Effect of air temperature and relative humidity on the thin-layer drying of celery leaves (*Apium graveolens* var. *secalinum*)

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Abstract: The thin-layer drying of celery leaves was studied under different conditions of air temperature (20-50℃) and relative humidity (10%-60%) in a through-flow laboratory dryer. Both parameters influenced the drying time, although the effect of air temperature was more pronounced. The effect of air relative humidity was practically negligible at 50℃. The experimental data was fitted to six thin-layer drying models and their goodness of fit was tested, being the Two-term exponential model the one that showed the best fit in the majority of treatments. The relationship between drying conditions and regression parameters of this model was analyzed to include it in the model. Parameter *a* had a negligible effect on the drying curves and was set constant. For parameter *k* a piecewise function was used in two parts, one for the temperatures between 20 and 40℃ and the other for 40 to 50℃, resulting in a good fit overall. The color of the dried leaves did not appreciably change at temperatures between 20 and 40℃, except at very high levels of relative humidity, which should be avoided when air recirculation is used. At 50℃ color was negatively affected.

Keywords: celery leaves, thin-layer drying, modeling, *Apium graveolens*, non-linear regression

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1 Introduction

Celery (*Apium graveolens* L.) is a kind of plant species of the family Apiaceae. It is one of the oldest cultivated plants to be used for medicinal and dietary purposes. Today it is mostly used as food and condiment (Mielke and Schöber-Butin, 2007). Leaf celery (*Apium graveolens* L. var. *secalinum*), also known as cutting celery, is a variety in which the usable parts are the dark-green, glossy leaves on long, thin leaf petioles, presenting a strong celery flavor. They may be eaten fresh or processed, mainly frozen or dried, with which their aroma is not lost (Mielke and Schöber-Butin, 2007; Rozek, 2007). Although not as well known as celeriac (*Apium graveolens* L. var. *rapaceum*), it is an economically important herb in Germany, with 136 ha of cultivated area in 2003 (Hoppe, 2005).

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In general leaves require less time and energy for drying than other parts of plants, which makes celery leaves more suitable for the drying process compared to the stalk or root parts commonly used in the other varieties.

The thin-layer drying behaviour of many medicinal plants and spices has been studied. No literature is however found for celery leaves. Moreover, the majority of studies have considered only temperature as a drying parameter. Only a few have studied the effect of air relative humidity, mostly within small ranges and at relatively high temperatures, finding in most cases a negligible effect of this parameter (Hosseini, 2005; Madamba et al., 1996; Phupaichitkun, 2008).

The objective of this study was to determine the effect of air temperature and relative humidity on the thin-layer drying of celery leaves; find a best mathematical model which includes these effects and determine the difference in color between treatments.

2 Materials and methods

2.1 Plant Material

Leaf celery (*Apium graveolens* var. *secalinum*) was sowed in pots in mid March and kept in a greenhouse until late May, when the plants were moved into the research field of the Agricultural Engineering Department of Kassel University in Witzenhausen in the year 2009. Plants were harvested manually and leaves were separated immediately before each trial.

2.2 Drying apparatus

The laboratory dryer consisted of a drying chamber, an axial fan and an air preconditioning chamber. The drying chamber contained two trays whose bottoms where made of metal mesh to allow air to flow vertically through the product. Each tray had dimensions 0.25 m × 0.16 m and rested over air channels with two perforated plates placed in series to improve the air distribution. Air flow delivered by the fan was controlled using a laboratory power supply and an Airflow TA-5 hot-wire anemometer. The heating system in the preconditioning chamber allowed the air temperature to be fixed at the desired level, whereas the air relative humidity was increased when needed using an air humidifier. The humidity was controlled using a relative humidity regulator and measured with an Ahlborn Therm 2286-2 electronic psychrometer.

2.3 Experimental procedure

Drying experiments were carried out at different combinations of air temperature and relative humidity as shown in Table 1. Since the drying apparatus did not incorporate an air dehumidifier, the treatments carried out were limited by the normal ambient conditions in Germany.

Table 1 Thin-layer drying treatments

Temperature/℃	20	30	40	50
Relative humidity/%			15	10
		30	30	30
	45	45	45	45
			60	

Air speed was set to 0.2 m/s. The drying system was set to the experimental conditions and left running empty for at least one hour so that it could reach stable conditions in the drying chamber. Trays were loaded with 50 g of fresh celery leaves, initially making a layer 4 to 5 cm thick. The samples were weighted at variable intervals, more often at the beginning and at the end of the trials than in the middle, and depending on the drying temperature. The weighing was done in a Sartorius E2000D electronic scale with a resolution of 0.01 g. The trials were stopped when the sample mass did not change anymore. The dry weight of the samples was determined using the oven method (105℃ for 24 h), from which the moisture content at every weighing time was derived. Two replications were carried out for each treatment.

At the end of each trial the color of a sample of ten leaves was measured with a colorimeter Konica Minolta CR-400, using the L^*C^*h color space, where L^* is the lightness from 0 (black) to 100 (white); h is the hue angle, where 0° corresponds to the positive a-axis of the CIELAB color space (red), 90° to the positive b-axis (yellow), 180° to the negative a-axis (green) and 270° to the negative b-axis (blue); and C^* is the chroma or saturation from 0 to 60.

2.4 Model fitting

Six equations widely used to describe the thin layer drying behavior of food products were fitted to the experimental data of each individual trial using non-linear regression (Table 2).

Table 2 Thin-layer drying models fitted to the experimental data

Model	Equation	Number of parameters	Source
Lewis	$MR=\exp(-kt)$	1	Lewis (1921)
Page	$MR=\exp(-kt^n)$	2	Brooker et al. (1992)
Modified Page	$MR=\exp[-(kt)^n]$	2	Panchariya et al. (2002)
Henderson and Pabis	$MR=a\exp(-kt)$	2	Wang et al. (2007)
Two-term exponential	$MR=a\exp(-kt)+(1-a)\exp(-kat)$	2	Phupaichitkun (2008)
Logarithmic	$MR=a\exp(-kt)+c$	3	Yaldiz et al. (2001)

Note: where MR is the moisture ratio; t is time; and a, k, n and c are the models' parameters.

The dimensionless moisture ratio, MR, was obtained using the following formula:

$$MR = \frac{M_t - M_{eq}}{M_o - M_{eq}}$$

Where M_t is the moisture content at time t; M_o is the initial moisture content, and M_{eq} is the equilibrium moisture content at the respective drying conditions, all in dry basis. The equilibrium moisture content at the different drying conditions was estimated from the modified Peleg equation (Román and Hensel, 2010):

$$M_{eq}=(0.5006+0.0052T)RH^{4.5595}+(0.1299-0.0013T)RH^{0.2102}$$

For the treatment at 20℃ in this study, the equilibrium moisture content was extrapolated.

The fitting was done using the non-linear regression function of SPSS 18. The coefficient of determination (R^2) and the standard error of estimate (SEE) were used to evaluate the goodness of fit of the equations. The equation for the SEE is as follows:

$$\text{SEE} = \sqrt{\frac{\sum_{i=1}^{e}(M - \hat{M})^2}{e - p}}$$

Where e is the number of experimental points and p the number of regression parameters.

The best model was chosen for further analysis, which attempted to include in the model the relationship between the drying variables studied and the regression parameters.

3 Results and discussion

Figure 1 presents the drying curves of celery leaves at the following combinations of air temperature and relative humidity: 20℃ and 45%, 30℃ and 30%, 40℃ and 15%, and 50℃ and 10%, all of which fall into an absolute humidity range between 6.5 to 8.0 g/kg of dry air.

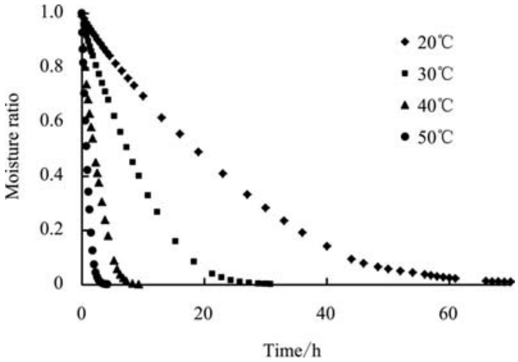

Figure 1 Moisture ratio versus time at different temperatures

The effect of drying air temperature is obvious, especially at low temperatures. Its relationship with drying time is not linear: higher temperatures translate into decreasingly shorter drying times. Although Figure 1 shows the drying curves until M_{eq} is reached, this equilibrium varies for each treatment, being lower as temperature increases. Since overdrying of food products should be avoided and a safe moisture content of 12% dry basis is to be aimed for celery leaves (Román and Hensel, 2010), Figure 2 shows drying times to reach this value (horizontal line).

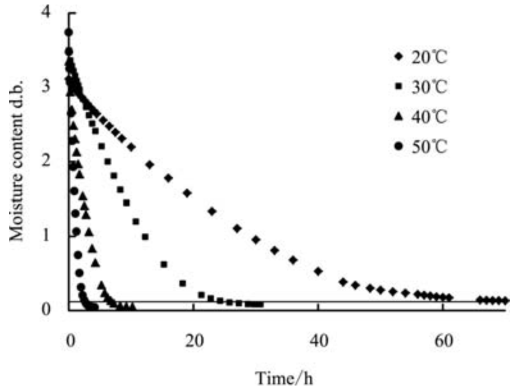

Figure 2 Moisture content versus time at different temperatures

Figure 3 shows the effect of air relative humidity on the drying curves at 30, 40 and 50℃. At 30 and 40℃ this effect seems to be important throughout the range of relative humidity considered. At 30℃ the effect on absolute drying time is much higher than at 40℃, which was to be expected since it is in proportion to its scale. It must be noticed that at 40℃ and 60% relative humidity, the safe moisture content of 12% d.b. was not reached since the equilibrium moisture content for these conditions is 13.8% d.b. At 50℃ the effect of air relative humidity is barely noticeable.

Müller (1992) reported an almost linear increase in the drying time of common sage at 50℃ when the air relative humidity increased from 10% to 50%, followed by an exponential increase from 50% onwards. In the same work he reported a very small influence in the drying of chamomile flowers at 60℃ when relative humidity increased from 10% to 40% and from 50%

onwards the drying time was more strongly affected. Phupaichitkun (2008) reported a negligible influence of this parameter on drying rate of longan fruits, however the range of humidity studied was only from 4% to 20% and at 80℃. Hosseini (2005) studied the thin-layer drying behaviour of tarragon and concluded that the relative humidity range applied did not have a significant effect in the drying rate. However his study was done at temperatures from 40 to 90℃, and the relative humidity level within each temperature was varied in up to 15 percentage points, which could explain their conclusion. Although more information is needed, it could be hypothesized that in general, as air temperature increases, the air relative humidity plays a less important role in the drying of celery leaves and other agricultural products.

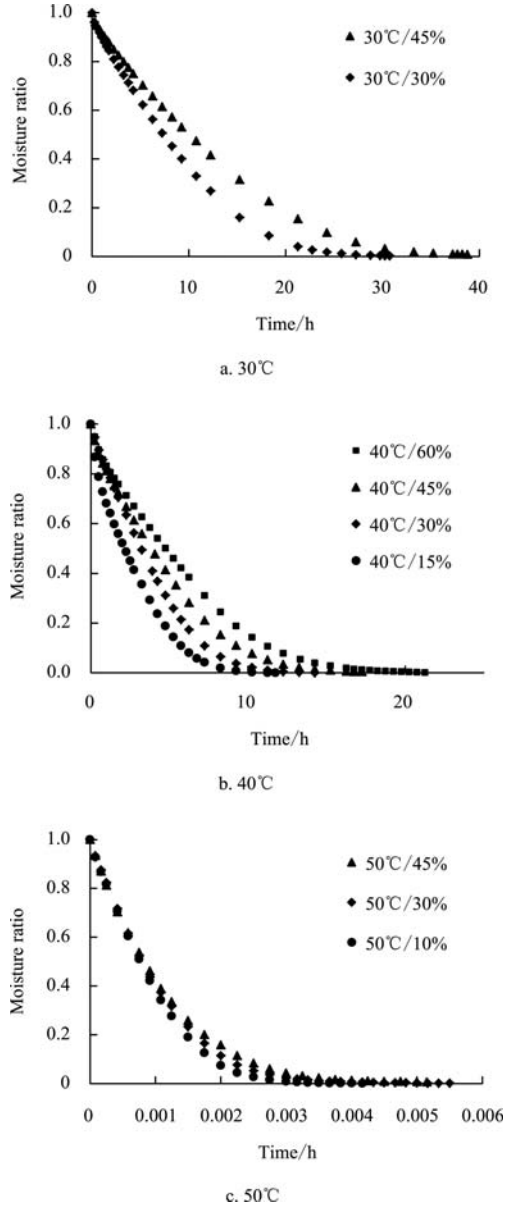

Figure 3 Effect of air relative humidity on drying curves at 30℃, 40℃, and 50℃

3.1 Model fitting

The results of the non-linear regression procedure show that the logarithmic model, although providing a very good fit with the data as determined by both R^2 and *SEE*, resulted for all trials in negative predicted values of *MR* for the last stage of the drying experiments. This is due to its third parameter, which allows the *MR* to acquire negative values, unlike the other five equations. Thus, the logarithmic model was not considered for further analysis.

Table 3 presents a summary of the regression results, showing the number of times a given equation had the highest R^2 or lowest *SEE*.

Table 3 Comparison of the fitted models by their best fit frequency

	Lewis	Page	Modified page	Henderson and pabis	Two-term exponential
R^2	0	6	6	0	13
SEE	0	6	6	0	13

It is obvious from both parameters, that the Two-Term Exponential model offered the best fit for a higher number of trials. The Page and Modified Page were the next best models. It was also noticed that the Page and the Modified Page models resulted for all trials in exactly the same predicted values, and hence in exactly the same goodness of fit, which means that this modification to the Page model is unnecessary. This is further confirmed from the results by Phupaichitkun (2008) and Ojediran and Raji (2010), who also fitted both versions of the model.

Tables 4 and 5 show the average regression and goodness of fit parameters for the Page and Two-Term Exponential models respectively.

Table 4 Regression results for the Page model

		k	*n*	R^2	*SEE*
20℃	45%	0.02347	1.19594	0.9970	0.02162
30℃	30%	0.07070	1.18231	0.9960	0.02398
	45%	0.05604	1.15683	0.9962	0.02347
40℃	15%	0.31722	1.11485	0.9934	0.02633
	30%	0.15213	1.30284	0.9970	0.01976
	45%	0.13092	1.22948	0.9958	0.02359
	60%	0.13501	1.12896	0.9942	0.02667
50℃	10%	1.02684	1.24092	0.9985	0.01402
	30%	0.98552	1.18408	0.9991	0.00994
	45%	0.91445	1.08411	0.9995	0.00735

Table 5 Regression results for the Two-term exponential model

		a	*k*	R^2	*SEE*
20℃	45%	1.72182	0.05971	0.9972	0.02098
30℃	30%	1.70318	0.14432	0.9964	0.02278
	45%	1.67470	0.11020	0.9967	0.02165
40℃	15%	1.57907	0.44778	0.9939	0.02561
	30%	1.82259	0.34260	0.9961	0.02230
	45%	1.73605	0.25839	0.9955	0.02445
	60%	1.62397	0.21904	0.9949	0.02501
50℃	10%	1.77367	1.45204	0.9983	0.01473
	30%	1.71451	1.35235	0.9992	0.00963
	45%	1.55313	1.13916	0.9997	0.00584

Although as already mentioned the Two-Term Exponential model gave the best results more often, the difference to the Page model is minor and therefore their predicted drying curves are very close to one another. Since both models have two regression parameters, the Two-Term Exponential model (Table 2) was chosen to try to incorporate the effect of the studied drying variables into a single equation. Figure 4 shows the relationship between the regression parameters for this model and the drying variables.

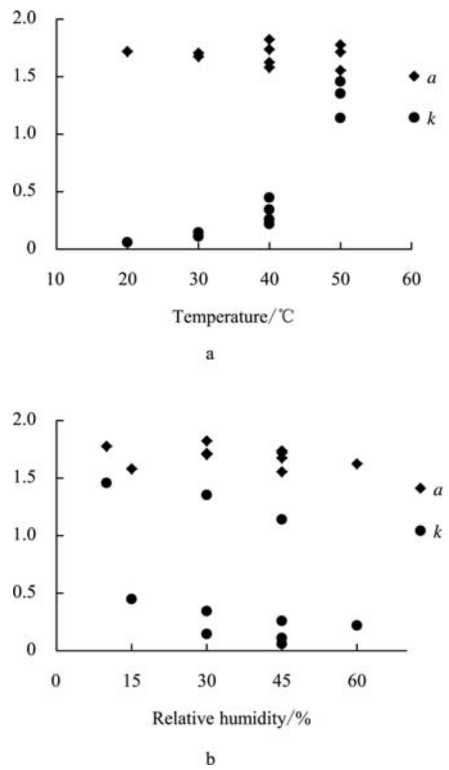

Figure 4 Plots of the Two-term exponential model parameters versus temperature (a) and relative humidity (b)

k shows a clear and strong relationship to temperature and to a lesser extent to relative humidity, while *a* does not. The value that *a* takes varies between 1.55 and 1.82, which has little effect on the drying curves. Using a parameter *a* related linearly to temperature and relative humidity showed only a small improvement compared to a fixed, average value, which was evident from a very low R^2 of 0.119, while introducing two more parameters to the model. Therefore it was decided to use a fixed parameter *a* equal to 1.69027, average of the values in Table 5.

For *k* as function of temperature and relative humidity several equations were tried. A quadratic function of relative humidity and exponential function of temperature resulted in a coefficient of determination of 0.995. However, it was seen that qualitatively the resulting general equation was not satisfactory at describing the drying behaviour at some drying conditions within their studied range. It was thus tried to apply a piecewise function, one part for temperatures between 20 and 40℃ and the second for 40 to 50℃. This solution had a good fit with the data throughout the studied range. The result was as follows:

$$k=0.06949RH^{-0.49763}\exp(0.08064T) \text{ if } 20℃\leq T\leq 40℃ \quad (R^2 = 0.994)$$

and

$$k=0.3986-0.0111RH+7.283\times10^{-5}RH^2+2.399\times10^{-4}\exp(0.1696T) \text{ if } 40℃<T\leq 50℃ \quad (R^2 = 0.996)$$

3.2 Color measurement

Figure 5a shows the effect of drying air temperature on *L**, *C** and h while keeping the absolute humidity of ambient conditions. The average values for fresh leaves are shown as dashed grey lines. For 20 to 40℃ there is an increase in the *L** and *C** values while for 50℃ these values are reduced with respect to fresh leaves. The hue angle decreased equally in all cases. Visually, samples at 20 through 40℃ showed an acceptable vivid green color, barely differentiable from each other. At 50℃, although the hue angle did not change, the color was generally negatively affected being a duller, darker green (lower *C**), while some leaves presented areas with light browning (Figure 6).

At 40℃ relative humidity showed no effect on color up to 45% (Figure 5b). At 60%, although the background color of the leaves remained, small dark areas appeared, reducing the product quality (Figure 6). This sets a quality limit to the degree of drying air recirculation. From the economic and drying time point of view however, this limit is lower, due to the only marginal energy consumption reductions at the expense of drying time (Heindl and Müller, 1997).

Finally, for treatments at 50℃ (Figure 5c) the value of *C** was reduced from the range of 25.8-31.0 for all other treatments to about 21.0, which was also visually evident. An air relative humidity of 45% drastically damaged the color by reducing the hue angle to an average of 108.85°, whereas the value for all other treatments ranged from 118.75° to 121.91°. This means a shift into the yellow region of the color space, which appreciably reduces the quality of the final product.

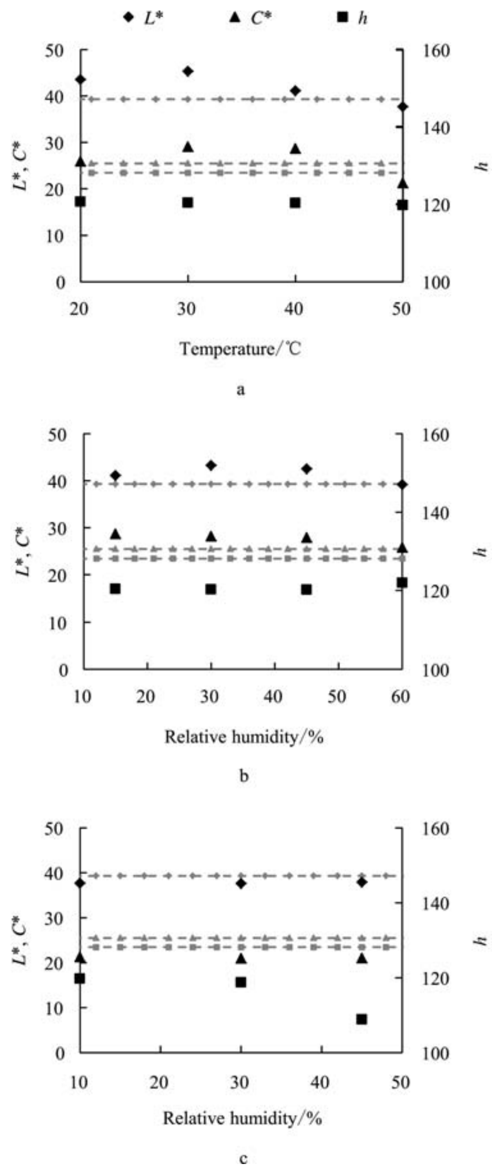

Figure 5 Color parameters of dried leaves

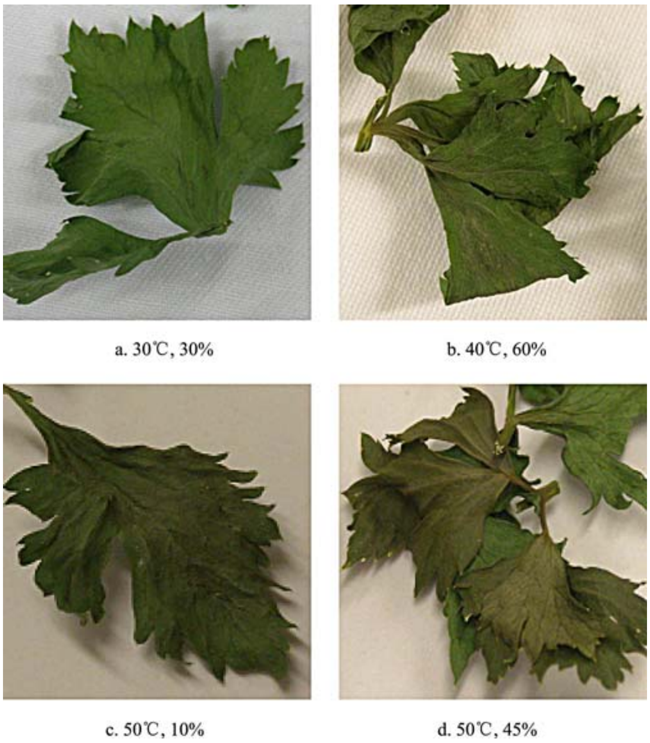

Figure 6 Celery leaves after drying at different air temperature and relative humidity

4 Conclusions

The thin-layer drying experiments on celery leaves showed a significant effect of air temperature at the low to moderate levels studied, resulting in considerable drying time reductions. An important effect of air relative humidity was also observed up to 40℃. At 50℃ this effect was negligible.

The Two-Term Exponential model fitted the experimental data best. The dependency of its parameters on air temperature and relative humidity was studied and parameter *a* was set constant due to its small range of variation between treatments and its negligible effect on the drying curves. For parameter *k* a piecewise function of air temperature and relative humidity was employed, resulting in a good fit.

The Modified Page model resulted in the same predicted values as the original Page equation, and thus only one of them needed to be fitted to data.

Color measurements also showed an effect of air temperature and relative humidity on the color of dried celery leaves. Temperatures of 50℃ and above would damage color and reduce the acceptability of the product. Moreover, at temperatures below 50℃ the mixing of fresh and recirculated air should be carefully controlled to avoid an excessive relative humidity, which also affects color negatively.

Nomenclature

a, c, k, n	regression parameters of the models
C^*	chroma in L^*C^*h color space (dimensionless)
e	number of experimental points
h	hue angle in L^*C^*h color space (dimensionless)
L^*	lightness in L^*C^*h color space (dimensionless)
M_{eq}	equilibrium moisture content (kg/kg, dry basis)
M_o	initial moisture content (kg/kg, dry basis)
MR	moisture ratio (dimensionless)
M_t	moisture content at time *t* (kg/kg, dry basis)
p	number of parameters of the models
RH	air relative humidity (%)
t	time (h)
T	air temperature (℃)

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