International Food Research Journal 25(6): 2620-2627 (December 2018)
Journal homepage: *http://www.ifrj.upm.edu.my*

Drying and pulverization processes affect the physico-chemical properties of kaffir lime leaves (*Citrus hystrix* DC)

[1]Setiyoningrum, F., [2*]Lioe, H. N., [2]Apriyantono, A. and [3]Abbas, A.

[1]Research Center for Biotechnology, Indonesian Institute of Sciences, Jl Raya Bogor KM. 46, Cibinong Bogor, Indonesia 16911
[2]Department of Food Science and Technology, Faculty of Agricultural Engineering and Technology, Bogor Agricultural University, IPB Campus Darmaga, Bogor, Indonesia 16680
[3]Center for Appropriate Technology Development, Indonesian Institute of Sciences, Jl KSA Tubun, Subang, Indonesia 41213

Article history

Received: 5 October 2017
Received in revised form: 6 February 2018
Accepted: 9 February 2018

Keywords

Citronellal
Essential oil
Kaffir lime leaves
Pulverization
Spices
Herbs drying

Abstract

The leaves of Kaffir lime (*Citrus hystrix* DC) are marketed in dried or powdered form. In this research, the effect of oven drying at a relatively low temperature but in a long time with or without the following pulverization on the characteristics of kaffir lime leaves was investigated. The change of moisture analyzed by vacuum oven method, the color analyzed by a chromameter, the yield of essential oils and the volatiles composition analyzed by GC-MS were observed for fresh, dried and dried-pulverized kaffir lime leaves. A moisture content desired for storage, 7%, was achieved when the leaves were dried for 12 hours using a cabinet dryer at 50°C. The yields of essential oils of the dried leaves were decreasing after their pulverization process into powder. Their color characteristics according to *L, a* and *b* values tend to increase. The volatiles composition also changed. Among the essential oil components, citronellal, an important volatile in the fresh leaves, was found to decrease, but on the other hand the oxidized compounds such as oxygenated sesquiterpenes were dominantly present in the yields.

Introduction

Kaffir lime (*Citrus hystrix* DC) is one of citrus varieties which is widely used as a spice or natural flavor in food and beverages in some Asian countries due to its favorable characteristic aroma (Sato *et al*., 1990; Nor, 1992). Kaffir lime leaves give an oriental aroma on Thai tom yam soup, laksa and curry. In Indonesia, kaffir lime leaves are widely used to mask the stench of fish or meat. Rendang (beef cooked with coconut milk), rawon (beef soup), soto (beef soup with coconut milk), pecel (vegetable mix with peanut sauce) and urap (vegetable mix with grated coconut) are traditional Indonesian foods that uses kaffir lime leaves as typical flavoring. Sliced fresh kaffir lime leaves are often added into tempe mendoan (fried tempeh) and peanut brittle.

Several studies have reported the components contributing to the flavor of kaffir lime leaves (Sato *et al*., 1990; Wijaya, 1995; Pudil *et al*., 1998). Their main components are citronellal, citronellol and nerol. Furthermore, Sato *et al.* (1990) suggested that the L-citronellal is a key compound of kaffir lime leaves that gives an aroma like a mixture of lemongrass and citrus aroma. Other volatile components contained in the kaffir lime leaves are α-pinene, champene, β-pinene, limonene, copaene, linalool, β-cubebene, isopulegol, caryophyllene, and citronellyl acetate (Wijaya, 1995; Tinjan and Jirapakkul, 2007).

Herbs and spices are often sold in dry form. Several studies using dried herbs and spices as research objects, mentioned the moisture content of less than or equal to 10% (Arslan *et al.*, 2010; Sellami *et al*., 2011; Usai *et al*., 2011) to inhibit microbiological and biochemical changes during storage (Argyopoulos and Muller, 2011). Thus, drying process could increase their shelf life (Diaz-Maroto, 2003). Unfortunately, drying can alter their essential properties, such as moisture content, color, nutrients and volatile components especially when dried at temperatures above 50°C (Baritaux *et al*., 1992; Diaz-Maroto *et al.*, 2003; Arslan *et al*., 2010; Szummy *et al.,* 2010; Ding *et al*., 2011; Raksakantong *et al*., 2011; Sellami *et al*., 2011). Some drying techniques using cabinet dryer, dryer with air blower, and dryer with FIR (far infra red) have been applied for drying herbs (Chua and Chou, 2003). Oven and hot air dryer are widely used for drying the herbs in developing countries because they are relatively inexpensive (Chua and Chou, 2003).

*Corresponding author.
Email: *hanifahlioe@apps.ipb.ac.id*

The drying of kaffir lime leaves with hot air of 50°C using cabinet dryer could increase the concentration of citronellal compared to FIR dryer or dryer with low relative humidity (Raksakantong *et al*., 2011). When the leaves dried with fluidized bed dryer at a constant temperature of 50°C, it gave no significant loss in volatiles composition (Tasirin *et al.,* 2014). In this research, cabinet dryer with a temperature of 50°C was chosen for the drying experiment of kaffir lime leaves. The objective of this research was to evaluate the effect of oven drying at relatively low temperature but in a long time, with or without the following pulverization process, on the changes of physicochemical characteristics of fresh kaffir lime leaves, which included moisture content, color, essential oil yield and the composition of essential oil or volatile compounds.

Materials and Methods

Plant materials

Kaffir lime leaves were obtained from Tulung Agung, East Java, Indonesia at the age of trees approx. 5 years, and the age of leaves was 3 – 4 months from the bud. Leaf size was ranged from 9 × 5 cm (length × width, ± 2 cm). Leaves were picked at the stem segments; length of petioles was ranged from 0.3 to 0.5 cm. After harvested, fresh kaffir lime leaves were collected and put in a porous plastic bags, 5 kg/plastic. Transportation of fresh kaffir lime leaves to the laboratory was taken less than 24 hours at room temperature (22 - 24°C).

Drying and pulverization process

Fresh kaffir lime leaves were dried by a cabinet dryer with a temperature controller JC/DO/L/100 (Jenn Chiang Machinery Works Co., China), set at temperature of 50 ± 2°C. The number of cabinet dryer trays was 20 pieces; each metal tray (size L.100 × W.50 cm) was filled with 400 grams of fresh kaffir lime leaves. The moisture content of kaffir lime leaves were observed every 4 hours, until their moisture content reached less than or equal to 10%. The moisture content was analyzed by a vacuum oven method (AOAC, 2000). To produce kaffir lime leaves powder, dried kaffir lime leaves were pulverized by a discmill, then sieved by 40 mesh FSRG00 (Enterprise Co., Taiwan). The dried kaffir lime leaves, either the whole form or the powder form was packaged on a PP plastic package, not in vacuum condition, and kept at a refrigerator, 1-2 days prior to distillation.

Color analysis

Color analysis was performed by a Minolta CR 310 chromameter (Minolta Co., Japan) following the procedure from Serpen and Gokmen (2009). The L^*, a^*, and b^* values were measured by the instrument. The measurements were taken at 3 random points for each sample and each measurement was done in 3 replicates. The value of L^* shows the brightness or lightness of sample, ranged 0 (black) to 100 (white). The value of a^* states a reflected light that produces chromatic color of mixture of red-green, ranged from -80 (greenness) to +100 (redness). The b^* value states a chromatic color of mixture of blue-yellow, ranged from -70 (blueness) to +70 (yellowness). Overall color changes (ΔAE) can be calculated as follows:

$$\Delta AE = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2}$$

Remarks:

$\Delta L = L^*$ sample – L^* standard

$\Delta a = a^*$ sample – a^* standard

$\Delta b = b^*$ sample – b^* standard,

ΔAE value > 1 indicates that the color change can be distinguished by human eye (Gonnet, 1998).

Essential oil yield measurement by distillation

The fresh, whole of dried kaffir lime and their powder form were separately steam-distilled for 3 hours. Amount of 260 g each sample were used for the distillation. This distillation was done in duplicate. The sodium sulphate anhydrous GR (Merck, Germany) was added on the distillate to separate water phase, then filtered by whatman paper. The essential oils were weighed and stored at -20°C (under nitrogen atmosphere) prior to GC/MS analysis.

Essential oil component analysis by GC/MS

The essential oils were analyzed for their volatiles composition by a modified method of volatile analysis (Pudil *et al*., 1998). Two replicates of essential oils were analyzed with a Agilent Technologies 7890A-5975 c inert XLEI/CI gas chromatograph (Agilent Technologies Co., USA) equipped with a mass spectra detector (MS) and fitted with a HP-5MS capillary column (length 30 m, 0.25 mm i.d, 0.25 µm film thickness). The mass units (m/z) were monitored from 33 to 550 at 70 eV. The samples were added with n-decana (Merck, Germany) as internal standard (5 mg/100µL samples). Amount of 0.2 µL each samples were injected into GC-MS using split 1:100 and helium as carrier gas. The injector temperature was 250°C and the detector temperature was 280°C. The column temperature program was set as follows: initially at 60°C, held 5 mins, increased 3°C/min to 220°C, then increased 6°C/min until the final temperature 260oC and held for 10 mins.

LRI (Linear Retention Index) was determined by comparing the retention time of all contituents of the samples with the retention time of homologous series of n-alkanes (C8-C33) (Sigma - Aldrich, Singapore) on the same column and conditions above. Identification of each component was made by matching their recorded mass spectra with reference spectra in the data base (NIST) and compared their LRI and mass spectra with published data in the literature (Adams, 2009).

Statistical analysis

Data were subjected by a one-way analysis of variance (ANOVA, p=0.05) using a SPPS software (SPSS, Chicago, II, USA). Differences of samples were determined by Duncan test.

Results and Discussion

The change of moisture content

Moisture content of fresh kaffir leaves was 62.22% (w/w). The content decreased significantly during 12 hours of drying at temperature of 50 ± 2°C (Table 1). After drying, moisture content of the dried ones was less than 10%, i.e. 7.17%. Temperature used in this research was comparable to various drying application for aromatic or herbal leaves, such as fixed bed drying of long pepper leaves (Braga *et al.*, 2004), and convective drying or hot air drying of mulberry leaves (Tao *et al.,* 2016), yerba leaves (Pilatti *et al.*, 2016), thyme (Lahnine *et al*., 2016) and peppermint leaves (Torki-Harchegani *et al*., 2016). The drying process allowed the water in the leaves to move diffusely to their surface and then the water was evaporated (Braga *et al.*, 2004). The required drying time for materials varied, depending on the type, dimensions, initial moisture content, various transfer rates on the surface and the equilibrium moisture content. In fact, the drying time was faster than that in other spice research. The drying time for thyme to reach moisture content 6.90% was 18 hours, by oven at the same temperature as in this study, 50°C (Sarosi *et al*., 2013).

Essential oil yield of kaffir lime leaves decreased significantly during 12 hours of drying (Table 1). The yield of fresh kaffir lime leaves oil was 3.51% (dry basis). After 12 hours of drying, the oil yield did decrease to 1.83% (dry basis), almost 50% of the oil yield of fresh leaves. Similar to our result, Sarosi *et al*. (2013) reported a decrease of thyme oil yield, from 1.77% to 0.69%, after drying at a temperature of 50°C for 18 hours. The decrease of oil yield might be due to the oil evaporation during drying. Heat of drying process induced cracking of trichomes (Diaz-Maroto *et al*., 2002; Diaz-Maroto *et al*., 2003). Further, essential oils move onto the leaf surface along with water and then evaporated by heat (Braga *et al*., 2004).

Table 1. Moisture contents and oil yield of kaffir lime leaves during oven drying

Drying time (hours)	Moisture content (% wb)	Oil yield (% db)	Decreasing of oil yield (%)
0	62.22±1.50^a	3.51±0.11^a	-
4	41.56±1.23^b	3.15±0.07^b	10.26
8	25.51±1.46^c	2.34±0.08^c	33.33
12	7.17±0.04^d	1.83±0.03^d	47.86

Different superscript letters in the same column indicate significantly different means among samples according to Duncan test at P<0.05.
Decreasing of oil yield was calculated based on fresh oil yield (0 hours).

The change of color

The L^* value of whole dried kaffir lime leaves was higher than the fresh form, indicated the color of whole dried was brighter than the fresh form. The same trend occurred to the powder form. Increasing of L^* value indicate that sample is brighter than standard. The result was similar with Naidu *et al.* (2016) that revealed the increasing of L^* value of dill leaves after drying process. This phenomenon was reinforced by increasing of b^* of either whole dried and powder form significantly. Increasing of b^* means the sample is more yellow than standard (McGuire, 1992; Serpen and Gokmen, 2009).

The a^* value of dried whole leaves did not differ from the fresh ones, this indicated there was no changes during drying process. Otherwise, pulverization process induced weakening of the green intensity, characterized by a^* value of powdered dried leaves lead to positive (Table 3). The results were in agreement with previous findings (Buchaillot *et al*., 2009; Arslan *et al.*, 2010).

Increasing of b^* and a^* values might be due to the degradation or the decrease of chlorophyll. Chlorophyll degradation has been known to be induced by light, exposure to oxygen (Llewellyn *et al*., 1990) and heating (Buchaillot *et al.*, 2009). Futhermore, Schwartz and Lorenzo (1991) revealed that chlorophyll retention was affected by temperature and length of heat treatment. In this research, the heat exposure during drying might change the chlorophyll into pheophytin and gave a brownish color (Gross,

Table 2. Changes of kaffir lime leaves attribute color analyzed by chromameter

Form of kaffir lime leaves	Value		
	L	a	b
Fresh	29.07±0.12^a	-8.08±0.66^a	8.07±0.48^a
Whole dried	31.80±0.23^b	-9.49±0.39^a	15.43±0.05^b
Powder	41.71±0.62^c	-2.88±0.20^b	9.66±0.47^c

Different supercript letters in the same column indicate significantly different means among samples according to Duncan test at P<0.05

Table 3. Changes of oil yield

Form of kaffir lime leaves	Oil yield (% b/b)	Decreasing of oil yield (%)
Fresh	4.16±0.15^a	-
Whole dried	1.94±0.06^b	53.36
Powder	1.22±0.07^c	70.67

Different capital letter within each column indicate significantly different means among samples according to Duncan test at P<0.05.
Decreasing of percentage oil yield was calculated based on oil yield of fresh kaffir lime leaves.

1991; Singh and Sagar, 2010). High temperature could lead to the replacement of magnesium in the chlorophyll by hydrogen, thereby converting chlorophylls to pheophytins (Rudra *et al*., 2008).

Essential oil yield

Essential oil yield of kaffir lime leaves did decrease significantly after drying ($p<0.05$), given by 50% of that observed in the fresh form (Table 3). Pulverization process also induced the decrease of the oil yield significantly ($p<0.05$), giving the 70.67% loss of the initial yield. Similar results were also reported by Buchaillot *et al.* (2009), that the decrease of the oil yield of lemon myrstle leaves up to 50%, was occurred after dried by fluidized bed dryer at temperatures of 30°C, 40°C or 50°C. The same phenomenon was reported by Ebadi *et al.* (2015) that a decrease of essential oil yield of lemon verbena leaves was occurred during drying process, either with an oven dryer or a vacuum dryer. However in peppermint leaves drying by hot air or convective dryer,maximum oil yield was obtained at 70°C (Torki-Harchegani *et al.*, 2016). A damage of grandular trichomas by drying process was suspected to be the cause of a decrease of essential oil yield, causing the release of volatile compounds (Ebali *et al.*, 2015).

In fact, the decrease of oil yield in powder form was higher than whole dried form. This phenomenon was allegedly happened due to the pulverization process. Damaged cells or oil glands resulted from the process could induce a higher rate of the oil evaporation in powder form materials.

Changes of essential oil

The essential oil of fresh kaffir lime contained 18 volatile compounds (Table 4). They were grouped into 11 hydrocarbon monoterpenes, 4 hydrocarbon sesquiterpenes, 1 oxygenated sesquiterpene and 2 esters. The dried whole leaves contained 20 volatiles, consisted of 6 hydrocarbon monoterpenes, 6 oxygenated monoterpenes, 4 hydrocarbon sesquiterpenes, 1 oxygenated sesquiterpene, 2 esters and 1 diterpene. The essential oil of the dried leaves in powder form had 19 volatiles, divided into 7 oxygenated monoterpenes, 8 hydrocarbon sesquiterpenes, 1 oxygenated sesquiterpene, 1 ester and 2 diterpenes.

Citronellal was the major component of fresh leaves essential oil. Its concentration was 468.40 µg/g (dry basis), almost 78% of essential oil content. This result was in agreement with other researchers (Sato *et al*., 1990; Wijaya, 1995; Jantan *et al*., 1996; Tinjan and Jirapakkul, 2007; Wijaya, 2010). Citronellal gives a unique aroma which is close to a mix of citrus and lemongrass aroma (Sato *et al*., 1990). Other volatile compounds in the essential oil of fresh leaves were citronellol (4.14%), linalool (3.02%), and sabinene (2.10%).

The volatiles concentration in dried whole leaves decreased to approximately 50% the level in fresh leaves, except phytol derivatives compounds. This can be happened due to the evaporation during drying, which was supported by the reduction of leaves size into powder by pulverization (Table 4). Therefore, the pulverization of kaffir lime leaves induced the decrease of 61% of essential oil concentration, even though the concentration of several sesquiterpene hydrocarbons and phytol derivates compounds increased. The highest decrease was occurred on monoterpene hydrocarbons.

The monoterpene hydrocarbons detected in fresh kaffir lime leaves were sabinene, β-pinene, myrcene, limonene, β-(E)-ocimene, and isoterpinolen. Drying process induced the decrease of each compound significantly ($p<0.05$), reached 50% compared to that from the fresh form (Table 4). The decrease of total monoterpene hydrocarbons in the dried whole leaves was 58.39%, compared to those in the fresh leaves. On the other hand, the monoterpene hydrocarbons were not detected in the powder dried leaves. Other research conducted by Sellami *et al.* (2011)

Table 4. Volatiles composition in fresh leaves, dried whole leaves and powdered dried leaves

No	Compounds	LRI exp	LRI ref	Concentration (µg/g db)		
				Fresh leaves	Whole dried leaves	Powdered dried leaves
1	Sabinene	978	975	12.63±1.32^a	5.55±1.92^b	n.d.^{c}
2	*β*-Pinene	992	980	0.66±0.07^a	0.28±0.12^b	n.d.^{c}
3	Myrcene	993	990	4.18±0.42^a	1.62±0.50^b	n.d.^{c}
4	Limonene	1032	1030	0.58±0.01^a	0.23±0.11^b	n.d.^{c}
5	*β*-(*E*)-Ocimene	1051	1050	2.05±0.32^a	0.77±0.21b	n.d.^{c}
6	*cis*-Linalool oxide	1087	1072	0.92±0.61^{ab}	0.28±0.16^c	1.63±0.29^a
7	Isoterpinolene	1103	1088	0.52±0.25^a	0.14±0.04^{ab}	n.d.^{c}
8	*trans*-Linalool oxide	1093	1088	n.d.^{b}	n.d.^{b}	0.71±0.14^a
9	Linalool	1104	1098	18.16±0.31^a	9.51±2.87^c	14.54±0.12^{ab}
10	*cis*-Rose oxide	1115	1111	n.d.^{b}	n.d.^{b}	0.17±0.02^a
11	Isopulegol	1153	1146	0.41±0.03^b	0.54±0.12^b	2.38±0.04^a
12	Citronellal	1160	1153	468.40±2.67^a	180.13±27.16^b	127.45±51.31^c
13	iso-Isopulegol	1165	1159	n.d.^{a}	0.61±0.66^a	n.d.^{a}
14	Citronellol	1232	1228	24.86±6.64^a	9.04±1.89^c	15.77±0.75^{ab}
15	Citronellyl acetate	1356	1352	6.45±0.13^a	3.38±0.30^b	6.24±0.06^a
16	Neryl acetate	1367	1361	1.87±0.12^a	0.90±0.06^b	n.d.^{c}
17	*α*-Copaene	1386	1376	n.d.^{b}	n.d.^{b}	3.50±0.11^a
18	*β*-Cubebene	1399	1388	n.d.^{b}	n.d.^{b}	1.95±0.25^a
19	(*E*)-Caryophyllene	1432	1418	3.62±1.44^b	2.35±0.49^b	14.81±1.85^a
20	*α*-Humulene	1466	1454	0.43±0.11^b	0.19±0.06^b	1.59±0.29^a
21	Germacrene D	1493	1481	n.d.^{b}	n.d.^{b}	0.88±0.22^a
22	Bicyclogermacrene	1508	1500	2.10±0.69^b	1.14±0.03^b	6.48±1.33^a
23	*α*-(*E*,*E*)-Farnesene	1512	1505	n.d.^{b}	n.d.^{b}	2.09±0.41^a
24	*δ*-Cadinene	1533	1523	0.43±0.17^b	0.34±0.05^b	3.28±0.77^a
25	(*E*)-Nerolidol	1569	1563	1.71±0.02^{ab}	0.52±0.05^c	2.69±1.06^a
26	(6*E*,10*Z*)-Pseudophytol	2025	2018	n.d^b	0.32±0.20^b	8.18±4.53^a
27	(6*Z*,10*E*)-Pseudophytol	2036	2031	n.d^b	n.d.^{b}	1.19±0.81^a
	Monoterpene hydrocarbons			20.62±2.38^a	8.58±2.81^b	n.d.^{c}
	Oxygenated monoterpenes			521.07±3.64^a	200.09±0.20^b	162.66±0.81^b
	Sesquiterpene hydrocarbons			6.58±2.40^b	4.01±0.90^b	34.57±5.25^a
	Oxygenated sesquiterpenes			1.71±0.25^{ab}	0.51±0.05^b	2.69±1.06^a
	Esters			8.32±0.25^a	4.28±0.36^b	6.24±0.06^a
	Diterpene (phytol derivatives)			n.d.^{b}	0.32±0.20^b	9.37±5.34^a

Different capital letters in the same row indicate significantly different means among samples according to Duncan test at P<0.05; n.d : not detected (mark as "0" for statistical analysis); data were obtained from 2 replicates
Linear retention index experiment (LRI exp) from GC-MS determination on DB-5 column.
Linear retention index reference (LRI ref) from literature (Adams, 2009).

also reported a decrease in the concentration of monoterpene hydrocarbons from 7.55% to 5.26% on *L. nobilis* leaves which were dried by oven at 45°C.

The same trend was occurred on the concentration of oxygenated monoterpenes, either in the whole or powder form of dried kaffir lime leaves. The concentrations of oxygenated monoterpenes in both types of dried leaves decreased by 60.78% and 67.28% respectively. The significant decrease ($p<0.05$) was occurred on linalool, citronellal, and citronellol (Table 4), and on the additional compounds, detected only in the powder dried leaves, i.e. trans-linalool oxide and cis-rose oxide.

Citronellal as the main component in the fresh kaffir lime leaves decreased by 61.54% after drying. This finding is different from that was investigated by Jirapakkul *et al.* (2013) and Tasirin *et al*. (2014), with no significant change. After pulverization process, the citronellal had a further decrease by 29.24% compared to that of the dried whole leaves or by 72.79% to the fresh ones. This condition could occur due to the evaporation and chemical changes. The chemical changes of citronellal might be due to cyclization to isopulegol, iso-isopulegol, neoiso-isopulegol and hydrogenation to citronellol (Sell, 2003; Leonardao *et al*., 2007). This is proven by the

significant increase of isopulegol in the powdered dried leaves, if compared to the dried whole leaves. On the other hand iso-isopulegol only detected in the dried whole ones.

The sesquiterpene hydrocarbons detected in the fresh kaffir lime leaves were β-caryophyllene, α-humulene, bicyclogermacrene, δ-cadinene. The concentration of each compound in the whole dried was not changed significantly ($p>0.05$). This was probably due to the drying process did not significantly affect the sesquiterpene hydrocarbons. This was reinforced by the concentration of sesquiterpene hydrocarbons which were not significant between in the fresh leaves and in the dried whole leaves (Table 4). This phenomenon was suggested that the membranes might selectively be more permeable to certain volatiles or separate compartments for the synthesis of emitted volatiles and stored substances (Gershenzon *et al.*, 2000). Otherwise, pulverization may caused the increase of sesquiterpene hydrocarbons (Table 4), reached 7.62 times compared to the whole dried leaves. Several novel sesquiterpene hydrocarbons compound were found, those were α-copaene, β-cubebene, germacrene D and α-(E,E)-farnesene. The changes of volatile compounds were due to the formation of new compounds through oxidation, esterification, glycoside hydrolysis, and/or other processes during drying and pulverization (Ghasemi Pirbalouti *et al.*, 2013).

It is interesting that nerolidol was the only oxygenated sesquiterpene, present either in the fresh, the whole or the powder forms of dried leaves. It seems that the drying process did not influence the concentration of nerolidol. The concentration of nerolidol in the powder ones increased significantly by 4.27 times compared to the whole ones.

The phytol derivates, such as (6E,10Z)-pseudo phytol and (6Z,10E)-pseudo phytol, were detected either in the dried whole leaves or in the powder dried leaves. Their concentrations were not significantly different in both dried leaves.

The above description proves that the decrease of volatile compounds in the dried leaves could be happened due to the evaporation and/or the thermal oxidation process occurred during drying. Further, Moyler (1994) suggested that the mechanism that allows volatile compounds dragged to the leaves surface and subsequently evaporated simultaneously with water contained in the leaves. The fact that the concentration of monoterpene hydrocarbons decreased higher than other terpenes in the dried herbs could be explained by the evidence of monoterpene hydrocarbons storage in cell at the gland oil near the leaves surface (Asekun *et al.*, 2007). Furthermore, the oxidation and re-arrangement of the component could be happened more susceptible in the powder form of dried leaves due to the cell and oil glands damage by the size reduction process during pulverization, and this induced its loss by evaporation or the formation of novel compounds by oxidation (Baritaux *et al.*, 1992; Sellami *et al.*, 2011).

Conclusion

The oven drying at 50°C for 12 hours and pulverization process altered the yield and the composition of volatile compounds of fresh kaffir lime leaves. Citronellal as a main volatile of fresh leaves was significantly decreased, whereas oxigenated volatile compounds were present after the drying and their number were increased after the following pulverization such as cis-rose oxide, trans-linalool oxide, α-copaene, β-cubebene, α-(E,E)-farnesene. Furthermore, the pulverization gave the increase of sesquiterpene hydrocarbons due to the formation of several novel sesquiterpene hydrocarbons detected in this study. This volatiles composition change may alter the flavor of fresh kaffir lime leaves as a result of drying process at a relatively low temperature in a long time with the following pulverization process.

Acknowledgments

Authors gratefully thank to the Center for Appropriate Technology Development, Indonesian Institute of Sciences and Indesso Aroma Co. in Indonesia for a support.

References

Adams, R. P. 2009. Identification of Essential Oil Components by Gas Chromatography/Quadrupole Mass Spectrometry. 4th ed., p. 54-788. Carol Stream: Allured Publishing Corporation.

AOAC. 2000. Official methods of analysis of AOAC international, 17th ed., p.1. Maryland, USA: AOAC International.

Argyropoulos, D. and Muller, J. 2011. Effect of convective drying on quality of lemon balm (*Melissa officinalis* L.). Procedia Food Science 1: 1932-1939.

Arslan, D., Ozcan, M. M. and Menges, H. O. 2010. Evaluation of drying methods with respect to drying parameters, some nutritional and colour characteristics of peppermint (*Mentha x piperita* L.). Energy Conversion and Management 51: 2769-2775.

Asekun, O. T., Grierson, D. S. and Afolayan, A. J. 2007. Effects of drying methods on the quality and quantity of the essential oil of *Mentha longifolia* L. Subsp. *capensis*. Food Chemistry 107: 995-998.

Baritaux, L. O., Richard, H. and Touche, J. M. 1992. Effects of drying and storage of herbs and spices on the essential oil. Part I. Basil, *Ocimum basilicum* L. Flavour and Fragrance Journal 7: 267-271.

Braga, N. P., Cremasco, M. A. and Valle, R. C. C. R. 2004. Effects of fixed bed drying on the yield and composition of essential oil from long pepper (*Piper hispidinervium* C. DC.) leaves. In Silva, M. A. and Roscha, S. C. S. (Eds). Proceedings of the 14th International Drying Symposium, p. 1906 – 1913. Sao Paulo, Brazil: UNICAMP.

Buchaillot, A., Caffin, N. and Bhandari, B. 2009. Drying of Lemon Myrtle *(Backhousia citriodora)* Leaves: Retention of Volatiles and Color. Drying Technology 27: 445-450.

Chua, K. J. and Chou, S. K. 2003. Low-cost drying methods for developing countries. Trends Food Science Technology 14: 519-528.

Diaz-Maroto, M. C., Pérez-Coello, M. S. and Cabezudo, M. D. 2002. Effect of drying method on the volatiles in bay leaf (*Laurus nobilis* L.). Journal of Agricultural Food Chemistry 50: 4520-4524.

Diaz-Maroto, M. C., Pérez-Coello, M. S., Vinas, G. M. A. and Cabezudo, M. D. 2003. Influence of drying on the flavour quality of spearmint (*Mentha spicata* L.). Journal of Agricultural Food Chemistry 50: 1265-1269.

Ding, S. H., An, K. J., Zhao, C. P., Li, Y., Guo, Y. H. and Wang, Z. F. 2011. Effect of drying methods on volatiles of Chinese ginger (*Zingiber officinale* Roscoe). Food Bioproducts Processing 273: 1-10.

Ebadi, M.T., Azizi, M., Sefidkonc, F. and Ahmadi, N. 2015. Influence of different drying methods on drying period, essential oil content and composition of *Lippia citriodora* Kunth. Journal of Applied Research on Medicinal and Aromatic Plants 2: 182-187.

Ghasemi Pirbalouti. A., Mahdad, E. and Craker, L. 2013. Effects of drying methods on qualitative and quantitative properties of essential oil of two basil landraces. Food Chemistry 141: 2440-2449.

Gershenzon, J., McConkey, M.E. and Croteau, R.B. 2000. Regulation of monoterpene accumulation in leaves of peppermint. Plant Physiology 122: 205-214.

Gonnet, J. F. 1998. Colour effects of co-pigmentation of anthocyanins revisted—1. A colorimetric definition using the CIELAB scale. Food Chemistry 63: 409-415.

Gross, J. 1991. Pigments in Vegetables, p. 47-48. New York: An Avi Book, Van Nostrand Reinhold.

Jantan, I., Ahmad, A. S., Ahmad, A. R., Ali, N. A. and Ayop, N. 1996. Chemical composition of some citrus oils from Malaysia. Journal Essential Oil Research 8: 672-682.

Jirapakkul,W., Tinchan, P. and Chaiseri, S. 2013. Effect of drying temperature on key odourants in kaffir lime (*Citrus hystrix* D.C., Rutaceae) leaves. International Journal of Food and Science Technology 48: 143-149.

Lenardao, E. J., Botteselle, G. V., de Azambuja, F., Perin, G. and Jacob, R. G. 2007. Citronellal as key compound in organic synthesis. Tetrahedron 63: 6671-6712.

Llewellyn, C. A., Mantoura, F. C. and Brereton, R. G. 1990. Products of chlorophyll photodegradation: 2. Structural identification. Photochemistry Photobiology 52: 1043-1047.

McGuire, R. G. 1992. Reporting of objective colour measurement. Hortic Science 27: 1254-1255.

Moyler, D. A. 1994. Spices-Recent advances. In Charalambous, G. (Ed). Spices, Herbs and Edible Fungi, p. 1-70. London: Elsevier Science.

Naidu, M. M., Vedashree. M., Satapathy. P., Khanum. H., Ramsamy. R. and Hebbar, H. U. 2016. Effect of drying methods on the quality characteristics of dill *(Anethum graveolens)* greens. Food Chemistry 192: 849-856.

Nor, M. O. 1992. Development and application of Citrus hystrix flavor for food. Proceedings of Asean Workshop on the Production of Natural Flavours for Food. Kuala Lumpur: Malaysian Agricultural Research and Development Institute.

Pilatti, D., Palú, F., da Silva, E. A. and Johann, G. 2016. Evaluation of a concentrated parameters mathematical model applied to drying of yerba mate leaves with variable mass transfer coefficient. Applied Thermal Engineering 105: 483-489.

Pudil, F., Wijaya, C. H., Janda, V., Volfova, J., Valentova, H. and Pokomy, J. 1998. Changes in *Citrus hystrix* oil during autooxidation. In Contis, E.T. (Ed). Food Flavors: Formation, Analysis and Packaging Influences, p. 707-718. Amsterdam: Elsevier Science.

Raksakantong, P., Siriamornpun, S. and Meeso, N. 2011. Effect of drying methods on volatile compounds, fatty acids and antioxidant property of Thai kaffir lime (*Citrus hystrix* D.C.). International Journal Food Science Technology 47: 1-10.

Rudra, S. G., Singh, H., Basu, S. and Shivhare, U. S. 2008. Enthalpy entropy compensation during thermal degradation of chlorophyll in mint and coriander puree. Journal of Food Engineering, 86: 379-387.

Sarosi, Sz., Sipos, L., Kokai, Z., Pluhar, Zs., Szilvassy, B. and Novak, I. 2013. Effect of different drying techniques on the aroma profile of *Thymus vulgaris* analyzed by GC–MS and sensory profile methods. Industrial Crops and Products 46: 210-216.

Sato, A., Asano, K. and Sato, T. 1990. The chemical composition of *Citus hystrix* DC (Swangi). Journal of Essential Oil Research 2: 179-183.

Schwartz, S. J. and Lorenzo, T. V. 1991. Chlorophyll stability during continuous aseptic processing and storage. Journal of Food Science 56: 1059-1062.

Sell, C. S. 2003. A Fragrant Introduction to Terpenoid Chemistry, p. 60-62. Cambridge: The Royal Society of Chemistry.

Sellami, I. H., Wannes, W. A., Bettaieb, I., Berrima, S., Chahed, T., Marzouk, B. and Limam, F. 2011. Qualitative and quantitative changes in the essential oil of *Laurus nobilis* L. leaves as affected by different drying methods. Food Chemistry 126: 691-697.

Serpen, A. and Gokmen, V. 2009. Evaluation of the Maillard reaction in potato crips by acrylamide, antioxidant

capacity and color. Journal of Food Composition and Analysis 22: 589-595.

Singh, U. and Sagar, V. R. 2010. Quality characteristics of dehydrated leafy vegetables influences by packaging materials and storage temperature. Journal of Science Industrial Research 69: 785-789.

Szumny, A., Figiel, A., Ortiz, A. G. and Carbonell-Barrachina, A. A. 2010. Composition of rosemary essential oil *(Rosmarinus officinalis)* as affected by drying method. Journal Food Engineering 97: 253-260.

Tao, Y., Wang, P., Wang, Y., Kadam, S. U., Han, Y., Wang, J. and Zhou, J. 2016. Power ultrasound as a pretreatment to convective drying of mulberry (*Morus alba* L.) leaves: Impact on drying kinetics and selected quality properties. Ultrasonics Sonochemistry 31: 310-318.

Tasirin, S. M., Puspasari, I., Lun, A. W., Chai, P. V. and Lee, W. T. 2014. Drying of kaffir lime leaves in a fluidized bed dryer with inert particles: Kinetics and quality determination. Industrial Crops and Products 61: 193-201.

Tinjan, P. and Jirapakkul, W. 2007. Comparative study on extraction methods of free and glycosidically bound volatile compounds from Kaffir lime leaves by solvent extraction and solid phase extraction. Kasetsart Journal 41: 300-306.

Torki-Harchegani, M., Ghanbarian, D., Pirbalouti, A. G. and Sadeghi, M. 2016. Dehydration behavior, mathematical modeling, energy efficiency and essential oil yield of peppermint leaves undergoing microwave and hot air treatments. Renewable and Sustainable Energy Reviews 58: 407-418.

Usai, M., Marchetti, M., Foddai, M., del Caro, A., Desogus, R., Sanna, I. and Piga, A. 2011. Influence of different stabilizing operations and storage time on the composition of essential oil of thyme (*Thymus officinalis* L.) and rosemary (Rosmarinus officinalis L.). Food Science and Technology 44: 244-249.

Wijaya, C. H. 1995. Oriental natural flavor: Liquid and spray—dried flavor of "Jeruk purut" (*Citrus hystrix* DC) leaves. In Charalambous, G. (Ed). Food Flavors: Generation, Analysis and Process Influence, p. 235-248. Amsterdam: Elsevier Science.

Wijaya, C. H. 2010. Compositional analysis: Asia-Indonesia. In Sawamura M (Ed.). Citrus Essential Oils (Flavor and Fragnance), p. 96-104. New Jersey: John Wiley and Sons Inc.