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Modeling of the Equilibrium Moisture Content (EMC) of Tarragon (*Artemisia Dracunculus L.*)

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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: Russian and French tarragon. Experimental curves of moisture sorption isotherms were fitted by modified Henderson, modified Halsey, modified Oswin, modified Chung-Pfost and GAB equations 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 Russian and French tarragon.

KEYWORDS: Absorption, Artemisia dracunculus, desorption, drying, storage, tarragon

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1 INTRODUCTION

Tarragon (*Artemisia dracunculus* L.) is an aromatic, perennial plant of the Asteraceae family. The long narrow leaves have a strong, spicy anise flavour. 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 flavour for foods, herbal vinegars, oils and mustards. At least two cultivars 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, (Simon, Chadwick and Craker 1984). 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 elemicine in Russian tarragon. (Anonymous. 1997; Yaichibe, Masanori, and Kenichi 1997)

Moisture sorption isotherm defines the relation between Equilibrium Relative Humidity (ERH) and Equilibrium Moisture Content (EMC) (Soysal and Oztekin 1999). 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 (Hamer, Knight et al. 2000).

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 (Silakul and Jindal 2002). The relative humidity in this condition is known as the Equilibrium Relative Humidity (ERH) (Soysal and Oztekin 1999). 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. 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 Russian and French 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 30 mm long pieces. The samples of 10-12 g fresh leaves and 15-20 g 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 56×36×26 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 800 ml contains 150 ml 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 (Menkov 2000a). The cryostat was adjusted to 25, 50 and 70°C. Every three days, the samples were weighed with an accuracy ± 0.1 mg. Equilibrium was acknowledged when three consecutive weight measurements showed a difference less than 1 mg.

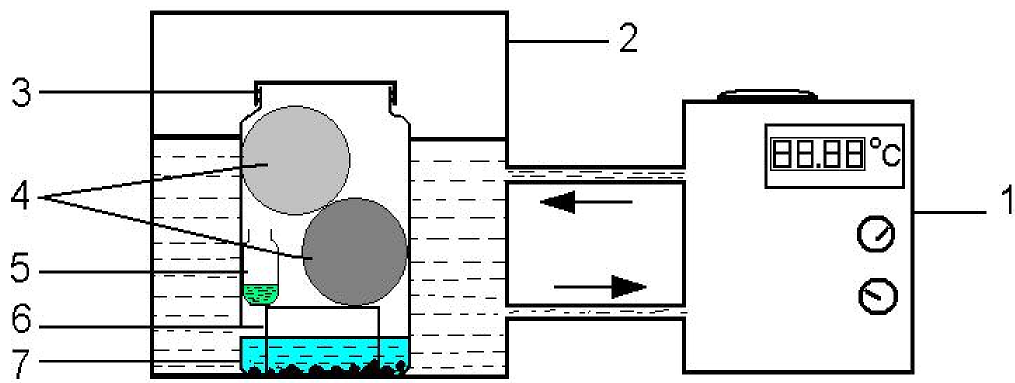

Fig. 1. Equipment for the sorption isotherm measurement: 1) cryostat; 2) water bath; 3) jar with cap; 4) netballs including samples; 5) thymol; 6) base; 7) saturated salt solution.

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 24 hr) (Park, Vohnikova, and Brod. 2002). The EMCs were determined by triplicate measurements of each sample.

2.3 Salt solution

The saturated salt solutions method is based on the fact that ERHs of specific salt solutions are a well known physical property (Greenspan 1976). 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 condensation gradient in the liquid phase, the salt solution was agitated from time to time (Kouhila et al. 2001).

Table 1 Saturated Salt solutions and Equilibrium Relative Humidity at different temperatures (Greenspan 1976); (Labuza, Kaanane, and Chen 1985).

No	Salt Formula	Equilibrium Relative Humidity (ERH) (%)		
		T=25°C	T=50°C	T=70°C
1	K2SO4	97.30	95.82	94.5
2	KNO3	93.58	84.78	n.f.*
3	KCl	84.34	81.20	79.49
4	KBr	80.89	79.02	79.07
5	NaCl	75.29	74.43	75.06
6	NaNO3	74.25	69.04	66.04
7	KI	68.86	64.49	61.93
8	NaBr	57.57	50.93	49.70
9	K2CO3	43.16	40.91	37.37
10	MgCl2	32.78	30.54	27.77
11	LiI	17.56	12.38	7.23
12	LiCl	11.30	11.10	10.7
13	ZnBr2	7.75	7.70	8.72
14	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 (Chen 2002) have been adopted by the American Society of Agricultural Engineers as standard equations for describing sorption isotherms (ASAE 2003). We transformed the equations to get EMC as dependent variable and ERH as independent variable.

Modified Henderson

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

Modified Halsey

$$EMC=\left(\frac{-\exp(C_1+C_2T\,)}{\ln(ERH)}\right)^{1/C_3} \tag{2}$$

Modified Oswin

$$EMC=(C_1+C_2T\,)\left(\frac{ERH}{1-ERH}\right)^{1/C_3} \tag{3}$$

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

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

EMC (dry basis) and ERH are given dimensionless. C1, C2 and C3 are the equation coefficients and T is temperature in °C. The parameters C2 and C3 in the GAB equation are correlated with temperature using the following equations (Lahsasni, Kouhila, and Mahrouz 2004):

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

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

C4, C5, C6 and C7 are coefficients and Ta 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 is evaluated by using residual sum of square (RSS), standard error estimation (SEE) and mean relative deviation (MRD) (Sun 1999). 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.:

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

The smaller the values of these statistical parameters 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 (Chen and Morey 1989).

3 RESULTS AND DISCUSSION

3.1 Experimental results

Leaves of French 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.

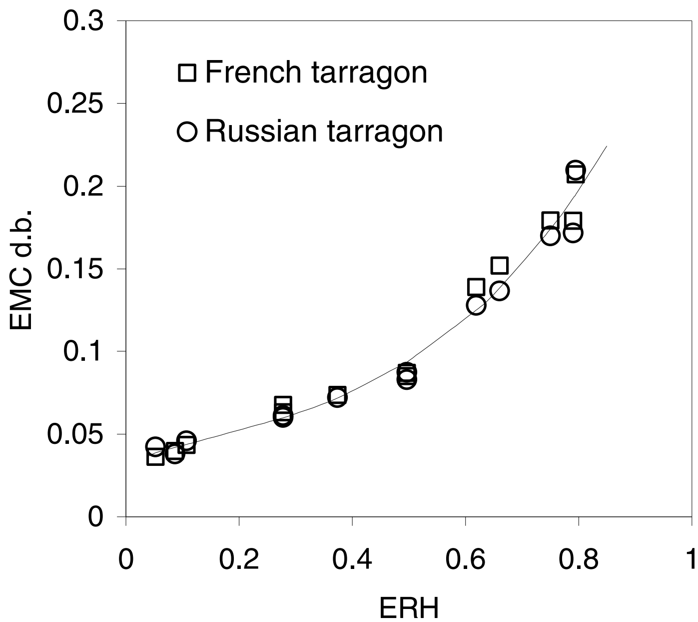

Fig. 2. Desorption data of the French and Russian tarragon leaves at 70 °C and fitted isotherm of the Halsey equation.

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 25ºC and also around 50% relative humidity at 70ºC.

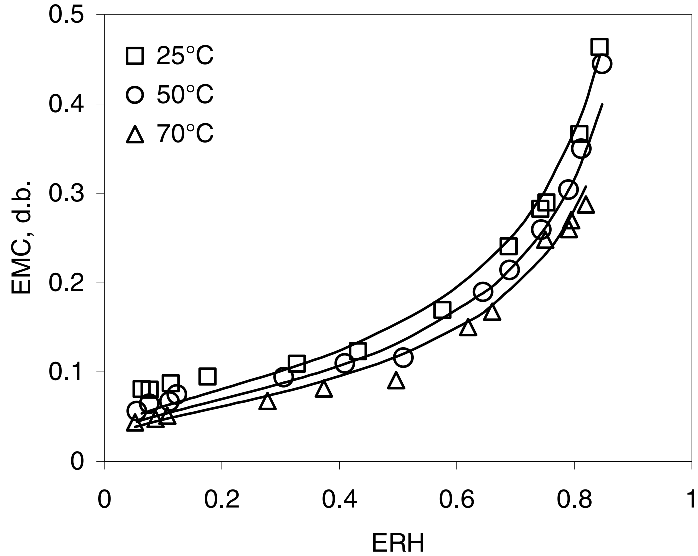

Fig. 3. Desorption isotherm data of tarragon leaves at 25, 50 and 70 °C and fitted curves of the Halsey equation.

Desorption data and the fitted modified Halsey equation for tarragon stems at 25, 50 and 70°C are shown in Fig.4. Again typical sigmoid 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.

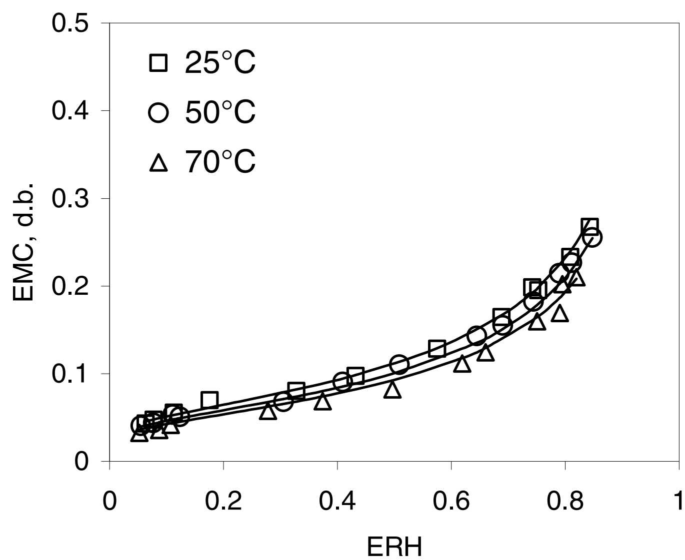

Fig. 4. Desorption isotherm data of tarragon at 25, 50 and 70 °C and fitted curves of the Halsey equation.

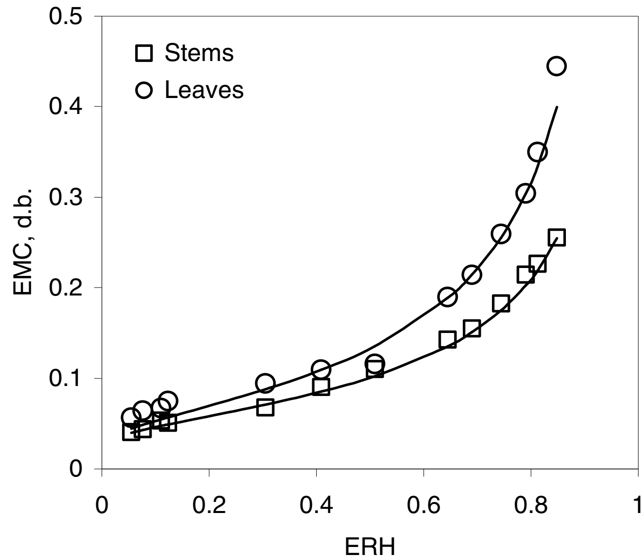

Fig. 5. Desorption isotherm data of tarragon stems and leaves at 50 °C and fitted curves of the Halsey equation.

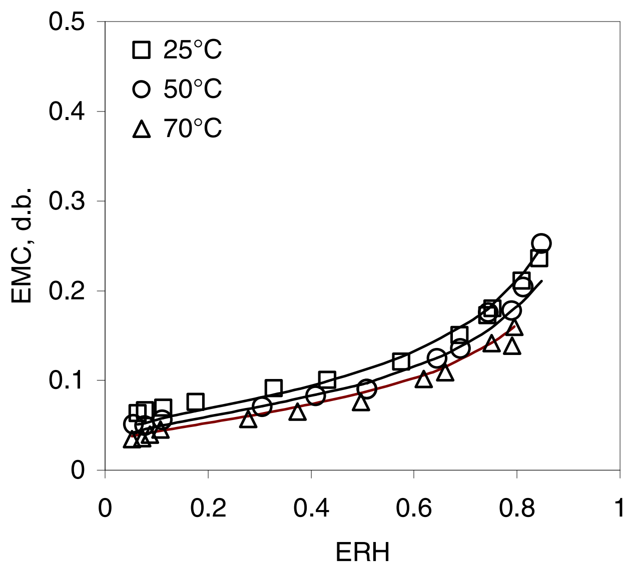

Fig. 6. Adsorption isotherm data of tarragon leaves at 25, 50 and 70 °C and fitted curves of the Halsey equation.

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 70ºC. Again fitting was suboptimal for low ERH at 25ºC. 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.

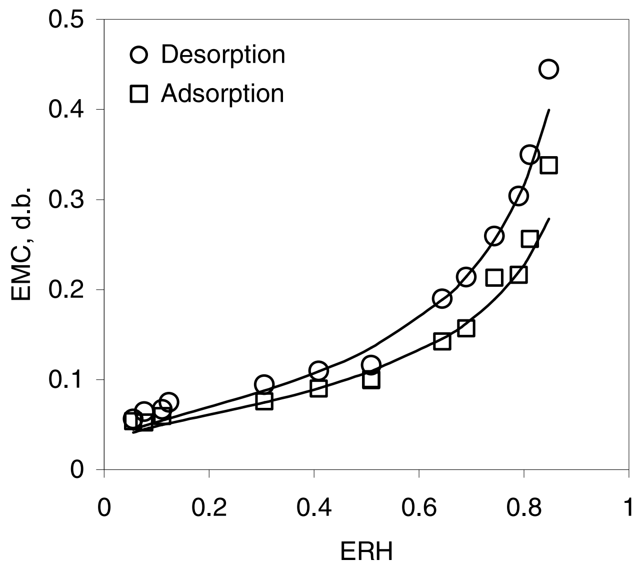

Fig. 7. Adsorption and desorption isotherm data of tarragon leaves at 50 °C and fitted curves of the Halsey equation.

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. The results of the adsorption and desorption of the first four equations are shown for leaves (Table 2) and stems (Table 3). Table 4 presents the results of the adsorption and desorption data for the leaves and stems of the GAB equation.

Table 2 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).

Parameters	Estimated values and the variance of the equations and statistical 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	$-9.62 \pm 1.59\times10^{-3}$	$-8.20 \pm 1.59\times10^{-4}$	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	$-7.76\pm0.97 \times10^{-3}$	$-8.89 \pm1.61\times10^{-4}$	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 3 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	$-7.20 \pm 1.52\times10^{-3}$	$-5.63 \pm 1.19\times10^{-4}$	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	$-6.29 \pm 0.68\times10^{-3}$	$-4.70\pm0.68\times10^{-4}$	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

Table 4 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	
	Stems	Leaves	Stems	Leaves
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	$5.65 \pm 4.08\times10^{-3}$	$1.74 \pm 0.78\times10^{-3}$	$4.25 \pm 2.03\times10^{-3}$	$2.23 \pm 0.50\times10^{-3}$
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

The residuals of EMC of the selected equations at the three temperatures were plotted against the experimental data. The pattern were 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 (see Fig.9 as an example).

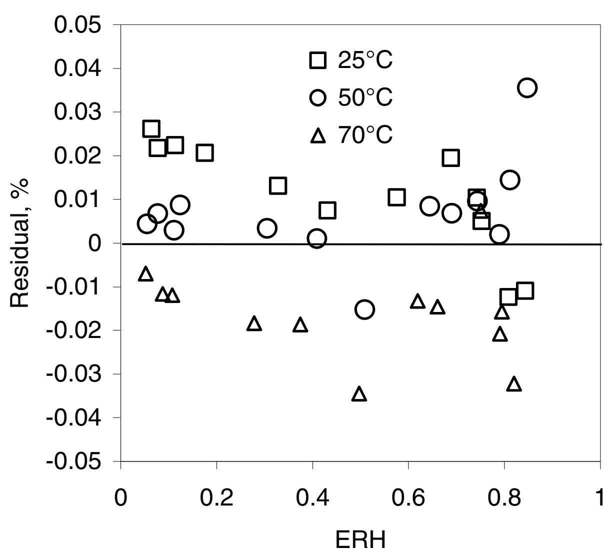

Fig. 8. Residual plot for the GAB equation for desorption data of tarragon leaves at 25, 50 and 70 °C.

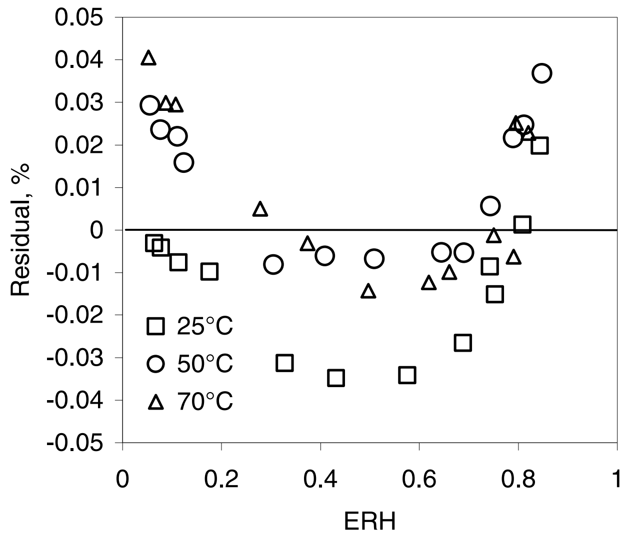

Fig. 9. Residual plot for the modified Chung-Pfost equation for desorption data of tarragon stems at 25, 50 and 70 °C.

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 (*Lippa citriodora*)(Kouhila et al. 2001) and garden mint (*Mentha crispa L.)* (Park, Vohnikova, and Brod. 2002). 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 10ºC, 0.136 at 20ºC and 0.129 at 30ºC storage temperature (Fig.10). Drying to a lower moisture content would cause additional operation costs and mass losses without increasing storage safety.

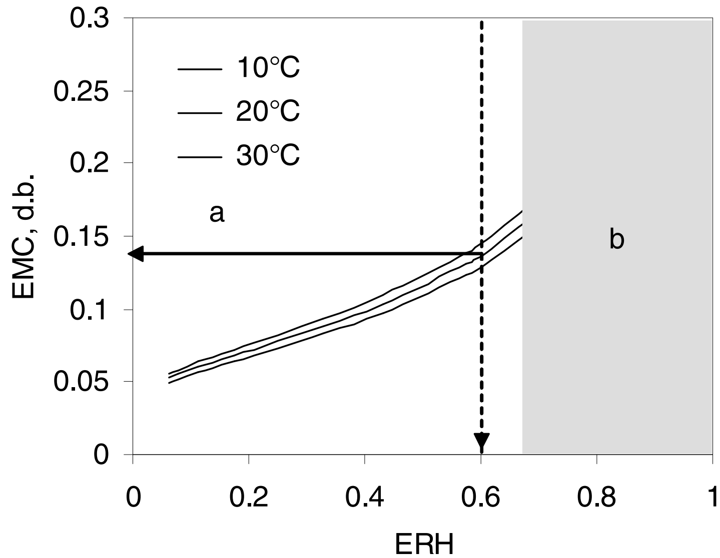

Fig. 10. EMC domain for storage of dried tarragon a) optimum for storage b) mould growth.

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 of 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 (Yang and Siebenmorgen 2003).

Since the variance of the coefficient C7 in the GAB equation was too wide in the first calculations, this parameter was fixed at a value of C7=25000. The value is at the same level as given in literature for other agricultural products (Menkov 2000a; 2000b).

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 equations 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 the 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.

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