ABSTRACT: The objectives of this work were to determine the effect of the drying air temperature and fruit fragmentation on the essential oil yield of Brazilian peppertree (Schinus terebinthifolius) fruits, to model drying curves, and evaluate the energy consumption of the drying process. The study was conducted in Viçosa, MG, Brazil, in May 2018. The experiment was conducted using a completely randomized design, in a 3 x 2 factorial arrangement (3 drying air temperatures and 2 fruit fragmentation types), with three replications. Whole fruits (350 g) were dried at 50, 60, and 70 °C until the water content reached a water content of 0.11 on dry basis; 90 g of these fruits where kept in the dryer until reaching an equilibrium water content for the modeling. The data of drying were fitted to 12 mathematical models, whose performances were evaluated by the coefficient of determination, mean relative error, mean estimated error, and residue distribution. The essential oil was extracted by hydro-distillation using whole or fragmented fruits. Fruits dried at 50 °C and fragmented before extraction had higher essential oil yield. The use of air temperature of 70 °C resulted in lower drying time and energy consumption. The data fitted to the Midilli model satisfactorily, regardless of the drying air temperature.

Key words: medicinal plant, post-harvest, mathematical modeling, temperature, fruit fragmentation

Secagem e extração de óleo essencial de pimenta-rosa (Schinus terebinthifolius Raddi)

RESUMO: Objetivou-se determinar a influência da temperatura do ar de secagem e da fragmentação dos frutos no rendimento de óleo essencial de pimenta-rosa (Schinus terebinthifolius), ajustar as curvas de secagem e avaliar o consumo energético do processo. O estudo foi conduzido em Viçosa, MG, em maio de 2018. Realizou-se o experimento no delineamento inteiramente casualizado e esquema fatorial 3 x 2 (três temperaturas de secagem e dois tipos de fragmentação do fruto), com três repetições. Foram secos 350 g de frutos inteiros a 50, 60 e 70 °C até atingir teor de água de 0,11 base seca. Para modelagem, 90 g permaneceram no secador até atingir teor de água de equilíbrio. Os dados experimentais de secagem foram utilizados para ajustar 12 modelos matemáticos, cujos desempenhos foram avaliados pelo coeficiente de determinação, erro médio relativo, erro médio estimado e distribuição de resíduos. Após a secagem, extraiu-se o óleo essencial por hidrodestilação com os frutos inteiros ou triturados. Frutos secos a 50 °C e triturados antes da extração tiveram maior rendimento de óleo essencial. A temperatura de 70 °C apresentou menor tempo de secagem e consumo energético. O modelo de Midilli se ajustou satisfatoriamente aos dados, independente da temperatura de secagem.

Palavras-chave: planta medicinal, pós-colheita, modelagem matemática, temperatura, fragmentação do fruto
**Introduction**

*Schinus terebinthifolius* Raddi (Brazilian peppertree) is a plant species native to Brazil, locally known as ‘areia-vermelha’. Its fruits are rich in essential oil and phenolic compounds, and their therapeutic properties had been confirmed in several studies (Rosas et al., 2015; Dannenberg et al., 2016).

Fruits of this plant should be dried after harvest to ensure their conservation and preserve their chemical compounds. The drying, in this case, is a complex process due to the characteristics of this plant and water transport phenomena involved, which also affect the drying rate. Thus, mathematical modeling and simulations can be used to overcome this complexity and obtain adequate operational conditions (Castro et al., 2018).

This process can represent up to 40% of the energy consumption required by the industry (Mujumdar & Devahastin, 2000). Increasing the drying air temperature can affect the drying rate and time (Silva et al., 2015) and, consequently, the energy consumption and operational costs. However, a continuous high-temperature air supply can cause structural and physico-chemical changes in the plant part used (Borsato et al., 2008; Gasparin et al., 2014).

In addition, essential oil yield and chemical composition are also affected by the extraction method (Memarzadeh et al., 2015), and the plant part used (Cavalcanti et al., 2015) and fragmentation (Rosado et al., 2011). The fragmentation can increase yield of extraction of compounds of interest due to the larger contact surface area (Vinatouru, 2001).

In this context, the objectives of this study were to determine the effect of the drying air temperature and fruit fragmentation on the essential oil yield of Brazilian peppertree fruits, to model drying curves, and to evaluate the energy consumption of the drying process.

**Material and Methods**

The experiment was conducted using a completely randomized design in a 3 x 2 factorial arrangement, with three replications, totaling 18 experimental units. The factors consisted of 3 drying air temperatures (50, 60, and 70 °C) and 2 fruit fragmentation types before the extraction of the essential oil (dried whole fruits, and fragmented fruits in a knife mill after drying).

The Brazilian peppertree (*Schinus terebinthifolius* Raddi) fruits were collected in São Mateus, ES, Brazil, provided by the AgroRosa company (18°43'5''S, and 39°51'26''W, and altitude of 37 m). The samples were placed in plastic containers and taken to Viçosa, MG, Brazil, in May 2018. The fruits were cleaned, homogenized, and stored in a BOD (Biochemical Oxygen Demand) chamber at 3.5 °C until the drying. The initial content of water in fruit was 0.62 on dry basis (d.b.), which was determined using 10 g of fruits dried in a forced-air circulation oven at 105 ± 2 °C until constant weight (ANVISA, 2010).

The fruits were dried at the Medicinal Plant Drying Laboratory of the Universidade Federal de Viçosa, in Viçosa, MG, Brazil; 350 g of fresh whole fruits were subjected to continuous drying in an electric dryer with air temperatures of 50, 60 and 70 °C and air speed of 2.0 m s⁻¹. The mean environmental air temperature and relative humidity were 22.9 °C and 65.8%, respectively; they were determined using a digital thermohydropeter (HOBO datalogger, Onset) and the drying air relative humidity was determined using the GRAPSI 7.0 program (Melo et al., 2004), considering a constant mix ratio during the warming.

The samples were weighed every 5 min and the drying ended when the water content reached 0.11 d.b. (Gasparin et al., 2014; 2017). However, 90 g of fruits were kept in the dryer for the mathematical modeling, forming a thin layer until an equilibrium water content was reached. The hygroscopic equilibrium occurred when three consecutive weights had no variation. The water content ratio (RX) was calculated during the drying for the different temperatures using Eq. 1:

\[
RX = \frac{X - X_{e}}{X_{i} - X_{e}}
\]  

(1)

where:

- RX - water content ratio, dimensionless;
- X - product water content at weighing time, decimal, d.b.;
- Xₐ - product initial water content, decimal, d.b.; and,
- Xₑ - product equilibrium water content, decimal, d.b.

The drying rate, product water evaporated per unit of dry matter (d.m.) per unit of time, was estimated at every weighing using Eq. 2 (Park et al., 2007):

\[
DR = \frac{X_{0} - X_{i}}{\Delta T}
\]  

(2)

where:

- DR - drying rate, kg water kg⁻¹ d.m. h⁻¹;
- Xₐ₀ - product water content in the previous weighing, decimal, d.b.;
- Xᵢ - product water content in the current weighing, decimal, d.b.; and,
- ΔT - time interval between weighing, h.

The drying data were fitted to 12 mathematical models (Eq. 3 to 14) used in the description of drying of agricultural products (Table 1).

**Table 1. Non-linear regression models used to predict the drying phenomenon in the thin layer formed from whole fruits of Brazilian peppertree**

| Model                  | Formula                                      |
|------------------------|----------------------------------------------|
| Page                   | RX = exp(-kₜt)                              |
| Page modified           | RX = exp[-(kₜt)²]                            |
| Thompson               | RX = exp(-a(t)² + 4b t(t)½)/2b               |
| Newton                 | RX = exp(-k t)                              |
| Midilli                | RX = a exp(-k t) + b t                      |
| Verma                  | RX = a exp(k(t) + (1-a) exp(-k₁ t))          |
| Henderson and Pabis    | RX = a exp(-k t) + b exp(-k₂ t) + c exp(-k₁ t) |
| Henderson and Pabis    | modified                                     |
| Two terms              | RX = a exp(-k t) + b exp(-k₂ t) + c exp(-k₁ t) |
| Approximation of diffusion | RX = a exp(-k t) + (1-a) exp(-k a t)      |
| Wang and Singh          | RX = 1 + a t + b t                           |

RX - product water content ratio, dimensionless; t - drying time, min; k, k₁, k₂, kₚ - constants of drying, min⁻¹; a, b, c, n - coefficients of the models; Source: Ertekin & Firat (2017)
The Quasi-Newton method was used to estimate the parameters of the non-linear regression models. The fitted coefficient of determination ($R^2$), mean relative error (P), mean estimated error (SE), and residue distribution (RD) were considered for the fit level analysis. P and SE were calculated according to the Eqs. 15 and 16:

$$P = \frac{100}{n} \sum_{i=1}^{n} \left( \frac{Y_i - \hat{Y}_i}{Y_i} \right)$$  (15)

$$SE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left( \frac{Y_i - \hat{Y}_i}{DF} \right)^2}$$  (16)

where:
- P - mean relative error;
- n - number of experimental observations;
- Y - experimental value found;
- $\hat{Y}$ - estimated value by the model;
- SE - mean estimated error; and,
- DF - degrees of freedom of the model.

The consumed electric energy and specific energy consumption were calculated using Eq. 17 (Park et al., 2007) and 18 and the data were analyzed through correlation:

$$CEE = \left[ (F_{air} \rho_{air} C_{p,air} \Delta T) + Ev \right] t$$  (17)

$$SEC = \frac{3.6CEE}{w_{ev}}$$  (18)

where:
- CEE - consumed electric energy, kWh;
- $F_{air}$ - air flow, m$^3$ s$^{-1}$;
- $\rho_{air}$ - air specific weight, kg m$^{-3}$;
- $C_{p,air}$ - air specific heat at constant pressure, kJ kg$^{-1}$ °C;
- $\Delta T$ - difference between environmental and drying air temperatures, °C;
- Ev - electric energy needed to move the fan, kW;
- t - dryer operation time, h;
- SEC - specific energy consumption, MJ kg$^{-1}$; and,
- $w_{ev}$ - evaporated water weight, kg.

The Brazilian peppertree fruits were cleaned for removal of impurity and green fruits before the extraction of the essential oil, since the maturation stage affect the yield and quality of the essential oil (Schimitberger et al., 2018). The essential oil was extracted by hydro-distillation, in a Clevenger device, using 100 g of fruits for the dried whole fruits and 40 g for the dried fruits fragmented in a knife mill with a 2 mm mesh sieve (Fritsch Pulverisette 14, Oberstein, Germany).

The fruits were added to a volumetric flask containing 1000 mL of distilled water and this mixture was subjected to essential oil extraction for three hours from the beginning of ebullition.

The essential oil was then separated from the condensed water by subjecting the hydrolat to a partition process with 20 mL of pentane p.a. for three times. Anhydrous sodium carbonate p.a. was added to the organic fraction (essential oil and pentane) to remove residual water. The solution was filtered and concentrated in a rotative evaporator at 40 °C under low pressure to obtain the pure essential oil. The essential oil yield was determined by weighing and expressed in percentage in relation to the dry matter (% d.m.).

The essential oil yield as a function of drying air temperature was analyzed through regression analysis. The Tukey’s test at $p \leq 0.05$ was used to compare the means between the fruit fragmentation types.

### Results and Discussion

All models analyzed presented coefficient of determination ($R^2$) above 99%, except the Wang and Singh model for the temperature of 50 °C (Table 2); however, the $R^2$ is not an adequate index when used singly to select non-linear models. Thus, the model was satisfactory when presented value of $R^2$ close to 100, P lower than 10%, and low SE (Chen & Morey, 1989; Madamba et al., 1996).

The Verma and Approximation of diffusion models had better fit for the temperatures of 50 and 60 °C; and the Midilli

| Model                  | T (°C) | P (%) | SE (decimal) | $R^2$ (%) | RD |
|------------------------|--------|-------|--------------|-----------|----|
| Page                   | 50     | 4.355 | 0.012        | 99.791    | Tendentious |
| Page modified          | 60     | 3.948 | 0.012        | 99.830    | Tendentious |
|                      | 70     | 1.592 | 0.005        | 99.976    | Random |
| Thompson               | 50     | 4.355 | 0.012        | 99.791    | Tendentious |
|                      | 60     | 3.948 | 0.012        | 99.830    | Tendentious |
|                      | 70     | 1.592 | 0.005        | 99.976    | Random |
| Newton                 | 50     | 8.860 | 0.023        | 99.241    | Tendentious |
|                      | 60     | 5.109 | 0.015        | 99.685    | Tendentious |
|                      | 70     | 1.889 | 0.009        | 99.926    | Tendentious |
| Midilli                | 50     | 0.890 | 0.004        | 99.982    | Random |
|                      | 60     | 0.734 | 0.004        | 99.988    | Random |
|                      | 70     | 0.969 | 0.004        | 99.981    | Random |
| Verma                  | 50     | 0.594 | 0.002        | 99.994    | Random |
|                      | 60     | 1.889 | 0.009        | 99.985    | Tendentious |
|                      | 70     | 7.602 | 0.020        | 99.425    | Tendentious |
| Henderson and Pabis    | 50     | 4.841 | 0.015        | 99.715    | Tendentious |
|                      | 60     | 1.709 | 0.007        | 99.944    | Tendentious |
|                      | 70     | 0.933 | 0.004        | 99.982    | Random |
| Henderson and Pabis modified | 50  | 0.581 | 0.002        | 99.995    | Random |
|                      | 60     | 1.709 | 0.011        | 99.944    | Tendentious |
|                      | 70     | 0.933 | 0.004        | 99.982    | Random |
| Two-term               | 50     | 0.598 | 0.002        | 99.994    | Tendentious |
|                      | 60     | 1.709 | 0.009        | 99.944    | Tendentious |
|                      | 70     | 3.003 | 0.009        | 99.889    | Tendentious |
| Two-term exponential   | 50     | 4.462 | 0.014        | 99.742    | Tendentious |
|                      | 60     | 2.043 | 0.010        | 99.908    | Tendentious |
| Approximation of diffusion | 50 | 0.870 | 0.004        | 99.981    | Random |
|                      | 60     | 0.594 | 0.002        | 99.994    | Random |
|                      | 70     | 1.406 | 0.005        | 99.980    | Tendentious |
| Wang and Singh         | 50     | 10.006| 0.031        | 98.597    | Tendentious |
|                      | 60     | 5.212 | 0.019        | 99.538    | Tendentious |
|                      | 70     | 3.158 | 0.010        | 99.890    | Tendentious |

T - drying temperature; P - mean relative error; SE - mean estimated error; $R^2$ - coefficient of determination; RD - residue distribution
model had better fit for 70 °C. The models with better fit had
the lowest mean estimated error and mean relative error lower
than 5% for all temperatures. Silva et al. (2015) evaluated
Brazilian peppertree fruits and found that the best model to
describe the drying process was the Henderson; and the Wang
and Singh was the least adequate.

Despite several models have adequate fit of the data, a
model with satisfactory fit for all temperatures was chosen to
facilitate its use, since modeling is used for drying simulations
in thick layers and in the construction process of new dryers
(Berbert et al., 1995; Goneli et al., 2014b; Gasparin et al., 2017).
Therefore, the most adequate model was the Midilli. The result
of P, SE, and $R^2$ was acceptable, presenting random distribution
of residues for all temperatures. This model was also adequate
for the drying of leaves of Brazilian peppertree (Goneli et al.,
2014b), rosemary (Mghazli et al., 2017) and timbo (Serjania
marginata) (Martins et al., 2015) plants.

The increases in drying air temperature increased the $k$ and
$n$ values; and the variables $a$ and $b$ presented no trend to the
variations in temperature (Table 3).

The drying constant $k$ of the Midilli model is related to the
effective diffusivity in the decreasing period, which is similar
to the Henderson and Pabis model (Ertekin & Firat, 2017).
The $k$ value varies according to the drying air temperature and
the product initial water content (Mujumdar & Devahastin,
2000). The Brazilian peppertree fruits had no variation in initial
water content; thus, the temperature affected the $k$ value. No
trend was found for $a$ and $b$; the variations were related to
mathematical adjustments of the model and not to the drying
process (Goneli et al., 2014b).

The drying process occurred with decreasing rates (Figure
1); the resistance to migration of water from the interior to
the surface of the product increased as it was drying (Park
et al., 2001). The product temperature tends to increase in
this stage up to temperatures close to that of the drying air.
Studies with several medicinal plants also found drying curves
with decreasing rates (Martinazzo et al., 2010; Ali et al., 2014;
Gasparin et al., 2017; Mghazli et al., 2017).

Figure 2 shows the drying rate in the first 30 min of the
process and confirms the increase in resistance of migration
of water from the interior to the surface of the product as it
was drying.

The increases in drying air temperature increased the
drying rate, since a higher water removal from the product
was found, agreeing with several researchers (Martins et al.,
2015; Gasparin et al., 2017; Mghazli et al., 2017). A higher
inclination of the line was found for the 70 °C, indicating
a higher variation in the drying rate, making the product
to faster reach the water content desired than the other
temperatures.

Table 3. Coefficients of the Midilli model for each drying air
temperature

| Coefficients of the model | 50      | 60      | 70      |
|--------------------------|---------|---------|---------|
| $k$                      | 0.026003| 0.028339| 0.037470|
| $n$                      | 0.993336| 1.044275| 1.076749|
| $a$                      | 0.997523| 0.998492| 0.998579|
| $b$                      | 0.000575| 0.000872| 0.000475|

The effects of temperature on the drying time, specific
energy consumption, and consumed electric energy are shown
in Table 4.

The increase in drying air temperature from 50 to 70 °C
decreased the drying time in approximately 59%, since higher
temperatures result in higher water removal rates. This was
also found for Mentha piperita (Gasparin et al., 2017) and
Cordia verbenacea (Goneli et al., 2014a). The specific energy
consumption decreased as the drying air temperature was
increased, i.e., less energy was required to evaporate the same
quantity of water from the Brazilian peppertree fruits when the
drying air temperature was 70 °C, when compared to that at 50 °C.

Despite the dryer demands more energy when using
a drying air temperature of 70 °C, the drying time in this
temperature is lower than that in 50 °C, under the same
environmental conditions, and it decreased the energy
consumption.

The energy consumption of the drying operations can reach
40% of the total energy consumption of the industry, becoming
a very expensive operation (Mujumdar & Devahastin, 2000).
Therefore, increasing drying air temperature can significantly
decrease the operational cost of this process.
Drying and essential oil extraction of Brazilian peppertree (Schinus terebinthifolius Raddi) fruits

The correlations between drying air temperature and the other variables were significant ($p \leq 0.05$). Temperature presented negative correlation with drying time (-0.98), specific energy consumption (-0.95), and consumed electric energy (-0.95). Therefore, increasing drying air temperature tends to decrease other variables. This effect had been found for several agricultural products, such as rosemary, rice, and chrysanthemum (Mghazli et al., 2017; Tohidi et al., 2017; Wang et al., 2018).

According to the analysis of variance, the interaction between drying air temperature and fruit fragmentation was significant by the F test ($p$-value = 0.0163) for essential oil yield; therefore, these factors have dependent effect on essential oil yield.

Figure 3 shows the mean essential oil yield as a function of drying air temperature and fruit fragmentation.

The essential oil yield from whole fruits of Brazilian peppertree was maintained as the drying air temperature increased, but decreased linearly ($p$-value = 0.0037) for fragmented fruits.

Essential oils are substances sensitive to heat; therefore, increasing drying air temperature can volatilize these compounds (Ebadi et al., 2015; Ahmed et al., 2018). Differences in yield due to temperatures were not found for whole fruits, since the exocarp hindered the extraction of essential oil (Machado & Carmello-Guerreiro, 2001).

The essential oil yield of the fragmented Brazilian peppertree fruits was approximately 26-, 23-, and 18-fold those obtained with whole fruits for the drying air temperature of 50, 60, and 70 °C, respectively.

This may be due to the well-developed and complex secretor channels of the Brazilian peppertree seeds, which are protected by the exocarp (Machado & Carmello-Guerreiro, 2001). Thus, the fragmentation facilitated the extraction of essential oil, since it had exposed structures that contained the essential oil and increased the contact surface of the fruits with the water used in the hydro-distillation.

Schimitberger et al. (2018) found similar essential oil yield for fragmented Brazilian peppertree fruits. However, Oliveira Junior et al. (2013) found yields varying from 5.50 to 8.41%. This difference is not related only to the drying process, but to edaphoclimatic factors and factors inherent to the plants, which can affect secondary metabolites, such as water availability, and plant development (Gobbo-Neto & Lopes, 2007).

Ahmed et al. (2018) and Schimitberger et al. (2018) evaluated visually the color of essential oil obtained. The color of the essential oil was also affected by the fruit fragmentation. Visually, the essential oil extracted from whole fruits was yellowish and that of the fragmented fruits was translucid; this indicates differences in their chemical composition.

**Conclusions**

1. The Midilli model was the most adequate to describe the drying process of Brazilian peppertree fruits for all temperatures studied.
2. The drying air temperature of 70 °C decreased the drying time, consumed electric energy, and specific energy consumption.
3. The highest essential oil yield of Brazilian peppertree was found for the drying temperature of 50 °C and for fragmented fruits after the drying process.

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**Table 4.** Mean and standard deviation of drying time, specific energy consumption, and consumed electric energy as a function of drying air temperature

| Drying air temperature (°C) | Drying time (min) | Specific energy consumption (MJ kg$^{-1}$) | Consumed electric energy (kWh) |
|-----------------------------|-------------------|------------------------------------------|--------------------------------|
| 50                          | 108.33 ± 2.89     | 112.63 ± 2.74                           | 3.44 ± 0.09                     |
| 60                          | 71.67 ± 5.77      | 98.47 ± 7.84                            | 3.02 ± 0.24                     |
| 70                          | 45.00 ± 0         | 73.65 ± 0.62                            | 2.27 ± 0                        |

Means followed by same letter for same temperature are not different by the Tukey’s test at $p \leq 0.05$; Regression equation referring to fragmented fruits; CV - coefficient of variation; ** - Significant at $p \leq 0.01$ by t test

**Figure 3.** Brazilian peppertree fruit essential oil yield as a function of drying temperature and fruit fragmentation
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