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Physical and Chemical Changes of Seven Selected Herbs Used as Herbal Bath Affected by Different Drying Methods

Rose Fazila Md Zuki[1,2], Mohd Firdaus Ismail[1]* and Julia Abdul Aziz[3]

[1]*Department of Crop Science, Faculty of Agriculture, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia*

[2]*Department of Agriculture Malaysia, Presint 4 Pusat Pentadbiran Kerajaan Persekutuan, 62624 Putrajaya, Malaysia*

[3]*Biodiversity Unit, Institute of Bioscience, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia*

ABSTRACT

The effect of oven drying (OD) at 50±5°C, sun drying (SD), and fresh leaves (control) of seven selected herbs used in herbal preparation as herbal baths were evaluated. Herbal baths (HB) involve mixing herbs into water and boiling them or simply immersing them in the mixture during a regular bath. The herbs selected were leaves of torch ginger, greater galangal, pandan, citronella grass, henna, betel leaves, and kaffir lime leaves and fruits. The herbs were planted in a plot at the Institute of Bioscience, Universiti Putra Malaysia, Malaysia. Physical changes such as HB herbs' colour, aroma, and chemical composition were evaluated. The hydro distillation method was used for the extraction process of HB herbs, where it produced essential oils (EO), essential water (EW), and boiling water (BW), and their chemical composition was determined by gas chromatography-mass spectrometry. As a result, the OD herbs possessed brighter and more attractive colours compared to the SD method, which was dull and pale. The colour of EO was yellow, colourless for EW, and reddish for BW. Additionally, OD herbs possessed 80% and only 50% of the scent strength of the SD herbs from extracting fresh herbs. The fresh and dried HB herbs contained chemical constituents mostly from the terpene group. Herbal preparation developed using all treatments retained an appreciable amount of chemical composition studied, thus having the potential for commercial purposes.

Keywords: Chemical compositions, drying method, essential oil, herbal bath, hydro distillation method

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E-mail addresses:
rosefazila@doa.gov.my (Rose Fazila Md Zuki)
mohd.firdaus@upm.edu.my (Mohd Firdaus Ismail)
julia_a@upm.edu.my (Julia Abdul Aziz)
*Corresponding author

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INTRODUCTION

An herbal bath (HB) adds specific herbs to a hot bath with a specific goal. Many Malaysians believe HB could revitalise their bodies, improve blood circulation, and enhance their skin's beauty, radiance, and health. Most herbs used for HB are fragrant, common knowledge, and easily accessible (Tungsukruthai et al., 2018). Citronella grass, pandan, henna, betel leaf, torch ginger, greater galangal, and kaffir lime were the seven herbs employed in this investigation to produce this HB product (Zaman et al., 2007). Since most plant components can be used to extract or acquire volatile oils, the herb species listed here are also known as aromatic herbs (Solórzano-Santos & Miranda-Novales, 2012).

There are many different types of HB practises, including leaf baths, flower baths, and milk baths, that are dependent on ethnicity (Alsarhan et al., 2021; Hishamshah et al., 2010) as well as a variety of bathing techniques, including steam baths or saunas, regular baths with herbal water as the final wash, soaking the entire body in herbal water in the bathtub, mixing regular baths with herbal water, and gently massaging the entire body with EO (Mohamed & Hj. Bidin, 2012; Jamal et al., 2011; Li et al., 2006; Zaman et al., 2007). It requires 20 to 30 min to take a bath once or twice a day, three times a week (Panyaphu et al., 2011), so the nutrients from the herbs can be easily absorbed through the skin pores (Li et al., 2006). Although there has not been much scientific research on HBs, the chemical content of each herb convinces customers that HB practices are beneficial for enhancing health.

In HB practises, there are typically odd-numbered combinations of herbs, such as 5, 7, 9, or 11. The dosage of these many herbs varies depending on the practitioner and is not precise (Li et al., 2006; Zaman et al., 2007). Fresh herbs have a limited shelf life for commercial uses, and if they do not undergo processing immediately, they lose their stability. Therefore, dried herbs are also an alternative for producing HB because they are always accessible and usually cost less. However, they lack the aromatic qualities of fresh herbs because of the loss or oxidation of volatile oils through drying or other processing procedures (King, 2006).

To extend the shelf life of food products, people frequently dry foods, including meat, herbs, and fruits (Mashkani et al., 2018), to inhibit the activities of microbes and bacteria when there is no water present; this procedure employs air and heat to eliminate moisture from the product (Thamkaew et al., 2020). It also relies on the factors that affect drying, such as the climate, drying period, spread thickness, and relative humidity. Three categories of drying methods: (1) thermal, (2) chemical, and (3) special, can be used to dry leaves. Thermal drying refers to natural methods that use sunlight, shade, and wind. In contrast, special drying refers to artificial methods that use modern technologies like vacuum, microwave, dryer, and oven (Babu et al., 2018). The drying process will impact the product's chemical composition, colour, fragrance, and durability.

An optimised leaf drying process aims to maintain a high level of nutrients when the leaves are fresh while achieving the required end moisture content (Tasirin et al., 2014). So, choosing the correct drying method is crucial to guaranteeing the quality of the final product. This study used two different drying techniques: (1) oven drying (OD) and (2) sun drying (SD) to dry the plant leaves. In the OD technique, the sample is dried at a predetermined temperature using low-temperature convection air or thermos-gravimetric technology (Ahmed et al., 2013). Compared to the SD, this drying method is weather-independent, quicker, continuous, cleaner, and sanitary. However, the SD method approach is inexpensive and appropriate for bulk densities, particularly for commercial purposes (Özgüven et al., 2019).

According to several studies, dramatic drying alters the active components in fresh herbs' EO. However, the effect varies based on the plant species, drying temperature, and drying period. According to Mashkani et al. (2018), OD and vacuum obtained the highest EO content in *Thymus daenensis*. Additionally, according to Sefidkon et al. (2006), OD, shade drying, and SD increased the concentration of EO in *Satureja hortensis* L. leaves. Contrarily, Filho et al. (2018) claimed that the drying procedure did not impact the amount of EO in the dried parsley leaves.

The chemical composition in herbal leaves is believed to promote human health in several ways, including reviving the body, reducing stress, healing illnesses, and enhancing skin quality, even though the research on the impact of HB practises on human health is quite limited. Therefore, this experiment was conducted to determine the physical and chemical changes in HB herbs due to the impact of various drying processes to ensure and manage the herbs' quality. Additionally, this study will offer details on the results of using more than three herbs in a single product for potential applications in cosmetics.

MATERIALS AND METHODS

Description of the Study Area

The studied area for field and laboratory work was at the Institute of Bioscience, Universiti Putra Malaysia (IBS, UPM), Serdang, Selangor (30°00'56.3"N, 101°72'32.69"E). The climate of the location is mainly hot and damp; temperature ranges between 28–34°C with 65.5–87% relative humidity, received between 95.19–1,611.85 μmol of light density in the daytime and experiences rainfall annually about 2,000–2,500 mm means a month (Jabatan Meteorologi Malaysia [METMalaysia], 2020).

Collection and Preparation of Plant Materials

There were seven types of herbs used in the study: *Etlingera elatior* (torch ginger), *Alpinia galanga* (greater galangal), *Pandanus odours* (pandan), *Cymbopogon nardus* (citronella grass), *Lawsonia inermis* (henna), *Citrus hystrix* (kaffir lime), and *Piper betle* (betel leaves). The herbs were grown from cuttings at IBS, UPM. The herbs were selected based on their long-term usage as HB in traditional Malaysian practices. Fresh herbal leaves and fruits (kaffir lime only) were collected between 8–10 a.m. (Chan et al., 2008; Mashkani et al., 2018). After collection, the leaves and fruits were repeatedly washed with tap and distilled water and air-dried for 72 hr at room temperature (Gebrehiwot et al., 2016). The herbs were cut into ±2.5 cm (1") or sliced into small parts (Jamal et al., 2011).

Drying Treatments

Two drying methods were used: oven-dried (OD) at 50±5°C and sun-dried (SD), with fresh samples as control. For the SD, the sample was spread on the cloth, while for OD, the sample was spread on the tray. The herbs were dried until the moisture content reached ±10% (Mashkani et al., 2018) or until they lost ±85% of their weight (Babu et al., 2018) or the dried leaves felt crunchy when crushed using a hand.

Preparation of HB Herbs

Two samples of HB herbs were prepared: fresh and dried samples. For the fresh herbs sample, 255 g of combined herbs (41 g of pandan leaves, 14 g of henna leaves, 22 g of kaffir lime leaves, 25 g of kaffir lime fruit, 40 g of betel leaves, 25 g of torch ginger leaves, 22 g of greater galangal leaves, and 66 g of citronella grass leaves) were prepared. The sample was air-dried in a closed room for one whole night before undergoing the extraction process using the hydro distillation method. For the dried herbs sample, the OD and SD herbs were weight 2 g for each herb (pandan, henna, kaffir lime leaf, kaffir lime fruit, betel leaf, torch ginger, and greater galangal) and 6 g of citronella grass. The samples were combined, wrapped and tied neatly in muslin cloth. One pack of combination dried herbs equal to 20 g. The samples were replicated four times for each sample (Mahanom Jr. et al., 1999; Tamboli & Bhong, 2018).

Physical Properties of HB Herbs and EO

Two physical properties measured manually are colour and scent, with fresh samples as a control. For colour analysis, the Munsell Colour Chart was used as standard. In contrast, for scent analysis, the strength was recorded in percentages between 0–100% using the fresh herbal leaf as control (Prof. Ir. Dr Yus Aniza Yusof, Department of Process and Food Engineering, Faculty of Engineering, UPM, personal communication in October 2018). Ten IBS UPM employees, who have actively produced these HB herbs for over three years, assessed fresh and dried HB herbs for this sensory investigation. Each sample was examined, felt, and smelled individually by the analyst manually in an air-circulated chamber.

Extraction of HB Herbs by Hydro Distillation Method

Fresh and dried HB herb samples weighing 255 and 80 g were put in a 20 L round bottom glass flask separately. Next, the flask was filled with 5 L of distilled water and mixed thoroughly. Then, the flask was fitted with Clevenger's apparatus, a glass condenser, heated using a heating mantle, and hydro distilled at atmospheric pressure for 4 hr. The EO was isolated from the aqueous layer using 100 ml chloroform (Chemiz, Malaysia) and filtered using Whatman No. 1 filter paper. Finally, the extraction process products, EO, EW, and BW, were stored in the refrigerator at 4°C to keep them from being volatile. The chemical compositions of EO, EW, and BW were determined by gas chromatography-mass spectrometry (GC-MS).

$$\text{Yield (\%)} = \frac{\text{Volume of essential oil (ml)}}{\text{Weight of sample (g)}} \text{ X } 100\% \quad [1]$$

GC-MS Analysis of EO, EW, and BW of HB Herbs

EO, EW, and BW from fresh and dried HB herbs were analysed for their chemical compositions and quantity after dissolving in methanol using GC-MS by Shimadzu CG-17A (Shimadzu Corporation, Japan), which was directly coupled with QC-2010 high polar fused silica capillary column (Zebron™ ZB5-ms 30 m x 0.25 mm ID x 0.25 µm film thickness, Phenomenex Corporation, USA). The gas carrier was helium, delivered through split injection at a volume of 0.3 µl ratio, with a normal mode at a flow rate of 6.0 ml/min and head pressure of 49.7 kPa. The column temperature was held at 40°C for 5 min. The chromatographic condition was helium, 0.3 µl/min and 49.7 kPa; injector and interface temperatures were 220 and 250°C, respectively. Additionally, the column temperature was maintained at 40°C for 5 min. Methanol was used as the solvent to dissolve the oil.

EO, EW, and BW Identification and Quantification

The quality and quantity of EO, EW, and BW compounds that GC-MS detected were recorded in chromatograms. The quantitative data were determined from the peak total percentage areas of the analysed EO, EW, and BW extracted from fresh and dried HB herbs. The individual compounds were identified by comparing their relative retention indices, which were calculated using the *n*-alkanes formula and compared with the National Institute of Standards and Technology libraries (NIST).

Statistical Analysis

The results were statistically analysed using analysis of variance (ANOVA) in the statistical analysis system (SAS) programme (version 9.4). The data were expressed as means of four replications. Significant differences among the treatment mean at $p<0.05$ were determined by the least significant difference (LSD).

RESULTS AND DISCUSSION

Effect of Drying Method on Physical Properties of HB Herbs

The colour and scent of herb leaves are important for product marketing because HB herb is for external use. Therefore, product freshness and appearance are very important to meet consumers' demands.

Figure 1 shows the colour changes analysed in fresh and dried herbal preparations. The drying treatments caused significant colour changes for all herb leaves. Compared to fresh leaves, the herb's colour changes were reduced from slightly to significantly by the OD and SD methods.

The results showed that henna, torch ginger, betel leaves, and kaffir lime fruit did not significantly differ in colour between OD and SD methods but were darker than fresh samples. It indicated that OD and SD could not preserve the sample's green colouration. While there were significant colour differences between the OD and SD methods for greater galangal, kaffir lime leaves, citronella grass, and pandan leaves, in comparison to OD and fresh herbs, SD herbs exhibited a darker hue, indicating that OD herbs appeared more similar to the fresh samples.

The benefits of the OD perform over the SD method are supported by these results, which corroborated earlier reports in the literature on several types of herbs (Arslan & Özcan, 2010; Özgüven et al., 2019; Telfser & Galindo, 2019). The OD method at lower drying temperatures (<50°C) is particularly beneficial for herbs containing active compounds subjected to thermal degradation. Most nutritional and external properties are preserved since OD is performed at low temperatures and without oxygen (Laurence et al., 2019). In this study, OD herbs maintained a consistent and bright colour compared to SD herbs, which were dull and pale. It is a result of the colour degradation caused by the possible destruction of pigments like chlorophyll and anthocyanin during the drying process, showing that the SD approach has a bigger impact on colour degradation than the OD method (Thamkaew et al., 2020).

Figure 1. Herbal bath herbs colour changes before and after drying treatment: (A) fresh herbs, (B) oven-dried herbs and (C) sun-dried herbs

Figure 1. (Continue)

Table 1
Herbs' colour and scent changes before and after the drying process

No.	Analysis	Colour changes			Scent changes: Percentage (%) from the original smell of fresh leaves maintained	
	Drying process/ Herbal bath herbs	Fresh	OD	SD	OD	SD
1.	Kaffir lime fruit	5Y, 9/6	5Y, 7/10	5Y, 5/12	70–80	70
2.	Betel leaves	6GY,5/8	5YR, 1/4	5Y, 1/2		0

Table 1 *(Continue)*

No.	Analysis	Colour changes			Scent changes: Percentage (%) from the original smell of fresh leaves maintained	
	Drying process/ Herbal bath herbs	Fresh	OD	SD	OD	SD
3.	Greater galangal	6GY, 6/10	5GY, 6/10	5Y, 5/4		60
4.	Torch ginger	6GY, 7/10	5Y, 4/10	5Y, 3/6		60
5.	Kaffir lime leaves	6GY, 7/8	6GY,9/4	5GY, 3/8		60
6.	Citronella grass	6GY, 8/8	5GY, 7/6	5GY, 9/2		70
7.	Pandan	6GY, 8/12	5Y, 7/8	5GY, 6/4		30
8.	Henna leaves	6GY, 6/8	5YR, 4/8	5YR, 2/2		10

Note. Herbs colour analysis using Munsell Colour Chart for hue 5 yellow (5Y), 5 yellow-red (5YR), 5 green-yellow (5GY), and leaf hue 6 green-yellow (6GY), and scent analysis due to the effect of oven-dried (OD) and sun-dried (SD) methods

The colours of all dried herb leaves were compared using the Munsell Colour Chart for the hues 5Y, 5YR, 5GY, and leaf hue 6GY. The dried herbs were compared to the fresh herb leaves for the fragrance analysis, where they underwent slight to significant odour changes. Table 1 displays the colour data for fresh and dried HB herbs in terms of hue values and percentages of aroma before and after drying. According to this study, OD herbs retained their bright colour and had a strong, mild aromatic fragrance compared to SD herbs, which were dull and pale and had a less fragrant aroma. All dry herbs' leaves smelled, and compared to fresh herbs' leaves, the OD herbs had 70–80% of the scent of fresh herbs' leaves. Meanwhile, SD herbs also had a slightly smoky scent besides possessing 10–70% of the aromatic fragrance.

The aroma of herbs results from the chemical content found in EO. The higher the chemical content, the stronger the aroma of the resulting EO. The lack of EO aroma is caused by damage to the secretory cells, epidermal cells, or glandular trichomes that store the Eo in the plant (Thamkaew et al., 2020). Therefore, the result indicates that SD causes a higher damage effect than the OD method. Telfser and Galindo (2019) further claim that drying at low temperatures (40–60°C) is preferable for preserving volatile chemicals and effectively prevents the loss of scent. Thus, in their study, OD has been recommended as the optimum dehumidification and dehydration method for dry aromatic plant species by Özgüven et al. (2019).

Effect of Drying Method on the EO, EW, and BW of HB Herbs' Content

Hydro distillation is used in this study to extract EO, EW, and BW from HB herbs. Boiling HB herbs in a pot is a typical practice that produces EO and EW, but all these extracts are combined in BW. Therefore, boiling the HB herbs with most of the pot's top covered is recommended to trap the hot water vapour and ensure that it releases little heat into the atmosphere but returns to the BW instead. Figure 2 shows the colour of fresh and dried EO, EW, and BW of HB herbs obtained from the hydro-distillation extraction process. EO was yellowish, colourless for EW, and reddish for BW. The drying procedures did not produce significant colour changes when extracting HB herbs. The OD or SD methods did not significantly reduce the HB herb's colour changes in extraction compared to fresh leaves.

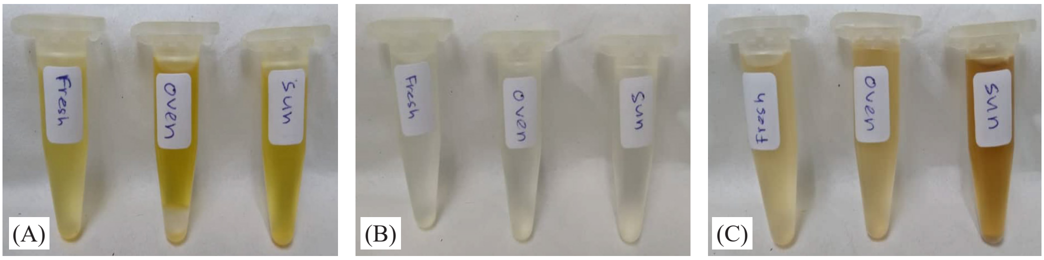

Figure 2. The extraction process of different types of drying of herbal bath herbs using the hydro distillation method produces (A) essential oil, (B) essential water, and (C) boiling water

The extraction was compared using the Munsell Colour Chart for hue 5Y. For the scent analysis, the HB herbs' extractions were compared to the fresh, where they went through slight to very significant odour changes. Table 2 compares the colours and scents of the extracted HB herbs. In contrast to SD herbs, which were darker, the EO and BW colours for fresh and OD herbs were the same. BW's reddish colour might be the result of the effects of henna and betel leaf. For all EW, there were no changes in colour. The BW possessed a strong aromatic fragrance of betel leaves, henna leaves, and a small amount of citronella grass. However, the EO and EW for all drying techniques possessed a mild aromatic, smell-like mixture of citronella grass and kaffir lime fruit. Although the EO and BW of SD herbs were darker, they had a scent similar to fresh and OD herbs but with different strengths.

OD herbs extraction possessed 80% scent strength, while SD herbs only had 50% scent strength from extracting fresh herbs. The drying treatments caused significantly different scents to strengthen for HB herbs' extraction. The OD and SD methods caused a slighter to a greater reduction in the HB herb's extraction of scent strength compared to fresh leaves. Table 3 shows the EO yields produced from the extraction of HB herbs using different drying methods.

Table 2
Comparison of herbal bath (HB) herbs extraction colour and scent

No.	Analysis	Colour changes			Scent changes: Percentage (%) of scent strength from fresh HB extract		
	HB extraction/ Drying process	EO	EW	BW	EO	EW	BW
1.	Fresh	5Y, 9/14	0	5YR, 8/6	100	100	100
2.	OD	5Y, 9/14	0	5YR, 8/6	80	80	80
3.	SD	5Y, 7/14	0	5YR, 6/10	50	50	50

Note. Essential oil (EO), essential water (EW), and boiling water (BW) colour analysis using Munsell Colour Chart for hue 5 yellow (5Y) and 5 yellow-red (5YR) and scent analysis due to the effect of oved dried and sun-dried methods, respectively

According to the result, the drying methods significantly affected EO yields. Fresh herbs produced a higher EO yield than OD and SD HB herbs. The lower production of EO yield was from the SD HB herbs. The yield of EO from the fresh and dried herbs was in the order of fresh herbs oil 88% (v/w) > OD herbs oil 63% (v/w) > SD HB herbs oil 392% (v/w).

Table 3
Effects of different drying methods on essential oil's yield from the herbal bath herbs' extract

Drying method	Yield (%)
Fresh (control)	88%[a]
OD	63%[b]
SD	39%[c]

Note. Essential oil yields from fresh, oven-dried (OD), and sun-dried (SD) herbal bath herbs because of drying methods. Means with the same letters are not significantly different at $p>0.05$ using the least significant difference

More oil content was present from fresh and dried HB herbs that could be extracted, but these compounds were lost or transformed during the processing of hydro distillation and evaporation (Aziz, 2015). So, to minimise EO losses and preserve the distinctive components, drying should be done immediately after harvest. Additionally, post-harvest activities, as well as agricultural practices, have an impact on oil content (Özgüven et al., 2019). The conclusion that the OD technique can boost the concentration of EO isolated by hydro distillation agrees with most prior investigations on various plants.

Additionally, according to Özgüven et al. (2019), different drying techniques result in varying concentrations of certain aromatic herbs, while Mashkani et al. (2018) found that the kind of tissue temperature duration and drying technique typically influenced variations in EO content after drying. Additionally, even if a plant is of the same species, its chemical composition varies due to internal and external factors such as the kind of media used for planting, the type and rate of fertilisation used, the timing of crop harvest, as well as topography and environmental conditions (Aziz, 2015; Tunjung et al., 2015).

In contrast to the OD approach, which employs hot air flows and merely dries the leaf surface, the SD method directly penetrates heat into the leaf cells, and prolonged exposure to it may damage nearly all plant cell walls (Thamkaew et al., 2020). The findings show that the lack of EO in SD herbs results from strong heat factors (>50°C) breaking the membrane cells of oil sacs in herbs. It suggests that, compared to the OD method, the SD method had a more significant impact on HB herbs.

Effect of Drying Method on the EO, EW, and BW of HB Herbs' Chemical Contents

The chemical contents of the plant can be determined by using EO, EW, and BW after they undergo determination techniques such as column chromatography, high-performance liquid chromatography, ultrafiltration, and many other techniques. In this study, the GC-MS technique was used to determine the chemical contents of HB herbs. The use of EO, also known as volatile oils, extracted or derived from most plant parts, including flowers, leaves, buds, fruits, twigs, bark, seeds, wood, and roots (Solórzano-Santos & Miranda-Novales, 2012) is quite common in aromatherapy (Jaradat et al., 2018). According to Patel and Patel (2016), phytochemicals are bioactive chemical compounds used as antimicrobial, antibacterial, and anticancer agents. Herbs are traditionally used for food or medicinal purposes and are known to be phytochemically rich (Guldiken et al., 2018).

The GC-MS analysis of the EO, EW, and BW in this study reveals 42 numbers (Table 4), 22 numbers (Table 5), and 12 numbers (Table 6) of identified compounds, respectively, by comparing their mass spectra with the National Institute of Standards and Technology (NIST) 2005 library of mass spectra and their retention (R) times. So, few unidentified components were present that either no mass spectra could be obtained or the spectrum was too weak to be interpreted. The drying process significantly affected EO compounds. The total oil of the EO chemical components (99.44%, 99.36%, and 99.15%) was isolated from fresh, OD, and SD plants (Table 4). Only 34 and 35 of the 42 identified chemical compounds from new extraction were discovered in OD and SD herbal leaves, respectively. Drying OD and SD herbs produced 8 and 7 newly discovered chemical compounds, respectively.

The highest compound percentage of HB's EO produced was 2,6-octadienal, 3,7-dimethyl-(E), another name is α-citral (fresh: 26.08%, OD: 35.79%, and SD: 24.32%), while the lowest percentage was contributed by tau-cardinal (fresh: 0.19%, OD: 0.31%, and SD: 0.14%). The fresh, OD, and SD HBs had different chemical compositions; while some compounds may be high in OD or SD plants, others have a high proportion in fresh herbs. For instance, OD and SD herbs have higher percentages of naphthalene,1,2,3,4,4a,5,6,8a-octahydro-4a,8-dimethyl-2-(1-methylethenyl)-[2R-(2alpha., 4a. alpha)] (9.86% and 7.91%, respectively) than fresh herbs

(3.01%). Additionally, certain substances, including 1,4-cyclohexadiene and 1-methyl-4-(1-methylethyl), remained the same in composition regardless of the drying process.

According to the results obtained, the five chemical contents that recorded the highest content composition in fresh HB herbs can be considered the main active ingredients of HB products, considering that the sample is a combination of seven types of herbs used as HB herbs. There were 2,6-octadienal,3,7-dimethyl-(E) (26.08%), copaene (14.52%), 1,6,10-dodecatrien-3-ol,3,7,11-trimethyl (7.71%), 2,6-Octadien-1-ol,3,7-dimethyl-acetate(E) (6.52%), and 1-naphthalenol,1,2,3,4,4a,7,8,8a-octahydro-1,6-dimethyl-4-(1-methylethyl) (5.48%). The most significant reduction in the composition of the main chemical content after the drying process was in 1,6,10-dodecatrien-3-ol,3,7,11-trimethyl, where only 0.91% and 0.74% were produced after undergoing the OD and SD methods, respectively.

Table 4
Percentage composition of the identified components in essential oil of herbal bath analysed by gas chromatography-mass spectroscopy

No.	Component	R. time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
1	β-pinene	6.300	0.74	0.68	0.49
2	Bicyclo[3.1.0]hexane,4-methylene-1-(1-methylethyl)-	6.857	0.36		0.27
3	B-myrcene	8.477	0.14	0.08	
4	D-Limonene	9.348	0.85	0.32	0.54
5	Eucalyptol	9.572		0.23	0.15
6	1,4-Cyclohexadiene,1-methyl-4-(1-methylethyl)	10.843	0.15	0.15	0.15
7	4-Nonanone	13.559	0.30	0.56	0.36
8	5-Hepten-2-one,6-methyl-	13.941		0.10	0.23
9	2H-Pyran,tetrahydro-4-methyl-2-(2-methyl-1-propenyl)	14.205			0.19
10	2-Furanmethanol,5-ethenyltetrahydro-.alpha.,.alpha.,5-trimethyl-,cis	16.751	0.53	0.52	0.37
11	Cyclohexanol,5-methyl-2-(1-methylethenyl)-	17.308		0.16	
12	2-Furanmethanol,5-ethenyltetrahydro-.alpha.,.alpha.,5-trimethyl-,trans	17.510	0.28	0.31	0.17

Table 4 *(Continue)*

No.	Compound	R. time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
13	Copaene	17.872	14.52	9.45	21.39
14	6-Octenal,3,7-dimethyl-,(R)-	18.250	0.30		
15	1-Cyclohexene-1-acetaldehyde, .alpha., 2-dimethyl	18.396	0.22		
16	Bicyclo[3.1.1]heptan-3-one,2,6,6-trimethyl-	19.215		0.14	
17	1,6-Octadien-3-ol,3,7-dimethyl	19.619	3.27	2.68	2.56
18	Cyclohexanol, 5-methyl-2-(1-methylethyl)-, (1.alpha.,2.beta.,5.beta.)	19.738	0.85	0.63	1.37
19	2-Cyclohexen-1-ol, 1-methyl-4-(1-methylethyl)-,cis-	19.883			0.13
20	Cyclohexanol,5-methyl-2-(1-methylethenyl)-, [1R-(1.alpha.,2.beta.,5.alpha.)]-	20.018	1.27	1.15	1.09
21	Bicyclo[7.2.0]undec-4-ene, 4,11,11-trimethyl-8-methylene	20.387	2.49	1.77	1.26
22	Bicyclo[7.2.0]undec-4-ene, 4,11,11-trimethyl-8-methylene-,[1R(1R*,4Z,9S*)]	20.579	0.17		0.38
23	Azulene, 1,2,3,4,5,6,7,8-octahydro-1,4-dimethyl-7-(1-methylethenyl)-,[1S (1.alpha.,4. alpha.,7.alpha.)]-	20.798	2.08	3.23	3.33
24	1H-Cyclopenta[1,3]cyclopropa[1,2] benzene,octahydro-7-methyl-3-methylene-4-(1-methylethyl)-,[3aS	20.983		0.08	
25	Naphthalene,1,2,4a,5,6,8a-hexahydro-4,7-dimethyl-1-(1-methylethyl)-	22.058	0.51	0.26	0.36
26	Naphthalene,1,2,3,5,6,8a-hexahydro-4,7-dimethyl-1-(1-methylethyl)-,(1S-cis)	22.157		0.25	0.22
27	α-Caryophyllene	22.251	1.55	1.97	2.18
28	1,6,10-Dodecatriene,7,11-dimethyl-3-methylene-	22.554	0.36	0.12	
29	2,6-Octadienal,3,7-dimethyl-,(Z)	22.771	4.21	5.55	8.06
30	1,6-Cyclodecadiene,1-methyl-5-methylene-8-(1-methylethyl)-,[s-(E,E)]	23.054	1.83	2.32	1.96
31	Cyclohexane, 1-ethenyl-1-methyl-2-(1-methylethenyl)-4-(1-methylethylidene)-	23.404	0.41	0.21	
32	γ-elemene	23.552	0.25	0.25	0.18

Table 4 *(Continue)*

No.	Compound	R.time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
33	2,6-Octadienal, 3,7-dimethyl-,(E)	23.956	5.93	7.68	11.03
34	α-farnesene	24.147	1.37	1.88	1.20
35	2,6-Octadien-1-ol, 3,7-dimethyl-,acetate,(E)	24.431	6.52	2.62	1.76
36	Naphthalene, 1,2,3,4,4a,5,6,8a-octahydro-4a,8-dimethyl-2-(1-methylethenyl)-,[2R-(2.alpha.,4a.alpha.,	24.674	3.01	9.86	7.91
37	2,6-Octadienal,3,7-dimethyl-,(Z)	25.369	0.16	0.36	0.22
38	2,6-Octadienal,3,7-dimethyl-,(E)	26.571	26.08	35.79	24.32
39	1-Dodecanol	28.559	1.21		
40	Caryophyllene oxide	28.909	0.87	0.97	0.55
41	1,6,10-Dodecatrien-3-ol,3,7,11-trimethyl	30.238	7.71	0.91	0.74
42	Cyclohexanemethanol,4-ethenyl-.alpha.,.alpha.,4-trimethyl-3-(1-methylethenyl)-,[1R-(1.alpha.,3.alpha	30.939	0.13	0.72	0.67
43	2-Naphthalenemethanol,decahydro-.alpha.,.alpha.,4a-trimethyl-8-methylene-,[2R-2.alpha.,4a.alpha.,8	31.408	0.19	0.29	0.75
44	1-Naphthalenol,decahydro-1,4a-dimethyl-7-(1-methylethylidene)-,[1R-(1.alpha.,4a.beta.,8a.alpha.)]-	31.755		0.18	0.25
45	tau.-Cadinol	32.555	0.19	0.31	0.14
46	2-Naphthalenemethanol,1,2,3,4,4a,5,6,7-octahydro-.alpha.,.alpha.,4a,8-tetramethyl-,(2R-cis)	32.663	0.35	0.68	0.35
47	1-Naphthalenol,1,2,3,4,4a,7,8,8a-octahydro-1,6-dimethyl-4-(1-methylethyl)	33.156	5.48	3.29	1.26
48	α-cadinol	33.495	0.65	0.49	
49	trans-.alpha.-Bergamotene	35.730	0.60		0.11
50	2,6,10-Dodecatrien-1-ol,3,7,11-trimethyl,acetate,(E,E)-	36.192		0.16	0.10
51	5-(1-Bromo-1-methyl-ethyl)-2-methyl-cyclohexanol	36.516	0.15		0.24
52	Phytol	40.259	1.20		0.16
	Total (%) of identification of compound		99.44	99.36	99.15
	No. of components		42	42	42

Note. [a] Retention time; [b] Percentage of oil (%)

Table 5 shows the percentage composition of the identified components in EW of HB analysed by GC-MS. The chemical compounds of EW extracted from fresh, OD and SD herbs made up 99.71%, 96.87%, and 88.06% of the total essential water, respectively. Out of the 18 numbers of identified chemical compounds produced from fresh extraction, only 10 and 12 numbers of identified compounds were found in OD and SD herbs, respectively. The drying process produced another 4 and 3 new numbers of identified chemical compounds from OD and SD herbs, respectively. The drying process significantly affected on EWs compounds.

The highest compound percentage of HB's EW produced was phenol, 2-methoxy-3-(2-propenyl)- (33.30%) for fresh, 2-furanmethanol,5-ethenyltetrahydro-alpha, alpha,5-trimethyl-, trans (37.81%) for OD, and cyclohexanol,2-(2-hydroxy-2-propyl)-5-methyl- (31.36%) for SD. Meanwhile, the lowest compound percentage found in fresh (0.21%) and SD herbs (0.55%) was contributed by 5-hepten-2-one,6-methyl, and for OD herbs (0.36%) was phenol,2-methoxy-4-(2-propenyl)-acetate. The compound composition among the fresh, OD and SD of HB's EW was varied, and there was no obvious pattern like EO.

According to the results obtained, the three chemical contents that recorded the highest content composition in fresh HB herbs can be considered as the main active ingredients of HB products, considering that the sample is a combination of seven types of herbs used as HB herbs and there were: phenol, 2-methoxy-3-(2-propenyl)- (33.30%), trans-geraniol (12.31%), and 2-furanmethanol, 5-ethenyltetrahydro-.alpha.,.alpha.,5-trimethyl-, trans (9.87%). After the drying process, the most significant reduction in the composition of the main chemical content was 1,6,10-dodecatrien-3-ol, 3,7,11-trimethyl, where only 5.34% and 3.71% are produced after undergoing the OD and SD methods, respectively. For most of the chemical contents contained in fresh EW, it was found that there were no significant changes after undergoing the SD method; on the other hand, the significant changes were with the OD method.

Table 5
Percentage composition of the identified components in essential water of herbal bath analysed by gas chromatography-mass spectroscopy

No.	Component	R. time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
1	5-Hepten-2-one,6-methyl	13.758	0.21		0.55
2	2-Furanmethanol,5-ethenyltetrahydro-.alpha.,.alpha.,5-trimethyl-,trans	16.732	9.87	37.81	11.29
3	Acetic acid	17.363	8.10		12.65
4	β-linalool	19.968	0.88	2.52	0.56

Table 5 *(Continue)*

No	Component	R. time [a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
5	Cyclohexanol,5-methyl-2-(1-methylethenyl)-,[1R-(1.alpha.,2.beta.,5.alpha.)]	20.219	0.35	2.77	0.79
6	3-Cyclohexen-1-ol,4-methyl-1-(1-methylethyl)((-)-4-Terpineol)	21.126	1.10	5.79	2.10
7	2,6-Octadienal,3,7-dimethyl-	23.133	2.43		
8	3-Cyclohexene-1-methanol,.alpha.,.alpha.,4-trimethyl-,	23.275	1.15	3.68	2.00
9	Cyclohexene,3-acetoxy-4-(1-hydroxy-1-methylethyl)-1-methyl-	23.942	2.72		
10	β-citronellol	24.813	0.39	1.82	1.20
11	trans-Geraniol	26.519	12.31	19.08	12.46
12	Cyclohexanol,2-(2-hydroxy-2-propyl)-5-methyl-	31.458	9.17	11.17	31.36
13	Phenol,2-methoxy-4-(2-propenyl),acetate	32.726	8.25	0.36	1.71
14	Phenol,2-methoxy-3-(2-propenyl)-	33.188	33.30	5.34	3.71
15	Phenol,4-(2-propenyl)-	35.773	3.49		
16	5-(1-Bromo-1-methyl-ethyl)-2-methyl-cyclohexanol	36.058	0.86		
17	1,5,9-Decatriene,2,3,5,8-tetramethyl	36.843	3.59		
18	4-Allyl-1,2-diacetoxybenzene	38.718	1.54		
19	Eucalyptol			0.43	
20	α-citral			2.65	2.87
21	β-Citral			2.63	3.20
22	Furfural			0.82	1.61
	Total (%) of identification of compound		99.71	96.87	88.06
	No. of components		18	14	15

Note. [a] Retention time; [b] Percentage of water (%)

Table 6 shows the percentage composition of the identified components in BW of HB analysed by GC-MS. The chemical compounds of BW extracted from fresh, OD, and SD herbs made up 99.15%, 99.99%, and 99.38% of the total boiled water, respectively. The drying process significantly affected BW compounds. Out of the 12 identified chemical compounds produced from fresh extraction, only 7 and 6 identified compounds were found in OD and SD herbal leaves, respectively. The drying process produced another identified chemical compound from SD herbs.

The highest compound percentage produced was acetic acid (fresh: 86.74%, OD: 92.52%, and SD: 93.25%), while the lowest percentage was contributed by 2-butanone, 3-hydroxy (fresh: 0.3%, OD: 0.39%, and SD: 0.41%). It was found that there were no significant changes in the chemical content composition in fresh BW after undergoing the OD and SD methods. The chemical content found in fresh herbs significantly reduces content, almost half after drying by the OD and SD methods. It indicates that the exposure of herb leaves to thermal factors significantly affects some chemical contents that cause EOs decomposition and destruction of storage cells.

According to the results, only one chemical content recorded the highest composition in BW. It can be considered the main active ingredient of HB products because of the dominance of the chemical content over other substances. Therefore, nine chemical contents that can be considered active ingredients of HB products were 2,6-octadienal, 3,7-dimethyl(E), copaene, 1,6,10-Dodecatrien-3-ol, 3,7,11-trimethyl, 2,6-Octadien-1-ol, 3,7-dimethyl-, acetate,(E),1-Naphthalenol, 1,2,3,4,4a,7,8,8a-octahydro-1,6-dimethyl-4-(1-methylethyl), phenol, 2-methoxy-3-(2-propenyl)-, trans-Geraniol, 2-Furanmethanol, 5-ethenyltetrahydro-.alpha., alpha.,5-trimethyl-, trans and acetic acid.

Table 6
Percentage composition of the identified components in boiling waters of herbal bath analysed by gas chromatography-mass spectroscopy

No.	Component	R. time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
1	2-Butanone,3-hydroxy	12.908	0.30	0.39	0.41
2	2-Propanone,1-hydroxy	13.390	3.25	1.80	
3	Acetic acid	17.229	86.74	92.52	93.25
4	2(5H)-Furanone, 3-methyl	23.998	0.46		
5	Cyclohexanol, 2-(2-hydroxy-2-propyl)-5-methyl	31.538	2.76	3.49	1.26
6	2-Hydroxy-gamma-butyrolactone	32.995	0.62	0.58	0.66
7	Phenol, 2-methoxy-3-(2-propenyl)	33.282	0.83	0.43	
8	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	34.584	0.39	0.78	
9	Phenol, 4-(2-propenyl)	35.785	0.69		
10	Benzofuran, 2,3-dihydro	36.817	1.35		0.63
11	4-Allyl-1,2-diacetoxybenzene	38.693	1.76		
12	Acetol				3.17

Table 6 *(Continue)*

No.	Component	R. time[a]	Area (%)[b] / Types of drying		
			Fresh	Oven	Sun
	Total (%) of identification of compound		99.15	99.99	99.38
	No. of components		11	7	6

Note. [a]Retention time; [b]Percentage of water (%)

Table 7 shows the chemical class distribution in EO, EW, and BW of fresh, OD, and SD HB herbs. The chemical class distribution of EO revealed the dominance of oxygenated monoterpenes for drying methods (fresh: 43.1%, OD: 55.3%, and SD: 49.89%). Next was the sesquiterpene hydrocarbons class, while the lowest percentage of EO was the monoterpene hydrocarbons, fresh 2.24%, OD 1.23%, and SD 1.45%. The compound 2,6-octadienal,3,7-dimethyl-(E) was the substance that contributed the highest percentage in the oxygenated monoterpenes class, while for sesquiterpenes, hydrocarbons were copaene.

The chemical class distribution of EW revealed the dominance of oxygenated monoterpenes for drying methods (fresh: 81.53%, OD: 91.17%, and SD: 68.85%), and the second highest chemical class was the monoterpene hydrocarbons (fresh: 3.98%, OD: 4.88%, and SD: 4.40%). Unfortunately, no percentage of compound composition was found in the sesquiterpene hydrocarbons class. Substances that contributed the highest percentage in the oxygenated monoterpenes class varied for each drying method.

The chemical class distribution of BW revealed the dominance of the other groups compared to the terpene group for all types of drying (fresh: 92.06%, OD: 95.29%, and SD: 97.49%). Other group constituents were substances from terpene esters, terpene aldehydes, terpene alcohols, aliphatic acids, and ketone. Next were in oxygenated monoterpene (fresh 5.33%, OD: 4.7%, and SD: 1.89%). No percentage of compound composition was found in the monoterpene hydrocarbons and sesquiterpene hydrocarbons class.

The drying process did not significantly affect the chemical class distribution in EO, EW, and BW of fresh, OD, and SD HB herbs. After the HB herbs were dried, the chemical content quantity and type were not significantly different. The only aspect of the chemical content that changed significantly after drying was its composition. Drying processes significantly decreased monoterpene hydrocarbons and oxygenated sesquiterpenes while increasing the content of oxygenated monoterpenes and sesquiterpene hydrocarbons.

According to the results, the HB herb combinations contain chemical compounds, mostly from the terpene group. Terpenes are aromatic compounds found in many plants. Thus, the herbs are suitable to become HB because one of the functions of having HB is to get therapy from the herbs' aroma or "aromatherapy". This therapy relaxes the

Table 7
The chemical class distribution in essential oil (EO), essential water (EW), and boiling water (BW) of fresh, oven-dried, and sun-dried herbal bath herbs

Constituent	EO						EW						BW					
	Types of drying																	
	NC	Fresh	NC	Oven	NC	Sun	NC	Fresh	NC	Oven	NC	Sun	NC	Fresh	NC	Oven	NC	Sun
		% area		% area		% area		% area		% area		% area		% area		% area		% area
Terpenes																		
Monoterpene hydrocarbons	5	2.24	4	1.23	4	1.45	4	3.98	3	4.88	2	4.40	-	-	-	-	-	-
Oxygenated monoterpenes	9	43.10	11	55.30	11	49.89	12	81.53	10	91.17	10	68.85	4	5.33	3	4.70	2	1.89
Sesquiterpene hydrocarbons	13	29.15	13	31.65	12	40.48	-	-	-	-	-	-	-	-	-	-	-	-
Oxygenated sesquiterpene	8	15.57	9	7.84	8	4.71	1	1.54	-	-	-	-	1	1.76	-	-	-	-
Other	7	9.38	5	3.34	7	2.62	5	12.66	1	0.82	3	14.81	6	92.06	4	95.29	4	97.49
Total	42	99.44	42	99.36	42	99.15	22	99.71	14	96.87	15	88.06	11	99.15	7	99.99	6	99.38

Note. NC = Number of compounds of herbal bath herbs analysed by gas chromatography-mass spectroscopy

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body, mind, and soul. Inhaling aromatic herbs can also significantly increase sleep quality, and massage can improve mood and relax people (Edris, 2007).

In this study, important chemical compounds from single herbs such as β-pinene, D-limonene, copaene, and 6-octenal, 3,7-dimethyl-(R) from kaffir lime (Warsito et al., 2017), β-pinene, caryophyllene oxide, β-farnesene, 1,1-dodecanediol diacetate, 1-dodecanol, dodecanoic acid, myrcene, camphene from torch ginger (Juwita et al., 2018), α-farnesene, α-bergamotene, β-pinene, and 1,6,10-dodecatriene,7,11-dimethyl-3-methylene from greater galangal (Subramanian & Nishan, 2015), and linalool, geraniol, myrcene, 2,6-octadienal, 3,7-dimethyl-(E), D-limonene from citronella grass (Muttalib et al., 2018) were still detectable even though the herbs went through an extraction process in combination form.

It indicates that drying and combinations did not significantly affect chemical compounds' important and highest content. The drying process also causes many chemical compound losses. However, the most important and highest content of volatile compounds and the highest intensity of most aroma attributes were still found (Nöfer et al., 2018). Ashafa et al. (2008) reported that different drying methods have no significant effect on Felicia muricata leaves' quality and chemical composition.

CONCLUSION

The proper method and technique for drying fresh herbal leaves and fruit are critical to maintaining their medical value and benefiting all parties. Our result revealed that an oven is the best method to dry herb species because the herbs are bright and attractive and possess mild to strong aromatic fragrances. It is recommended that HB herbs be produced using the OD method because the product is uniform, and the production process is hygienic and continuous (weather-independent). However, this study did not consider the cost of drying the herbs because it is important to retain the chemical compounds as much as possible during the dried herbal preparation. Other than that, from this study, it was observed that the hydro distillation method could be used to extract the herbal bath herbs and provide an aromatic essential oil with a better yield. Further study of combining many herbs in one product is required to determine the effect on human health, especially for those who consume it directly in the body. Therefore, the emphasis is on the antimicrobial activities of the combination herb product extraction.

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