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Original Article

Mathematical modeling of thin layer drying using open sun and shade of *Vernonia amygdalina* leaves

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ABSTRACT

The drying behavior of *Vernonia amygdalina* leaves was investigated using open sun and shade drying. The effective diffusivity coefficients for both methods were evaluated. Eight drying kinetics models were fitted to the experimental data obtained from the two drying methods; the goodness of fit was evaluated using three statistical tools, and the best model was selected. The Midilli and Kucuk model, which gave higher values of the coefficient of determination and lower values of the reduced chi-square and root mean square error was considered the best for predicting the drying behavior of open sun-dried and shade-dried *V. amygdalina* leaves with values of *R* of 0.99951 and 0.99981, RMSE of 0.00243 and 0.00253 and reduced chi-square of 0.000511 and 0.000428, respectively. The values of the effective diffusivity coefficients for open sun and shade drying were 26.58×10^{-10} m^2/s and 52.77×10^{-11} m^2/s, respectively. Furthermore, open sun drying resulted in severe deformity of the leaf morphology which may lead to degradation of the phytochemicals. Thus, shade drying was the better way of drying *V. amygdalina* leaves to preserve the nutrients compared to open sun drying.

Introduction

Bitter leaf (*Vernonia amygdalina*) is one of the medicinal plants or herbs that grows in Africa and Asia. It is rich in vitamins, minerals, flavonoids (luteolin and luteolin 7-O- glucosides), sesquiterpene lactones (vernolide, vernolepin, vernodalin, vernodalol and vernonide), terpenoids, and steroidal glycosides (Farombi and Owoeye, 2011; Jisaka et al., 1993; Mwanauta et al., 2014; Toyang and Verpoorte, 2013). These bioactive compounds result in the leaves having several pharmacological properties such as antioxidant, anti-diabetic, anti-inflammatory, and anti-cancer (Alara et al., 2017; Farombi and Owoeye, 2011; Ngatu et al., 2012). All the parts of this plant are rich in medicinal properties; the leaves have been used in treating diabetes, gonorrhoea, malaria, stroke, inflammation, and cancer (Osinubi, 2008; Owu et al., 2008; Adetunji et al., 2013). The stems and roots have been used in the treatment of stomach aches and skin infections such as itching, ringworm, eczema, and rashes (Ijeh and Ejike, 2011; Oduah, 2012).

Fresh medicinal plants including bitter leaf may deteriorate a few days after harvesting; therefore, there is a need for preservation to extend their shelf-life (Ali et al., 2014; Gumus, 2015). Drying is one of the ancient preservative methods used in extending a herb's shelf-life. In this method, moisture is removed from the grain, fruits or leaves, resulting in a reduction in their weight (Doymaz, 2006; Ali et al., 2014). Likewise, drying prevents the growth of yeast, bacteria and molds on the food. Most importantly, drying causes a reduction in shipping weight and in packaging requirements (Akpinar, 2006).

In recent years, different methods of drying have been studied to examine the effect of the drying rate and kinetics models on the plant matrix. Open sun and shade drying are well-known for reducing the moisture content in perishable products and thereby preventing deterioration (Toğrul and Pehlivan, 2003; Akpinar, 2006). In spite of many disadvantages (such as probable contamination) of these methods, tropical and sub-tropical countries of the world still use open sun and shade drying to preserve agricultural products (Akpinar and Bicer, 2008). Solar energy which is the source of energy for open sun drying has

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been a preferred energy source because it is abundant, renewable, environmentally friendly, cheap, inexhaustible, and a non-pollutant (Akpinar and Bicer, 2008).

A series of studies have been conducted on the drying process and mathematical models developed for different leaves and fruits such as *Moringa oleifera* (Ali et al., 2014), curry leaves, mint, and bitter gourd (Vyankatrao, 2014), barbunya bean (Kayisoglu and Ertekin, 2011), orange seed (Rosa et al., 2015), spearmint leaves (Antal et al., 2011), banana (da Silva et al., 2014), and mango, guava and aonla (Kumar and Sagar, 2014). Although the drying process and mathematical kinetics of *V. amygdalina* leaves using open sun drying have been studied (Sobukola et al., 2007) whereby the Midilli and Kucuk model was reported best in describing the behavior, however, to the best of the authors' knowledge, there has been no study reported on the drying behavior of *V. amygdalina* leaves by comparing the open sun and shade drying methods. Therefore, this study was conducted to provide a comprehensive investigation on the drying behavior and model development of *V. amygdalina* leaves under open sun and shade drying.

Materials and methods

Materials

Fresh leaves of *V. amygdalina* were harvested from a garden in Gambang, Malaysia (3° 42′25.183″N; 103°6′8.982″E). The matured leaves were selected to ensure a homogenized sample.

Drying experiment

The open sun drying process was carried out in December 2016 at the Faculty of Chemical and Natural Resources Engineering laboratory, Universiti Malaysia Pahang, Malaysia. Fresh leaves of *V. amygdalina* were separated from the stalks manually and rinsed with clean tap water. Thereafter, the elliptical-shaped leaves were cut into smaller pieces of approximately 1 cm in length and the gross weight of the sample (50 ± 1 g) was determined using a laboratory scale (513-1S; Secura; New York, NY, USA) with readability to 0.001 g. Afterwards, the average initial moisture content of the 50 g samples was recorded following AOAC (1990). Before exposing the samples to open sun, the ambient air temperature, average relative humidity and average wind speed were recorded. The samples were spread uniformly on a single layer sample tray and then exposed immediately to the open sun. The experiment started about 09:00 h and continued until 19:00 h. The loss in moisture content from the samples was determined using the weight changes at 1 h intervals throughout the drying process. The samples were dried until their moisture content was less than 0.11 g water/g sample. The dried samples were stored in air-tight low-density polyethylene (LDPE) bags and sealed thermally to prevent moisture absorption. The experimental procedures were repeated thrice and the results were expressed as mean values.

For shade drying, the fresh leaves of *V. amygdalina* were rinsed with clean tap water. The elliptical-shaped leaves were cut into small pieces of approximately 1 cm in length. Then, the gross weight of the sample (50 ± 1 g) was determined using a laboratory scale (513-1S; Secura; New York, NY, USA) with readability to 0.001 g. The average initial moisture content of the 50 g samples was recorded following AOAC (1990). The samples were spread uniformly on a single layer sample tray and were placed on a well-ventilated laboratory bench. Before drying, the average air room characteristics of 26 ± 1 °C and 33 ± 5% relative humidity were recorded. The loss in moisture content was recorded at 1 h intervals until less than 0.11 g water/g sample was attained. The dried samples were stored in air-tight LDPE bags and sealed thermally to prevent moisture absorption. The experimental procedures were repeated thrice and the results were expressed as mean values.

Mathematical modeling

The moisture ratio (MR) of *V. amygdalina* leaves was determined using Equation (1):

$$MR = \frac{M_t}{M_o} \tag{1}$$

where M_t and M_o are the moisture content at a particular time t and the initial moisture content of the sample, respectively. Equation (1) was used due to continuous fluctuations in the relative humidity of the drying air during the open sun and shade drying processes (Toğrul and Pehlivan, 2003).

Table 1 shows the eight models used in selecting the best drying kinetics for describing the drying curve equation of *V. amygdalina* leaves using open sun and shade drying. The Statistica computer program (STATISTICA 12.0; Dell Technologies; United States) was used to perform the regression analysis. The correlation coefficient R for Equation (2), the root mean square error (RMSE) from Equation (3), and χ^2 from Equation (4) as the mean square of the deviations between the experimental and calculated values of the models were used to determine the quality of the fit.

$$R = 1 - \frac{\sum_{i=1}^{N}\left(MR_{pre,i} - MR_{\exp,i}\right)^2}{\sqrt{\left[\sum_{i}^{N}\left(MR_{\exp,i} - MR_{\exp\ mean}\right)^2\right]}} \tag{2}$$

Table 1
Theoretical and semi-theoretical models for thin-layer drying.

Number	Model name	Model equation	References
1	Approximation of diffusion	$MR = a\exp(-kt) + (1-a)\exp(-kbt)$	Sobukola et al. (2007)
2	Henderson and Pabis	$MR = a\exp(-kt)$	Henderson and Pabis (1961)
3	Logarithmic	$MR = a\exp(-kt) + c$	Toğrul and Pehlivan (2003)
4	Midilli and Kucuk	$MR = a\exp(-kt^n) + bt$	Midilli et al. (2002)
5	Newton	$MR = \exp(-kt)$	Sobukola et al. (2007)
6	Page	$MR = \exp(-kt^n)$	Page (1949)
7	Two-term	$MR = a\exp(-kt) + b\exp(-k_o t)$	Henderson (1974)
8	Two-term exponential	$MR = a\exp(-kt) + (1-a)\exp(-kat)$	Sharaf-Eldeen et al. (1980)

where a, b, c, n = coefficient; k, ko = drying constant.

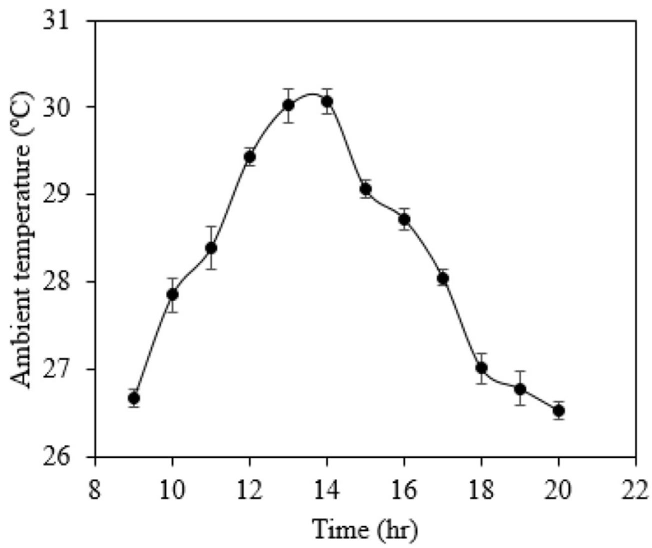

Fig. 1. Ambient temperature during the open sun drying experiment at Gambang, Malaysia, December 2016.

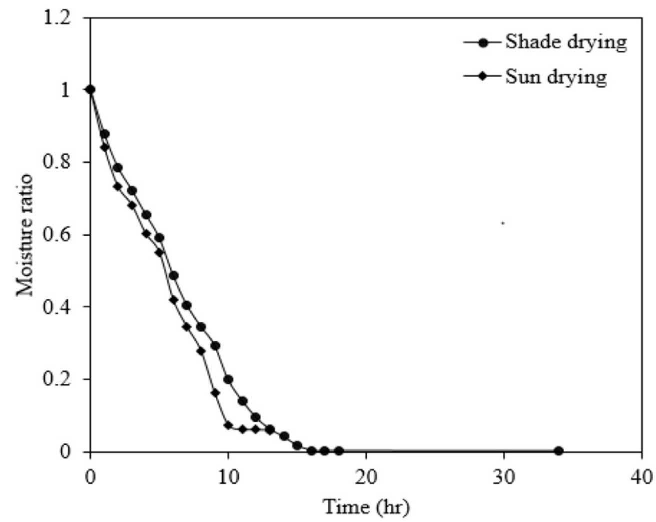

Fig. 3. Change in moisture ratio with drying time of *V. amygdalina* leaves.

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}(MR_{pre,i} - MR_{\exp.i})^2\right]^{\frac{1}{2}} \tag{3}$$

$$\chi^2 = \sum_{i=1}^{N}\frac{(MR_{\exp,i} - MR_{pre,i})^2}{N-n} \tag{4}$$

where $MR_{exp,i}$ is the experimental moisture ratio, $MR_{pre,i}$ is the predicted moisture ratio, N is the number of observations and n is the number of constants in the drying model (Toğrul and Pehlivan, 2003; da Silva et al., 2014; Rosa et al., 2015).

Sample morphological and elemental characterization

The sample appearance before and after drying was examined using scanning electron microscopy (SEM; TM3030Plus; Hitachi; Ibaraki, Japan). The sample was mounted on an SEM stub and the morphology appearance was observed. This analysis was carried out under a high vacuum condition at an accelerated voltage of 15 keV, 100 × magnification and an analytical working distance of 1 mm. The elemental analysis of the sample before and after drying was analyzed using energy-dispersive X-ray spectroscopy. The detection of the elements in the *V. amygdalina* leaves was carried out using a silicon (lithium) detector cooled with liquid nitrogen. The EDX Shimadzu software package (Shimadzu EDX-7000/8000; Hitachi; Japan)was used to determine the intensity of each element in counts per second from the sample X-ray spectrum deconvolution.

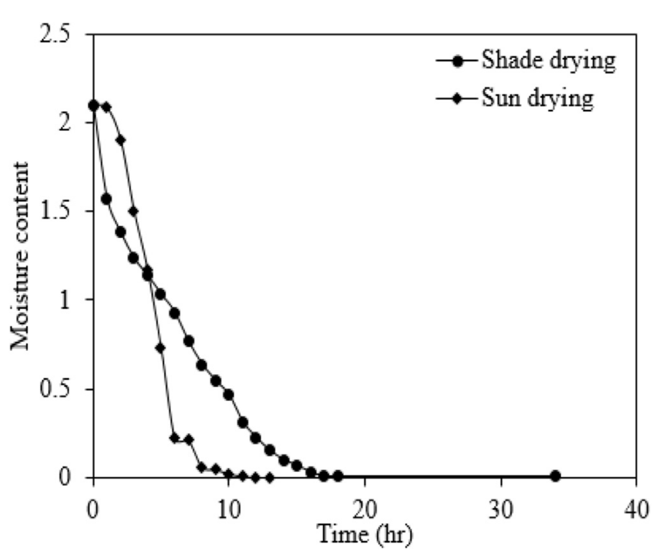

Fig. 2. Change in moisture content with drying time for open sun and shade drying.

Results and discussion

The average values recorded for changes in ambient temperatures during the open sun drying of *V. amygdalina* leaves are shown in Fig. 1. These ranged from 26.53 to 30.07 ± 1 °C with the highest ambient temperature (30.07 ± 0.15 °C) recorded at 1400 h. The average relative humidity and average wind speed were recorded as 75% and 2.28 m/s, respectively.

Drying characteristics

Drying curves showing the behavior of *V. amygdalina* leaves under both open sun and shade drying are presented in Fig. 2. The decrease in moisture content over time for both drying methods indicates no constant rate in the drying of *V. amygdalina* leaves. This was similar to the reported results from other herbs and food materials, showing a rapid moisture removal from the leaves in the initial stage that later decreased with the drying time (Ali et al., 2014; Vyankatrao, 2014; Rosa et al., 2015). Thus, the time required to dry *V. amygdalina* leaves under open sun and shade drying conditions from the initial moisture content of 2.0842 ± 0.1 g water/g dry sample on a dry basis (d.b.) to the final moisture content of 0.011 ± 0.001 g water/g dry sample d.b. and 0.011 ± 0.001 g water/g dry sample d.b. was 13 h and 58 h, respectively. In Fig. 2, it can be clearly seen that the time required to dry the sample was lower (13 h) under open sun. The results obtained using both methods showed that internal mass transfer occurred by diffusion (Akpinar, 2006). This finding is in agreement with results obtained from drying mint leaves, long green

Table 2
Estimated parameters and comparison criteria of moisture ratio for open sun drying of *V. amygdalina* leaves.

Drying method	Model number	Coefficients	R	RMSE	χ^2
Open sun	1	a = 0.256437; b = 0.376544; k = 0.000163	0.95252	0.04834	0.010002
	2	a = 1.000012; k = 0.000163	0.97850	0.03763	0.007644
	3	a = 0.654320; k = 0.100003; c = 0.254673	0.96074	0.03067	0.008617
	4	a = 0.122883; b = 0.000436; k = 1.134270; n = 0.000234	0.99951	0.00243	0.000511
	5	k = 0.000178	0.96992	0.04832	0.009210
	6	k = 0.011322; n = 0.012126	0.99684	0.00262	0.000601
	7	a = 0.220021; k = 0.002228; b = 1.000023; k_o = 1.033651	0.98981	0.03762	0.007231
	8	a = 1.000025; k = 0.000512	0.95004	0.04302	0.010045
Shade	1	a = 0.222654; b = 0.315534; k = 0.000132	0.99644	0.01362	0.000758
	2	a = 1.000038; k = 0.000155	0.95992	0.01054	0.002153
	3	a = 0.633254; k = 0.100000; c = 0.249983	0.96031	0.00984	0.008215
	4	a = 0.119947; b = 0.000356; k = 1.112201; n = 0.000212	0.99981	0.00253	0.000428
	5	k = 0.000146	0.94900	0.01902	0.005320
	6	k = 0.010225; n = 0.010132	0.99592	0.00339	0.000622
	7	a = 0.211062; k = 0.001164; b = 1.000019; k_o = 1.010456	0.95858	0.01612	0.002643
	8	a = 0.000013; k = 0.000423	0.95325	0.01672	0.003103

pepper, carrot, aloe vera, garlic, black tea, and king of bitters (Madamba et al., 1996; Simal et al., 2000; Panchariya et al., 2002; Doymaz, 2004, 2006; Akpinar and Bicer, 2008).

Fitting of the thin layer drying curves

In order to evaluate the moisture content as a function of drying time, the eight models (Table 1) were fitted to the experimental data (Fig. 3) by converting the moisture content data at the different drying time to moisture ratio expressions. The obtained statistical results from these eight models for both drying methods are presented in Table 2. Accordingly, all the tested models had high coefficient of determination (R^2) values in the range 0.95004–0.99951 and 0.95325–0.99981 for open sun and shade drying, respectively. These showed that all the models can actually describe the open sun and shade drying of *V. amygdalina* leaves but the Midilli and Kucuk model had the highest R^2 value and the lowest χ^2 and RMSE values for the open sun and shade drying of *V. amygdalina* leaves, as shown in Table 2. From the Midilli and Kucuk model, the open sun drying of *V. amygdalina* leaves resulted into $R = 0.99951$, RMSE = 0.00243 and $\chi^2 = 0.000511$ while that of shade drying of *V. amygdalina* leaves resulted in $R = 0.99981$, $RMSE = 0.00253$ and $\chi^2 = 0.000428$. In similar research, the Midilli and Kucuk model was used to predict the drying behavior of *V. amygdalina* under open sun conditions (Sobukola et al., 2007). Therefore, this model may be used in the prediction of both open sun and shade drying behavior of *V. amygdalina* leaves.

Validation of the model

The validation of the predicted model was carried out by comparing the predicted with experimental moisture ratio values. The linear correlation between the predicted moisture ratio generated and the experimental moisture ratio (Fig. 4) showed that the Midilli and Kucuk model could best describe the drying behavior of *V. amygdalina* leaves under open sun and shade drying conditions. Likewise, the higher values of R from the correlations for open sun (0.9931) and shade drying (0.9934) indicated the best fit of the model and showed that the predicted model provided good agreement between the experimental and predicted results. Therefore, the Midilli and Kucuk model can be used to predict the thin layer open sun or shade drying behavior of *V. amygdalina* leaves.

Effective diffusivity coefficients determination

The drying behavior of leafy plants can best be described using Fick's second diffusion model (Doymaz, 2006; Sobukola et al., 2007; Akpinar and Bicer, 2008). Equation (5) gives the analytical solution of Fick's second diffusion law in a slab geometry by assuming

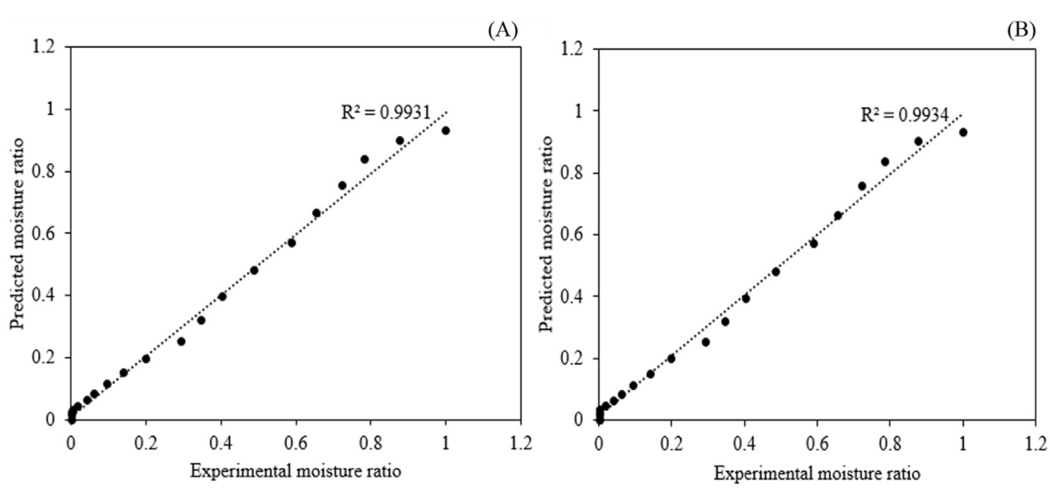

Fig. 4. (A) Experimental and predicted Midilli and Kucuk model moisture ratio values for open sun drying and (B) shade drying.

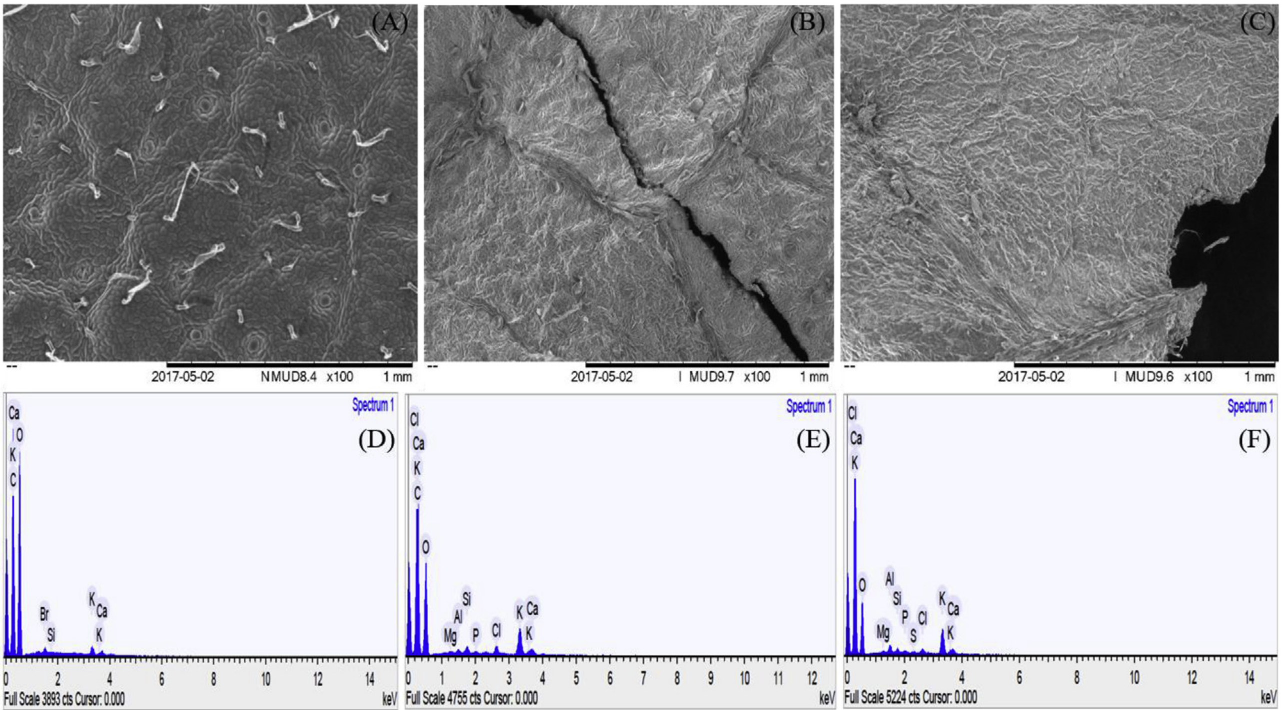

Fig. 5. Morphological appearance of fresh (A); open sun-dried (B); shade-dried *V. amygdalina* leaf (C); and the elemental analysis of fresh (D); open sun-dried (E); shade-dried *V. amygdalina* leaf (F).

uniform initial moisture distribution, negligible shrinkage, simplification of the moisture movement by diffusion and the temperature and constant diffusion coefficients (Crank, 1975).

$$MR = \frac{8}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{(2n+1)^2}\exp\left[\frac{-(2n+1)^2\pi^2 D_{eff}t}{4L^2}\right] \quad (5)$$

where D_{eff} is the effective diffusivity (square meters per second), L is the half thickness of slab (m) of drying from both sides, and n is the number of terms taken into consideration.

For long drying periods, equation (5) can be simplified to the first term ($n = 1$) of the series only (Akpinar, 2006; Doymaz, 2006). The diffusion coefficients for each mode of drying can be calculated from the slope obtained by plotting the natural logarithm of MR against drying time.

During the open sun and shade drying, the effective diffusivity coefficients were 26.58×10^{-10} m^2/s and 52.77×10^{-11} m^2/s, respectively. These values fall within the general range of 1×10^{-9} m^2/s to 1×10^{-11} m^2/s for food samples and agricultural crops (Toğrul and Pehlivan, 2003; Rosa et al., 2015). Related results have been reported for *V. amygdalina* leaves and mint leaves under open sun drying (Doymaz, 2006; Sobukola et al., 2007), carrot under convective air drying (Doymaz, 2004), black tea (Panchariya et al., 2002), garlic (Madamba et al., 1996), and aloe vera (Simal et al., 2000). However, variation in the values of the effective diffusivities may be due to the drying conditions, implying that the open sun drying has a higher D_{eff} value than shade drying.

Morphological changes and elemental analysis of the V. amygdalina *leaves*

The morphological appearance of *V. amygdalina* leaves before and after drying is presented in Fig. 5A_1-A_3. Glandular and non-glandular trichomes are present on the epidermal surface of the fresh leaf (Fig. 5A_1). It can be clearly seen that the drying conditions affected the epidermal surfaces and caused the shrinkage of glandular trichomes. These may be attributed to the increased drying temperature. More shrinkage was seen in the sun-dried sample compared with the shade drying as higher temperature caused a more violent evaporation of moisture as well as melting of starch granules (Fig. 5A_2-A_3). Furthermore, this phenomenon suggests that drying affects the physical structure of the cell wall which might lead to breakage of vegetal tissue. Likewise, it has been

Table 3
Nutrient composition (%) in *V. amygdalina* leaf before and after drying.

Element	Weight (%)	Sigma weight (%)	Atomic (%)
Fresh leaf			
C	47.176	0.620	54.806
O	51.257	0.609	44.704
Si	0.080	0.026	0.040
K	0.673	0.042	0.240
Ca	0.391	0.039	0.136
Br	0.423	0.059	0.074
Open sun-dried			
C	57.537	0.604	65.849
O	37.608	0.572	32.312
Si	0.345	0.031	0.169
K	2.806	0.073	0.986
Ca	0.598	0.046	0.205
Mg	0.118	0.029	0.067
Al	0.152	0.027	0.077
P	0.185	0.031	0.082
Cl	0.649	0.041	0.252
Shade-dried			
C	53.123	0.775	66.232
O	39.382	0.709	30.799
Si	1.500	0.061	0.744
K	3.289	0.103	1.171
Ca	0.680	0.065	0.236
Al	0.459	0.040	0.237
P	0.240	0.045	0.108
Cl	0.663	0.053	0.261
Fe	0.413	0.109	0.103

reported that higher drying temperature tends to reduce the yields and phytochemicals present in plants (Antal et al., 2011). In the current study, less shrinkage was recorded in the shade-dried sample. Therefore, it is imperative to select an appropriate drying method to preserve the sample's phytochemical nutrients.

The mineral contents in *V. amygdalina* leaves before and after drying are shown in Fig. 5B_1-B_3 and Table 3. Carbon, oxygen, silicon, phosphorus, potassium, calcium, and bromine were present in the fresh leaf (Fig. 3). There was a larger percentage of oxygen followed by carbon and traces of other elements. The presence of these elements in the plant was responsible for curing several diseases (Mushtaq et al., 2012). After drying, the percentage of carbon residues increased and of oxygen declined which showed the burning effect that had occurred on the sample. In comparison, the shade-dried samples had less carbon residue and a higher oxygen value. Likewise, after drying the samples, the presence of other elements was recorded, including magnesium, aluminum, phosphorus, chlorine, and iron which might have been in trace amounts in the fresh leaf. The presence of toxic elements such as Hg, Cd and Sb was not detected in the samples before and after drying. Other elements detected in the shade-dried samples were higher than in the sun-dried samples. Therefore, it can be deduced that shade drying has a higher capacity to preserve more nutrients compared to sun drying.

The calcium and potassium found in the *V. amygdalina* leaves showed the leaf potency in muscle contraction and the synthesis of some proteins. In addition, calcium helps in teeth and bone development, blood clotting, regulating heart rhythm, and lowering blood pressure (Ragavendran et al., 2012). Potassium is an important component that regulates the blood pressure and heart rate. Furthermore, chlorine acts as an electrical charge when dissolving body fluids and regulates the body pH (Ragavendran et al., 2012). Therefore, *V. amygdalina* leaves can be used for pharmaceutical and therapeutic agents.

In this study, the drying behavior of *V. amygdalina* leaves was studied under open sun and shade drying conditions. The drying curves did not show a constant rate of drying under the experimental conditions though a falling rate with time was observed. Eight thin-layer-drying kinetics models were used to explain the drying behavior and three statistical tools were used to quantify the goodness of fit. The Midilli and Kucuk model, which gave higher values of coefficient of determination and lower values of reduced chi-square and RMSE was considered the best for predicting the drying behavior of open sun and shade dried *V. amygdalina* leaves with values of R of 0.99951 and 0.99981, RMSE of 0.00243 and 0.00253, *and* χ^2 of 0.000511 and 0.000428, respectively. The values of effective diffusivity coefficients for open sun and shade drying were 26.58×10^{-10} m^2/s and 52.77×10^{-11} m^2/s, respectively. In addition, the open sun drying caused more severe damage on the morphological appearance of the leaf which resulted in the reduction of its mineral contents. Thus, shade drying would be preferable in preserving the phytonutrients of the *V. amygdalina* leaves.

Conflict of interest

The authors report no conflicts of interest.

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