were found to be the most suitable for describing the relationship among equilibrium moisture content, relative humidity and temperature. There was no significant difference between the equilibrium moisture content of the French and Russian Tarragon.

Keywords: Adsorption; *Artemisia dracunculus*; desorption; drying; storage; tarragon.

1 Introduction

Tarragon (*Artemisia dracunculus* L.) is an aromatic, perennial plant of the Asteraceae family. Its long narrow leaves have a strong, spicy anise flavor. The reported life zone of tarragon is 7 to 17°C with an annual precipitation of 300 to 1300 mm and a soil pH of 4.9 to 7.8. Tarragon grows best in warm, sunny locations on dry soils with good drainage. It can be used fresh, dried and frozen as a flavor for foods, herbal vinegars, oils and mustards. At least two varieties of *A. dracunculus* are known in Europe: “French Tarragon” (sometimes also called “German Tarragon” or “True Tarragon”) and “Russian tarragon”, which is native in Russia and Western Asia [1]. Tarragon also is considered as a medicinal plant. The major components of essential oil are methyl chavicol, limonene, sabinene, β -ocimene in French Tarragon and sabinene, methyl eugenol, β -ocimene, γ -terpinene and elemicin in Russian Tarragon. [2, 3]

Moisture sorption isotherms define the relation between Equilibrium Relative Humidity (*ERH*) and Equilibrium Moisture Content (*EMC*) [4]. This information is required for drying and storage of agricultural and food products, for instance to maintain the quality in the storage period. This knowledge is also required to stop the drying process at the aimed moisture content to avoid quality losses and to save energy [5].

EMC is defined as the moisture content of a hygroscopic material in equilibrium with a particular environment in terms of temperature and relative humidity. *EMC* of the product is the final result of moisture exchange between the product and the air surrounding the sample. In this condition, the water in a product is in balance with the moisture in the surrounding atmosphere [6]. The relative humidity in this condition is known as the Equilibrium Relative Humidity (*ERH*) [4]. Moisture sorption isotherms are either measured during desorption (starting from the wet state) or during adsorption (starting from the dry state).

Not much information is available about the sorption isotherms of tarragon. Therefore the objective of this study is to find the desorption and adsorption isotherm curve of tarragon (stem and leaf) at temperatures between 25 and 70°C for *ERH* values in the range of 5 to 90% and to find appropriate equations for it. The main part of the tarragon is leaf. In practice separation of stems and leaves will be done either before or after drying process. Therefore, stems and leaves were evaluated separately. As the moisture desorption isotherm is one of the basic parameters of the drying process and the adsorption isotherm is important for storage, this work will give an important contribution to the optimization of the drying of tarragon.

2 Materials and Methods

2.1 Plant material

Artemisia dracunculus of the varieties French and Russian Tarragon has been used for this research. The Russian variety was planted at Wageningen University (The Netherlands) and the French variety was planted in Elburg (The Netherlands) in 2003 and 2004. The plants were harvested just before flowering. Stems and leaves were separated immediately after harvesting. The stems were chopped to 20 to 30mm long pieces. The samples of 10-12g fresh leaves and 15-20g fresh stem were used for desorption tests. For the adsorption process, first the separated leaves and chopped stems were stored in room condition to reach a low moisture content. Then the dry materials were dehydrated in a desiccator over P_2O_5 at room temperature for three weeks. The moisture content of the samples was less than 2% at the end of this period.

2.2 Experimental procedure

The apparatus for providing various *ERHs* consists of an insulated water bath of dimensions $56 \times 36 \times 26$ cm with 14 glass jars and a cryostat, which includes a cooling and heating system with an accuracy of 0.1°C . The water circulates between cryostat and water bath to provide a constant temperature of water around the glass-jars. The glass jars are closed with a cap. Every glass jar of 800ml contains 150ml of saturated salt solution. A plate with base prevents contact of sample and salt solution. Stainless steel netballs were used to put the stems and leaves separately in the jars. In tests with relative air humidity above 60% crystalline thymol was placed in the bottle to prevent microbial spoilage [7a]. The cryostat was adjusted to 25, 50 and 70°C . Every three days, the samples were weighed with an accuracy $\pm 0.1\text{mg}$. Equilibrium was acknowledged when three consecutive weight measurements showed a difference less than 1 mg.

When equilibrium has been reached (approximately after 3 weeks depending on the temperature and relative humidity), *MC* of the samples was determined gravimetrically using the oven method (105°C for 24h) [8]. The *EMCs* were determined by triplicate measurements of each sample.

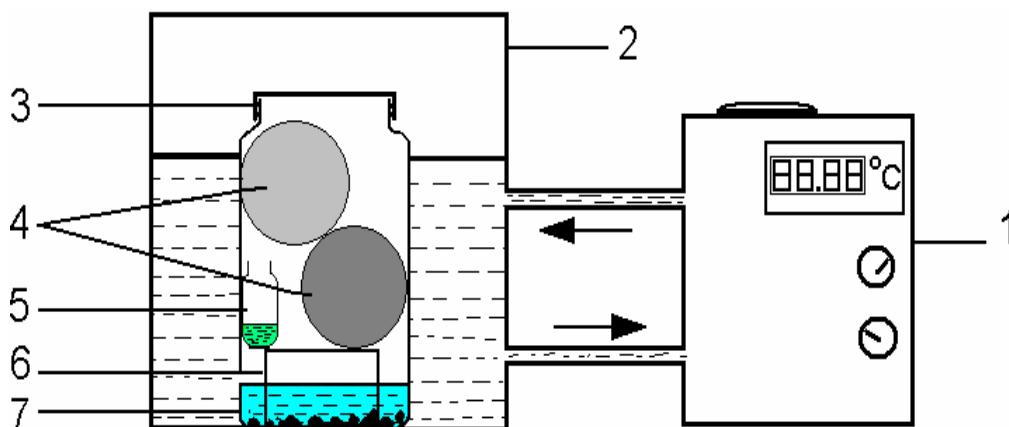


Fig. 1 Equipment for the sorption isotherm measurement: 1) cryostat; 2) water bath; 3) jar with cap; 4) netballs including samples; 5) thymol; 6) base; 7) saturated salt solution.

2.3 Salt solutions

The saturated salt solution method is based on the fact that *ERH* of specific salt solutions is a well known physical property [9]. Table 1 gives the equivalent of relative humidity of the selected salt solutions at three temperatures. Salt solutions were provided by mixing and dissolving of the salt crystals in distilled water at a 20°C higher temperature than the later one in the water bath, to guarantee a saturated solution. To avoid the formation of a concentration gradient in the liquid phase, the salt solution was agitated from time to time [10].

Table 1 Saturated Salt solutions and Equilibrium Relative Humidity at different temperatures [9]; [11].

| Salt formula | Equilibrium Relative Humidity (ERH) (%) | | |
|--------------------------------|---|--------|--------|
| | T=25°C | T=50°C | T=70°C |
| K ₂ SO ₄ | 97.30 | 95.82 | 94.50 |
| KNO ₃ | 93.58 | 84.78 | n.f.* |
| KCl | 84.34 | 81.20 | 79.49 |
| KBr | 80.89 | 79.02 | 79.07 |
| NaCl | 75.29 | 74.43 | 75.06 |
| NaNO ₃ | 74.25 | 69.04 | 66.04 |
| KI | 68.86 | 64.49 | 61.93 |
| NaBr | 57.57 | 50.93 | 49.70 |
| K ₂ CO ₃ | 43.16 | 40.91 | 37.37 |
| MgCl ₂ | 32.78 | 30.54 | 27.77 |
| LiI | 17.56 | 12.38 | 7.23 |
| LiCl | 11.30 | 11.10 | 10.70 |
| ZnBr ₂ | 7.75 | 7.70 | 8.72 |
| LiBr | 6.37 | 5.53 | 5.23 |

* n.f.: not found in literature, so not applied.

2.4 Equations for sorption isotherms

A number of equations have been suggested in literature to describe the relationship between *EMC* and *ERH*. The modified Henderson, modified Oswin and modified Halsey, modified Chung-Pfost and GAB equation [12] have been adopted by the American Society of Agricultural Engineers as standard equations for describing sorption isotherms [13]. We transformed the equations to get *EMC* as dependent parameter and *ERH* as independent variable:

Modified Henderson

$$EMC = \left(-\frac{1}{C_1(T+C_2)} \ln(1-ERH) \right)^{1/C_3} \quad (1)$$

Modified Halsey

$$EMC = \left(\frac{-\exp(C_1 + C_2 T)}{\ln(ERH)} \right)^{1/C_3} \quad (2)$$

Modified Oswin

$$EMC = (C_1 + C_2 T) \left(\frac{ERH}{1-ERH} \right)^{1/C_3} \quad (3)$$

Modified Chung-Pfost

$$EMC = \frac{1}{C_1} \ln \left(\ln(ERH) \frac{(C_2 - T)}{C_3} \right) \quad (4)$$

GAB equation

$$EMC = \frac{C_1 C_2 C_3 (ERH)}{[1 - C_2 (ERH)][1 - C_2 (ERH) + C_2 C_3 (ERH)]} \quad (5)$$

EMC (dry basis) and *ERH* are given dimensionless. C_1 , C_2 and C_3 are the equation coefficients and T is temperature in °C.

The parameters C_2 and C_3 in the GAB equation are correlated with temperature using the following equations [14]:

$$C_2 = C_4 \exp \left(\frac{C_6}{RT_a} \right) \quad (6)$$

$$C_3 = C_5 \exp \left(\frac{C_7}{RT_a} \right) \quad (7)$$

C_4 , C_5 , C_6 and C_7 are coefficients and T_a is the absolute temperature (K) and R is the universal gas constant ($R=8.314$ kJ/kmolK).

Nonlinear regression was used to fit the five equations to the experimental data. The quality of the fitted curves were evaluated by using residual sum of square (RSS), standard error estimation (SEE) and mean relative deviation (MRD) [15].

RSS is defined as:

$$RSS = \sum_{i=1}^m (EMC - \overline{EMC})^2 \quad (8)$$

SEE, which is the conditional standard deviation of *ERH*, represents the fitting ability of the equations for the given data points:

$$SEE = \sqrt{\frac{\sum_{i=1}^m (EMC - \overline{EMC})^2}{df}} \quad (9)$$

The value of *MRD* shows the fitting of the curves.

$$MRD = \frac{1}{m} \sum_{i=1}^m \frac{|EMC - \overline{EMC}|}{EMC} \quad (10)$$

The smaller the values of these statistical parameters the better fits the equation.

The residuals of the *EMC*, obtained for each equation, were also plotted against the measured values and assessed visually as random or patterned. If the residual plots indicate a clear pattern, the equation should not be accepted because a systematic error is involved [16].

3 Result and discussion

3.1 Experimental results

Leaves of French Tarragon and Russian Tarragon were examined under the same conditions to identify potential impact of varieties (Fig.2). At higher *ERH* values the level of French Tarragon was a little higher than Russian Tarragon but there were no significant differences between the two varieties.

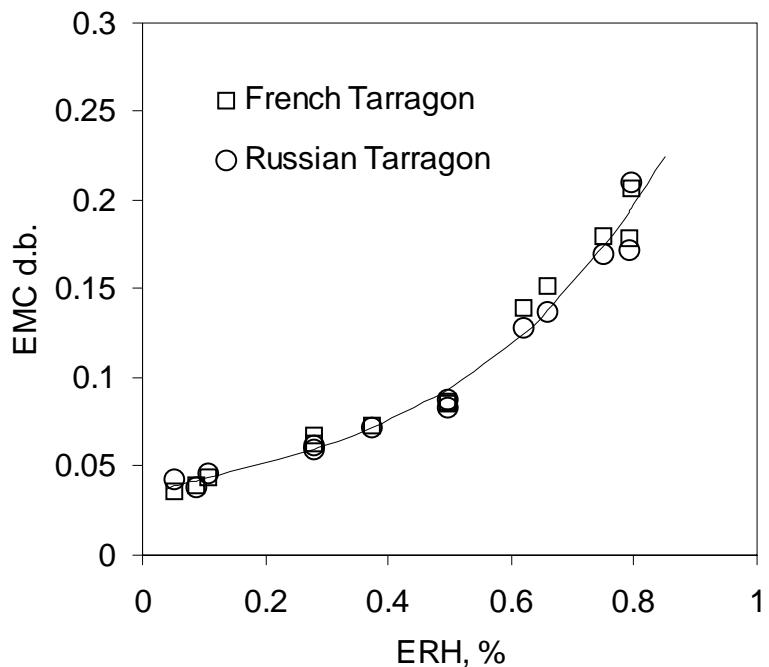


Fig. 2 Desorption data of the French and Russian Tarragon leaves at 70 °C and fitted isotherm of the Halsey equation.

Fig.3 shows the experimental results for desorption of tarragon leaves and the modified Halsey equation fitted to the data at 25, 50 and 70°C. The typical sigmoid curves were found for all three temperatures. The comparison of the three isotherms shows that at a given relative humidity, *EMC* increases at decreasing temperature. The modified Halsey equation was fitted and showed a good fit. Suboptimal fitting was found for relative humidities below 20% at 25°C and also around 50% relative humidity at 70°C.

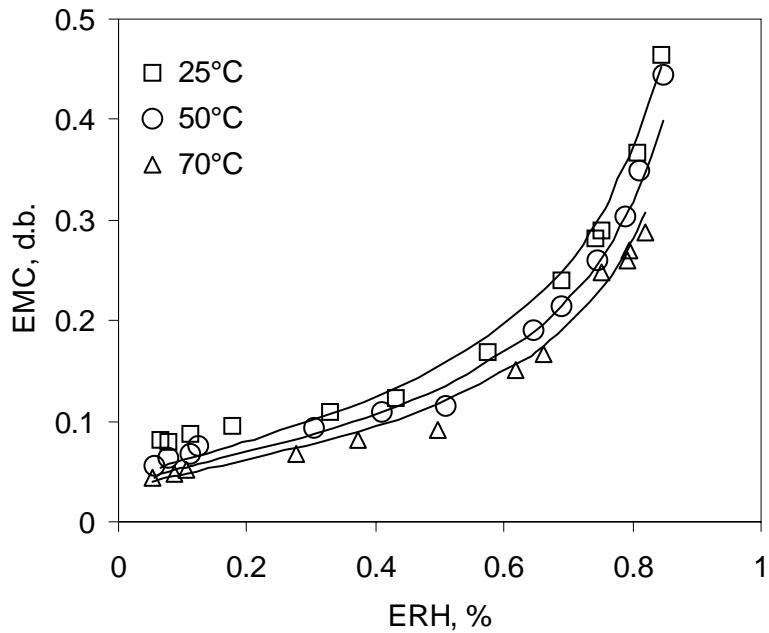


Fig. 3 Desorption isotherm data of tarragon leaves at 25°C (□), 50°C (○) and 70°C (△) and fitted curves of the Halsey equation.

Desorption data and the fitted modified Halsey equation for tarragon stems at 25, 50 and 70°C are shown in Fig.4. Again typical S-shape curves were found, but the level of *EMC* is lower for stems than for leaves (Fig.5). The difference was more distinct for higher *ERH* than lower ones.

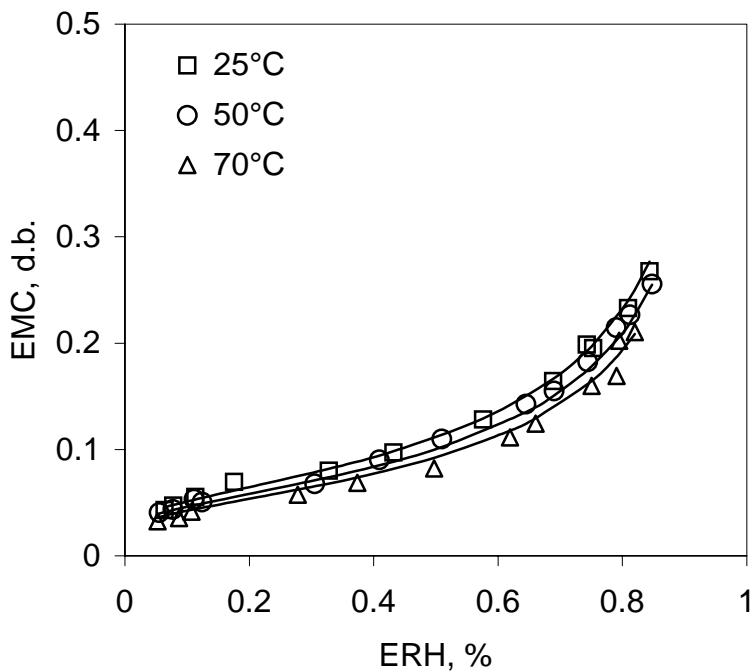


Fig. 4 Desorption isotherm data of tarragon stems at 25°C (□), 50°C (○) and 70°C (△) and fitted curves of the Halsey equation.

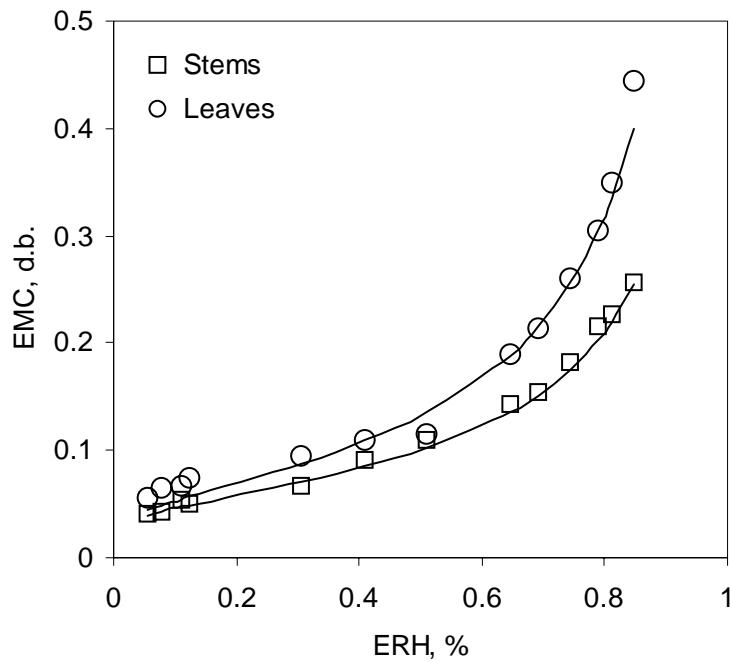


Fig. 5 Desorption isotherm data of tarragon stems and leaves at 50 °C and fitted curves of the Halsey equation.

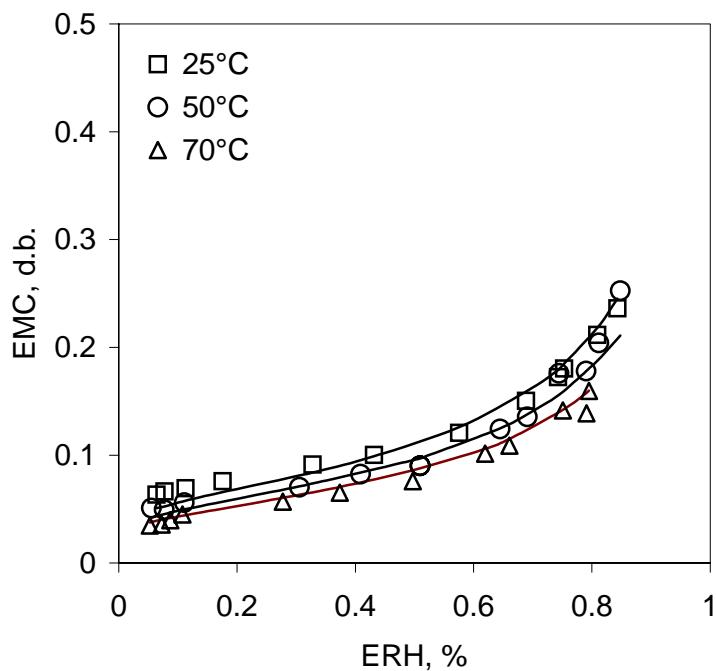


Fig. 6 Adsorption isotherm data of tarragon leaves at 25°C (□), 50°C (○) and 70°C (△) and fitted curves of the Halsey equation.

Fig.6 presents the experimental data of adsorption and the fitted curve of the modified Halsey equation for the tarragon leaves at 25, 50 and 70°C. Again fitting was suboptimal for low ERH at 25°C.

Fig. 7 shows the experimental data of adsorption and desorption of tarragon leaves at 50°C. A hysteresis effect was visible: at a given ERH, the EMC of adsorption was always lower than of desorption.

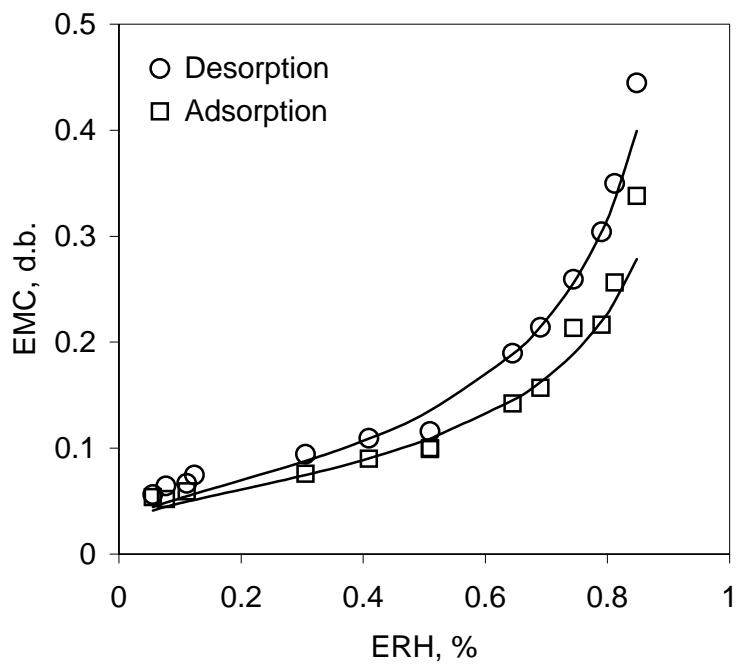


Fig. 7 Adsorption and desorption isotherm data of tarragon leaves at 50 °C and fitted curves of the Halsey equation.

Table 2 Coefficients and error parameters of the GAB equation fitted to adsorption and desorption isotherm data of tarragon stems and Leaves at three temperatures 25, 50 and 70°C.

| Parameters | Adsorption | | Desorption | |
|----------------|------------------------------|------------------------------|------------------------------|------------------------------|
| | Leaves | Stems | Leaves | Stems |
| C ₁ | 0.054 ± 0.003 | 0.049 ± 0.004 | 0.067 ± 0.003 | 0.058 ± 0.002 |
| C ₄ | 0.695 ± 0.087 | 0.652 ± 0.090 | 0.669 ± 0.045 | 0.626 ± 0.044 |
| C ₅ | 5.65 ± 4.08×10 ⁻³ | 1.74 ± 0.78×10 ⁻³ | 4.25 ± 2.03×10 ⁻³ | 2.23 ± 0.50×10 ⁻³ |
| C ₆ | 1022 ± 323 | 997.96 ± 335.25 | 1040 ± 165 | 1010 ± 174 |
| C ₇ | 25000 | 25000 | 25000 | 25000 |
| RSS | 0.0099 | 0.0076 | 0.0077 | 0.0019 |
| SEE | 0.017 | 0.015 | 0.015 | 0.008 |
| MRD | 0.106 | 0.084 | 0.090 | 0.063 |
| Residual | Random | Random | Random | Random |

3.2 Fitting of sorption equations to experimental data

The resulting coefficients of the equations are shown in Table 2, Table 3 and Table 4 along with residual sum of squares (*RSS*), standard error of estimation (*SEE*), mean relative deviation (*MRD*) and the visual judgment of the residual plots. Table 2 presents the results of the adsorption and desorption data for the leaves and stems of the GAB equation. The results of the adsorption and desorption of the first four equations are shown for leaves (Table 3) and stems (Table 4).

Table 3 Coefficients and error parameters for Henderson, Halsey, Oswin and Chung-Pfost equations fitted to adsorption and desorption isotherm data of tarragon leaves at three temperatures (25, 50 and 70°C).

| Estimated values and the variance of the equations and statistical parameters | | | | |
|---|--------------------|-------------------------------|-------------------------------|----------------|
| Parameters | Sorption equations | | | |
| | Henderson | Halsey | Oswin | Chung-Pfost |
| Adsorption | | | | |
| C1 | 0.1151 ± 0.0495 | -3.235 ± 0.107 | 0.156 ± 0.009 | 13.37 ± 0.96 |
| C2 | 65.15 ± 31.25 | -9.62 ± 1.59×10 ⁻³ | -8.20 ± 1.59×10 ⁻⁴ | 13.27 ± 12.43 |
| C3 | 1.387 ± 0.131 | 1.499 ± 0.065 | 2.053 ± 0.130 | 234.27 ± 52.54 |
| RSS | 0.0264 | 0.0083 | 0.0145 | 0.0323 |
| SEE | 0.028 | 0.016 | 0.021 | 0.031 |
| MRD | 0.228 | 0.089 | 0.159 | 0.251 |
| Residual | Systematic | Random | Systematic | Systematic |
| Desorption | | | | |
| C1 | 0.034 ± 0.012 | -2.606 ± 0.059 | 0.185 ± 0.011 | 9.155 ± 0.624 |
| C2 | 115.67 ± 44.82 | -7.76±0.97 ×10 ⁻³ | -8.89 ± 1.61×10 ⁻⁴ | 42.03 ± 27.26 |
| C3 | 1.074 ± 0.091 | 1.30 ± 0.04 | 1.713 ± 0.092 | 305.44 ± 97.64 |
| RSS | 0.0409 | 0.0087 | 0.0206 | 0.0632 |
| SEE | 0.035 | 0.016 | 0.025 | 0.043 |
| MRD | 0.266 | 0.102 | 0.185 | 0.284 |
| Residual | Systematic | Random | Systematic | Systematic |

Table 4 Coefficients and error parameters for Henderson, Halsey, Oswin and Chung-Pfost equations fitted to adsorption and desorption isotherm data of tarragon stems at three temperatures (25, 50 and 70°C).

| Parameters | Estimated values and the variance of the equations and statistical parameters | | | |
|-------------------|---|-------------------------------|-------------------------------|----------------|
| | Sorption equations | | | |
| | Henderson | Halsey | Oswin | Chung-Pfost |
| Adsorption | | | | |
| C ₁ | 0.073 ± 0.032 | -3.37 ± 0.11 | 0.123 ± 0.007 | 14.51 ± 0.93 |
| C ₂ | 117.23 ± 53.86 | -7.20 ± 1.52×10 ⁻³ | -5.63 ± 1.19×10 ⁻⁴ | 21.11 ± 14.78 |
| C ₃ | 1.24 ± 0.10 | 1.38 ± 0.06 | 1.88 ± 0.11 | 229.67 ± 52.21 |
| RSS | 0.0156 | 0.0067 | 0.009 | 0.0224 |
| SEE | 0.021 | 0.014 | 0.016 | 0.026 |
| MRD | 0.224 | 0.094 | 0.131 | 0.284 |
| Residual | Systematic | Random | Systematic | Systematic |
| Desorption | | | | |
| C ₁ | 0.086 ± 0.028 | -3.596 ± 0.057 | 0.131±0.004 | 13.80 ± 0.68 |
| C ₂ | 127.93 ± 44.06 | -6.29 ± 0.68×10 ⁻³ | -4.70±0.68×10 ⁻⁴ | 21.56 ± 11.37 |
| C ₃ | 1.42 ± 0.08 | 1.543 ± 0.031 | 2.104 ± 0.067 | 241.82 ± 43.52 |
| RSS | 0.0081 | 0.0015 | 0.0031 | 0.0148 |
| SEE | 0.016 | 0.006 | 0.009 | 0.021 |
| MRD | 0.172 | 0.050 | 0.099 | 0.218 |
| Residual | Systematic | Random | Systematic | Systematic |

The residuals of EMC of the selected equations at the three temperatures were plotted against the experimental data. The pattern was visually judged to be either randomly or systematically distributed. In most of the cases the GAB and the modified Halsey equations had a random distribution in the residual plots, shown exemplarily in Fig.8. Other equations showed systematic distribution in the residual plots. The of the residual plots for (Fig. 9).

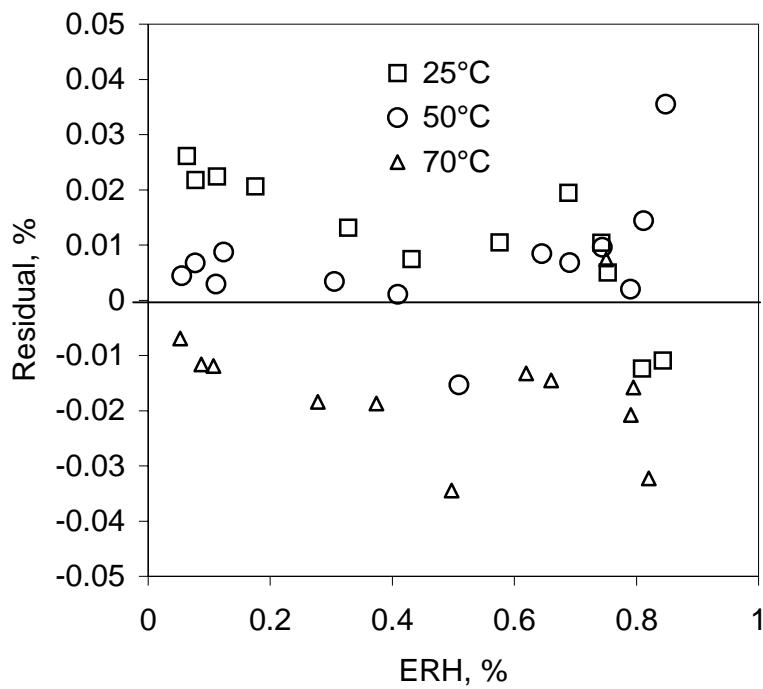


Fig. 8 Residual plot for the GAB equation for desorption data of tarragon leaves at 25°C (□), 50°C (○) and 70°C (△).

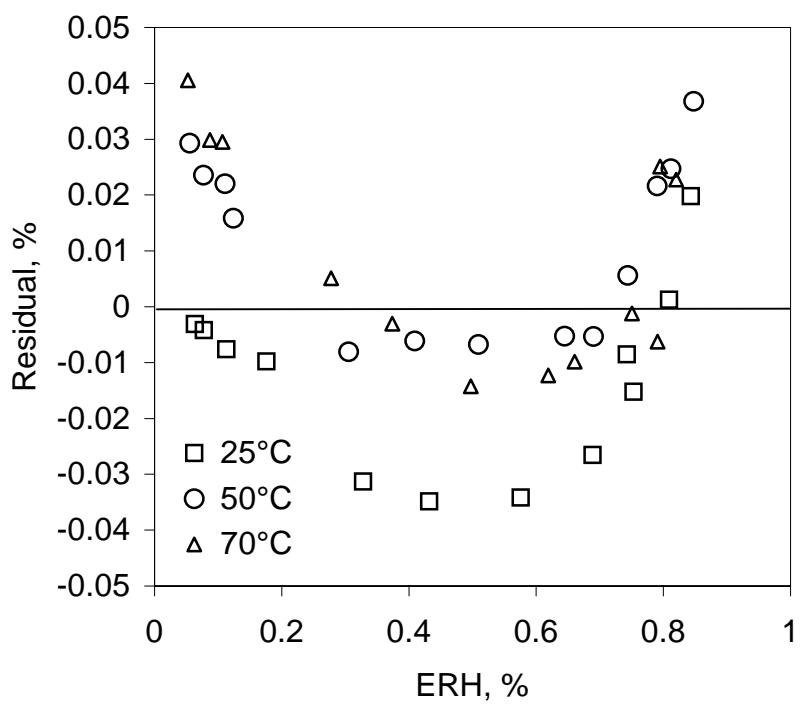


Fig. 9 Residual plot for the modified Chung-Pfost equation for desorption data of tarragon stems at 25°C (□), 50°C (○) and 70°C (△).

3.3 Discussion

As the sorption isotherms of tarragon were not found in literature, the results were compared with the available data of mint (*Mentha viridis*), sage (*Salvia officinalis*), verbena (*Lippia citrodora*)[10] and garden mint (*Mentha crispa L.*) [8]. The comparison showed that the *EMC* of tarragon leaves is similar to mint and garden mint but higher than that of sage and verbena.

This indicates that the sorption behaviour is a characteristic property of a species. In contrast, the difference in the sorption behaviour of French and the Russian Tarragon leaves was not significant.

Desorption and adsorption isotherms showed the typical hysteresis effect, i.e. *EMC* was higher for desorption of fresh material than for adsorption of dried material. *EMC* of the desorption isotherm is an essential parameter for modelling the drying process. To decide on safe storage conditions, the adsorption isotherm is relevant. Mould growth occurs at relative humidities above 70%. To be on the safe side it is recommended to set a relative humidity of 60% as threshold to derive the required moisture content for storing dried tarragon. Fig.10 helps to find out the relevant moisture contents based upon the storage conditions (temperature and relative humidity of the air). For instance if the crops has to be stored at 60% relative humidity, they must be dried to a *MC* of 0.145 at 10°C, 0.136 at 20°C and 0.129 at 30°C storage temperature (Fig.10). Drying to a lower moisture content would cause additional operation costs and mass losses without increasing storage safety.

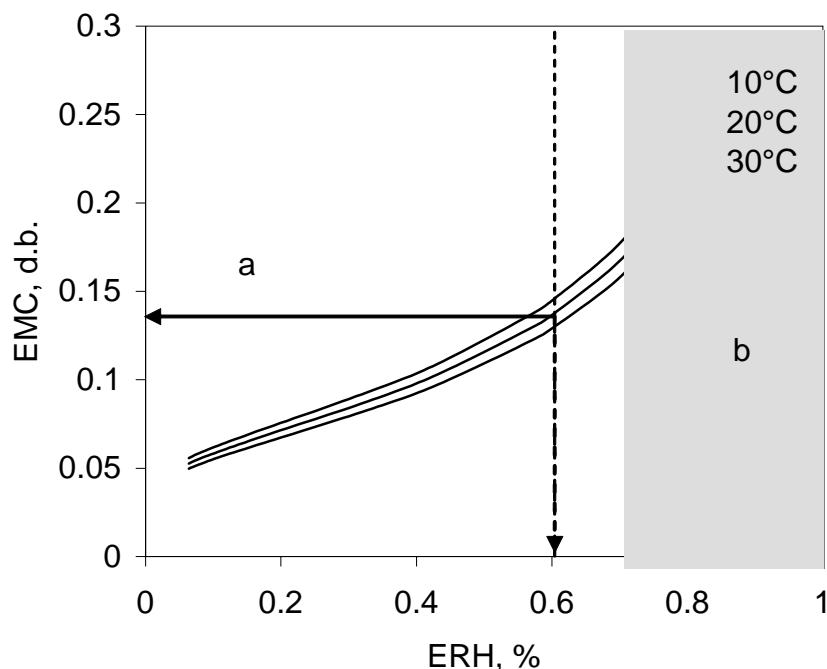


Fig. 10 **EMC domain for storage of dried tarragon a) optimum for storage b) mould growth.**

Both, the GAB and Halsey equations are suitable as they had the lowest values for *RSS*, *SEE* and *MRD*. The results are in line with the results from literature, in which was found that the GAB equation is appropriate for the isotherms of most agricultural and food materials and the modified Halsey equation tends to approximate the isotherms of oil-rich materials better [17].

Since the variance of the coefficient C_7 in the GAB equation was too wide in the first calculations, this parameter was fixed at a value of $C_7 = 25000$. The value is almost at the same level in literature for other agricultural products [7, 18].

4 Conclusion

Differences in the sorption behaviour of French and Russian Tarragon leaves are not significant. Therefore, the sorption behaviour of both varieties can be described with the same model. The moisture isotherms of tarragon stems are on a lower level than that of leaves. That means that tarragon has to be dried to lower moisture content if stems are not separated from leaves before storage. Out of five commonly used moisture sorption equations, the GAB and Halsey equation showed the best fitting. The Halsey equation was preferred because it needs only three coefficients instead of four like the GAB equation. EMC of tarragon can now be calculated for any temperature in a range of 20-70°C using our coefficients. For safe storage of tarragon leaves at a temperature of 20°C, a moisture content of 0.136 will be required. Knowledge about the required final moisture content will prevent over-drying and thus decrease drying time, energy costs, mass losses and the risk of quality deterioration.

Acknowledgment

Financial support from Ministry of Science and Research and Technology of Iran is gratefully acknowledged.

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3

Modeling of drying of tarragon (*Artemisia dracunculus* L.)

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This chapter submitted for publication

Abstract

The drying behavior of tarragon was evaluated. Tarragon leaves as well as chopped plants were dried at air temperatures ranging from 40 to 90°C at various air relative humidities and a constant air velocity of 0.6 m/s. The experimental data was fitted to a number of thin layer drying equations from the literature. Preliminary evaluation was performed to classify the equations and to select the more relevant equations. Finally three drying equations remained. The coefficients of the equations were compared by two statistical parameters as residual sum of squares and standard error of estimate. The effect of temperature and relative humidity on the coefficients of the three selected equations was evaluated by non linear regression. The results show that higher temperature cause shorter drying time, but no significant effect of relative humidity on drying rate was found. Separated leaves have shorter drying time compared to chopped plants. Although the Diffusion approach equation showed the best fit, the Page equation was selected, since it had almost a similar performance but the equation is simpler with 2 parameters instead of 3. Then a function was derived describing the relationship between the two parameter values and drying temperature.

Keywords: *Artemisia dracunculus*; drying; modeling; tarragon.

1 Introduction

Drying is the most common food preservation method used in practice [1-3]. Preservation is needed for food safety and security. Drying helps to achieve longer shelf life, lighter weight, less storage space, lower packing and transportation costs and encapsulates original flavor [4, 5].

Tarragon (*Artemisia dracunculus* L.) is mainly produced with two varieties: French Tarragon and Russian Tarragon [6, 7]. It is a strong aromatic plant, and is also considered as a medicinal plant [8]. Tarragon can be consumed as fresh, dried and frozen product. Drying is the main step in the preparation of tarragon for marketing. Since the main flavor components are in the leaves, the stems have to be separated. In practice the separation can be done either before or after the drying process. For this reason both, leaves and chopped plants (stems and leaves) were examined to evaluate the drying behavior separately. This research was based on thin layer drying with heated air.

The main objective of this research was to evaluate the effect of temperature and Relative Humidity (RH) of the drying air on the drying behavior. Furthermore another goal was to select the best drying equations, in order to use them for the calculation of drying time and energy consumption.

2 Material and method

2.1 Plant material

Russian and French Tarragon were used for the drying experiments. The French variety was collected from a research farm in Elburg (The Netherlands) and the Russian one was planted at Wageningen University (The Netherlands). Both were harvested just before flowering in July, August and September 2004. “Leaves” and “chopped plants”, were used for experiments: leaves were separated manually just before the experiment and whole plants

were chopped in length of 5 to 7 cm. For subsequent experiments the extra samples were kept in closed plastic bags in a cold room at 4°C to keep the plant as fresh as possible for maximum three days.

2.2 Experimental dryer

Drying experiments were performed at the Department of Agrotechnology and Food Science of Wageningen University. The dryer is shown in *Fig.1*. Calibrated sensors (Vaisala HMP-233A) were used to measure temperature and RH in the dryer and also to measure and control temperature and RH in the air conditioner. The accuracy of the temperature sensor was $\pm 0.2^\circ\text{C}$ and the accuracy of the RH sensor was $\pm 2\%$ in the range of 0 to 90% RH and $\pm 3\%$ for 90 to 100% RH. A balance with an accuracy of 0.01 g was used to measure the weight of the samples. Two trays of 14×28 cm were used for drying. The air velocity in the tubes just before the trays was 0.6 m/s.

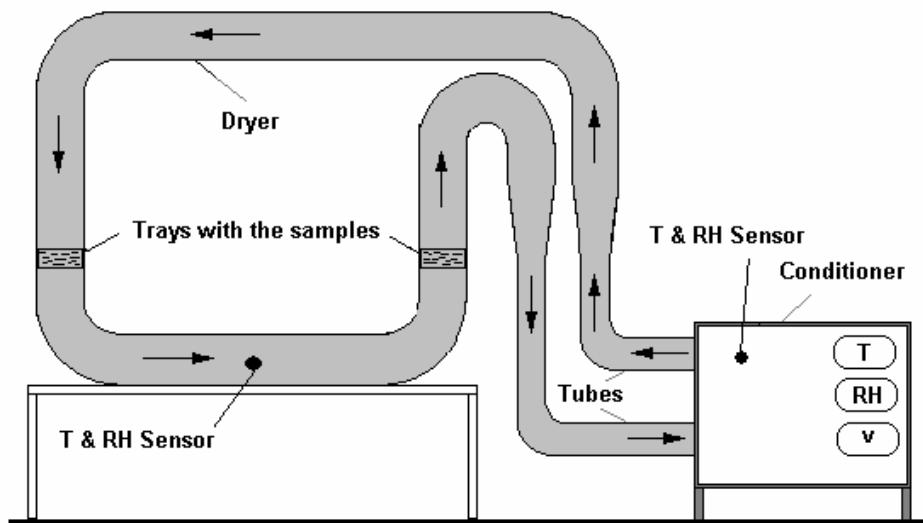


Fig.1 Lab dryer.

2.3 Experiments

Experiments were performed at air temperatures of 40, 45, 50, 55, 60, 70, 80 and 90°C. At each temperature three RH values were adjusted: low, medium and high, which were determined from the psychrometric chart following a special procedure (*Fig.2*): Ambient air condition was taken as 20°C and 70% RH (10.25 g water vapor per kg of air) (a). Heating this air to the target drying temperature will result in a RH that was referred to as “low RH” (b). Assuming, that the exhaust air is completely adiabatically saturated (c) and heated again to the target drying temperature (d) leads to a condition referred to as “high RH”. Finally, the “medium RH” was calculated as arithmetic mean of “low RH” and “high RH” (e). The selected treatments for the drying experiment are shown in Table 1.

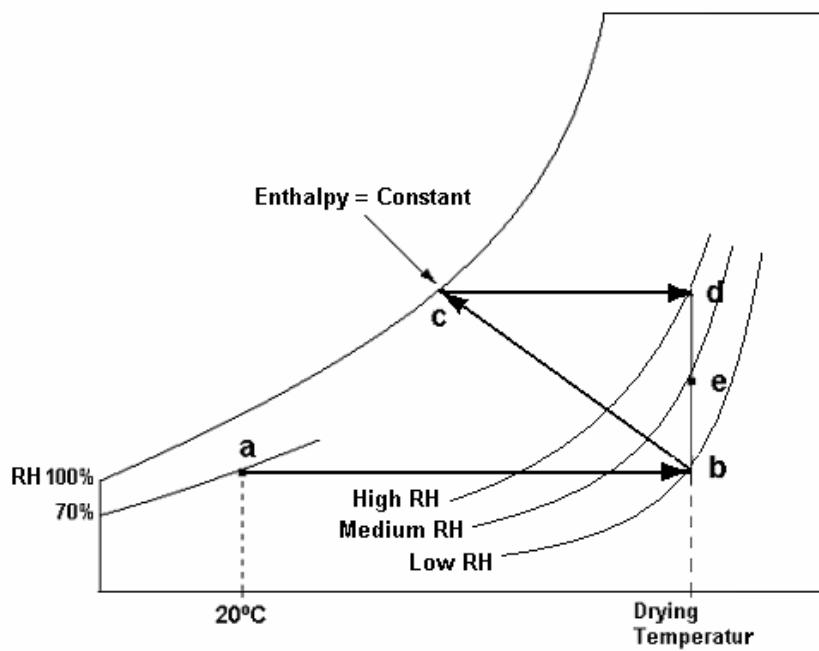


Fig. 2 Selection procedure for “low RH”, “medium RH” and “high RH” in the psychrometric chart: Ambient air (a) was heated to the target drying temperature (b). The RH at this point is as “low RH”. The exhaust air was adiabatically saturated (c) and heated to the target drying temperature (d) leads to a condition referred to as “high RH”. The “medium RH” was calculated as arithmetic mean of “low RH” and “high RH” (e).

Table 1 Treatments of the drying experiment. RH means relative humidity of the air.

| Temperature, °C | Low RH, % | Medium RH, % | High RH, % |
|-----------------|-----------|--------------|------------|
| 40 | 20 | 27 | 35 |
| 45 | 15 | 22 | 30 |
| 50 | 12 | 18 | 25 |
| 55 | 10 | 15 | 20 |
| 60 | 7 | 12 | 18 |
| 70 | 5 | 9 | 13 |
| 80 | 3.5 | 7 | 10 |
| 90 | 2.5 | 5 | 7.5 |

After setting the conditioner at the selected combinations of temperature and RH, the dryer was first run without samples to reach stable air conditions. For the drying experiments the trays were loaded with 100 g of fresh leaves or 120 g of fresh chopped plants to have similar airflow resistance. During the drying process, the weight of the samples was measured by taking the trays out of the dryer. The time interval for weighing was adapted to the drying velocity: shorter intervals have been chosen at the beginning of each test than at the end, and longer intervals have been chosen for lower temperatures. For instance at 90°C the time interval was 1 min at the beginning and 3 min at the end, and at 40°C the time interval was 5 min at the beginning and 30 min at the end. All the tests were continued to reach to a constant weight of the samples. After drying, the final Moisture content (M) of the samples was measured using the oven method (105°C, 24 hr). The final dry matter mass of a sample was used to derive the temporal course of M during the drying experiment:

$$M_t = \frac{W_t - DM}{W_t} \quad (1)$$

In which M_t is the moisture content in decimal during the drying process at time t , W_t is the weight of the sample in kg at time t and DM is the dry matter weight in kg.

As a check, also the initial M of the fresh sample was measured before each drying test.

2.4 Drying equations

The moisture ratio (MR) of the tarragon during the drying experiments was calculated using Eq. 2 [9, 10]:

$$MR = \frac{M_t - M_e}{M_0 - M_e} \quad (2)$$

In which M_t is moisture content (dry basis) at time t , M_e is equilibrium moisture content and M_0 is initial moisture content. M_e was calculated according to the Halsey equation [11].

$$M_e = \left(\frac{-\exp(C_1 + C_2 T)}{\ln(ERH)} \right)^{1/C_3} \quad (3)$$

In which T is temperature in °C, ERH is equilibrium relative humidity in %, and C_1 , C_2 and C_3 are coefficients.

A list of drying equations was made based on the literature (Table 2).

All equations are monotonic decreasing equations, which is the property of drying, except the Wang and Singh, and Midilli and Kucuk equations. The Wang and Singh equation as well as Midilli and Kucuk equation are rejected because these equations may lead to an increase of MR at long term.

Table 2 Mathematical equations given by various authors for the drying curves and grouping of the equation based on the mathematical similarity.

| No | Model name | Model equation | Coefficients | Reference |
|----|---|--|--------------|------------|
| 1 | Lewis | $MR = \exp(-kt)$ | 1 | [12, 13] |
| 2 | Henderson and Pabis | $MR = a \exp(-kt)$ | 2 | [2, 14] |
| 3 | Simplified Fick's diffusion equation (SFFD) | $MR = a \exp[-c(t/L^2)]$ | 2 | [13, 15] |
| 4 | Two-term exponential | $MR = a \exp(-kt) + (1-a)\exp(-kat)$ | 2 | [2, 5, 13] |
| 5 | Page | $MR = \exp(-kt^n)$ | 2 | [16, 17] |
| 5a | Mod. Page I | $MR = \exp[-(kt)^n]$ | 2 | [2, 18] |
| 5b | Mod. Page II | $MR = \exp[-k(t/L^2)^n]$ | 2 | [15, 17] |
| 6 | Yagcioglu et al. | $MR = a \exp(-kt) + c$ | 3 | [13, 19] |
| 7 | Diffusion approach | $MR = a \exp(-kt) + (1-a)\exp(-kbt)$ | 3 | [5, 13] |
| 7a | Verma et al. | $MR = a \exp(-kt) + (1-a)\exp(-gt)$ | 3 | [13, 20] |
| 8 | Two-term | $MR = a \exp(-k_0 t) + b \exp(-k_1 t)$ | 4 | [2, 13] |
| 9 | Mod. Henderson and Pabis | $MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$ | 6 | [13, 21] |
| 0 | Wang and Singh | $MR = 1 + a t + b t^2$ | rejected | [5, 22] |
| 0 | Midilli and Kucuk | $MR = a \exp(-kt^n) + bt$ | rejected | [3, 5] |

The remaining equations were arranged in the order of complexity. The complexity was defined based on number of coefficients, number of terms, and power of time. For instance the Lewis equation, which is the simplest among the equations, has one coefficient, one term and the power of time is one. The Page, modified Page-I and modified Page-II equations are mathematically similar, so the Page model was selected as the representative of these equations. The equation of Diffusion approach and the one of Verma have three coefficients, two terms and the power of t is one. The results of these two equations are presented with the name of Diffusion. Finally we can conclude that there are nine different equations for fitting to the data (Table 2).

The moisture content data obtained at different drying air temperatures and relative humidities were converted to the MR . Then the selected MR equations were fitted to the experimental data of tarragon leaves and chopped plants separately.

The fitting steps for both leaves and chopped plants were as follows:

- A nonlinear regression program in Matlab was used to fit the equations to the experimental data to find the coefficients of the equation and the error parameters residual sum of square (RSS) and standard error of estimation (SEE) [23].

$$RSS = \sum_{i=1}^m (MR - \bar{MR})^2 \quad (4)$$

$$SEE = \sqrt{\frac{\sum_{i=1}^m (MR - \bar{MR})^2}{df}} \quad (5)$$

In the Eq.5, MR is moisture ratio (dimensionless) and df is degree of freedom.

- The best equation was selected based on the values of the statistical parameter and simplicity (less parameters).
- The parameters in the equations were fitted as a function of temperature.

3 Results and discussion

3.1 Experimental results

Fig.3 presents the drying curves of the French and Russian Tarragon leaves, which were dried at the same air conditions (40°C and 25% RH). They showed almost a similar behavior. Leaves and chopped plants were also compared. It was found that drying of leaves was almost two times faster than that of chopped plants (*Fig.4*).

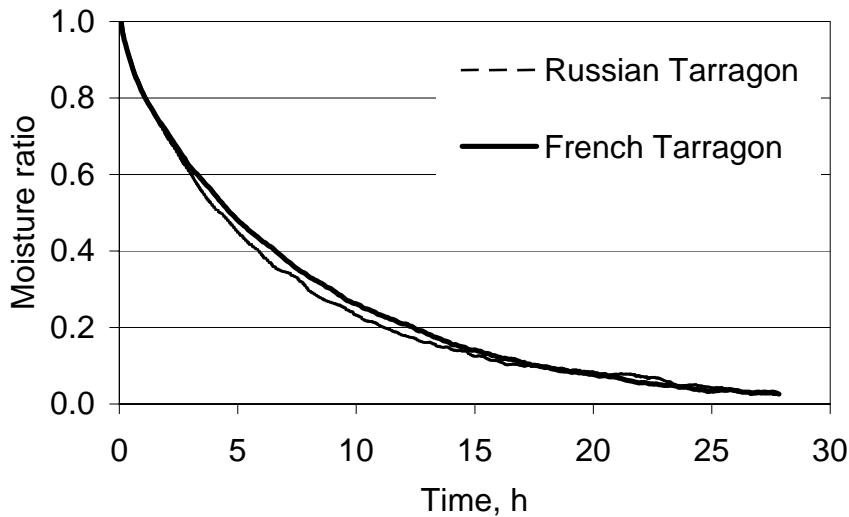


Fig. 3 Moisture ratio changes of chopped plants of French and Russian Tarragon at 40 °C and 25% RH.

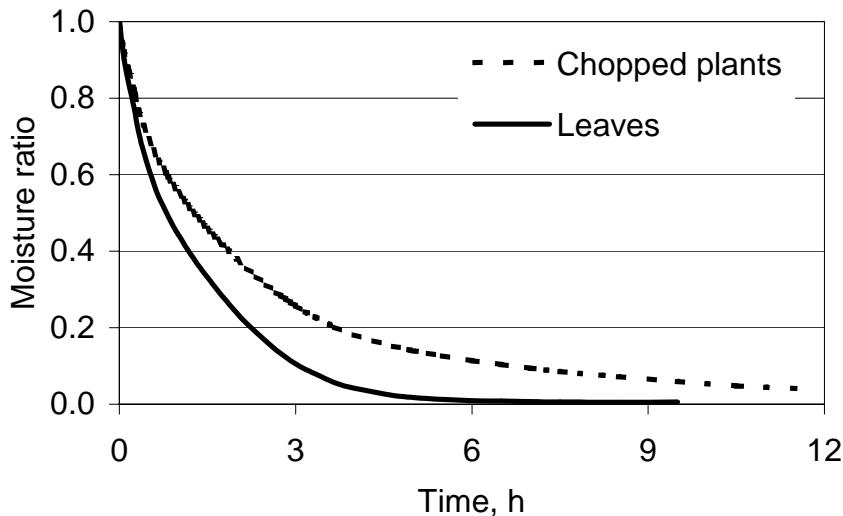


Fig. 4 Moisture ratio changes of chopped plants and leaves of Russian Tarragon at 45 °C and 15% RH.

3.2 Selection of the best equation

All the data series (42 data sets for leaves and 27 data sets for chopped plants) were fitted to the remaining nine equations. The average values of the *RSS* and the *SEE* for all equations at different temperature are shown in *Fig.5* for leaves and in *Fig.6* for chopped plants. The lower value of *RSS* or *SEE* shows a higher accuracy of the fitting. The prospect was having higher accuracy for the more complex equation. The result was against of this expectation for Yagcioglu, Modified Henderson and Two-term equations. The Page and Diffusion equations showed almost the same level of *RSS* and *SEE* for tarragon leaves. For chopped plants, the Diffusion equation showed slightly higher accuracy for fitting compared to the Page equation as it was expected because of more complexity for Diffusion equation.

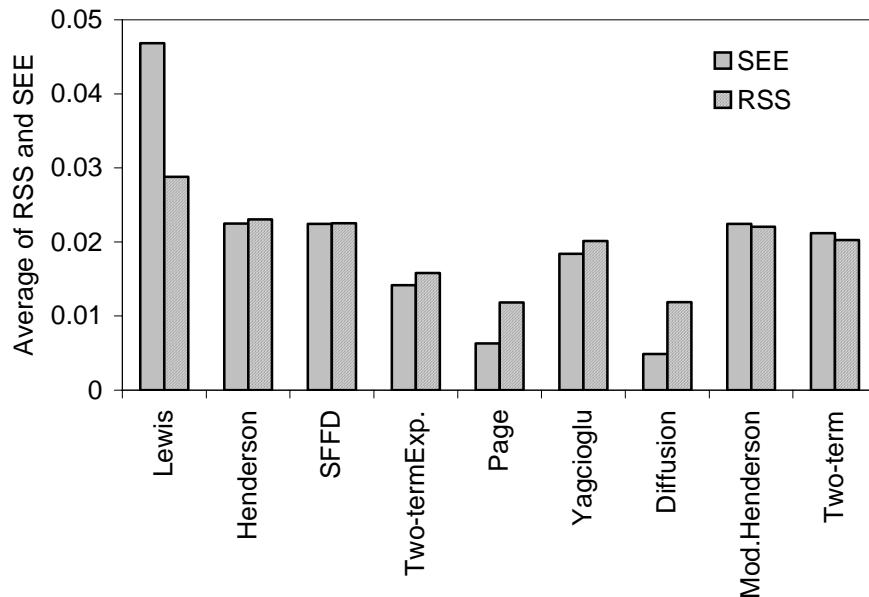


Fig.5 The average level of RSS and SEE of all data series for different drying equations for tarragon leaves.

The results are also given as a function of the drying temperature in Table 3 for leaves and in Table 4 for chopped plants. For all experiments the number of times that an equation showed the best fit, based on the *RSS* and *SEE* parameters is presented in these tables. The Diffusion equation showed the best fit for most of the data series of leaves especially at the temperatures 40, 45, 55°C. The Page equation was the best for the leaves at the temperatures 70 and 80°C (*Fig.6*). The level of the *SEE* as well as *RSS* for the Diffusion and the Page equations were always very close to each other. For chopped plants the Diffusion equation shows to be the best.

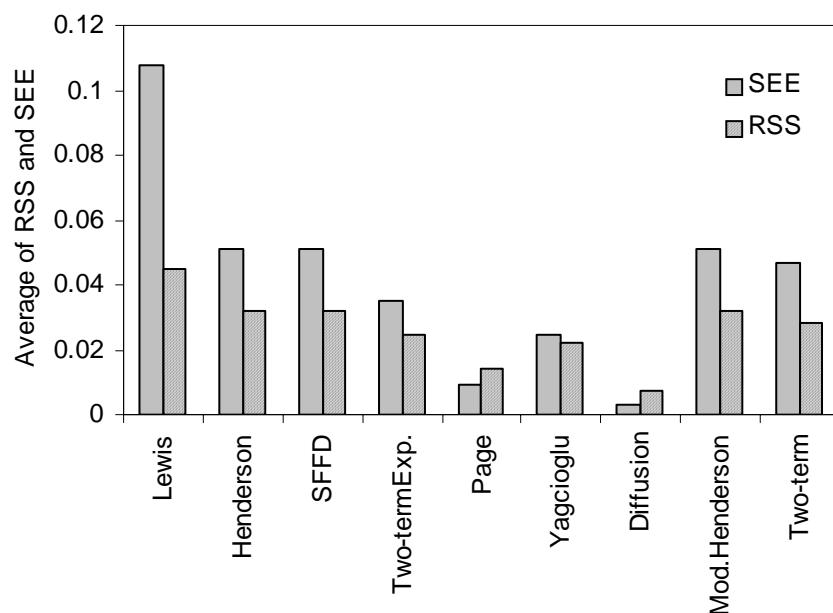


Fig.6 The average level of RSS and SEE of all data series for different drying equations for chopped plants.

Although the Diffusion showed the best fit among the selected equations, the Page equation was selected as the appropriate equation for this research because the levels of the Page equation and Diffusion equation were very similar, specially for leaves, but the Page equation is more simple with two parameters while Diffusion has three parameters. The lower numbers of parameters is preferable to find meaningful relations behavior the parameter value and the drying conditions.

Table 3 The winning results of drying equations for leaves and whole plant based on residual sum of square (RSS).

| Material | Temperature, °C | Equation | | | |
|----------------|--------------------|-----------|-----------|----------|----------|
| | | Page | Diffusion | Two-term | Others |
| Leaves | 40 | 0 | 6 | 0 | 0 |
| | 45 | 0 | 6 | 0 | 0 |
| | 50 | 0 | 5 | 1 | 0 |
| | 55 | 3 | 3 | 0 | 0 |
| | 60 | 1 | 0 | 5 | 0 |
| | 70 | 4 | 1 | 1 | 0 |
| | 80 | 2 | 1 | 1 | 0 |
| | 90 | 0 | 2 | 0 | 0 |
| | Total score | 10 | 24 | 8 | 0 |
| | | | | | |
| Chopped plants | 40 | 0 | 6 | 0 | 0 |
| | 45 | 3 | 3 | 0 | 0 |
| | 50 | 1 | 3 | 0 | 0 |
| | 55 | 0 | 5 | 0 | 0 |
| | 60 | 0 | 3 | 0 | 0 |
| | 70 | 0 | 1 | 2 | 0 |
| | Total score | 4 | 21 | 2 | 0 |

Table 4 The winning results of drying equations for leaves and whole plant based on standard error of estimation (SEE).

| Material | Temperature, °C | Equation | | | |
|----------------|--------------------|-----------|-----------|----------|----------|
| | | Page | Diffusion | Two-term | Others |
| Leaves | 40 | 0 | 6 | 0 | 0 |
| | 45 | 0 | 6 | 0 | 0 |
| | 50 | 0 | 5 | 1 | 0 |
| | 55 | 3 | 3 | 0 | 0 |
| | 60 | 1 | 0 | 5 | 0 |
| | 70 | 4 | 1 | 1 | 0 |
| | 80 | 2 | 1 | 1 | 0 |
| | 90 | 0 | 2 | 0 | 0 |
| | Total score | 10 | 24 | 8 | 0 |
| | | | | | |
| Chopped plants | 40 | 0 | 6 | 0 | 0 |
| | 45 | 3 | 3 | 0 | 0 |
| | 50 | 1 | 3 | 0 | 0 |
| | 55 | 0 | 5 | 0 | 0 |
| | 60 | 0 | 3 | 0 | 0 |
| | 70 | 0 | 1 | 2 | 0 |
| | Total score | 4 | 21 | 2 | 0 |

The independent variables of the drying experiments were temperature, RH and the material (leaf or chopped plants). The experimental data were fitted to the Page equation to find the k and n values simultaneously at each temperature. The coefficients of leaves and

chopped plants at different temperatures are shown in *Fig.7* for k and in *Fig.8* for n . Both, n and k increase with temperature.

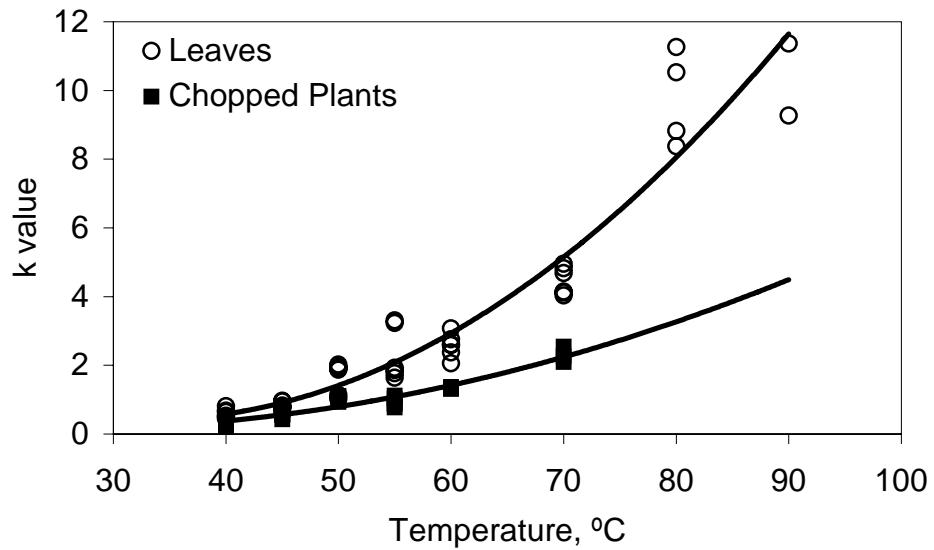


Fig. 7 The k value of the Page equation ($MR = \exp(-kt^n)$) for tarragon leaves and chopped plants at different temperatures.

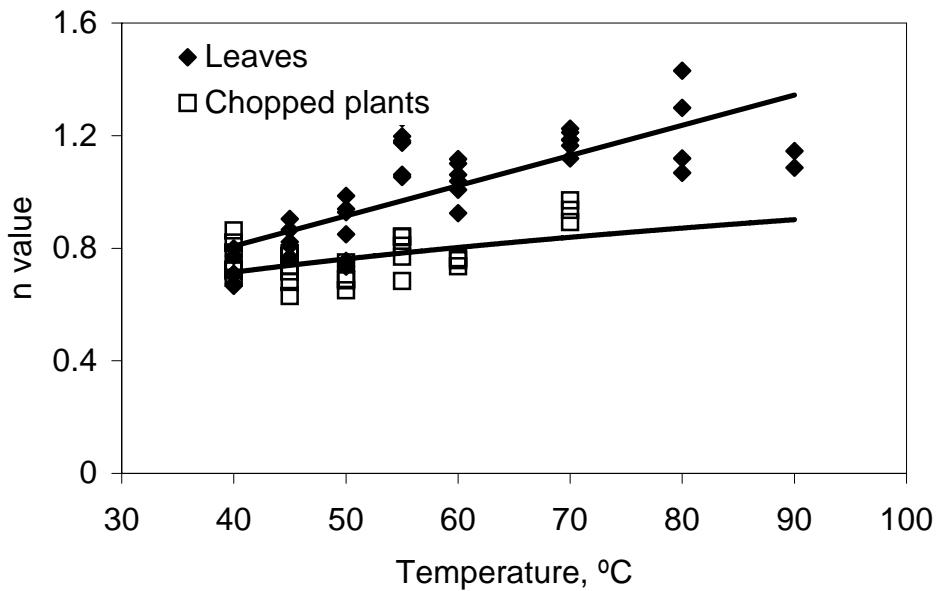


Fig. 8 The n value of the Page equation ($MR = \exp(-kt^n)$) for tarragon leaves and chopped plants at different temperatures.

Fig. 9 presents the k values for the Page equation as a function of relative air humidity. The k values at each temperature are nearly constant for the different relative humidities.

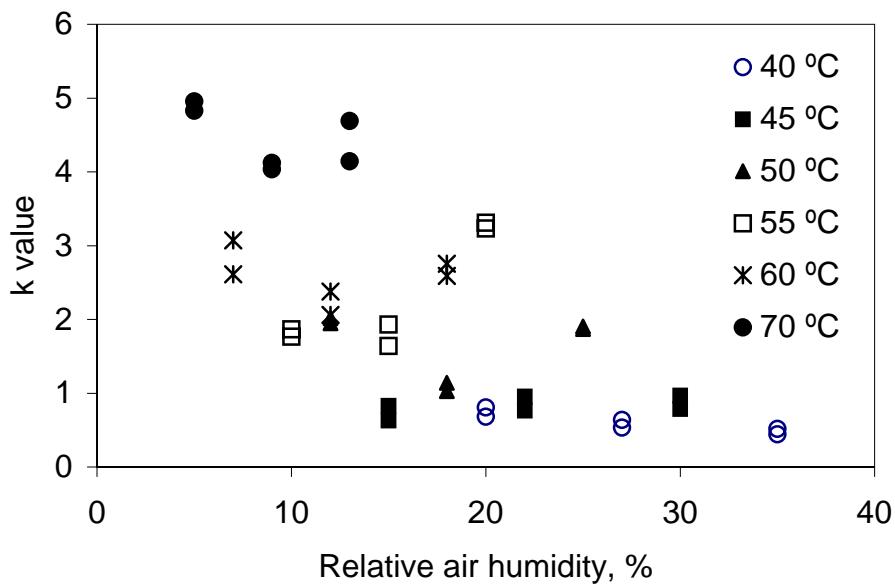


Fig.9 The k value in the Page equation ($MR = \exp(-kt^n)$) of the experiments with leaves at different relative air humidity and temperature levels.

For the n value at different temperatures a linear equation was fitted for leaves as well as chopped plants using all data series. The results are given in equation 6 for leaves (L) and equation 7 for chopped plants (CP), T represents the temperature in °C.

$$n_L = 0.3784 + 0.0107 T \quad (6)$$

$$n_{CP} = 0.5189 + 0.0049 T \quad (7)$$

Using these expressions in the Page equation for leaves (Eq.8) and chopped plants (Eq.9), the following correlation of k with temperature was found.

$$MR_L = \exp(-kt^{(0.3784 + 0.0107T)}) \quad (8)$$

$$MR_{CP} = \exp(-kt^{(0.5189 + 0.0049T)}) \quad (9)$$

Several expressions were applied for the relation of k and temperature. An exponential equation was chosen to find the best fit for the k values as a function of temperature (T in °C). Finally the following equations were found for drying of tarragon leaves (Eq. 10) and chopped plants (Eq. 11).

$$k_L = 0.0456 \times \exp(0.0665T) \quad (10)$$

$$k_L = 0.0307 \times \exp(0.0633T) \quad (11)$$

The complete equation become therefore for leaves (Eq.12) and for chopped plants (Eq.13).

$$MR_L = \exp(-(0.0456 \times \exp(0.0665T))t^{(0.3784 + 0.0107T)}) \quad (12)$$

$$MR_{CP} = \exp(-(0.0307 \times \exp(0.0633T))t^{(0.5189 + 0.0049T)}) \quad (13)$$

Some of the experimental MR values as a function of time are presented in *Fig.10* for leaves and in *Fig.11* for chopped plants together with the predicted values using the above given exponential equations. We can see that these equations are good enough to estimate the M at different drying time for our purpose of calculating energy and drying time.

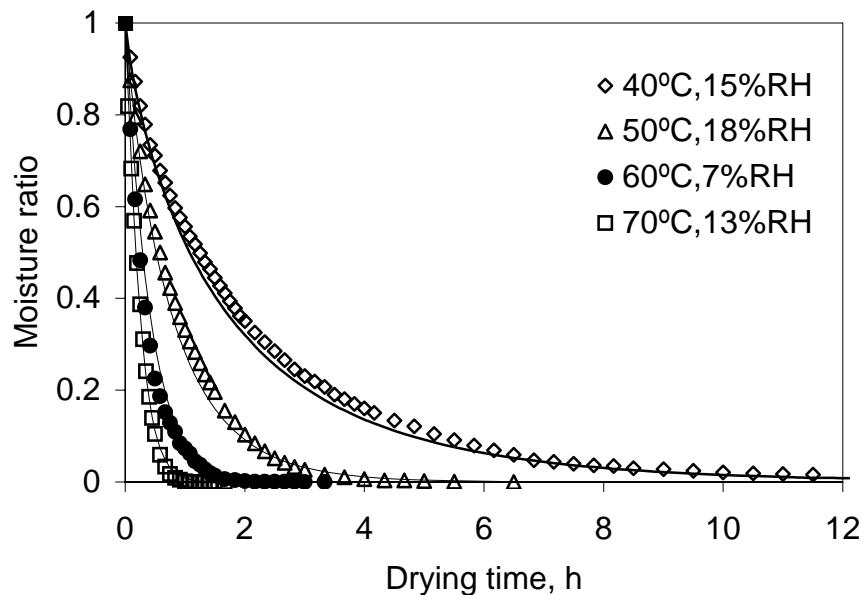


Fig.10 Experimental data of moisture ratio of tarragon leaves at different temperatures and the fitted curves to Eq.12.

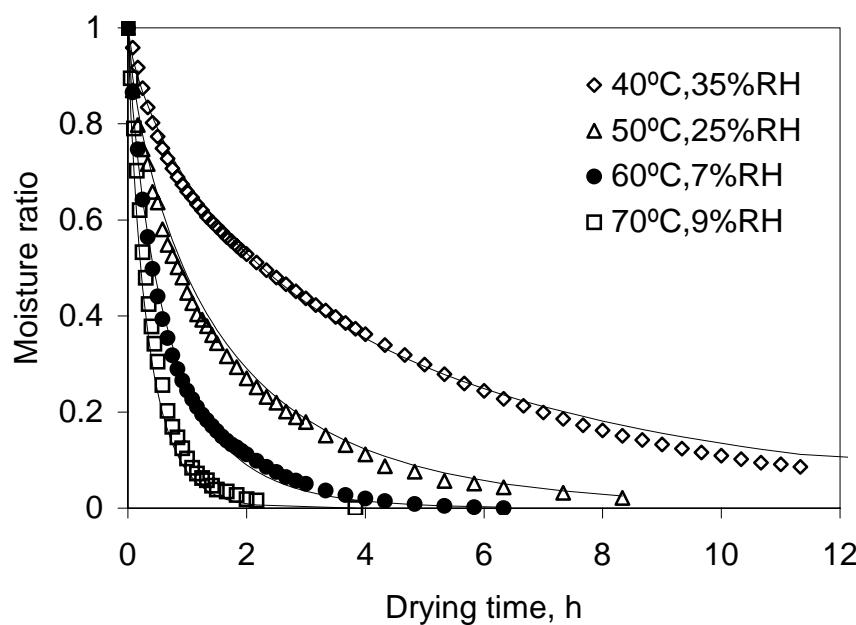


Fig.11 Experimental data of moisture ratio of chopped tarragon at different temperatures and the fitted curves to Eq.13.

3.3 Discussion

The drying rate for the chopped plants and the leaves was different and the required drying time for chopped plants was almost two times than for the leaves. When drying chopped plants, the leaves will be over-dried so efficiency of the dryer will be less and the energy consumption will be higher. Separation of the leaves before drying or even during the drying process will help to optimize the process.

The experiments were done using a batch dryer. As the fundamental theory of the batch and continuous dryers are similar, the equations that were found in this research can be used to evaluate both dryer types.

It is expected that a higher RH causes longer drying time because of raise in Me and hence the smaller driving force of humidity between the air surrounding the material and the material itself. The effect of RH on Me is not that strong and it causes that the differences in drying time for varying RH in the appearance at a fixed temperature are moderate. The levels of relative humidities, applied in this research, were 35% and lower. The selected level was not high enough to illustrate significant effects of RH on drying time. It was found in literature on herb drying that the drying time for sage and chamomile becomes significant different for above 50% [24].

4 Conclusion

As the drying behavior of the French and Russian Tarragon was similar, the same drying model can be used for both varieties. The drying time was about two times larger for chopped plants compared to the leaves.

Among the drying equations, which were evaluated in this research, the Page and Diffusion approach equations showed the best fit. The difference between the Page equation and the Diffusion approach equation is very small. The Page equation has two variables, which makes it easier to find the dependency of the parameters with temperature and RH. Therefore the Page equation is selected as a relevant equation for drying of tarragon. The relative humidities applied in this research have no significant effect of on drying rate of tarragon.

Relevant equations as well as n and k values were found as function of temperature for leaves and chopped plants. Equations show small difference with experiments. The equations can be used at temperatures 40°C to 90°C and relative humidities lower than 35%. They are suitable to estimate the moisture content during drying in order to calculate energy consumption, drying time and cost estimations. They are also usable in dryer design.

Acknowledgment

Financial support from Ministry of Science and Research and Technology of Iran is gratefully acknowledged.

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4

Effect of drying on the essential oil in tarragon (*Artemisia dracunculus* L.) leaves

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This chapter submitted for publication

Abstract

The effect of hot air-drying on the essential oil constituents and yield in French and Russian Tarragon (*Artemisia dracunculus* L.) leaves was studied. The tarragon leaves were dried at air temperatures ranging from 40 to 90°C. Mostly the drying stopped when the moisture content of the samples reached to 10% or for some of the treatments at 7, 20 and 30%. The essential oil of the fresh and dried leaves was isolated by hydrodistillation and analyzed by capillary gas chromatography and gas chromatography-mass spectrometry. During the drying process total oil recovery decreases. The decrease was highest at 60°C drying temperature. For French Tarragon the decrease of oil recovery was significantly lower at 90°C. The effect of the relative humidity of the drying air at each temperature was not significant. The main compounds were estragole in French Tarragon with 69% and sabinene in Russian Tarragon with 40%. The drying process changed the relative percentage of the constituents in the oil, for instance the relative percentages of estragole decreased and sabinene increased in French Tarragon.

Keywords: *Artemisia dracunculus*; drying; essential oil; French Tarragon; Russian Tarragon.

1 Introduction

The use of spices has increased significantly over the past years, partly due to renewed interest in dishes that use a wide variety of spices [1]. Consumers prefer high quality, which is mainly defined by oil essential oil content in the material. Drying is needed for storage and loss of essential oil is unavoidable during drying processes but selection of the best drying parameters helps to minimize any loss of the oil during drying.

Correct drying of aromatic plants is necessary for high quality and stable products. The final Moisture Content (*MC*) must reach 5-10%. Drying prevents the growth of microorganisms which may cause spoilage or food poisoning [2]. The essential oil constituents can be adversely affected during drying of herbs. The effect of drying on the flavor and fragrance of different crops has been studied intensively. Examples are: Australian-grown ginger [3], basil [4-6], bay leaf [7], parsley [8, 9], roman chamomile [10], spearmint [1], thyme and sage [11] and aromatic plants in general [2]. These studies show that the changes in the concentration of essential oil constituents during the drying process depend on several factors like temperature and Relative Humidity of the drying air as well as the product properties [11]. Drying in a convection oven resulted in losses of volatile compounds that fluctuated with the drying temperature and drying time employed [9].

Tarragon, *Artemisia dracunculus* L., is a herbaceous plant belonging to the Asteraceae. Two varieties of tarragon can be distinguished [12], namely French Tarragon of South European origin and Russian Tarragon of Siberian origin [13]. Both species are morphologically similar. French Tarragon is mainly used as a culinary herb in oil, sauces, vinegars, mustard and spices. French people call it the 'King of Herbs'. Russian Tarragon has an inferior flavor and is more used as medicine [14].

Different researchers have studied the chemical composition of the essential oils of Russian and French Tarragon [12, 14-20]. The principal compounds are estragole and β -ocimene for French Tarragon and sabinene, methyl eugenol and elemicin for the Russian Tarragon. The main compounds of essential oils of tarragon vary widely according to geographic location,

climate, day length, soil type and cultivar [21, 22]. The effect of drying on these compounds has not yet been investigated.

The present study examines the influence of the temperature and RH during air-drying on the essential oil constituents of French and Russian Tarragon. The main goal of this research is to establish the optimal conditions for drying of the leaves for French and Russian Tarragon. It is aimed also to evaluate the changes in the ratio of the main constituents of these two varieties at different drying conditions.

2 Materials and Methods

2.1 Plant material

Russian and French Tarragon leaves (fresh and dried leaves) were used for distillation experiments. The Russian variety was cultivated at a research farm in Elburg (The Netherlands) and the French one was cultivated by Unifarm (Wageningen University, The Netherlands). Both were harvested just before flowering in July, August and September 2004. Leaves were collected manually for drying experiments. No herbicides were used during cultivation.

2.2 Drying experiment

Thin layer drying experiments were performed at the Department of Agrotechnology and Food Science, Wageningen University in 2004. The *MC* of the material is considered as wet basis ($100 \times$ weight of water content / total weight). Samples of 100 g fresh tarragon leaves at 80% *MC* were put in a tray of 14 by 28 cm with a metal net at the bottom part. The samples were dried in a batch dryer by hot air drying. A fan was used for circulation of the air at 0.6 m/s. The experiments were done at four different combinations of temperature and RH (Table 1). The dried material was collected at *MCs* ranging from 5 to 30% in three replicates. A *MC* of 10% was considered as optimal. As it was not possible to measure the *MC* of the samples during the drying process exactly at the given values in Table 1, the samples were taken at times around these values.

Table 1 Selected treatments for drying experiments.

| Temperature, °C | RH of circulating drying air, % | Final MC of the material, % (w.b.) | | | | |
|-----------------|---------------------------------|------------------------------------|---|----|----|----|
| | | 5 | 7 | 10 | 20 | 30 |
| 45 | 17 | | × | × | × | × |
| 60 | 7 | | × | × | × | × |
| 60 | 18 | | × | × | × | × |
| 90 | 2.5 | × | × | × | × | |

×: Each treatment (× in the table) was performed in three replicates.

2.3 Distillation

A Lickens-Nickerson apparatus was used for hydrodistillation [23]. Twenty grams (dry matter) of dried tarragon leaves was added to a flask and it was mixed with 500 ml of distilled water. The flask was then heated by immersion in an oil bath of 140°C for two hours counted from the time after condensation of the first drop of vapor in the calibrated tube [24]. After

the condensation phase separation took place. The amount of oil in the calibrated tube was measured and afterwards the oil was collected. The oil was stored in closed glass vials in the refrigerator at 5 °C.

2.4 Gas Chromatography (GC)

The oil compounds were analyzed with a Hewlett-Packard 5890 gas chromatograph, coupled to a flame ionization detector (FID). The conditions were: split 1:100; carrier gas, hydrogen at 5 ml/min (5 psi). A DB-5 capillary column (30 m × 0.32 mm, 1.5 µm film thickness) was used for separating the compounds. The following temperature program was used: 65 to 225°C at 8°C/min. The oil sample was dissolved in t-butyl methyl ether in a concentration of 1% and 1 µl was injected in the GC. Three major compounds were selected to examine any changes in oil composition as a function of the various drying parameters. The selected compounds were estragole (main constituent), E-β-ocimene (typical of a highly volatile constituent) and methyl eugenol (typical of a high boiling constituent).

2.5 Gas Chromatography-Mass Spectrometry (GC-MS)

GC-MS analyses were carried out on a Varian 3400 gas chromatograph equipped with a DB5 capillary column (60 m, 0.25 mm ID, 0.25 µm film thickness) directly coupled to the ion source of a Finnigan MAT 95 mass spectrometer. The samples were injected in split mode (split ratio 1:30) and the column temperature was programmed from the initial temperature of 60°C to 260°C at a rate of 3°C/min. The mass spectrometer was operated in the 70 eV EI mode with exponential scanning from m/z 24 to 300 at 0.5 s/decade, resulting in a cycle time of 0.68 s/scan.

Identification of the compounds was performed by comparing the obtained mass spectra with those in the Wageningen Collection of Mass Spectra of Natural Compounds and by checking the relative retention index of the proposed compound.

Quantitative analyses were performed on a HP5890 gas chromatograph, equipped with an autosampler and a DB5 column (30 m, 0.32 mm ID, 0.25 µm film thickness), using the same temperature program. The relative concentration of each compound was calculated as percentage of the total GC peak area, assuming equal response/gram for all compounds, and then the absolute amount was estimated based on this percentage and the total amount of the oil recovered for each treatment.

3 Result and discussion

3.1 Oil recovery

The amount of essential oil, recovered from the fresh leaves was 1.06% before drying for French Tarragon and 0.60% for Russian Tarragon. On a dry weight basis these values are 5.3% for French and 3% for Russian Tarragon leaves. Since the *MC* of the fresh leaves was in the range of 78 to 81% for both varieties. These yields are shown in *Fig.1* and *Fig.2* at a *MC* value of 80%. These values are significantly higher than those reported in literature: 0.31% for French Tarragon and 0.14% for Russian Tarragon [12] and 0.50% for the French Tarragon dried at 40°C [2]. Simon et al. reported a range of 0.25 to 2.4% for the essential oil content of French Tarragon [25]. The available information on the essential oil yield of tarragon (in the literature) is related to the whole plant and the data in this research is related to the leaves. As

the majority of the essential oil is in the leaves, the level of the recovered oil is higher than the data in literature.

During drying of the tarragon, the samples from which the oil was obtained by hydrodistillation were taken at MCs between 40% and 5%. The oil recovery is shown in *Fig.1* for French Tarragon leaves and in *Fig.2* for Russian Tarragon leaves. The leaves dried at 45°C and 90°C showed higher yields for both French and Russian Tarragon compared to the 60°C treatments. *Fig.1* and *Fig.2* illustrate a trend that the oil content decreases with a decreasing final MC.

The oil yield for French Tarragon leaves was higher at 90°C than other treatments in the range of 5 to 30% MC and at this range the oil yield was decreasing for lower final MCs. The reason for relatively high oil yields at 90°C could be the shorter drying time at this temperature. The required time for drying of tarragon leaves to reach 10% final MC was 0.3 hr at 90°C, 1.5 hr at 60°C and 5.5 hr at 45°C. The oil yield at 60°C was slightly higher at 18% RH of the drying air than at 7%. Loss of oil occurred mainly before the leaves reached 30% MC.

The oil recovery for Russian Tarragon leaves was almost the same at 45 and 90°C and the levels were higher than at 60°C. Comparison of the treatments at 60°C showed a higher level at 18% compared to 7% RH of the drying air. At 60°C most oil losses occurred before the leaves reached 35% final MC when dried with air of 7% RH and before 25% MC for the leaves dried at 18% RH.

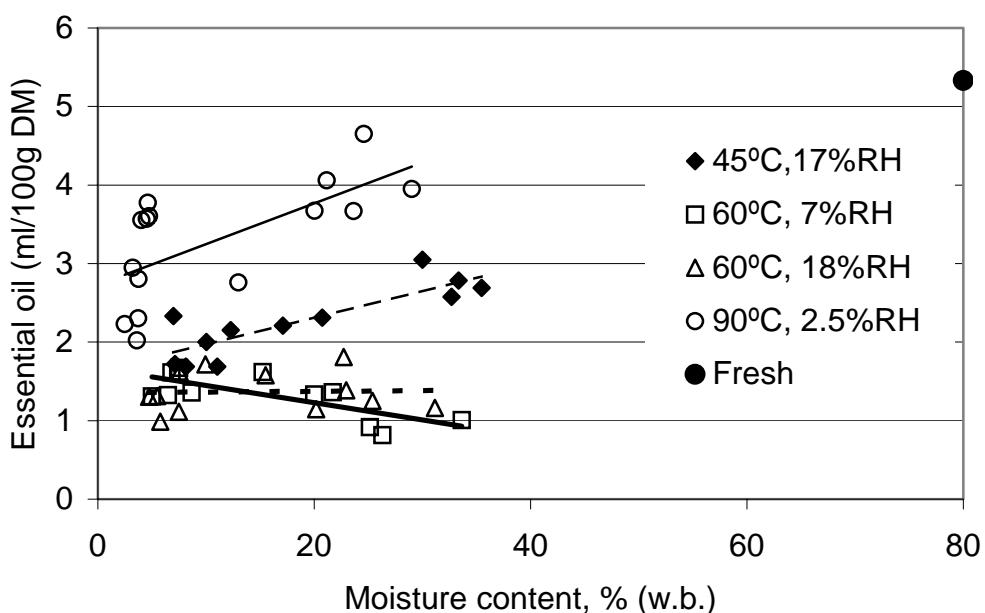


Fig. 1 Oil yield from French Tarragon leaves dried to different final MCs.

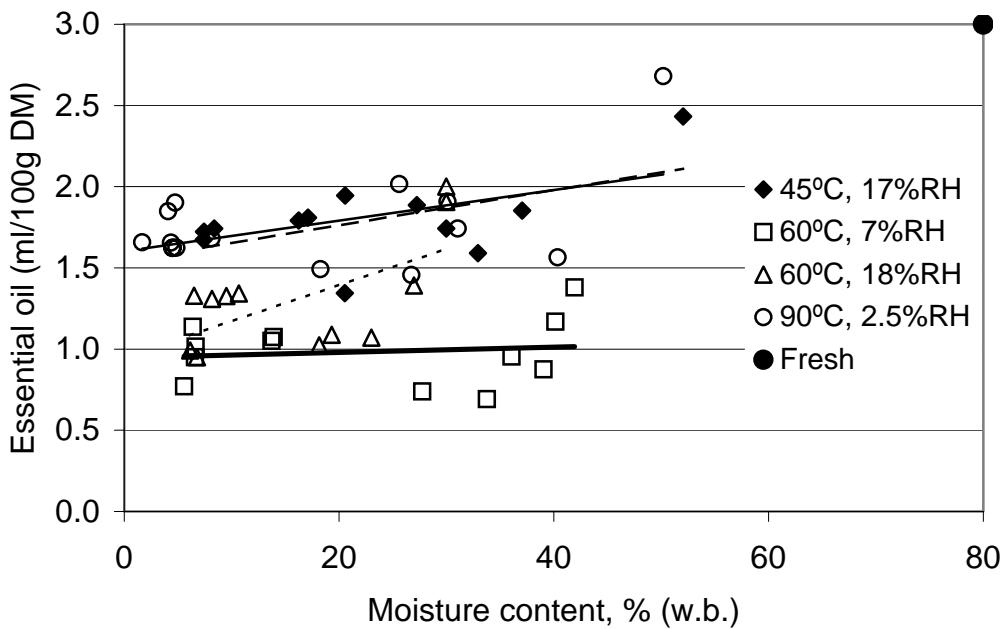


Fig. 2 Oil yield from Russian Tarragon leaves dried to different final MCs.

The oil yield at different final MCs was analyzed by an ANOVA tests fro both varieties. The differences were not significant for MCs in the range of 5 to 40% at the 95% probability level. Neither did the drying time at a specific temperature significantly affect the oil yield from the leaves, when dried to a MC of 40% or lower. Since the main goal of the drying of tarragon leaves is to extend the shelf life and a final MC of 10% is desirable for storage, the oil yields of the leaves dried at 10% final MC were used for further analysis. The ANOVA result is shown in Table 2. The oil recovery was significantly different at 90°C for the French Tarragon leaves and at 90°C and 45°C for the Russian Tarragon leaves, compared to 60°C. The two relative humidities at 60°C showed no significant difference.

Table 2 Oil yield at 10% final MC (ml/100 g DM).

| Variety | 45°C, 17%RH | 60°C, 7%RH | 60°C, 18%RH | 90°C, 2.5%RH |
|------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| French Tarragon | 1.95 ± 0.24 ^a | 1.13 ± 0.47 ^a | 1.48 ± 0.32 ^a | 3.39 ± 0.52 ^b |
| Russian Tarragon | 1.85 ± 0.08 ^a | 1.09 ± 0.04 ^b | 1.33 ± 0.02 ^b | 1.77 ± 0.21 ^a |

^aOil yields (mean ± standard deviation) followed by different letters are significantly different at $P<0.05$.

The losses of oil for the tarragon leaves dried to 10% final MC are shown in Table 3. The difference of the oil recovery between the two levels of 7 and 18% relative humidities at 60°C was not significant.

Table 3 Losses of essential oil when tarragon leaves are dried to 10% final MC.

| Variety | 45°C | 60°C | 90°C |
|------------------|------|------|------|
| French Tarragon | 63% | 75% | 36% |
| Russian Tarragon | 38% | 60% | 41% |

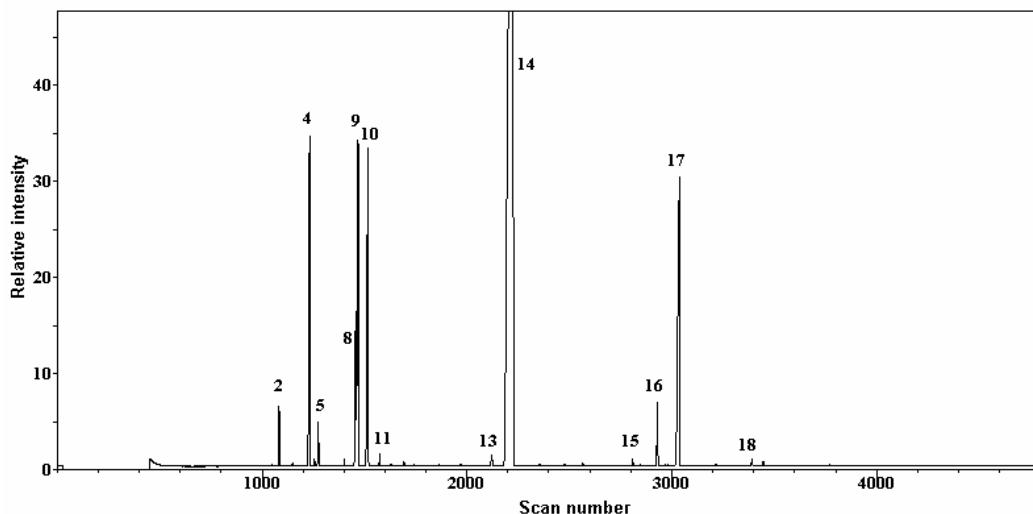


Fig. 3 Chromatogram of essential oil from French Tarragon leaves.

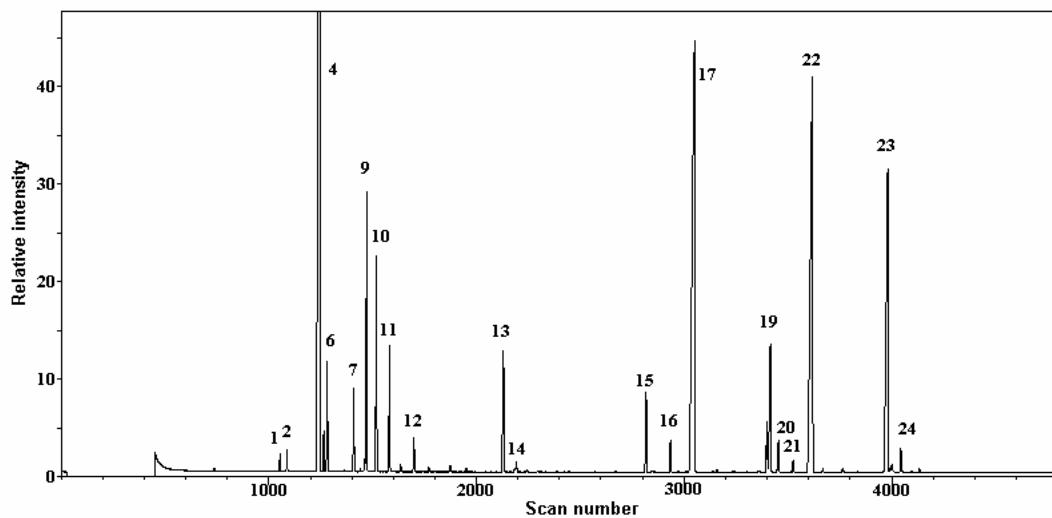


Fig. 4 Chromatogram of essential oil from Russian Tarragon leaves.

3.2 Constituents

The gas chromatographic analysis identified different organic constituents in the two types of the oil. Typical chromatograms are shown in *Fig.3* for French Tarragon and in *Fig.4* for Russian Tarragon. Although many compounds could be reproducibly detected, several of them were only present in trace amounts. Some of the oil samples were analyzed by GC coupled to mass spectrometry to identify the compounds present. Table 4 presents the percentages of the main compounds in the fresh leaves of French and Russian Tarragon. The major compound of the essential oil was estragole for French Tarragon (68.6%) and sabinene for Russian Tarragon (39.4%).

Table 4 Relative amount of constituents present in the oil from fresh leaves of French and Russian Tarragon.

| Peak No | Compound | Relative amount, % | |
|-----------------|-----------------------|--------------------|------------------|
| | | French Tarragon | Russian Tarragon |
| 1 | α -Thujene | 0.02 | 0.19 |
| 2 | α -Pinene | 0.69 | 0.25 |
| 3 | Camphene | 0.36 | 0.00 |
| 4 | Sabinene | 4.93 | 39.44 |
| 5 | β -Pinene | 0.08 | 0.49 |
| 6 | Myrcene | 0.50 | 1.45 |
| 7 | α -Terpinene | 0.08 | 1.11 |
| 8 | Limonene | 2.40 | 0.16 |
| 9 | (Z)- β -Ocimene | 6.04 | 4.09 |
| 10 | (E)- β -Ocimene | 5.97 | 3.06 |
| 11 | γ -Terpinene | 0.16 | 1.95 |
| 12 | Terpinolene | 0.00 | 0.47 |
| 13 | Terpinen-4-ol | 0.17 | 1.91 |
| 14 | Estragole | 68.60 | 0.16 |
| 15 | Citronellyl acetate | 0.09 | 1.30 |
| 16 | Geranyl acetate | 0.98 | 0.51 |
| 17 | Methyleugenol | 8.50 | 14.72 |
| 18 | Germacrene D | 0.10 | 0.87 |
| 19 | (E)-Methylisoeugenol | 0.00 | 2.15 |
| 20 | Bicyclogermacrene | 0.06 | 0.57 |
| 21 | δ -Cadinene | 0.00 | 0.21 |
| 22 | Elemicin | 0.01 | 15.97 |
| 23 | (E)-Isoelemicin | 0.00 | 7.67 |
| 24 | α -Cadinol | 0.00 | 0.43 |
| Total | | 99.74 | 99.13 |
| Other compounds | | 0.26 | 0.87 |

3.3 Variation of main compounds

The drying treatment could affect the absolute amount of constituent and relative concentration of every compound in a different manner. The absolute amount (Aa) of the constituent is given in ml/100 gr DM. The relative concentration is defined as the percentage of the constituents in the oil. To study the effect of drying on the absolute concentration of each compound the relative amount was calculated as:

$$\text{Relative amount} = \frac{\text{Aa}_{\text{dry}}}{\text{Aa}_{\text{fresh}}}$$

in which Aa_{dry} and Aa_{fresh} are the absolute amount of the compound in ml/100g dry matter (DM) in the dry and the fresh leaves.

The variation of the absolute amount of the major compounds was evaluated using Boxplots. This method was used to show the distribution of the variation for each compound. The box itself contains 50% of the data of all experiments. The upper edge (hinge) of the box indicates the 75th percentile of the data set, and the lower hinge indicates the 25th percentile.

The dot-line in the box indicates the medium value of the data. The whiskers extending the box represent the minimum and maximum data values, unless outliers are present in which case the whiskers extend to a maximum of 1.5 times the inter-quartile (box length) range. The points outside the ends of the whiskers (+) are outliers or suspected outliers.

The variation of the relative amount at 10% MC of the two main compounds at different temperatures is shown in *Fig.5* for estragole in French Tarragon and in *Fig.6* for sabinene in Russian Tarragon. The relative amount of the main compound of French Tarragon (estragole) was considerably reduced at 60°C. The losses at 90°C were small compared to those at 45°C and 60°C. In the Russian variety there were high losses of the main compound (sabinene) when the leaves were dried at 60°C, while the losses at 45°C and 90°C had almost the same median. Similar analyses were performed for all other oil compounds of both varieties. Most of them showed a greater loss at 60°C relative to 45°C and 90°C.

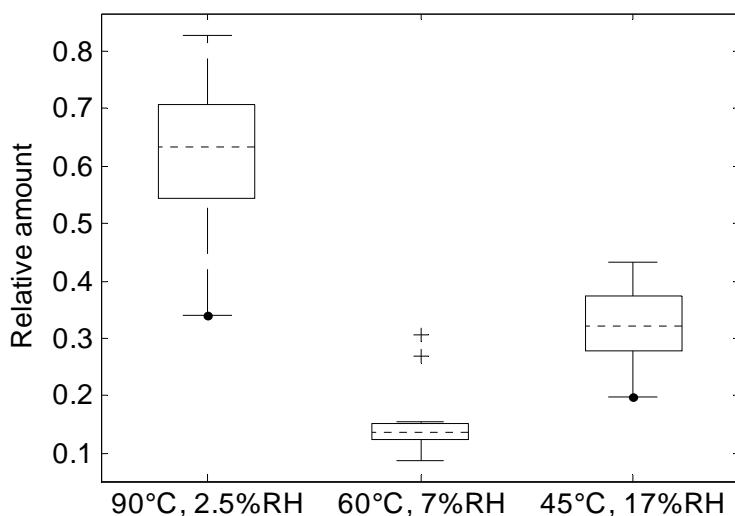


Fig. 5 Variation of the relative amount of estragole at temperatures 45, 60 and 90°C and a final MC of 10% in French Tarragon.

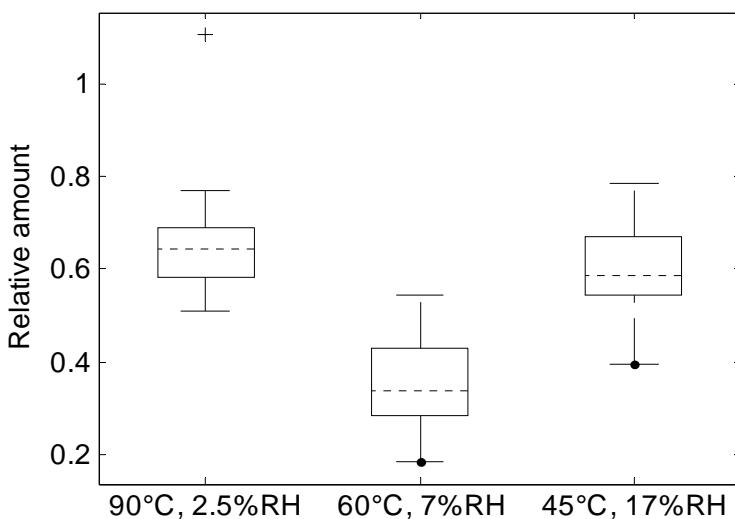


Fig. 6 Variation of the relative amount of sabinene at temperatures 45, 60 and 90°C and a final MC of 10% in Russian Tarragon.

The values of the relative amount at 10% *MC* for the major oil compounds and the variation in the compounds for all the treatments at 45, 60 and 90°C are shown for French Tarragon in *Fig. 7* and for Russian Tarragon in *Fig. 8*. The compounds on the y-axis are in the order of retention time. From the plots, it can be seen that the absolute amount of nearly all the major compounds of both varieties are reduced during the drying process, as is logical as oil yields also drop. However estragole and geranyl acetate behave clearly different. For those compounds, which show values above one, a larger variation in their relative amount was observed. A reason could be the low absolute amount of the constituent.

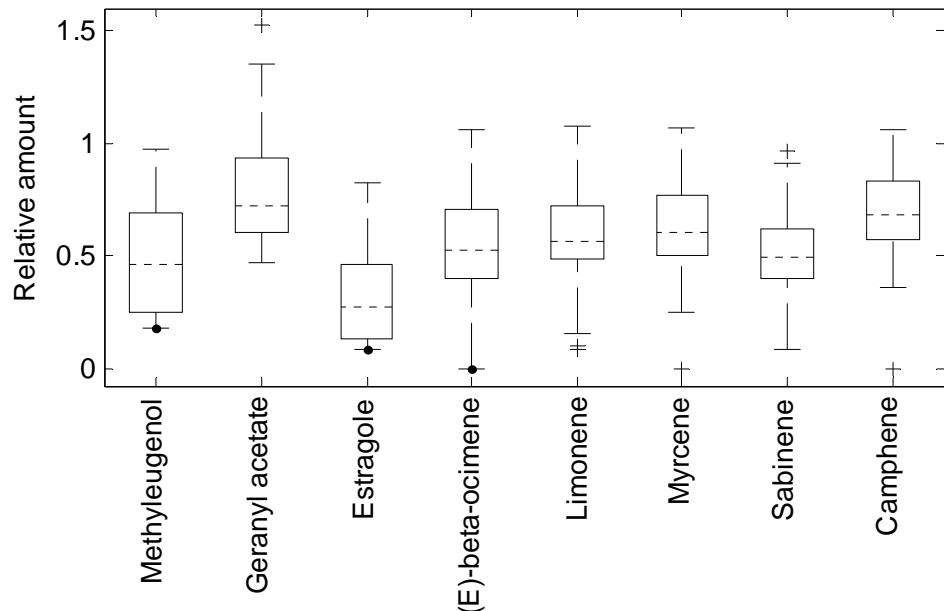


Fig. 7 Variation in oil compounds of French Tarragon after various drying treatments.

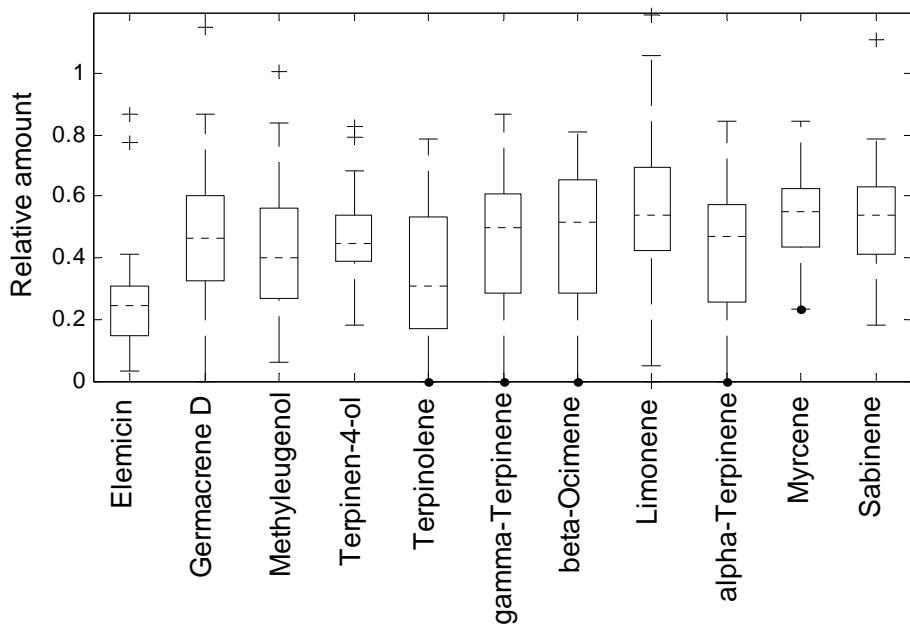


Fig. 8 Variation in oil compounds of Russian Tarragon after various drying treatments.

To study the changes in the relative concentration of the constituents, which is the percentage of the constituents in the oil, Table 5 was made for both varieties. The changes are categorized in five groups: \uparrow = more than 30% increase, \nearrow = 10 to 30% increase, \rightarrow = less than 10% changes, \searrow = 10 to 30% decrease and \downarrow = more than 30% decrease. Since estragole is the major compound (70%) in French Tarragon, we can expect that when the amount of estragole changes during drying, other compounds change in the opposite way. The results showed this indeed, except for methyleugenol. In Russian Tarragon the changes were not depended on the major compound (sabinene) because it consists of 40% of the total oil. The changes in the compounds of tarragon leaves dried at 60°C were very similar for the 7% RH and the 18% RH treatments. This means that the effect of RH on the level of these changes was not significant.

The changes in the relative concentration as a function of *MC* while drying are shown in *Fig.9* for estragole in French Tarragon and in *Fig.10* for methyleugenol in Russian Tarragon. Estragole decreased in the dried leaves at all drying temperatures. The largest change was observed at 60°C, 7%RH and the smallest change at 90°C. Sabinene increased at all temperatures during drying except at 45°C which remained almost at the same level as in the fresh leaves. The levels were slightly higher at 60 and 90°C compared to 45°C.

Table 5 The average changes of the relative amount of the main constituents in essential oil of French and Russian Tarragon leaves, dried at different drying temperatures. The changes are categorized in five groups: \uparrow = more than 30% increase, \nearrow = 10 to 30% increase, \rightarrow = less than 10% changes, \searrow = 10 to 30% decrease and \downarrow = more than 30% decrease.

| Variety | Constituent | Scan number | Drying condition | | | |
|------------------|-----------------------|-------------|------------------|---------------|---------------|---------------|
| | | | 45°C, 17%RH | 60°C, 7% RH | 60°C, 18% RH | 90°C, 2.5% RH |
| French Tarragon | Camphene | 1151 | \uparrow | \uparrow | \uparrow | \searrow |
| | Sabinene | 1233 | \nearrow | \uparrow | \uparrow | \rightarrow |
| | Myrcene | 1276 | \uparrow | \uparrow | \uparrow | \searrow |
| | (Z)- β -Ocimene | 1470 | \uparrow | \uparrow | \uparrow | \searrow |
| | (E)- β -Ocimene | 1518 | \uparrow | \uparrow | \uparrow | \searrow |
| | γ -Terpinene | 1575 | \uparrow | \uparrow | \uparrow | \downarrow |
| | Estragole | 2226 | \searrow | \downarrow | \downarrow | \rightarrow |
| | Geranyl acetate | 2930 | \uparrow | \uparrow | \uparrow | \uparrow |
| | Methyleugenol | 3037 | \searrow | \rightarrow | \rightarrow | \rightarrow |
| Russian Tarragon | Sabinene | 1233 | \rightarrow | \nearrow | \nearrow | \nearrow |
| | (Z)- β -Ocimene | 1470 | \nearrow | \searrow | \rightarrow | \nearrow |
| | (E)- β -Ocimene | 1518 | \nearrow | \searrow | \rightarrow | \nearrow |
| | Terpinen-4-ol | 2122 | \searrow | \rightarrow | \rightarrow | \nearrow |
| | Citronellyl acetate | 2810 | \uparrow | \uparrow | \uparrow | \uparrow |
| | Methyleugenol | 3037 | \rightarrow | \searrow | \searrow | \downarrow |
| | Germacrene D | 3391 | \searrow | \searrow | \downarrow | \nearrow |
| | Elemicin | 3598 | \downarrow | \downarrow | \downarrow | \downarrow |
| | (E)-Isoelemicin | 3961 | \uparrow | \downarrow | \downarrow | \uparrow |

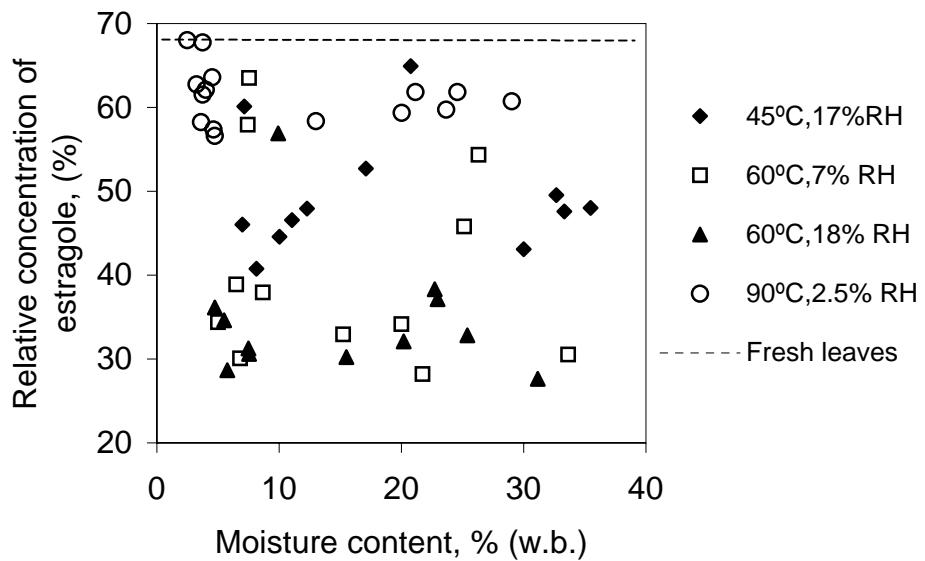


Fig. 9 Changes of the relative concentration of estragole in French Tarragon while drying leaves at different drying temperatures.

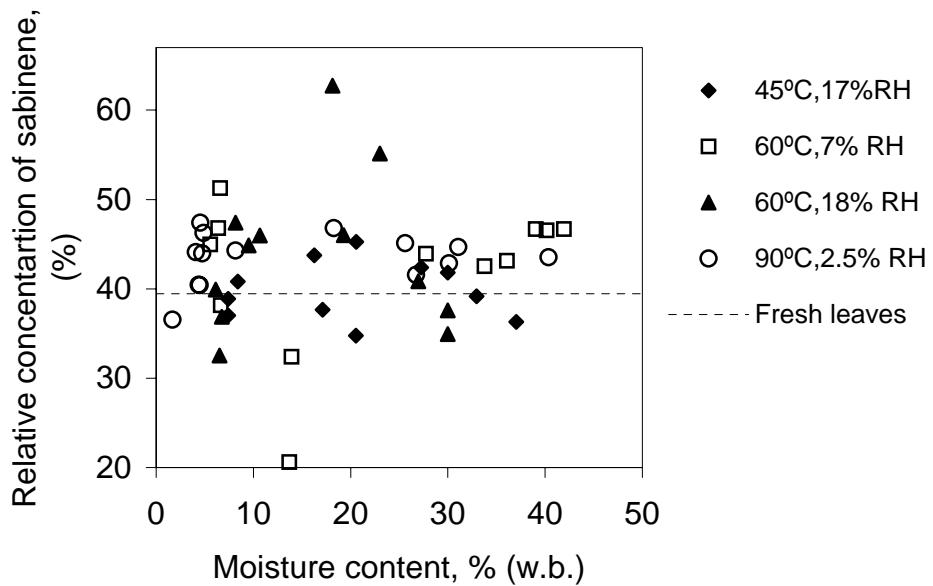


Fig. 10 Changes of the relative concentration of sabinene in Russian Tarragon while drying leaves at different drying temperatures.

3.4 Discussion

Table 6 presents the required time for drying of tarragon leaves to a certain *MC* for the selected treatments in this research. The oil yield of the tarragon leaves, dried at 60°C, was always lower than the yield of the treatments at 45°C and 90°C. Since the temperature and drying time are the main factors [7, 11], that affect the oil content of dried material, a lower temperature (at 45°C) and a shorter drying time (at 90°C) yield more oil. This is apparently

because at 45°C, the temperature is not high enough for evaporation of the oil, while at 90°C the time is not long enough to destroy oil glands in the leaves.

Table 6 Required time for drying of tarragon leaves at different final MCs.

| Drying Temperature, °C | RH of air, % | Final MC, wet basis (%) | | | | |
|------------------------|--------------|-------------------------|-----|-----|-----|----|
| | | 30 | 20 | 10 | 7 | 5 |
| Drying time, minutes | | | | | | |
| 45 | 17 | 170 | 220 | 330 | 610 | |
| 60 | 7 | 45 | 60 | 80 | 140 | |
| 60 | 18 | 50 | 65 | 90 | 160 | |
| 90 | 2.5 | 12 | 14 | 17 | 21 | 27 |

At 60°C there is enough force available to open the oil glands that causes a drop in oil yield. The changes are generally of a quantitative nature (loss or gain in some compounds), but they can also be qualitative (the formation of new compounds by oxidation, glycoside hydrolysis, esterification, etc) [6]. It can be questioned what will happen with the oil content after storage as a function of the different drying procedures. This will be the subject of further research.

4 Conclusion

Drying causes a loss of essential oil in French and Russian Tarragon leaves. The reduction of the amount of essential oil was significantly higher at 60°C than at 45°C and 90°C. For the drying at 60°C, the main oil losses occur in the initial phase of the drying process, i.e. when the moisture content is higher than 30%. The drying parameters have a similar effect on both French and Russian Tarragon. It appears that lower temperatures and shorter drying times yield more oil.

Acknowledgment

Financial support from Ministry of Science and Research and Technology of Iran is gratefully appreciated. The authors thank Elbert van der Klift of the Laboratory of Organic Chemistry, Wageningen University for technical support of the GC analyses.

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5

Effect of drying on the color of tarragon (*Artemisia dracunculus* L.) leaves

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This chapter submitted for publication

Abstract

The effect of drying conditions on the color of tarragon (*Artemisia dracunculus* L.) leaves was studied. Tarragon leaves were dried at temperatures of 40, 45, 50, 55, 60, 70, 80 and 90°C with a constant airflow of 0.6 m/s. At each temperature, three relative humidities were tested. The samples were collected at 7, 10, 20 and 30% moisture content wet basis for evaluation of the color change. For some of the treatments, color changes during drying from 80% (fresh leaves) to 5% moisture content wet basis were also tested. The color parameters of fresh and dried leaves were measured by a colorimeter. The individual parameters of $L^*a^*b^*$ and $L^*C^*h^*$ color systems were evaluated and h^* proved to be the best parameter to monitor color change. It was found that the young leaves are more sensitive for the temperature treatment and showed earlier color change compared to mature leaves. The smallest change of the color parameters was observed at 40°C, which temperature was low, and also at 90°C, when drying time was short. The biggest change occurred at the temperatures of 50 to 70°C. Most of the color change happened before the material reaches 35% moisture content. The combination of drying time and temperature defines the change of color.

Keywords: *Artemisia dracunculus*; color; drying; French Tarragon; Russian Tarragon.

1 Introduction

Color is an important component of quality throughout agriculture and food industry, because color is closely associated with factors such as freshness, ripeness, desirability and food safety. It is often the primary consideration of consumers when making purchasing decisions [1, 2]. The color kinetics of food is a complex phenomenon and there are not much reliable models to predict color change, which can be used in engineering calculations [1, 3].

Thermal processing is one of the most important methods of food preservation and it affects the food quality as measured by sensory evaluation or instrumental methods [4]. The effect of thermal processing on the color of food material has been studied by various researchers and different color systems have been used for describing color changes of food material [1, 3-11]. Color can be used to define adequate thermal processing conditions for maximizing final product quality if its degradation kinetics are determined [6]. It was shown that color change measured by tri-stimulus reflectance of a colorimeter may be used to predict quality change in food [10].

The CIELAB color system (Commission Internationale de l'Eclairage, 1986) is extensively used to evaluate food colors [12]. Numbers of researchers have already studied the color of food using the CIELAB color space for measurement of the color parameters [2, 12-15].

This research investigates the influence of temperature and Relative Humidity (RH) of the drying air on the color of tarragon. The main objective of this research is to establish the optimal drying conditions for tarragon leaves of two varieties: French Tarragon and Russian Tarragon.

2 Material and methods

2.1 Plant material

Fresh and dried leaves of two tarragon varieties (French Tarragon and Russian Tarragon) were used in the experiments. French Tarragon was collected at a research farm in Elburg (The Netherlands) and Russian Tarragon was planted at the farm of Wageningen University (The Netherlands). Both were harvested just before flowering in July, August and September 2004. To compare the influence of maturity, also young leaves were used. Young leaves have been harvested 2-3 weeks before flowering. Before drying leaves were separated from the stalks manually.

2.2 Drying

Thin layer drying experiments were performed in a laboratory dryer of the Department of Agrotechnology and Food Science, Wageningen University [16]. Samples of 100 g fresh tarragon leaves at a Moisture Content (*MC*) of 80% w.b. were put in a tray of 14 x 28 cm. An airflow of 0.6 m/s was used for all experiments.

Two types of experiments were performed. In the first stage samples were dried at temperatures of 40, 45, 50, 55, 60, 70, 80, and 90°C at “low”, “medium” and “high” RH (Table 1) [16]. The samples were dried to equilibrium moisture content. Color was measured before and after drying.

Table 1 Set values of drying air conditions in the first stage experiments.

| Temperature, °C | | 40 | 45 | 50 | 55 | 60 | 70 | 80 | 90 |
|----------------------|--------|----|----|----|----|----|----|-----|-----|
| Relative humidity, % | Low | 20 | 15 | 12 | 10 | 7 | 5 | 3.5 | 2.5 |
| | Medium | 27 | 22 | 18 | 15 | 12 | 9 | 7 | - |
| | High | 35 | 30 | 25 | 20 | 18 | 13 | - | - |

In the second stage, experiments were done to investigate color changes during the course of drying. Samples were dried at 45°C, 17% RH; 60°C, 7% RH and 90°C, 2.5% RH. These RHs are the result of heating ambient air of x g/kg abs. *MC*. The samples were collected during drying at decreasing *MCs* (Table 2). As it was not possible to measure *MC* during the drying process, the samples were taken at estimated *MCs* (based on drying time) as close as possible to the target values given in Table 2.

Table 2 Set values of drying air conditions and target *MC* for sample collection in the second stage experiments.

| Temperature, °C | RH, % | Moisture content, % w.b. | | | | | | | | |
|-----------------|-------|--------------------------|----|----|----|----|----|----|---|---|
| | | 70 | 60 | 50 | 40 | 30 | 20 | 10 | 7 | 5 |
| 45 | 17 | * | * | * | * | × | × | × | × | - |
| 60 | 7 | * | * | * | * | × | × | × | × | - |
| 90 | 2.5 | * | * | * | * | * | × | × | × | × |

* One sample

× three samples

2.3 Color measurement

Quantitative evaluation of the color changes in tarragon leaves was done by a portable tristimulus colorimeter (Minolta Chroma CR-300, Osaka Japan) for direct measurement of the color parameters of the samples. Calibration was done every day before color measurement. A white calibration plate ($Y = 92.40$; $x = 0.3137$; $y = 0.3194$) was used as reference for calibration of the chroma meter. The leaves were evenly distributed in a tray of 20×10 cm. The diameter of the measuring area was 8 mm. Measurements were taken at five random points for each sample and each measurement had three readings to compensate for the variation in the sensor of the measuring head.

The measurements were recorded in five color systems (Yxy , $L^*a^*b^*$, $L^*C^*h^\circ$, XYZ and Hunter Lab). The $L^*a^*b^*$ [4, 8, 10] and $L^*C^*h^\circ$ [17] system were selected because these are the most used systems for evaluation of dried food material.

The suitability of the various color parameters was based on the visual judgment of experts of two Dutch companies dealing with herbs (Euroma B.V. and VNK B.V.). A collection of 15 samples of dried tarragon leaves as well as the photographs of the same samples was arranged in decreasing order of the values of the color parameters L^* , a^* , b^* , C^* and h° . The parameter, which showed the best compliance between visual and measurement based ranking, was selected. Based on the expert judgment a threshold was defined to distinct acceptable and unacceptable color of dried tarragon leaves.

2.4 Data analysis

Statistical method of ANOVA with 95% confidence interval was applied to find the possible correlation between the data sets and also the significance differences among the experimental data sets.

3 Results and discussion

3.1 Evaluation of color parameters

Table 3 presents the photos of the dried tarragon leaves, which are arranged in decreasing order of L^* , a^* , b^* , C^* and h° values. The arrangement based on h° came closest to the experts visual ranking. The arrangements based on L^* and a^* values did not result in a continuous visual impression whereas b^* and the C^* values showed some similarity to h° in the arrangement. Based on this result, h° was selected as main parameter for evaluation of color change. The experts defined $h^\circ=95$ as threshold for acceptable color of dried tarragon leaves.

Table 3 Arrangement of dried tarragon leaves (variety) in descending order of L*, a*, b*, C* and h°.

| L* | a* | b* | C* | h° |
|----|----|----|----|----|
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3.2 Influence of temperature and RH

The h° value of fresh Russian Tarragon leaves was 123. After drying, h° was lower for all treatments. *Fig.1* shows h° of the dried Russian Tarragon leaves at drying temperatures between 40 and 90°C. In a temperature range from 40 to 70°C, h° decreased and increased again for 80°C and 90°C. In the temperature range of 55 to 80°C, h° was below the quality threshold level of 95. Although the level of h° for different RH at the same temperature varied, no systematic correlation was found.

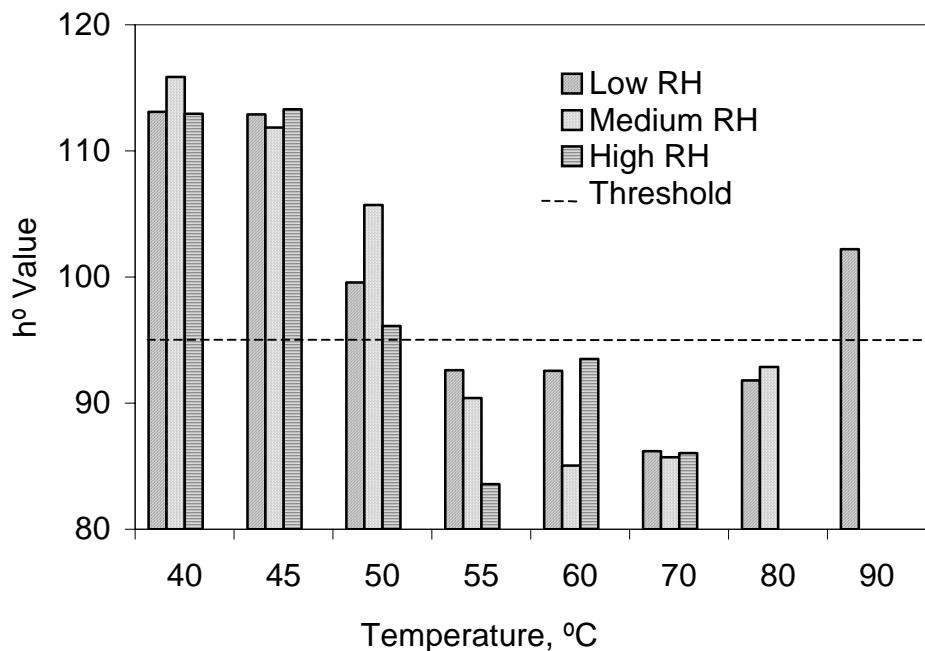


Fig. 1 Influence of temperature and RH of drying air on h° (hue) of Russian Tarragon leaves (h° of fresh leaves=123; quality threshold: $h^\circ \geq 95$).

The L^* value of fresh Russian Tarragon leaves was 48. L^* decreased after drying and the level was lower for higher temperatures. *Fig.2* shows L^* of the dried Russian Tarragon leaves at drying temperatures between 40 and 90°C. The level of lightness at different RH varied at random so no clear relation was found.

The C^* value of fresh leaves was 15. *Fig.3* represents C^* values of the dried Russian Tarragon leaves at drying treatments of 40 to 90°C. This value also decreased by increasing temperature. The C^* value was almost at the same (lower) level for the dried material at the temperature 55°C and higher. No correlation was found for different relative humidities.

Amongst h° , L^* and C^* , h° shows the clearest correlation to the treatments. As h° was also identified by the experts to express visual quality best, further results are only presented for h° .

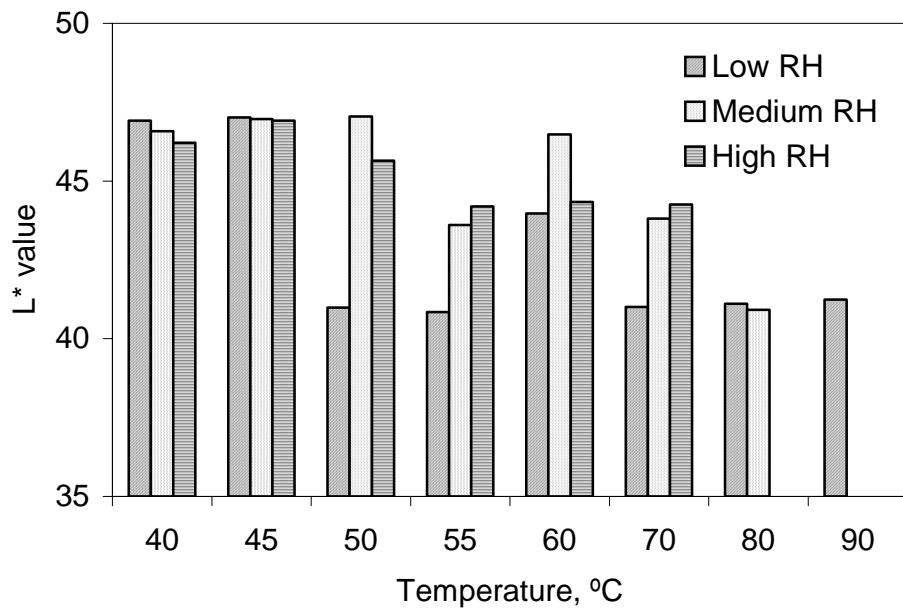


Fig. 2 Influence of temperature and RH of drying air on L* (lightness) of Russian Tarragon leaves; fresh leaves: L*=48.

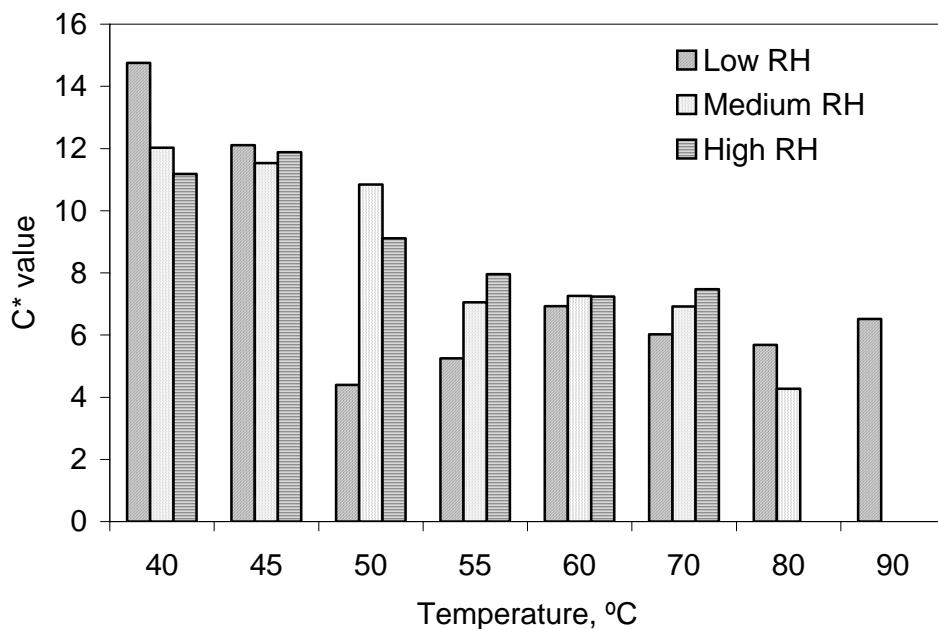


Fig. 3 Influence of temperature and RH of drying air on C* (saturation) of Russian Tarragon leaves; fresh leaves: C*=15.

3.3 Influence of variety

Table 4 presents the statistical results of ANOVA test for each variety. The effect of h^o values of leaves dried with different treatments.

Table 4 Statistical results of the color parameter (h^o) analysis of dried tarragon leaves

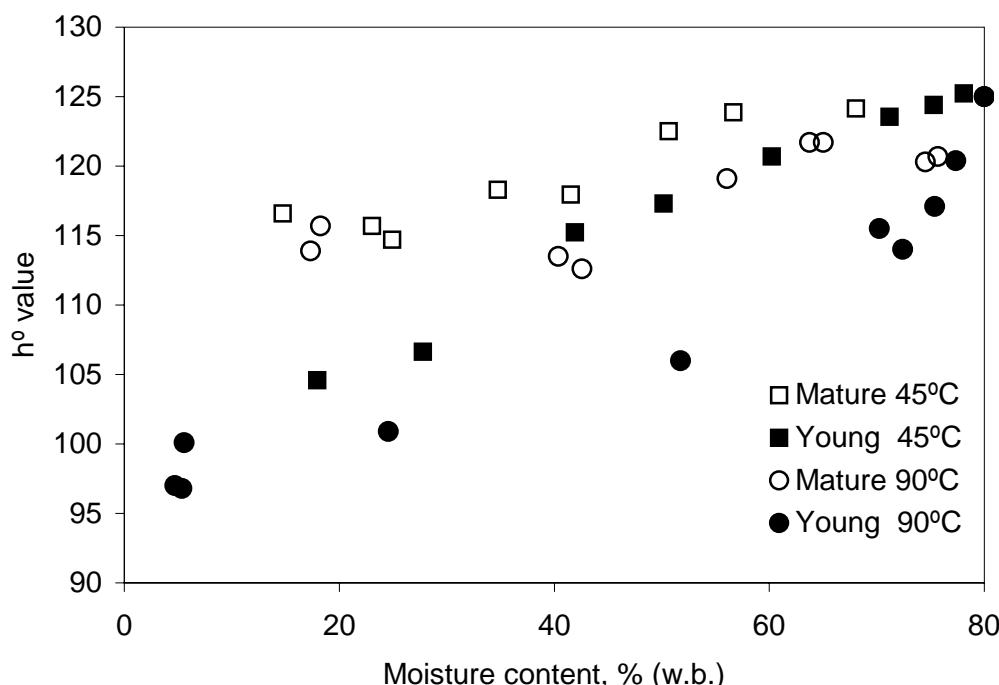
| Variety | Temperature and RH of drying air | | | |
|------------------|----------------------------------|----------------------------|---------------------------|-----------------------------|
| | Fresh | 45°C, 17% RH | 60°C, 7% RH | 90°C, 2.5% RH |
| French Tarragon | 124.5 ± 1.21 ^a | 104.05 ± 2.68 ^b | 91.24 ± 2.10 ^d | 106.21 ± 1.63 ^b |
| Russian Tarragon | 123.4 ± 1.07 ^a | 99.18 ± 1.15 ^c | 84.67 ± 1.52 ^e | 93.03 ± 7.20 ^{cde} |

Values (mean ± standard deviation) followed by different letters are significantly different at $P<0.05$.

There was significant difference between the fresh leaves and the leaves dried at any temperature at 95% probability. The results didn't show any significant differences between varieties for fresh leaves and between the leaves dried at 45°C and 90°C for each variety.

3.4 Influence of maturity

Fig.4 shows h^o of mature and young tarragon leaves at drying temperatures of 45°C and 90°C. Larger changes were observed at both temperatures for young leaves compared to mature leaves. Major changes happened for the young leaves dried at 90°C before they reach 40% MC whereas the color change continuously occurred for the mature leaves during the whole drying process.

**Fig. 4 Experimental h^o of mature and young tarragon leaves dried at 45 and 90°C.**

3.5 Color changes during drying

Fig.5 shows h^o of French Tarragon leaves at 80% MC to 5% dried at different temperatures. h^o of the fresh leaves of French Tarragon was 124.5. The color changes of the leaves dried at

45°C and 90°C are close to each other. Most changes were observed at the earliest drying phase for the drying temperature of 60°C. The level of leaves dried at 60°C was lower than the threshold when they reached the storage *MC* and for the leaves dried at 45°C and 90°C this was higher. The trend line of all treatments was almost horizontal in the measured range from 35% to 5% *MC* for fresh tarragon.

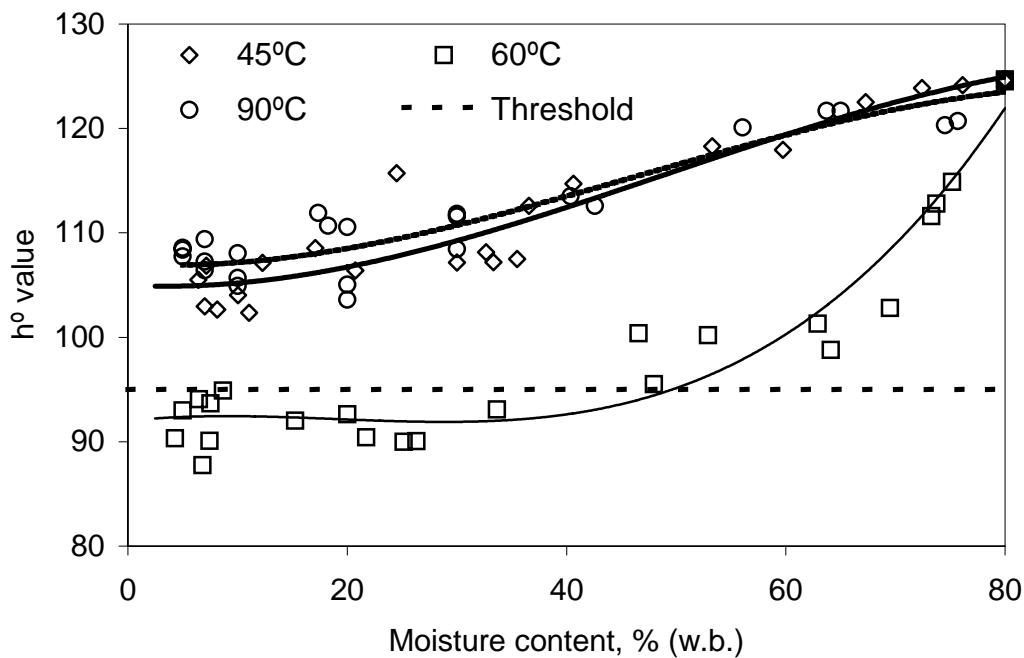


Fig. 5 Course of h° of French Tarragon leaves at decreasing MC during drying at a temperature of 45, 60 and 90°C (quality threshold: $h^\circ \geq 95$).

3.6 Color change as a function of drying time

The color change of French Tarragon leaves as defined by h° versus drying time is shown in *Fig.6*. The initial *MC* of the leaves was 80% and the drying process was continued to reach 7% *MC*, w.b. The figure shows fitted lines using the equation:

$$h^\circ = C_1 \exp(-C_2 t) + C_3 \quad (1)$$

In which C_1 , C_2 and C_3 are coefficients and t is drying time (hr). The coefficients at different drying treatments are shown in Table 5.

Table 5 Coefficients of Eq.1 for different temperatures of the drying air.

| Temperature | C_1 | C_2 | C_3 | R^2 |
|-------------|-------|-------|--------|--------|
| 45°C | 20.53 | 0.37 | 104.02 | 0.9768 |
| 60°C | 30.46 | 2.81 | 91.96 | 0.9698 |
| 90°C | 17.78 | 6.01 | 106.86 | 0.9228 |

The h° value of the material changed slower at 45°C compared to 60°C and 90°C drying temperature and it almost reached the final level after 6 hours drying time. The h° value decreased only to the level of about 108 for both the 45°C and 90°C drying temperatures

because at this time the material reached 7% *MC* and drying process was stopped so there was no time for more changes. h^o at 60°C drying temperature decreased to the level of about 92 after 2 hours. The decrease in h^o value for 60°C is twice the decrease of the 90°C.

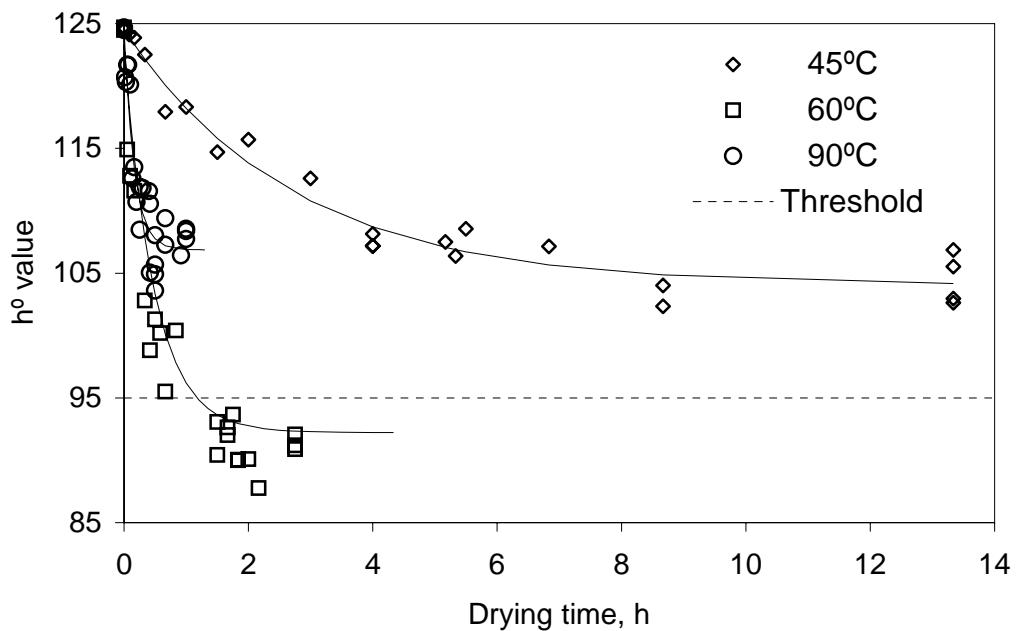


Fig. 6 Course of h^o of French Tarragon leaves during drying at a temperature of 45°C, 60°C and 90°C (h^o of fresh leaves=124.5; quality threshold: $h^o \geq 95$).

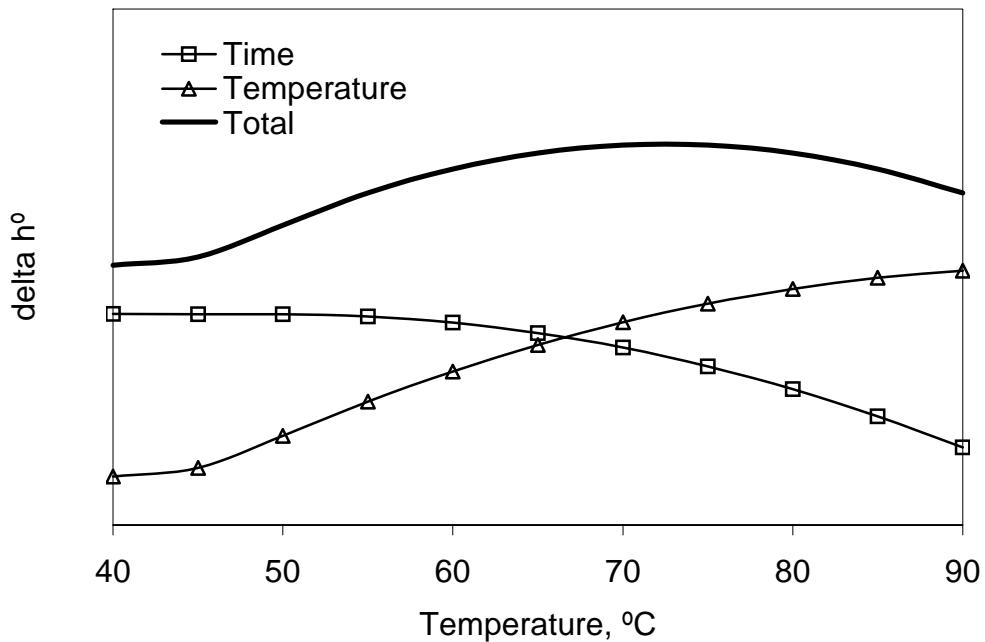


Fig. 7 Color changes affected by the drying time and the drying temperature during the drying process at temperatures of 40°C to 90°C.

3.7 Discussion

Drying of tarragon at 45°C and 90°C showed an acceptable level of h^o but the value was too low for the material dried at 60°C. It can be concluded that the temperature of 60°C is not good for keeping quality. The sum of the temperature effect and drying time effect, results in the total color changes during the drying process. To show these two effects looking together a model is shown in *Fig.7* based on the experimental color changes data, which are presented in *Fig.1*. The increasing temperature influences the change of h^o for the temperature range of 40°C to 60°C while the time effect is important from 70°C to 90°C.

More change was expected at 90°C than the measured level. The explanation could be that the actual temperature in the material was lower than 90°C during the drying process because of the large high evaporation during the earliest phases of the drying process and the drying time was not long enough to change the color.

It seems that 90°C is an acceptable drying temperature based on h^o if preservation of over-drying is guaranteed. However, further color changes could occur during storage. So further research is required to resolve this problem. As the main goal of the drying process is to extent the shelf life, color change during storage time is also very important. Drying time at 90°C is much shorter and also temperature stress is much more than at 45°C. Drying at 45°C can guarantee an acceptable level of color change during drying so higher temperatures can be used also at the beginning of the drying process but the temperature in the material must not exceed 45°C to avoid more color change.

The rate of color change decreases with the decrease of *MC* during the drying process at all temperatures.

4 Conclusion

The color of the tarragon leaves changes during the drying process. Based on expert evaluation, h^o was found as the best parameter, to show the color changes of tarragon leaves during drying process. Temperature and drying time are the main parameters that affect the color change. Higher temperature and longer drying time make more changes for color. The color parameters showed less changes at 40°C because of the low temperature and also at 90°C because the drying time was short. The main changes of color were observed before the materials reaches 40% *MC*. Young leaves showed more sensitivity to temperature than the mature leaves. Drying temperatures below 50°C are recommended for drying of tarragon leaves to avoid color degradation because the h^o was above the acceptable level.

Acknowledgment

Financial support from Ministry of Science and Research and Technology of Iran is gratefully acknowledged.

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6

Long term effects of drying conditions on the essential oil and color of tarragon (*Artemisia dracunculus* L.) leaves during storage

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This chapter submitted for publication

Abstract

The effect of storage on the essential oil content and color of French Tarragon (*Artemisia dracunculus* L.) leaves was studied. Tarragon leaves were dried at temperatures 45, 60 and 90°C, resulting in relative humidity of 17, 7 and 2.5%. In addition, for 60°C also relative humidity of 18% was applied. The air velocity was constant at 0.6 m/s. Oil content and color were measured for the fresh and dried leaves just after drying as well as after storage during 15, 30, 60 and 120 days. The essential oil compounds of the material were isolated by the hydro-distillation method and analyzed by GC-FID as well as GC-MS. A Chroma meter was used to measure the color of the samples. The results showed a reduction of the oil content and change of the color parameters during the storage period. The biggest change of the essential oil content (about 50% after 30 days) and color expressed in the hue value was found for the material, dried at 90°C compared to 45 and 60°C. Drying at 45°C was found as the best among the treatments.

Keywords: *Artemisia dracunculus*; color; drying; essential oil content; storage; tarragon.

1 Introduction

Over the past decades, herbs and medicinal plants gained in global importance. Tarragon *Artemisia dracunculus* L. is a herbaceous plant of the *Asteraceae* family. Two varieties of tarragon can be distinguished [1, 2]: French Tarragon and Russian Tarragon. French Tarragon of the South European origin is mainly used as a culinary herb in oils, sauces, vinegars, mustard and spices [3, 4]. This research deals with French Tarragon.

Post-harvest processing of plant material, such as drying is done to prevent the degradation of plant material during storage. However, hot air drying, in some cases, can cause degradation [5] in terms of color reactions and decomposition of active ingredients. . Color and essential oil content of the material have been in focus as the most important parameters, because essential oil content defines the taste and smell of the product and color is the most important parameter for visual appraisal by consumers. Some researchers have studied the chemical composition of the essential oils in fresh French Tarragon. The principal components found were estragole and E-β-ocimene [1].

Little information was found about the stability of dried tarragon and the color change after drying. [6] found a linear relation between drying temperature and losses of the essential oil in tarragon. However, in their application the drying time was 24 hr for all tests and therefore at high temperatures, the product has been over dried, which causes excessive losses of oil.

In previous research, the authors of this paper have studied the effect of drying conditions on the essential oil content and color of tarragon. The result showed that at the end of drying process the color (hue-value) and essential oil content were at drying temperatures of 45°C and 90°C significantly above that of 60°C. It was stated that drying time at 90°C was not long enough to impact color and essential oil content. Anyhow, quality deterioration could occur during the subsequent storage period.

The effect of drying conditions on the quality of other agricultural products during storage has been studied by a number of researchers. It has been reported that the amount of oil decreased during the storage time in mint species (*M. arvensis*, *M. spicata* and *M. pipertia*) [7], marjoram (*Majorana hortensis* M.) [8], tomato [9], *Alepidea amatymbica*, climbing onion (*Bowiea volubilis*), Savoy beet and amaranth leaves [10], wild ginger (*Siphonochilus aethiopicus*), (*Vernonia colorata*) [11] and shalgam (*Baucus carota* var. L.) [12]. Degradation

of the color during the storage time was found in chicory (*Cichorium intybus* L.) [13], tomato [9] and raspberry pulp [14].

The aim of this study was to evaluate the long-term effect of various drying conditions on color, essential oil content and oil composition during subsequent storage. The results of this research will be used for optimization of drying of tarragon to keep quality.

2 Materials and Methods

2.1 Plant material

French Tarragon was planted at Elburg (The Netherlands). The plants were harvested just before flowering in August 2004. Fresh tarragon leaves, separated from the stem manually, were used for the drying experiments.

2.2 Drying

Drying experiments were performed in the Department of Agrotechnology and Food Science of Wageningen University. The material was dried on trays in a hot air dryer an air velocity of 0.6 m/s. Ambient air was heated to 45, 60 and 90°C, resulting in RH of 17, 7 and 2.5%. In addition, for 60°C also RH of 18% has been applied to simulate drying at increased RH levels, e.g. in bulk drying or drying in recirculating mode. Drying was terminated when the material reached a moisture content of 10%, w.b.

2.3 Storage

The dried material was stored in airtight glass bottles [15]. The bottles were put in a box to protect the material against light [8] and kept in a refrigerator at a temperature of 5°C [16]. Samples were taken immediately after drying and after a storage time of 15, 30, 60 and 120 days.

2.4 Color

Quantitative evaluation of the color changes in tarragon leaves was done by a portable tri-stimulus colorimeter (Minolta Chroma CR-300, Osaka Japan) with a measuring area of 8 mm in diameter. Calibration of the colorimeter was done before each measurement series. The fresh as well as dried leaves were filled in a tray of 20 × 10 × 6 cm. Measurements were taken at five random points for each sample and each measurement was done in three replications.

Out of five available color systems, the L*a*b* [17-19] and L*C*h° [13] system were selected because these are the most used systems for evaluation of dried food material. To narrow down the number of parameters to be described, that parameter was looked for, reflecting best the visual appraisal of experts. Therefore, a panel of experts was formed from staff members of two Dutch companies that are active in the herbs and spices trade (Euroma B.V. and VNK B.V.). Fifteen samples the dried tarragon leaves together with photographs of the same samples were systematically arranged based on increasing values of the color parameters L*, a*, b*, C* and h°. The hue-value h° corresponded best to the visual ranking of the experts was selected. Based on the expert appraisal the hue-value of h°=95 was defined as threshold level to distinct between acceptable (h°>95) and unacceptable color for dried tarragon leaves.

2.5 Distillation

A Lickens-Nickerson apparatus was used for hydro-distillation [20]. Twenty grams (dry matter) of dried tarragon leaves was added to a flask and mixed with 500 ml of distilled water. The flask was then heated by immersion in an oil bath of 140°C for two hours counted from the time after condensation of the first drop of vapor in the calibrated tube [21]. The amount of extracted oil in the calibrated tube was measured and then collected in glass vials. The oil was stored in at 5°C.

2.6 GC-FID

The oil components were analyzed with a Hewlett-Packard 5890 gas chromatograph, coupled to a flame ionization detector (FID). The conditions were: split 1:100; carrier gas, hydrogen at 5 ml/min (5 psi). A DB-5 capillary column (30 m × 0.32 mm, 1.5 µm film thickness) was used for separating the components. The following temperature program was used: 65 to 225°C at 8°C/min. The oil sample was dissolved in t-butyl methyl ether in a concentration of 1% and 1 µl was injected in the GC. Three major components were selected to examine any changes in oil composition as a function of the various drying parameters. The selected compounds were estragole (main compound), E-β-ocimene (typical of a highly volatile compound) and methyl eugenol (typical of a high boiling compound).

2.7 GC-MS

GC-MS analyses were carried out on a Varian 3400 gas chromatograph equipped with a DB5 capillary column (60 m, 0.25 mm ID, 0.25 µm film thickness) directly coupled to the ion source of a Finnigan MAT 95 mass spectrometer. The samples were injected in split mode (split ratio 1:30) and the column temperature was programmed from the initial temperature of 60°C to 260°C at a rate of 3°C/min. The mass spectrometer was operated in the 70 eV EI mode with exponential scanning from m/z 24 to 300 at 0.5 s/decade, resulting in a cycle time of 0.68 s/scan.

Identification of the components was performed by comparing the obtained mass spectra with those in the Wageningen Collection of Mass Spectra of Natural Compounds and by checking the relative retention index of the proposed compound.

Three major components were selected to examine the changes of the relative and absolute value during the storage time.

3 Result and discussion

3.1 Color

The changes of the hue value after drying and during the storage time is shown in *Fig.1*. The value of the leaves just after drying is shown at time 0 in the figures.

The hue value, measured just after drying showed a lower level for all treatments compared to the value of fresh leaves (*Fig.1*). The difference between fresh leaves and the leaves after drying was smaller at 45°C and 90°C compared with the dried material at 60°C. During the storage time, the hue value was almost stable for the material dried at 45 and 60°C but it decreased for the material dried at 90°C in the first 30 days and remains then roughly at the same level as the dried at 60°C for the rest of the drying period. The values higher than the threshold show the acceptable level of hue value.

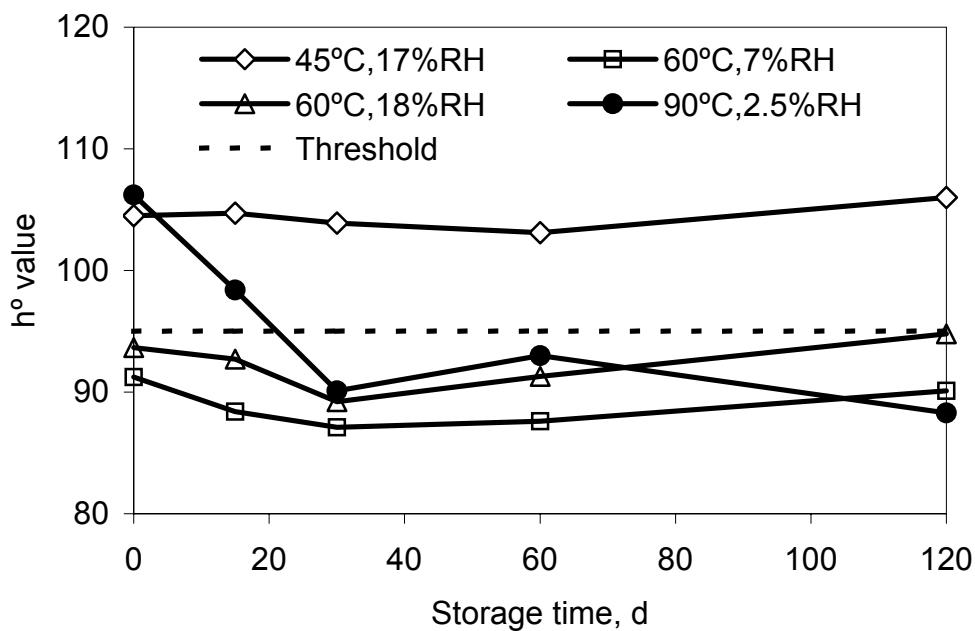


Fig. 1 Variation of the hue value h° of tarragon leaves, dried at different temperatures during storage (fresh tarragon: $h^\circ = 124.5$; threshold for dried tarragon: $h^\circ = 95$, higher values indicate acceptable quality).

The effect of the relative humidity of the drying air at 60°C was very small but for the driest air the level of hue was lower.

Only the condition of the material dried at 45°C remains above the quality threshold of $h^\circ = 95$ as defined by the experts.

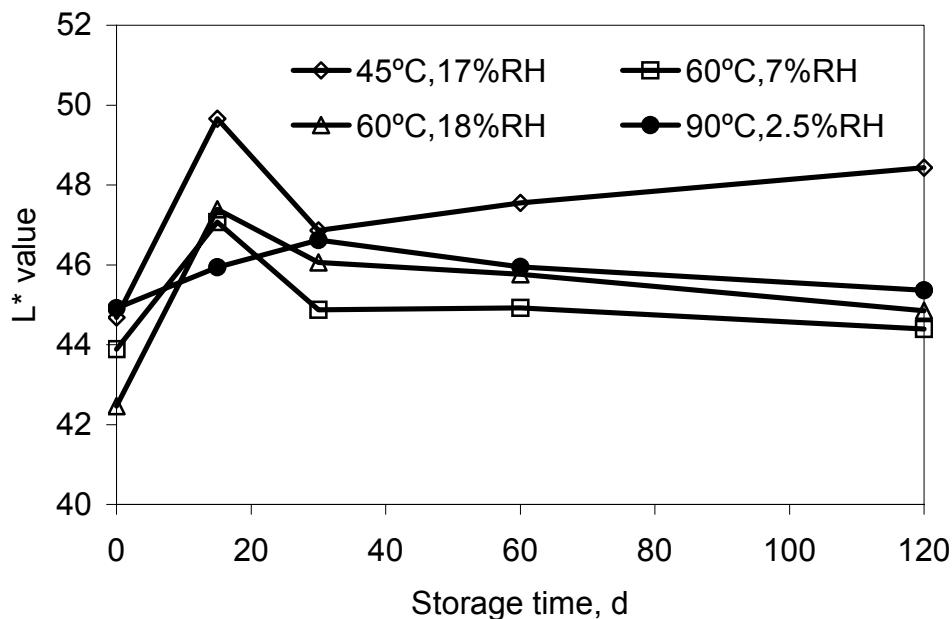


Fig. 2 Variation of the lightness value L^* of tarragon leaves, dried at different temperatures during storage (fresh tarragon: $L^* = 47$).

The L^* value represent the change in the lightness level of a sample so it is very useful to judge the browning and darkening lower values of leaves after drying. The change of the lightness is shown in *Fig.2*. The results show an increase after 15 days, then followed by a decrease for 30 days and longer, which continues for the later measurements except for the material dried at 45°C which increase again after 60 and 120 days.

Fig.3 presents the saturation (C^* value) with storage time. The graph shows that the saturation slightly tends to higher values for dried materials at 45 and 90°C and to the lower level for the dried leaves at 60°C compared with the fresh leaves.

Just like for hue, the material at 60°C shows lower values for 7% RH compared to 18% RH.

3.2 Oil recovery

Fig.4 presents the total amount of oil recovery for each treatment at different storage times. The amount of oil in the fresh leaves (MC=80%, w.b.) was 5.3 ml/100 g dry matter. The data at 0 show the amount of oil content of the dried leaves just after drying. Among the treatments the smallest losses of oil during drying was at 90°C and the largest was at 60°C. The losses at 60°C are more distinct at lower RH than at higher RH but the differences between the relative humidities are small. The results show that the total decay in oil content after 120 days is smallest at 90°C. The final oil content is at the same level as the oil content for drying at 45°C.

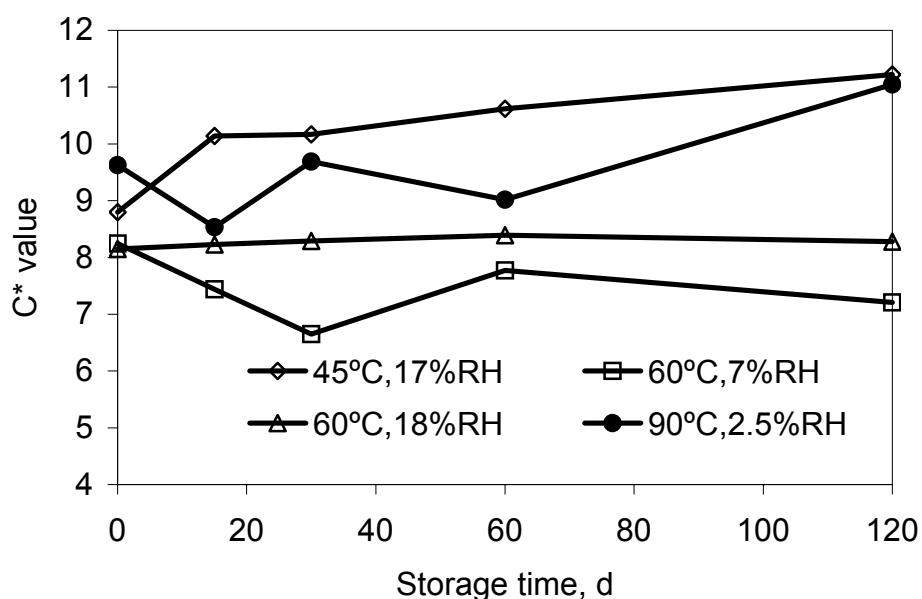


Fig. 3 Variation of the saturation (C^*) of the dried tarragon leaves at different temperatures during the storage time. (fresh tarragon: $C^* = 10$).

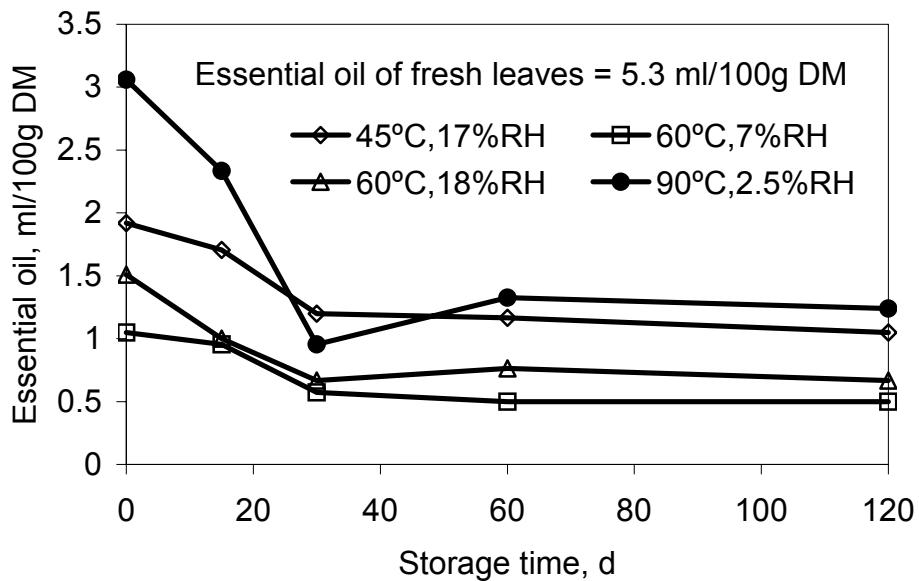


Fig. 4 Changes of the essential oil recovery of tarragon leaves dried at different temperature during the storage (essential oil content of fresh tarragon=5.3 ml/100g dry matter (DM)).

3.3 Compounds

The effect of storage was examined on the relative and absolute amount of estragole, E- β -ocimene and methyl eugenol. The variations of the absolute values are presented for the selected compounds in *Fig.5*. It was found that the reduction rate of the total oil recovery and of the three compounds was similar. The material dried at 60°C and 18% relative humidity showed the smallest change of the absolute value of estragole during the storage time. The level of the absolute value of estragole for the material dried at 60°C at 18% was higher than the one at 7% RH. The amount of E- β -ocimene of the material dried at 45°C was higher and showed less change than the other treatments after 120 days storage.

The level of methyl eugenol slightly decreased in the first 30 days and the largest changes were found for the material dried at 90°C. The rate of reduction was almost the same for all treatments after 30 days of storage. The methyl eugenol was most stable compound among these three compounds.

Note: The amount of estragole is about 10 times larger than that of the other two oils.

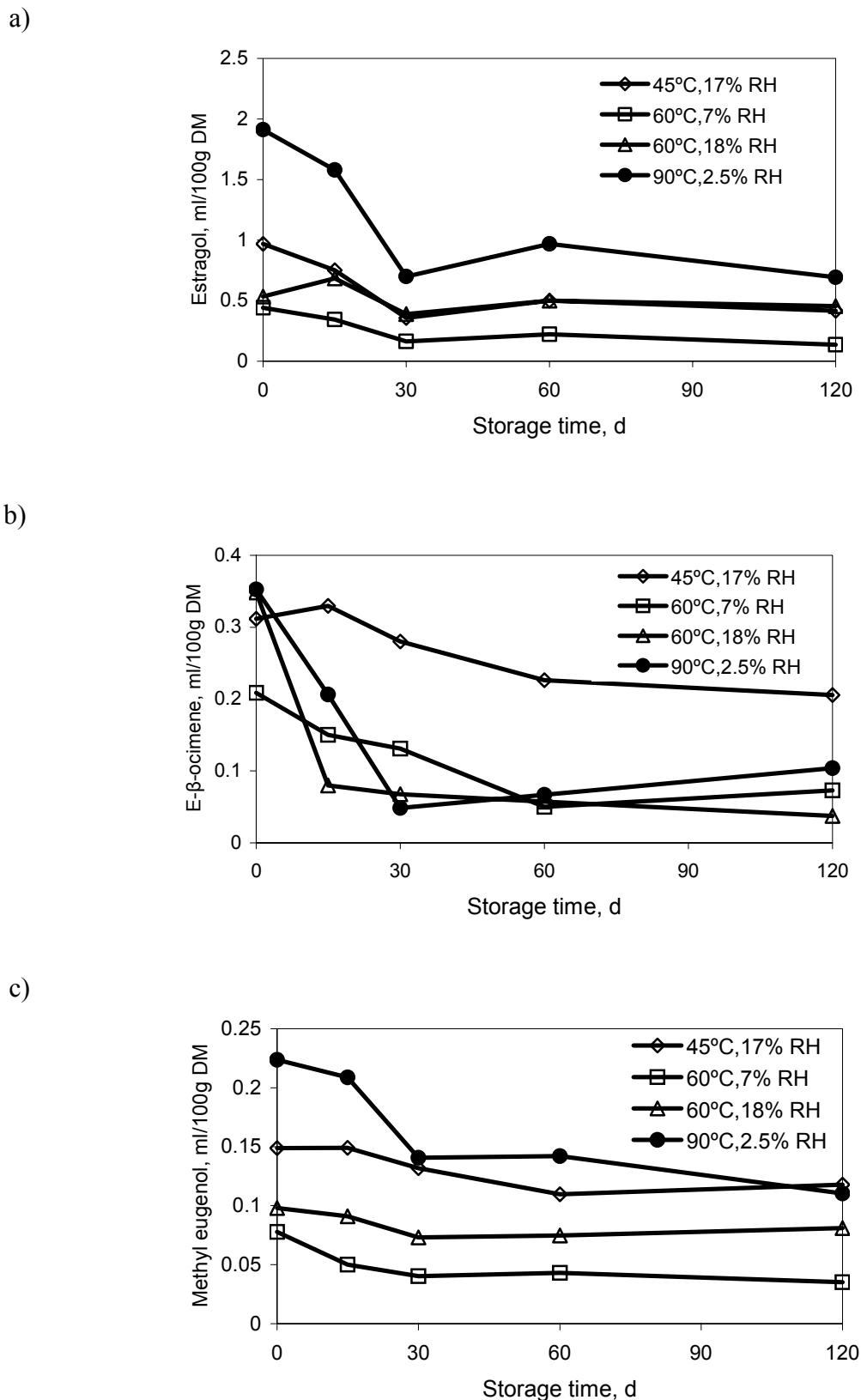


Fig. 5 Changes of absolute amount of estragole, E-β-ocimene and methyl eugenol in tarragon leaves dried at different temperatures during storage. (fresh tarragon leaves: estragole =3.63%, E-β-ocimene = 0.32% and methyl eugenol = 0.45%).

Table 1 variation of the relative amount of the main compounds in essential oil of French Tarragon leaves, dried at different drying temperatures during storage time. The changes are categorized in five groups: \uparrow = more than 30% increase, \nearrow = 10 to 30% increase, \rightarrow = less than 10% changes, \searrow = 10 to 30% decrease and \downarrow = more than 30% decrease.

| Drying condition | Compound | Scan number | Storage time, d | | | |
|------------------|---------------------|-------------|-----------------|---------------|---------------|---------------|
| | | | 15 | 30 | 60 | 120 |
| 45°C, 17%RH | Camphene | 1151 | \rightarrow | \rightarrow | \rightarrow | \nearrow |
| | Sabinene | 1233 | \rightarrow | \rightarrow | \rightarrow | \rightarrow |
| | Myrcene | 1276 | \nearrow | \nearrow | \nearrow | \nearrow |
| | E- β -ocimene | 1518 | \rightarrow | \rightarrow | \rightarrow | \rightarrow |
| | Estragole | 2226 | \rightarrow | \rightarrow | \rightarrow | \searrow |
| | Geranyl acetate | 2930 | \rightarrow | \nearrow | \nearrow | \nearrow |
| | Methyl eugenol | 3037 | \nearrow | \nearrow | \nearrow | \uparrow |
| 60°C, 7%RH | Camphene | 1151 | \rightarrow | \rightarrow | \nearrow | \uparrow |
| | Sabinene | 1233 | \rightarrow | \searrow | \rightarrow | \rightarrow |
| | Myrcene | 1276 | \nearrow | \rightarrow | \nearrow | \uparrow |
| | E- β -ocimene | 1518 | \searrow | \searrow | \downarrow | \downarrow |
| | Estragole | 2226 | \searrow | \searrow | \nearrow | \searrow |
| | Geranyl acetate | 2930 | \uparrow | \uparrow | \uparrow | \uparrow |
| | Methyl eugenol | 3037 | \rightarrow | \searrow | \nearrow | \rightarrow |
| 60°C, 18%RH | Camphene | 1151 | \searrow | \searrow | \rightarrow | \downarrow |
| | Sabinene | 1233 | \rightarrow | \rightarrow | \rightarrow | \downarrow |
| | Myrcene | 1276 | \downarrow | \downarrow | \downarrow | \downarrow |
| | E- β -ocimene | 1518 | \downarrow | \downarrow | \downarrow | \downarrow |
| | Estragole | 2226 | \uparrow | \uparrow | \uparrow | \uparrow |
| | Geranyl acetate | 2930 | \uparrow | \nearrow | \searrow | \downarrow |
| | Methyl eugenol | 3037 | \uparrow | \uparrow | \uparrow | \uparrow |
| 90°C, 2.5%RH | Camphene | 1151 | \searrow | \rightarrow | \rightarrow | \uparrow |
| | Sabinene | 1233 | \rightarrow | \downarrow | \rightarrow | \nearrow |
| | Myrcene | 1276 | \searrow | \searrow | \downarrow | \uparrow |
| | E- β -ocimene | 1518 | \downarrow | \downarrow | \downarrow | \downarrow |
| | Estragole | 2226 | \nearrow | \nearrow | \nearrow | \rightarrow |
| | Geranyl acetate | 2930 | \rightarrow | \rightarrow | \rightarrow | \uparrow |
| | Methyl eugenol | 3037 | \uparrow | \uparrow | \uparrow | \uparrow |

The relative amounts at different storage time were compared to the relative amount of the compounds in dried leaves just after drying. The variation of some of the main compounds for different drying conditions is shown in Table 1. The leaves dried at 45°C showed less changes for most of the compounds compared to 60°C and 90°C. Sabinene was the more stable and E- β -ocimene was the less stable one among the selected compounds.

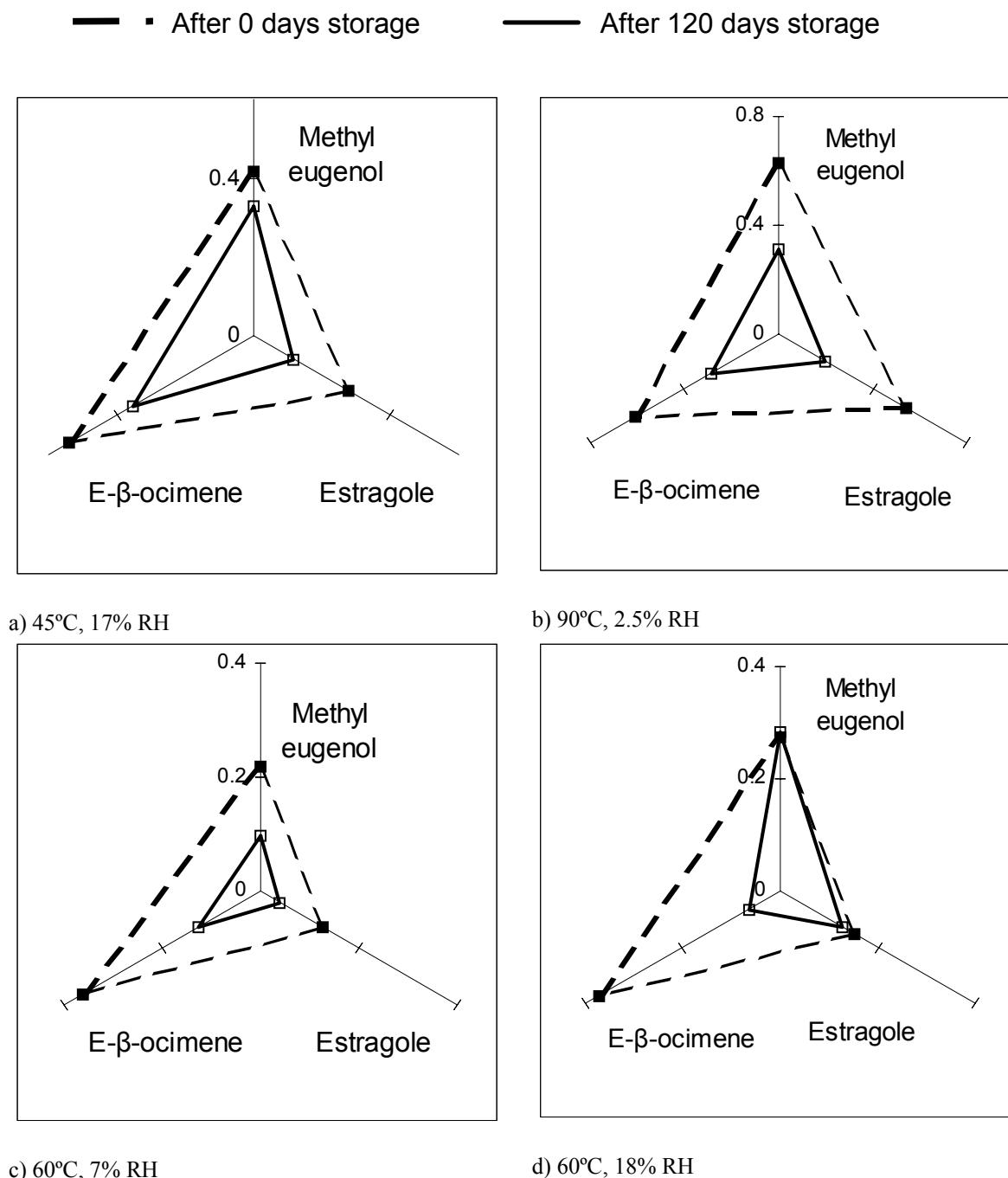


Fig. 6 Changes of three main components of French Tarragon leaves during storage (120 days) (The ratio of absolute amount of the compounds of the stored tarragon leaves over the absolute amount of the compounds of fresh tarragon leaves).

The reduction of the absolute amount of three main compounds for each drying treatment is shown in *Fig.6*. The three axes in the Radar graph are in one plane. The values on the three axes represent the percentage of the remaining oil compound compared to the amount in fresh leaves. The points on each axis represent the changes of the compound and the shape of the triangles show the shift of the composition of the selected compounds during storage time. The centre points of the triangles were calculated to find the possible shift of the compounds. In most of the cases the centre points were almost on the same place so the shift of the composition was negligible.

3.4 Discussion

Color and essential oil content change during storage due to the level of temperature and duration of the drying process. Low changes of color and essential oil were observed for the leaves dried at 45°C during the drying process as well as storage period. The changes for the leaves dried at 60°C were high by drying but low during storage time. The leaves dried at 90°C showed low changes by drying and high changes during storage.

For an equal duration of process more changes occur when the temperature is higher and at a constant temperature longer drying time causes more changes. As the required time to reach equilibrium moisture content is shorter at higher temperature, less change is expected because of a shorter time and more changes expect because of a higher temperature. The duration at 45 and 60°C is long enough for changes of color and essential oil. However during storage after drying no more changes in hue value occur in the material dried at 45°C, while for 90°C the color changes in 30 days to the level of 60°C drying temperature. The change in color at 60°C completely occurred during drying because drying time is long enough and temperature is high enough for changes.

It was assumed that moisture content also affects color changes during drying. When moisture content is higher more changes occur. That is the reason why more changes occur at the earlier steps of drying process. Color change also might stop earlier at 90°C compared to 60°C because material reaches faster lower moisture content.

Decrease of the total oil causes decrease of the absolute amount of each compound. But the change of the relative amounts can be different for different compounds because they have different sensitivity to the temperature.

The essential oil content and the hue value were at lower level for the dry air compared to the more humid drying air at 60°C. Humidity of the drying air could be a factor, which defines the level of degradation.

4 Conclusion

The essential oil content and the color of the dried tarragon leaves change during the storage time. The level of changes of the color and essential oil content during the storage time depend on the drying conditions. The largest change in color and essential oil content was observed for the material dried at 90°C and the smallest for the 45°C. For the material dried at 60°C, the oil compounds were more stable during storage for the material dried at 18% RH compared to 7% RH. During the storage time the shift of the three main oil compounds at 45, 60 and 90°C was negligible. It seems that the smaller changes for the material that occur during drying as at 90°C are compensated by larger changes at storage. Based on these results, drying at temperature of 45°C is recommended to have less change during storage period.

Acknowledgment

The financial support from Ministry of Science and Research and Technology of Iran was gratefully appreciated.

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7

Modeling of operation of drying tarragon (*Artemisia dracunculus* L.) leaves in batch dryer

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This chapter submitted for publication

Abstract

Cost calculations are an important aspect for drying herbal products. In this work a simulation model for batch drying of tarragon leaves is presented. It was found that thin layer drying models, which are explicit in time, are not useable for bed simulations. To solve this problem an extension is made to the standard Lewis model to calculate the drying rate in a batch dryer.

The simulation model is used to compare several drying scenarios, amongst others the effect of drying temperature, recirculation rate and bed height. Also the differences in drying costs in a West-European country (The Netherlands) and a country in the Middle-East (Iran) were evaluated.

It was found in previous research that for keeping quality, 50°C is the highest allowed temperature. To save energy, recirculation at a maximum humidity ratio of 0.015 kg_{water}/kg_{air} is recommended. Lower drying temperatures may give more energy savings, but then drying time increases significantly and thus the yearly capacity reduces and fixed costs increase.

Drying at 50°C was found as the best option to minimize the costs and keep the quality of the products. Cost estimation shows that the total costs per kg dry mass for the material dried at 50°C with an air velocity of 0.5 m/s for a bed height of 0.20m is about three times higher in The Netherlands compared to Iran conditions. Recirculation is profitable from an energy saving point of view and it is important for drying in West-Europe, but the effect is not that important for drying in the Middle-East.

Keywords: *Artemisia dracunculus*; cost; drying; modeling; cost optimization; quality; recirculation dryer; tarragon; thin layer.

1 Introduction

Tarragon, *Artemisia dracunculus* L., is a herbaceous plant belonging to Asteraceae. Two varieties of tarragon can be distinguished [1, 2], namely French Tarragon of South European origin and Russian Tarragon of Siberian origin. French Tarragon is mainly used as a culinary herb. Russian Tarragon has an inferior flavor and is more used as medicine. Preservation is needed to maintain the quality of the herbs during storage. Drying is one of the most important preservation methods of herbs to assure longer shelf life [3].

Foodstuffs are heat sensitive and a high drying temperature usually causes low quality of products [4]. Therefore the drying process must be carried out under specific conditions in order to obtain the desired final product quality.

Drying of herbal and medicinal products normally takes place in batch dryers in which heated air passes a bed with product. During drying a batch, there will be gradients for moisture content, temperature and relative humidity as a function of bed height, which change during time. The drying time for such a system depends on the bed height, air velocity, applied temperature and humidity of the inlet air. So for the operation of such system there are several decision variables, and the choice of these variables is also affected by the constraints of keeping quality in terms of product color and oil content (Ch-4, Ch-5). Moreover, the decisions on operation variables are also linked to the costs of the use of such equipment like thermal and electricity costs, costs for depreciation, interest and maintenance. The objective of this work is to determine from several scenarios the optimum conditions of drying of tarragon to minimize the costs and energy consumption per unit of dry mass of the product while taking care of the final product quality.

In previous work thin layer drying models for tarragon leaves were evaluated for drying of tarragon [5], and it was concluded that the Page model $MR = \exp(-kt^n)$ fitted the best to experimental data. It was the intention to use these models for the evaluation of several drying scenarios. For this work the approach as presented by Parry (1985) for deep bed drying models was followed [6]. However, in the Page model time is explicit in the equation, and as a result the drying rate will run out after a certain time. The use of the Page equation in the context of deep bed dryer models will have serious limitations and therefore in this work a shift has been made to an alternative drying model.

| List of symbols | | |
|-------------------------|-----------------------------------|--|
| C_1, C_2, C_3 and n | Coefficients | - |
| C_{el} | Electricity cost | €/kg dry matter |
| C_{thermal} | Thermal (fuel) cost | €/kg dry matter |
| $c_{p,a}$ | Heat capacity of dry air | kJ/kg _{air} .K |
| $c_{p,v}$ | Heat capacity of vapor | kJ/kg _{water} .K |
| E_{total} | Total energy consumption | MJ |
| ERH | Equilibrium relative humidity | (-) |
| F_u | Fuel consumption | l |
| H_{bed} | Bed height | m |
| h | Total enthalpy | kJ |
| h_{in} | Enthalpy of ambient air | kJ/h |
| h_{mix} | Enthalpy of mixed air | kJ/h |
| h_{return} | Enthalpy of return air | kJ/h |
| h_{vap} | Latent heat of vaporization | J/kg _{water} |
| k | Drying rate constant | 1/h |
| M | Moisture content of the material | -, dry base |
| M_0 | Initial moisture content | - |
| M_a | Mass flow rate | kg/h |
| MC | Moisture content | %, wet base |
| M_e | Equilibrium moisture content | - |
| MR | Moisture ratio | - |
| M_t | Moisture content at time t | - |
| M_{top} | Moisture content at the top layer | - |
| P_{el} | Price of electricity | €/kWh |
| P_{fu} | Price of fuel | €/l |
| RH_a | Ambient relative humidity | % |
| t | Drying time | h |
| t_{life} | Life time | yr |
| t_{max} | Maximum drying time | h |
| T | Drying temperature | °C |
| T_a | Ambient air temperature | °C |
| T_{crop} | Crop temperature | °C |
| v | Air velocity | m/s |
| W_{fan} | Power of fan | kW |
| X_a | Air humidity ratio | kg _{water} /kg _{air} |
| ρ_a | Density of the air | kg/m ³ |
| ρ_f | Density of the fuel | kg/l |
| ρ_p | Bulk density of product | kg/m ³ |

As the weather conditions and prices for equipment, energy and labor, which define the operation costs of dryers, differ over the countries, two situations are considered for the cost calculations. The situation in The Netherlands is considered as representative for West-Europe, while the situation of Iran is considered as representative for the Middle-East.

2 Methodology

First, a mass and energy balance model of the drying process was made in order to calculate drying time and energy requirements and then a cost model was used. This research used numerical simulation in Matlab for drying of tarragon at different temperatures, relative humidities, airflow rates and bed heights. A drying model was applied to find how to control quality and drying at different drying zones. The quality is taken into account in terms of maximum oil content (flavor) and minimum color change. Results of drying simulation were transferred to an Excel worksheet for cost evaluation.

2.1 System description

Conventionally, in most batch dryers for herb drying (8-10) air passes the product once and is exhausted directly. Main drawback of this method is that loss of energy potential occurs in the exhaust air. To realize energy savings, recirculation of air can be applied. However, after reach pass of the air through the dryer the moisture content increases and the drying capacity of the air lowers. In this work a recirculation mode is applied in which air humidity ratio and temperature at the dryer inlet are kept constant. The system is given in *Fig 1*.

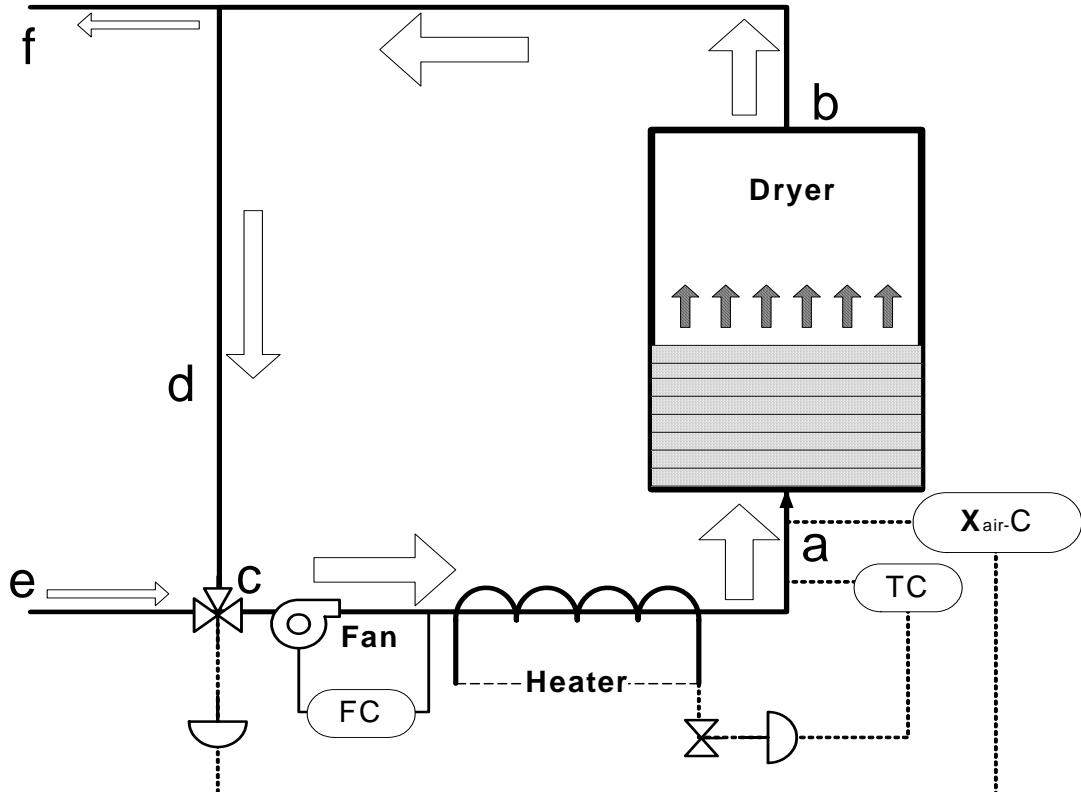


Fig. 1 Diagram of a recirculation dryer. TC , X_{air-C} and FC are the controllers of temperature, humidity ratio of the air and airflow, a is the set point, b is the outgoing air, c is the three-way-valve to regulate the flow rate of the fresh air e and return air d to the system. f is the exhaust air.

To create an accurate temperature of the air surrounding the material, the drying temperature is measured and controlled at set-point just before the dryer (a). Air is blown through the dryer at constant controlled flow rate. The outgoing air at (b) is partly exhausted at (f) and the other part passes the duct (d). The air from the duct (d) is mixed at (c) with fresh air from the duct (e). The humidity ratio (or relative humidity) at point (a) is controlled by a proper mixing ratio of flow (e) and flow (d). The mixed air is then heated to reach the selected temperature at point (a). Drying is continued until the desired moisture content is reached in the top layer. The controls given in this system can be done either automatically or by operator.

During the time of batch drying, the moisture content of the air leaving the bed will gradually decrease, and at the same time the temperature increases. By applying the humidity ratio controller, the amount of recirculated air increases, and more and more the drying potential of the exhaust air is used. As a result the required energy in the heater decreases. The recirculation rate is controlled by the humidity ratio before the dryer. In case the humidity ratio at this point is chosen to be equal to ambient, no recirculation is applied.

2.2 Assumptions and boundaries

Assumptions

The assumptions for modeling the drying process are shown in Table 1. Two ambient air conditions were taken as the average conditions of the inlet air during the harvesting season: The average conditions during the harvest season in The Netherlands as 20°C and 70% RH_a and those in Iran as 30°C and 30% RH. Most calculations are done at an air velocity of 0.6 m/s because the laboratory experiments also were performed at this velocity.

It was assumed that the dryer size is 12m² (3m×4m) and it will be used only for this crop. The fresh crops are available only 70 days per year for drying. The maximum drying time in a year, which is called *season*, was limited to 1150 hours (70 days and 24 hours per day and time efficiency of 80%).

The bulk density of the fresh material was measured as 250 kg/m³ based on wet material. At the start of drying the weight of the fresh material with a bed height of 0.20m in the batch will be 50 kg/m². The volume of the material in the dryer was taken as 2.4m³ (12m² × 0.20m) for most cost calculations.

The labor costs are left out of the calculation because we want to focus only on the costs of the drying process. Also the building costs are set at zero assuming that an existing building can be used for the drying floor.

Table 1 Assumptions for simulation and cost calculation.

| Parameter | Symbol | Value or range | | Unit |
|------------------------|------------------|-----------------|------|-----------|
| | | The Netherlands | Iran | |
| Ambient temperature | T _a | 20 | 30 | °C |
| Ambient RH | RH _a | 70 | 30 | % |
| Drying temperature | T | 35 to 90 | | °C |
| Air flow | v | 0.1 to 1.0 | | m/s |
| Working days | | 7 | | days/week |
| Working hours per year | | 1150 | | h/yr |
| Bed height | H _{bed} | 0.1 to 1.0 | | m |

Boundaries

Deep bed drying is not relevant for drying of heat-sensitive materials because drying starts fast at the inlet zone while the outlet zone is still wet and cool. The efficiency of the drying process will also decrease [7]. In previous work, it was found that drying at temperatures above 50°C does not provide an acceptable level of color and essential oil content. The top layer is always more wet than the layer at the bottom of the bed. A range of 0.1 to 1.0m was chosen to evaluate the effect of bed height on drying time, energy consumption and costs. The top layer was taken as the critical layer, to guarantee the necessary storage moisture content (MC). A final MC of 10% to be reached for the top layer was defined from the sorption isotherm of tarragon (Ch-2). The constraints are shown in Table 2.

Table 2 Boundaries for drying simulation.

| | | |
|------------------|------------|--------|
| MC of top layer | M_{top} | < 10% |
| Crop temperature | T_{crop} | < 50°C |

2.3 Simulation of batch dryer

The conditions of drying air passing a bed of product and the conditions of the product are described by mass and enthalpy balances. During drying of agricultural products the temperature difference between product and the air is small [8]. At a certain point in the dryer the mass and heat balance for the air is given by the following equations:

$$M_a \frac{dX_a}{dx} = \rho_p A \frac{dM}{dt} \quad (1)$$

$$M_a \frac{d(c_{p,a} + X_a c_{p,v})T_a}{dx} = h_{vap} \rho_p A \frac{dM}{dt} \quad (2)$$

This set of partial differential equations is solved by discretisation in time (time steps) and space (layers). *Fig 2.* represents a schematic view of the calculation scheme. First, at t_1 , and for the given inlet air conditions drying rate, product moisture content, temperature and moisture of the air are calculated for layer 1. Then air temperature and air moisture content leaving the first layer are considered as the input conditions for layer 2. Subsequently, the conditions in all layers are calculated. The air conditions in the top layer correspond to the recycle air.

Next a step forward in time is made and for the given air conditions at the bottom layer and the moisture content results in the bottom layer at the previous time step, the conditions in all layers are calculated. This procedure is repeated until the moisture content in the top layer equals to 10%. In this calculation scheme Euler-forward integration is applied. Step sizes were chosen by evaluation of the results for stepwise decreasing steps (in time and space) till no relevant improvement of the results occurred.

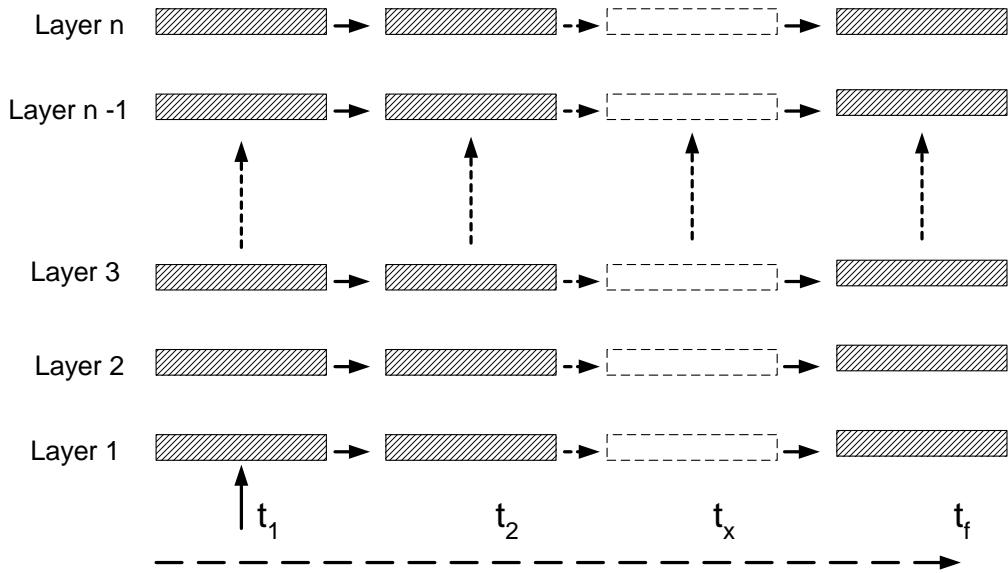


Fig. 2 Calculation scheme of the simulation model. The vertical arrows represent the air and crop conditions and the horizontal arrows the evolution in time. t_f is the final drying time.

2.4 Drying rate model

In previous work, thin layer models were evaluated to model the drying of tarragon [5]. From model fitting to experimental data, the Page equation was selected because of the lowest standard error and residual sum of square.

$$MR = \frac{M_t - M_e}{M_0 - M_e} = \exp(-kt^n) \quad (3)$$

or

$$M_t = (M_0 - M_e) \exp(-kt^n) + M_e \quad (4)$$

In which for k and n are derived equations dependent on temperature.

The drying rate follows from the derivative of the expression in time (dM/dt):

$$\frac{dM}{dt} = -(M_0 - M_e) kt^{n-1} n \exp(-kt^n) \quad (5)$$

Here time is explicit in the equation. A consequence is that the exponential term tends to zero if t , and so kt^n becomes large and thus drying stops. Late changes of drying conditions will not have any effect (in spite of change of M_e).

The model shows that in the initial phase of drying much water is removed from the lower layers and as a result in the higher layers temperature is lower and RH of the air is higher. As a consequence drying rate in the higher layers is lower which results in different moisture content for these layers. After some time the drying conditions in all layers come close to each other and the k and n value in the Page model are nearly equal in all layers. Then, as a consequence of the term $\exp(-kt^n)$ drying stops at all layers at the same time. This is not in correspondence to real drying systems where drying in the higher layers stop at a later moment than in the lower layers. So it is clear that the Page drying equation, just as all models

in which time is explicit in the equation, is not useable for deep bed drying simulation. An alternative is found by going back to the Lewis equation [5].

$$MR = \exp(-kt) \quad (6)$$

which originates by solving the differential equation

$$\frac{dM}{dt} = -k(M - M_e) \quad (7)$$

This differential equation is an autonomous model and not driven by time. However, previous work (Ch-3) showed that the accuracy of this model was rather poor compared to the Page equation. In order to improve the accuracy, the Lewis equation is modified into:

$$\frac{dM}{dt} = -k(M - M_e)^n \quad (8)$$

The desorption curve or the relationship of equilibrium moisture content and relative humidity is calculated as a function of temperature and relative humidity by using the modified Halsey equation (Eq.9) (Ch-2).

$$M_e = \left(\frac{-\exp(C_1 + C_2 T)}{\ln(ERH)} \right)^{1/C_3} \quad (9)$$

Table 3 shows the values of the constants used in the equations.

Table 3 Applied values in calculation.

| Parameter | Value | Reference | Parameter | Value | Reference |
|-----------|-----------------------|-------------|-----------|---|-------------|
| $C_{p,v}$ | 1.93 kJ/kg | [9] | C_1 | -2.606 | Ch-2 |
| $C_{p,a}$ | 1.0 kJ/kg | [9] | C_2 | 0.00776 | Ch-2 |
| ρ_p | 50 kg/m ³ | Experiments | C_3 | 1.30 | Ch-2 |
| ρ_a | 1.1 kg/m ³ | [9] | $M(t=0)$ | 4.0 kg _{water} /kg dry product | Experiments |
| h_{vap} | 2,500 kJ/kg | [9] | | | |

2.5 Energy consumption

The total drying energy consumption was calculated based on the enthalpy flows at three points in the air circulation. The required energy was calculated based on the algebraic accumulation of the enthalpies of the mixed, inlet and return air using Eq.10.

$$h = h_{mix} - h_{in} - h_{return} \quad (10)$$

in which h_{mix} is the enthalpy of the air just before the first layer, h_{in} is the enthalpy of the inlet (fresh) air and h_{return} is the enthalpy of the return air for recirculation. The enthalpy at each point was found using Eq.11.

$$h = M_a (c_{p,a} T_a + X_a (h_{vap} + c_{p,v} T_a)) \quad (11)$$

Total energy input (kJ) is:

$$\text{Thermal energy} = \int_0^t h \cdot dt \quad (12)$$

2.6 Cost model

A simple annual cost model was used for cost calculation of the drying system and all the prices are given based on the current conditions in The Netherlands and Iran in 2005. The total costs are calculated for drying during one season, while the amount of product that can be dried varies with the chosen drying scenarios.

The total costs of the drying system consists of constant and variable costs of the drying floor, heating and ventilation system.

The constant cost consists of depreciation, interest and maintenance (repair and service).

Table 4 Investment (dryer) costs in Euro (2005).

| Items | Price, € | Explanation |
|----------------------------|----------|---------------------------------------|
| Drying floor and ducts | 4,000 | Drying floor area = 12m ² |
| Fan | 3,000 | 7.5 kW, 700Pa, 25000m ³ /h |
| Heater and heat exchanger | 3,500 | Capacity, 200 kW |
| Installation of the system | 1,000 | |
| Adaptation of the building | 500 | |
| Total dryer cost | 12,000 | |

The investments for the dryer are presented in Table 4. The same investment costs were applied for both countries. All prices are valid for June 2005.

Depreciation is the total dryer cost divided by the lifetime because it was assumed that the residual value is equal to dismantling costs.

Interest was calculated over the average investment and maintenance was calculated over the total investment (Table 5).

The variable costs consist of thermal (fuel) and electricity (ventilation) costs.

The light oil used for heating has a Lower Heating Value (LHV) of 42 MJ/kg and the density of the oil is 0.88 kg/l so the fuel cost for each process was found as follow:

$$Fu = \frac{En_{total}}{LHV} \cdot \frac{1}{\rho_{fu}} \quad (13)$$

$$Co_{thermal} = Fu \cdot p_{fu} \quad (14)$$

The electricity costs of the fan is calculated as follows:

$$Co_{el} = t \cdot W_{fan} \cdot P_{el} \quad (15)$$

In which t is the required time for each drying process.

The variable costs were calculated using cost factors, which are considered to be different for The Netherlands and Iran. The lifetime of the dryer in Iran is longer because the labor

costs for repair are lower. The maintenance was considered as 5% for both countries because in Iran there will be more repairs with lower labor cost. The cost factors are shown in Table 5.

Table 5 Cost factors for The Netherlands and Iran in 2005.

| Factor | The Netherlands | Iran |
|---------------------|-----------------|------------|
| Fuel price | 0.65 €/l | 0.1 €/l |
| Electricity price | 0.1543 €/kWh | 0.01 €/kWh |
| Life time for dryer | 10 yr | 15 yr |
| Maintenance | 5% | 5% |
| Interest | 5% | 12% |

3 Results and discussion

3.1 Drying rate model

The modified Lewis equation (Eq.8) was fitted to all data sets obtained in previous work (Ch-3) to find the k and n values of the modified Lewis equation at different temperatures and relative humidities. The values of k (Eq.16) and n (Eq.17) of tarragon leaves in the modified Lewis model were modeled as a function of temperature (Ch-3):

$$k = (0.04237 \times \exp(0.06605 \times T_a)) \quad (R^2 = 0.91) \quad (16)$$

$$n = (0.000531 \times T_a^2) - (0.0819 \times T_a) + 3.8237 \quad (R^2 = 0.87) \quad (17)$$

where

k and n are the coefficients of the modified Lewis equation. No indication for a dependency of k and n with relative humidity was found.

The *SEE* and *RSS* values of these fits are given in Table 6 and compared to the *SEE* and *RSS* values obtained for the Page model in chapter three. It must be noticed that in the previous work the Page model was fitted to the *MR* which ranges from 0.0 to 1.0. In this work the data is fitted to the actual moisture contents, which have a 4 times higher range. As a result, fitting the Page equation to the moisture content values instead of to the moisture ratio gives a *RSS* value, which is 16 times higher, and a *SEE* value that is 4 times higher. Table 6 shows the values, corrected in this way.

From these results can be concluded that the modified Lewis equation is more accurate than the Page equation.

Table 6 Comparing of quality of fit between Page and modified Lewis equations.

| Equation | RSS | SEE |
|----------------------|--------|--------|
| Page model | 0.0063 | 0.0442 |
| Modified Lewis model | 0.0035 | 0.0388 |

As an illustration for the good agreement of this model to the data, a number of experimental data sets and the fitted curves are shown in *Fig.3*.

In order to show the progress of drying in 25 layers model *Fig.4* is shown. Dutch conditions were applied for temperature and relative humidity of the ambient air. In this figure, the drying process starts at the same time for the twelve lowest layers. The drying rate decreases

with increasing distance from the air inlet. At time zero the air humidity ratio in layer 13 and higher is that high that the difference between the product moisture and equilibrium moisture content is zero ($M-Me=0$) which results in a drying rate equal to zero. After a short period the moisture ratio for layer 13 decreased so far that Me decreases and the difference between the product moisture and equilibrium moisture content is big enough to cause drying in layer 13. With progress of time, at different moments new layers start to dry. The drying process stops as soon as the moisture content of the top layer reaches 10%.

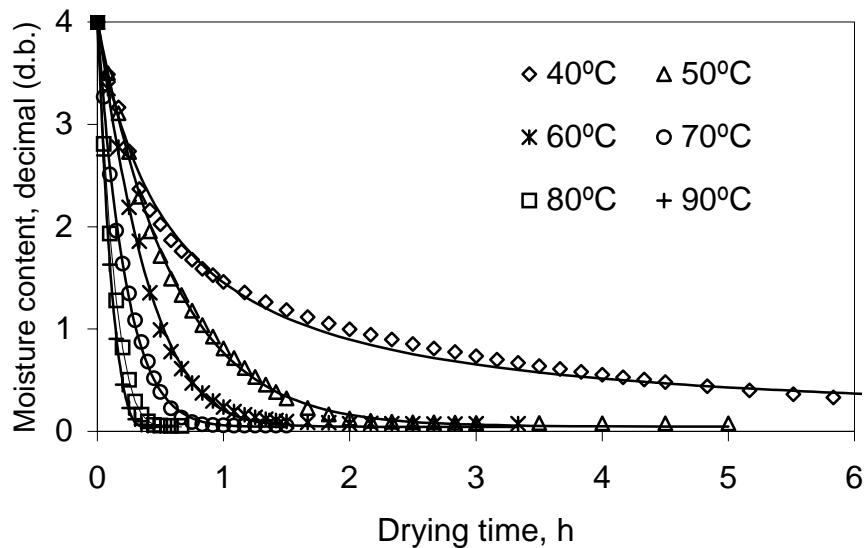


Fig. 3 Experimental data of the M of tarragon leaves at different temperatures and the fitted curves to $dM/dt=-k(M-Me)^n$.

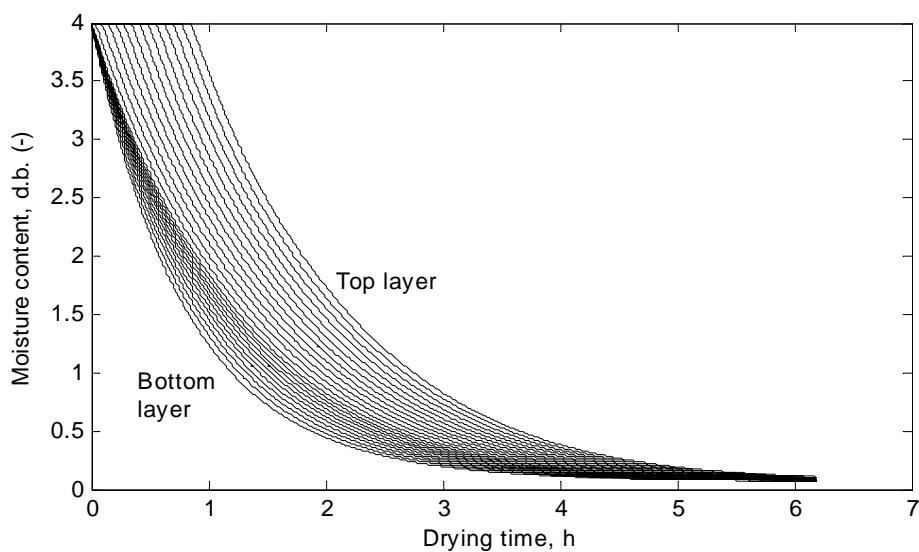


Fig. 4 The drying curves of the 25 layers of a batch of 0.20m height, dried at a temperature of 50°C, humidity ratio of 0.013 kg_{water}/kg_{air} and air velocity of 0.6 m/s.

The drying curve for the higher layers seems artificial. The main reason is not the nature of the model, but the available information used for the model. First reason is the fitting of the sorption isotherms was based upon data till a RH value of 90%, so from 90-100% extrapolation is applied. Second, and the most important aspect, is that the experimental data for the drying curves was restricted to RH values below 35%. In this range no dependency of

k and n with RH was found. Extrapolation of these results to RH-values in the range 50-99% in the top layers is not accurate (Mueller, 1991), but it is assumed that despite this omission the drying scenarios can be compared. It is recommended that for further implementation, the drying at high relative humidities will be validated.

Fig.5 shows the effect of humidity ratio on the drying time at temperature of 50°C and air velocity of 0.6 m/s for a batch of 0.20m height. The drying time is not much different for humidity ratios of 0.01 to 0.025 kg_{water}/kg_{air} and it increases much at humidities above 0.025 kg_{water}/kg_{air}. This in favor with the results of Mueller (1991) [10].

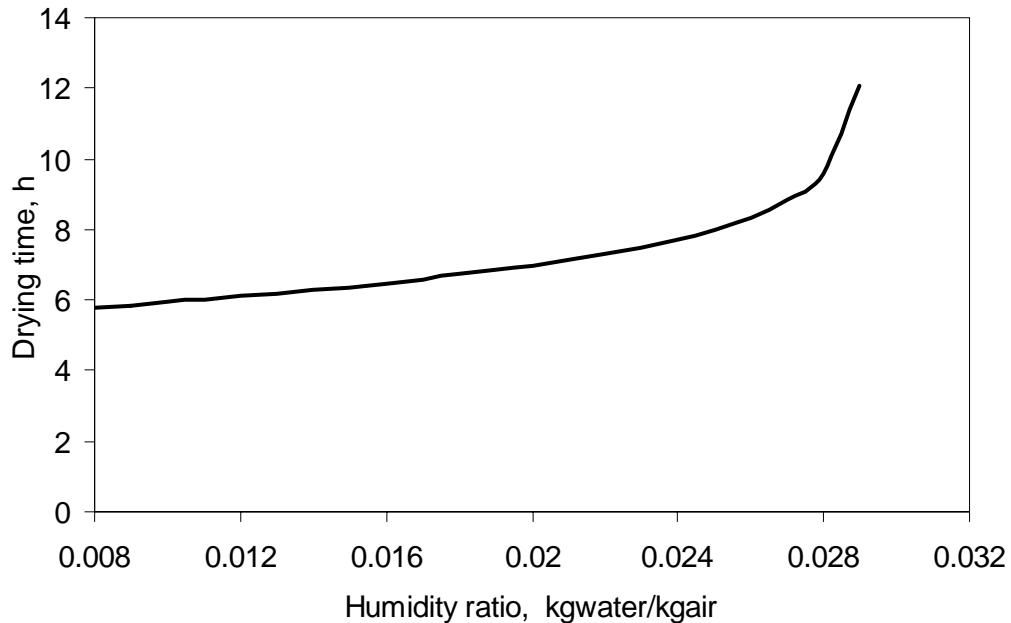


Fig. 5 Effect of humidity ratio on drying time when drying at a temperature of 50°C and air velocity of 0.6 m/s for a batch of 0.20m height.

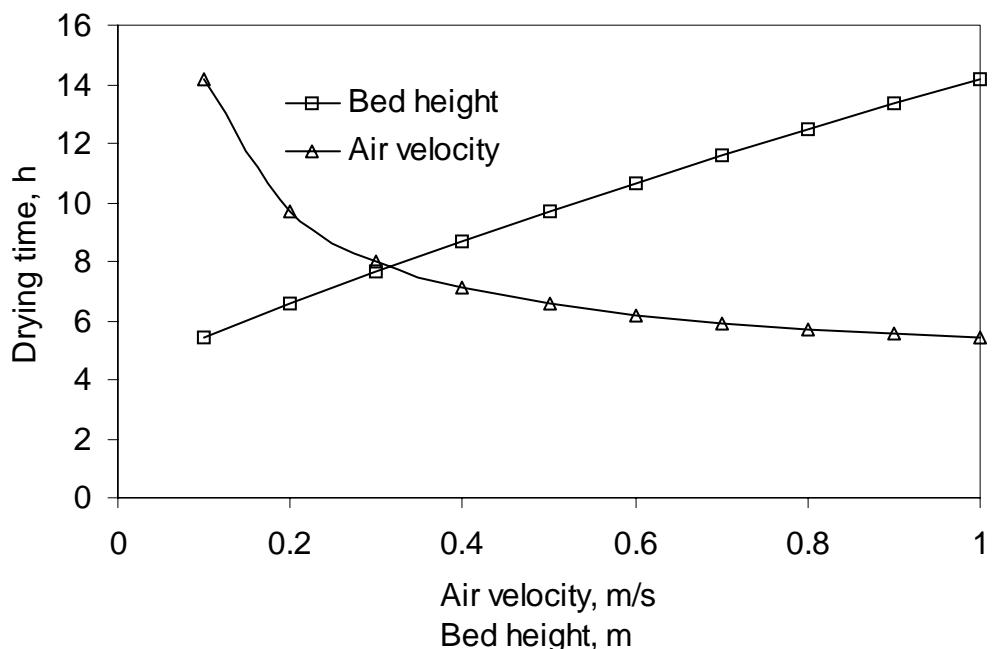


Fig. 6 Effect of air velocity and bed height on drying time when drying at a temperature of 50°C and humidity ratio of 0.013 kg_{water}/kg_{air}. The evaluation was done using an air velocity of 0.5 m/s for different bed heights and a bed height of 0.20m for different air velocities.

The effect of air velocity and bed height on drying time, when drying at a temperature of 50°C and a humidity ratio of 0.013 kg_{water}/kg_{air}, is shown in *Fig.6*. The air velocity of 0.5 m/s was used for evaluation of different bed heights while a bed height of 0.20m was applied at different air velocities. The drying time changed almost linear for the bed heights of 0.1 to 1.0m. The required time for drying of a bed height of 1.0m is about three times longer compared to the bed height of 0.1m while the amount of the product is 10 times more. It means that the efficiency of the dryer is higher at longer bed heights.

3.2 Energy

With the assumption that the crop temperature is nearly equal to the drying air temperature, the crop temperature during the drying process for each layer was calculated. *Fig.7* shows the temperature of the crop at different layers during the drying process, for the same conditions, which were shown in *Fig.4*. The temperature in the initial phase is at the level of saturated air, and the saw-tooth form is caused by the succeeding start of new drying layers. This graph can be used to control and to limit the crop temperature at an acceptable level to avoid degradation of color and oil content.

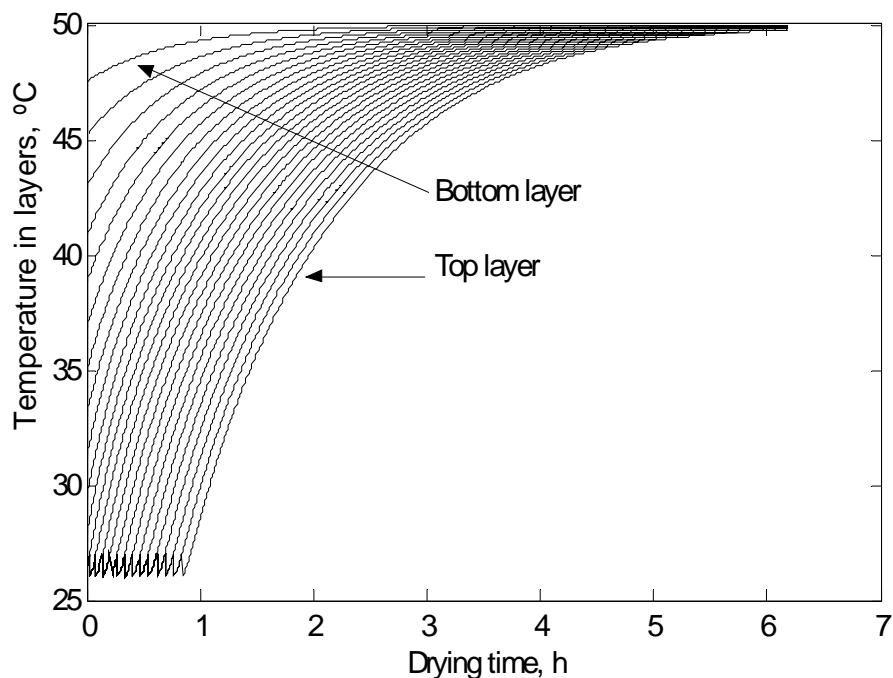


Fig. 7 The temperature of the 25 layers during the drying process of a batch of 0.20m height, dried at a temperature of 50°C, humidity ratio of 0.013 kg_{water}/kg_{air} and air velocity of 0.6 m/s.

The mass flow, temperature and humidity ratio of the drying air (just before the first layer) were controlled at their set points. As the moisture of the air increases during the pass of air through the dryer, the amounts of return and fresh air change continuously (*Fig.8*). In this figure the amounts are constant for the time that the exhaust air is saturated.

The enthalpy flows during the drying process are shown in *Fig.9* where the effects of combined humidity ratio and temperature control are given. The level of thermal enthalpy for the heater is constant till the drying starts at the top layer. Then the return enthalpy flow increases and amount of fresh air decreases. The enthalpy of mixed air is the set energy for drying and it is the sum of the inlet air, return and thermal enthalpies. The total energy that is used for the cost calculations is the accumulation of the thermal enthalpy for each process.

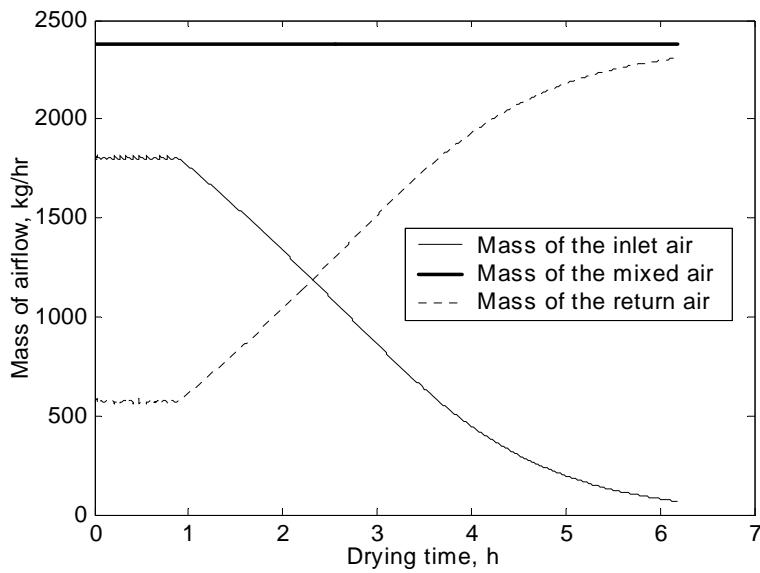


Fig. 8 Effect of humidity control. Mass of airflow (inlet, mixed and return) during the drying process at 50°C, humidity ratio of 0.013 kg_{water}/kg_{air} and air velocity of 0.6 m/s for a batch of 0.20m height.

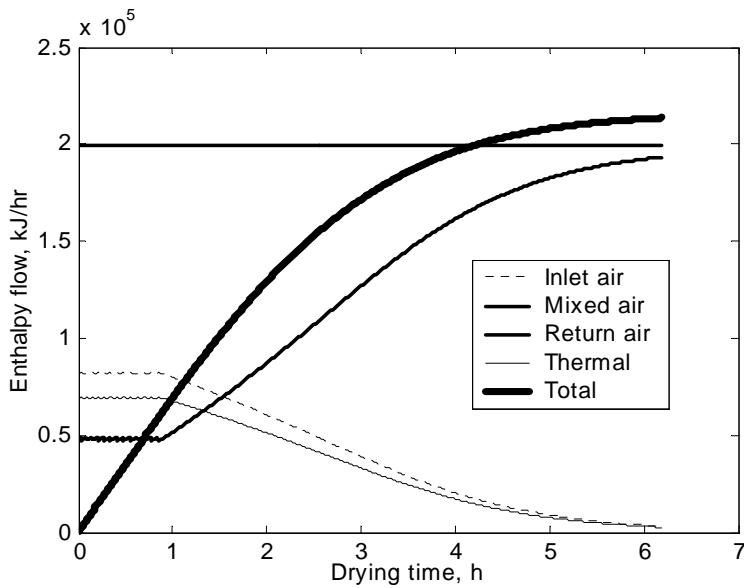


Fig. 9 Combined effect of humidity and temperature control during the drying process with a bed height of 0.20m dried at a temperature of 50°C, humidity ratio of 0.013 kg_{water}/kg_{air} and air velocity of 0.6 m/s.

Fig.10 shows the energy requirement for drying as a function of inlet air humidity ratio and inlet temperature. The humidity ratio of 0.01kg_{water}/kg_{air} considered for Dutch ambient air conditions and five steps of increasing humidity ratios, indicating the rate of recirculation, were applied for temperatures ranging from 40°C to 90°C. The humidity ratio of 0.01 kg_{water}/kg_{air} represents in this way the drying conditions without recirculation of drying air. From the graph follows that with increasing value of humidity ratio the energy requirement decreases. This result is obvious because with increasing humidity ratio the recirculation flow increases and thus more energy is recycled. There is a maximum in the

curve and these operation points should be avoided when energy minimization is aimed. For tarragon leaves a maximum temperature of 50°C is acceptable and from that point of view the humidity ratio of 0.015 kg_{water}/kg_{air} should be preferred or a lower temperature for humidity ratios that are a little lower when only the energy consumption is considered.

The energy consumption for a non-recirculation system at 50°C is about 31,000 kJ/kg dry material. Drying at an input humidity ratio of 0.015 kg_{water}/kg_{air} is to be preferred and will give energy savings of 45% compared to a non-recirculation system when also the drying time is taken into consideration, which is explained in § 3.3.

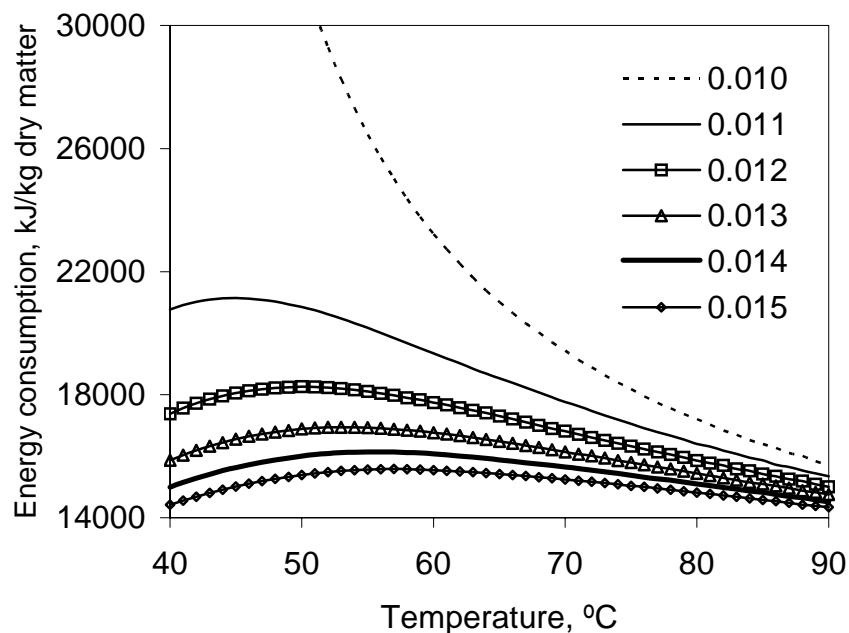


Fig. 10 Effect of drying temperature on energy consumption at different set points of humidity ratio for recirculation control with a bed height of 0.20m and air velocity of 0.5 m/s for Dutch conditions.

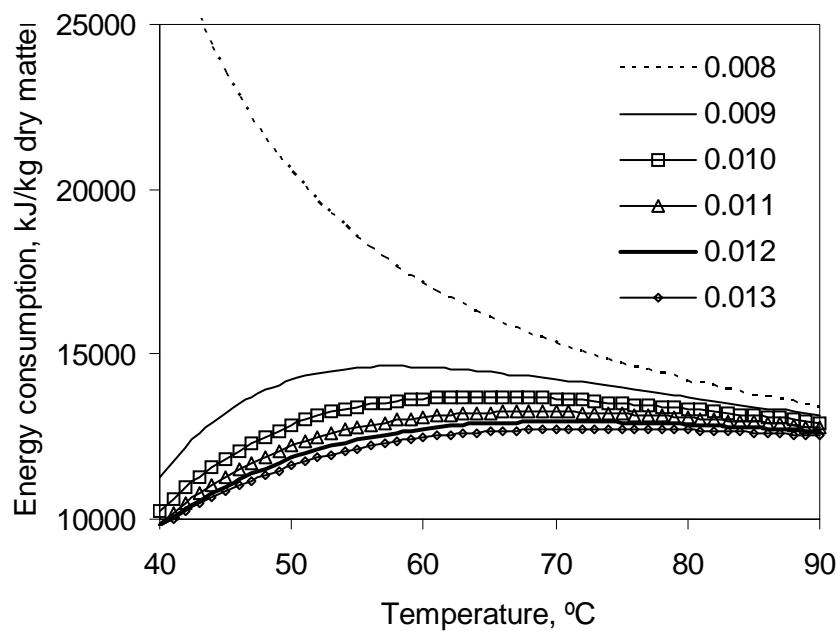


Fig. 11 Effect of drying temperature on energy consumption at different set point of humidity ratio for recirculation control with a bed height of 0.20m and air velocity of 0.5 m/s for Iran conditions.

The energy requirement for Iran conditions is shown in *Fig.11*. Here also recirculation helps to decrease the energy requirement for drying per kg dry matter. A temperature of 30°C and a humidity ratio of 0.008 kg_{water}/kg_{air} was considered for ambient air and the same ranges of temperatures and humidity ratios were applied for evaluation of energy requirement. The level of energy requirement was therefore always lower than for Dutch conditions. The humidity ratio showed almost the same affect as for the Dutch condition but at a lower level of energy consumption. The required energy per kg dry matter was minimum at 40°C and maximum around 60°C.

3.3 Costs

The total drying costs, considered in this work consists of the items fuel, electricity, depreciation, interest and maintenance. The individual contribution of the cost items per kg dried product for temperatures of 40°C to 60°C, air velocity of 0.6 m/s and a bed height of 0.20m are shown in *Fig.12*. The left figure shows the costs for the Dutch situation and a humidity ratio of the ambient air of 0.012 kg_{water}/kg_{air}. The right figure shows the Iran situation with a humidity ratio of ambient air of 0.010 kg_{water}/kg_{air}. These two levels of humidity ratios were selected for comparison because in both cases the humidity ratio is then 0.002 kg_{water}/kg_{air} higher than the ambient air. All the cost elements are higher for the Dutch condition except the interest, which is slightly higher for Iran. The fuel cost is the major cost factor for the Dutch conditions especially for higher temperatures because the price is high. The fuel and electricity costs in Iran are much lower than in the Dutch condition.

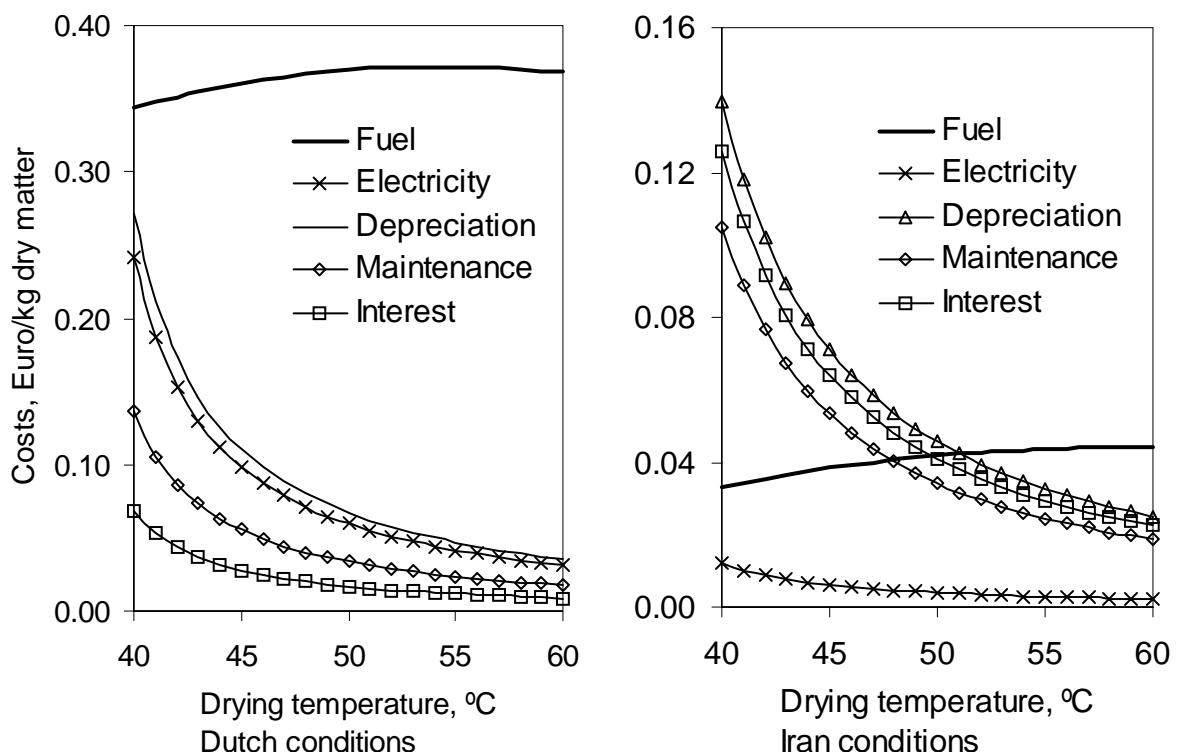


Fig. 12 Drying costs per cost item material dried at temperatures of 40°C to 60°C, air velocity of 0.5 m/s and humidity ratios 0.012 kg_{water}/kg_{air} for Dutch and 0.010 kg_{water}/kg_{air} for Iran conditions for a batch of 0.20m height.

The total drying costs calculated per unit of dry mass for the conditions in The Netherlands and Iran conditions for three humidity ratios, and for a bed height of 0.20m are shown in *Fig.13*. The ambient air conditions represent the scenarios without recirculation. The total costs decrease with temperature because of the decrease in drying time is larger than the increase in energy costs for the lower temperatures. The minimum costs therefore are found at the maximum temperature permitted for good quality, which is 50°C. The costs in The Netherlands were about three times higher compared to the costs in Iran at 50°C. For Dutch condition, recirculation decreased the total costs with almost 50% at 50°C whereas for Iran conditions there were no significant differences. The reason is the difference between the ambient air conditions and the lower energy price in Iran.

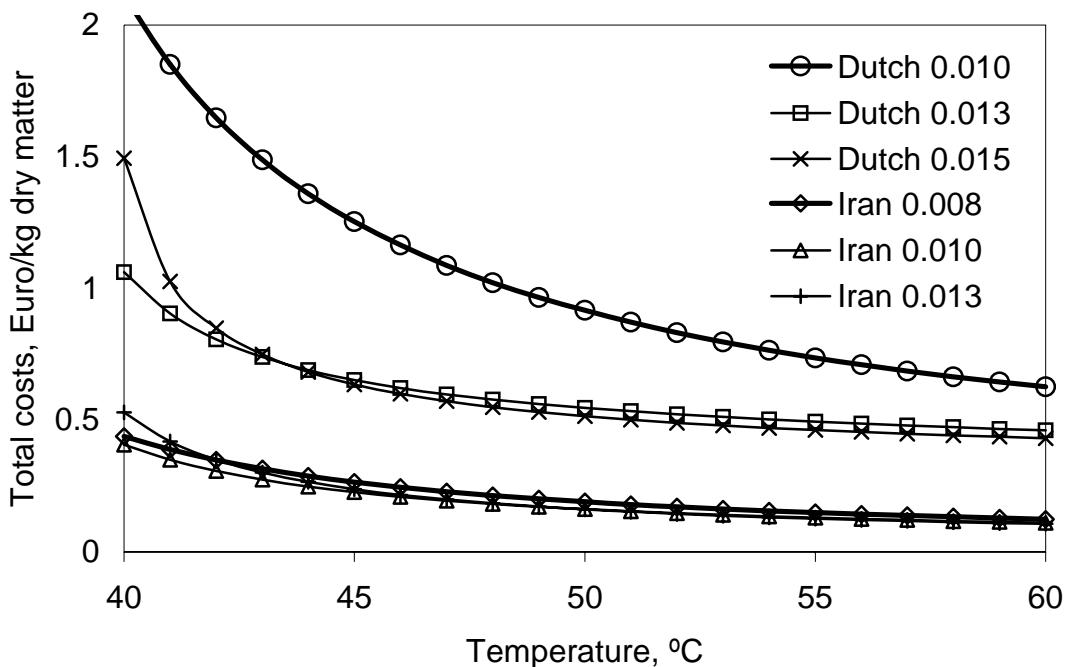


Fig. 13 Total cost of material dried at temperatures of 40°C to 60°C, three levels of humidity ratio and air velocity of 0.5 m/s for Dutch and Iran conditions for a batch of 0.20m height.

Effect of bed height on total cost per unit of dry matter is shown in *Fig.14*. Total cost is shown for the material dried at temperatures of 40°C to 60°C, humidity ratio of 0.012 kg_{water}/kg_{air}, air velocity of 0.5 m/s and three levels of bed height for Dutch conditions. The results showed that the bed height has more effect on costs at lower temperatures and smaller bed heights. The costs of a layer above 0.50m is almost the same for temperatures from 40°C to 60°C. Raising the bed height showed more decrease of the costs for the thin layers than for the thick layers. For instance the difference of the costs between the bed heights of 0.1m and 0.20m was much more than between 0.50m and 1.0m. From *Fig.14* it could be concluded that a bed height of 0.5m should be recommended, since the costs are about 30% lower than for 0.2m and drying time increases only from 6.5h to 9.5h at 50°C. The drying time depends on drying temperature, air relative humidity, air velocity and bed height. There will be a maximum acceptable time for a drying process based on mould growth on the top of the batch. Since in this research, no measurement was done on mould growth and our model is not accurate when rewetting occurs at the top layers, no recommendation can be given for the maximum drying time and so for the maximum bed height.

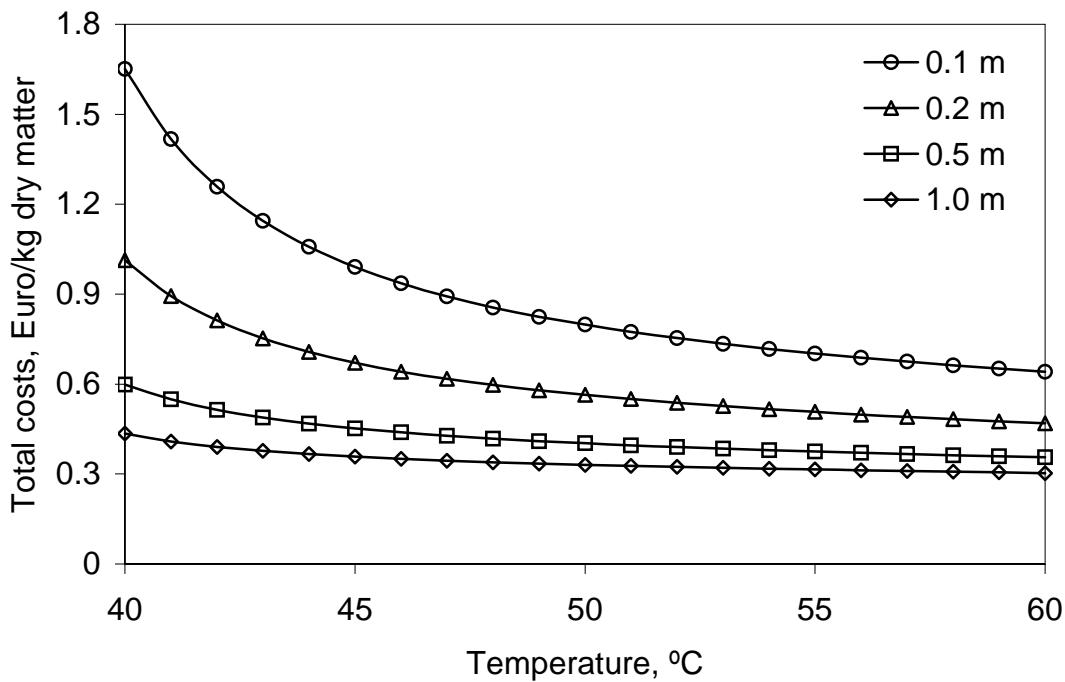


Fig. 14 Total cost of material dried at humidity ratio of $0.012 \text{ kg}_{\text{water}}/\text{kg}_{\text{air}}$, air velocity of 0.5 m/s and three levels of bed height for Dutch conditions.

4 Conclusions

This work concerns cost estimation of different scenarios for batch wise processing of tarragon leaves in a drying bed.

Thin layer drying models, which are explicit in time, were not useful for simulation of bed dryers. Therefore the drying rate is calculated from a modified Lewis model. This modified Lewis was fitted to experimental data obtained for drying experiments for tarragon leaves. The drying rate coefficients in this model are temperature dependent, but no relation with relative humidity was found.

Although the simulation model has accuracy limitations for the higher relative humidities, it gives a good indication for comparing drying scenarios.

The drying simulation showed that recirculation of the air is important to recover the energy potential in the exhaust air especially for the Dutch ambient air conditions. Despite a lower drying rate, and hence a longer drying time, the recirculation is still profitable from an energy saving point of view and cost minimization.

The drying at 50°C was found as the best option to minimize the costs and keep the quality of the product. The total costs for the product dried at 50°C were about three times higher for The Netherlands compared to Iran.

Acknowledgment

Financial support from Ministry of Science and Research and Technology of Iran is gratefully acknowledged.

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Summary in English

Summary

During the past years due to renewed interest in dishes that use a wide variety of spices the use of herbal plants has increased significantly. Correct drying of aromatic plants is necessary to achieve longer shelf life with high quality, preserving the original flavor. Drying is also needed for food safety and will result in less weight and storage space and so lower packing, storage and transportation costs.

Essential oil content and color are the most important parameters that define the quality of herbal and medicinal plants. However hot air drying affects the essential oil content and color during the drying process.

The quality of the product influences the financial output of the production chain since a better quality will be more appreciated by consumers and will result in a higher consumption and thus more income for the producers. So processing engineers need to know how they have to dry their products to obtain the best quality. In addition, energy consumption and total costs depend on the variables in the drying process. Therefore the objective of this research is to find the optimal conditions for the process with respect to quality and total costs. As different points of view are in the focus of the production chain, optimization of the total chain needs optimization of individual quality parameters and process stages.

The drying process of tarragon (*Artemisia dracunculus* L.) leaves and chopped plants was evaluated. Two varieties were tested: French Tarragon and Russian Tarragon. The drying air was heated to temperatures ranging from 40 to 90°C with three levels of relative humidity at each temperature (heated ambient air and two levels with higher relative humidity, assuming recirculation of dryer exhaust air). The airflow was 0.6 m/s for all experiments. Drying was continued until equilibrium moisture content was reached. A number of drying equations were selected from literature and fitted to the experimental data of drying of tarragon leaves and chopped plants. The "Page equation" was found as the most suitable model for drying of tarragon based on the good fit and simplicity of the equation.

Since tarragon leaves are much more important than the stems additional drying experiments were done on leaves. The changes of essential oil content as well as color of the tarragon leaves were evaluated during the drying process. For this purpose the leaves were collected while the moisture content decreased from 80% to 7% during the drying process at temperatures of 45, 60 and 90°C.

The essential oil of the fresh and dried leaves was isolated by hydro-distillation and analyzed by capillary gas chromatography and gas chromatography-mass spectrometry. During the drying process the total oil content of both varieties decreased. The decrease was highest at 60°C drying temperature. For French Tarragon the decrease of oil content was significantly lower at 90°C. The effect of relative humidities at each temperature was not significant. The main compounds were estragole in French Tarragon with 69% and sabinene in Russian Tarragon with 40% of the total oil content.

The color parameters of fresh and dried leaves were measured by a colorimeter. The individual parameters of the "L*a*b*" and "L*C*h°" color systems were evaluated and the hue (h°) value was selected as the best parameter to monitor color change of tarragon leaves during drying. Long drying times and drying at 60°C yielded the strongest decrease in color. The smallest change of color was observed at 40°C, where temperature was low enough, and also at 90°C, where drying time was short. The biggest change occurred at the temperatures of 55 to 70°C. Most of the color change happened before the material reached 35% moisture content wet basis.

The quality of the dried material can change during the storage time. To investigate this aspect the dried material was stored in airtight glass bottles, in darkness at a temperature of

5°C. Oil content and color were measured for the fresh leaves and just after drying as well as after storage of 15, 30, 60 and 120 days. The results showed a reduction of the oil content and change of color during the storage period. The biggest change of the essential oil content (about 50% after 30 days) and color was found for the material dried at 90°C. Drying at 45°C was found as the best among the treatments.

Sorption isotherms are necessary knowledge for modeling drying. Moreover they are necessary to determine the optimal moisture content after drying in order to avoid mold growth during storage. The equilibrium moisture contents of tarragon (stem and leaf separately) were determined using the saturated salt solutions method at three temperatures (25, 50 and 70°C) within a range of 5 to 90% relative humidity. Both adsorption and desorption methods were used for stem and leaf. Experimental curves of moisture sorption isotherms were fitted in different equations. The “modified Halsey” and “GAB” equations were found to be the most suitable to describe the relationship between equilibrium moisture content, relative humidity and temperature. There was no significant difference between the equilibrium moisture content of the Russian and French Tarragon.

The optimization of drying conditions for tarragon was studied by using individual results explained in the above paragraphs. Simulation models were made for the drying process, energy consumption and for cost calculation using the results of the earlier research steps like equations and parameter values. It was found that thin layer drying models, which are explicit in time, are not useable for bed simulations. To solve this problem a modification is made to the standard Lewis model to calculate the drying rate in a bed. The model was applied for the current conditions in The Netherlands and Iran. The model was run at selected ranges of temperature, humidity ratio, air velocity and various bed heights with and without recirculation of the drying air. The drying time, and thermal energy consumption was found and translated in total costs per kg dry mass. The various drying scenarios showed that recirculation of the air is important to recover the energy potential in the exhaust air, especially for Dutch drying conditions. Despite a lower drying rate, and hence a longer drying time, the recirculation is still profitable from an energy saving point of view. Drying at 50°C was found as the best option to minimize total costs and keep the quality of the products at an acceptable level. The total costs for the product dried at 50°C were about three times more expensive for The Netherlands compared to Iran. The recirculation is less important in Iran because the ambient air has a higher drying potential and energy prices are lower.

Summary in Dutch

Samenvatting

Dragon (*Artemesia dracunculus* L.) is een veelgebruikt culinair en medicinaal kruid. Om bewaren over langere tijd mogelijk te maken wordt het gewas gedroogd. Dit heeft ook als voordeel dat de transport- en opslagkosten dalen. Om bij de bewaring de oorspronkelijke kwaliteit zo goed mogelijk te behouden moet het op de goede manier gedroogd worden. Oliegehalte, als parameter voor de geur, en kleur zijn de belangrijkste kwaliteitsparameters. Drogen met warme lucht is de meest toegepaste droogmethode. De condities tijdens het droogproces bepalen niet alleen de kleur en het oliegehalte maar ook de kosten en de energieconsumptie. Om de opbrengsten in de productieketen te maximaliseren moeten procesingenieurs en boeren dus weten hoe ze moeten drogen. Het doel van dit werk is te onderzoeken wat de beste condities voor het drogen van dragon zijn in verband met de kwaliteit, kosten en energieconsumptie. Hiertoe zijn verschillende experimenten uitgevoerd.

Er zijn twee variëteiten dragon onderzocht. De z.g. Franse dragon is de culinaire soort, vormt geen zaad en wordt dus vermeerderd door plantenstekken. De Russische dragon wordt door zaad vermeerderd en is vooral van belang voor medisch gebruik.

Het droogverloop van dragonblaadjes en gehakselde hele planten is afzonderlijk bepaald in een dunne-laag-droger bij luchttemperaturen van 40-90°C en bij drie niveaus van luchtvochtigheid (opgewarmde omgevingslucht en twee niveaus hoger vergelijkbaar met recirculatie van de vochtige drooglucht). De luchtsnelheid was steeds 0.6 m/s en het drogen werd voortgezet tot het evenwichtsvochtgehalte was bereikt. Uit de bekende vergelijkingen uit de literatuur om het droogverloop in de tijd te beschrijven is de beste geselecteerd. De “Page vergelijking” gaf de beste beschrijving met het geringste aantal parameters.

Om de invloed van de temperatuur en luchtvochtigheid van de drooglucht op de kwaliteit van de dragon vast te stellen is het oliegehalte en de kleur is vastgesteld. Hiertoe werden tijdens het drogen met temperaturen van 45, 60 en 90°C monsters verzameld van de dragon bij productvochtgehaltes tussen 80% en 7%. Van de verse en de droge dragon werd de olie geïsoleerd door hydrodestillatie en geanalyseerd door capillaire gaschromatografie en gaschromatografie-massa spectrometrie. Het bleek dat tijdens het drogen de oliegehaltes van beide variëteiten daalden en wel het meeste bij 60°C. Voor Franse dragon was de daling bij 90°C significant lager. De invloed van de relatieve luchtvochtigheid van de drooglucht was niet significant. De belangrijkste oliecomponent was in Franse dragon estragole met 69% en in Russische dragon sabinene met 40% van de totale hoeveelheid olie in de monsters.

De kleur van de monsters werd gemeten met een colorimeter. Van de “L*a*b*” en “L*C*h°” kleursystemen werden de individuele parameters vergeleken en de hue ($h°$) waarde bleek de beste om de verandering van de kleur van dragon tijdens het drogen te beschrijven. Langdurig drogen en drogen bij 60°C gaf de grootste kleurverandering. De minste verandering trad op bij 40°C vanwege de lage temperatuur en bij 90°C vanwege de korte tijd die voor het drogen nodig is. De grootste kleurverandering trad op voordat de monsters een vochtgehalte van 35% hebben bereikt.

De kwaliteit van de dragon daalde ook tijdens de bewaring na het drogen. Om dit effect te onderzoeken werden monsters bewaard in luchtdichte flessen, in het donker bij 5°C. Van verse dragon en direct na het drogen alsmede na 15, 30, 60 en 120 dagen bewaring werden de oliegehaltes en de kleur gemeten. Het bleek dat de oliegehaltes en de kleur tijdens bewaring achteruit gingen en wel het meest bij dragon die bij 90°C was gedroogd. De daling van het oliegehalte was hierbij 50% na 30 dagen. Het materiaal gedroogd bij 45°C veranderde het minst tijdens de bewaring.

Adsorptie en desorptie experimenten zijn uitgevoerd om de evenwichtsvuchtgehaltes te bepalen bij temperaturen tussen 25°C en 70°C en luchtvochtigheden tussen 5% en 90%. Deze informatie is nodig om een model van het drogen te maken en om vast te stellen tot welke vuchtgehaltes gedroogd moet om schimmelvorming tijdens de bewaring te voorkomen. Monsters van dragon zijn daartoe in flessen bewaard boven een verzadigde zoutoplossing waardoor luchtvochtigheden van 5 – 90 % werden gecreëerd bij drie temperaturen: 25, 50 en 70°C. Na enkele weken werd het evenwichtsvuchtgehalte gemeten en gemodelleerd. Het bleek dat de “Modified Halsey” en “GAB” vergelijkingen de beste beschrijving geven van de relatie tussen het evenwichtsvuchtgehalte, de luchtvochtigheid en de temperatuur. Er was geen verschil tussen de twee dragon variëteiten.

Met de verzamelde gegevens zijn modellen gemaakt van het droogproces van een recirculatiedroger, de energieconsumptie bij het drogen en de droogkosten. Het bleek dat de gevonden vergelijkingen die het droogproces beschrijven als functie van de tijd niet geschikt waren voor het modelleren van een droogbed. Hierom werd de “Lewis vergelijking” aangepast zodat de droogsnelheid in een droger als functie van de bedhoogte kon worden gesimuleerd. De modellen zijn toegepast op diverse scenario’s voor drogen van dragon in Nederland en Iran. Droogtemperatuur, luchtvochtigheid (mate van recirculatie), luchtsnelheid en bedhoogtes zijn gevarieerd. Het blijkt dat recirculatie van de lucht van belang is om energiegebruik en kosten te verminderen, met name in Nederland. Ondanks een lagere droogsnelheid, dus langere droogtijd is bij recirculatie van de drooglucht de energiebesparing aanzienlijk. In Iran waar de omgevingslucht droger en warmer is en de energieprijzen veel lager zijn geldt dat veel minder. Een droogtemperatuur van 50°C is het beste wat betreft kwaliteit en kosten voor dragon die moet worden bewaard. De totale droogkosten daarbij zijn in Nederland drie keer zo hoog als in Iran.