Investigation and assessment of the quality of Japanese mint essential oil (*Mentha arvensis*) originated from various geographical regions in Vietnam

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Abstract

In this study, the effects of distillation methods including hydro-distillation (HD) and microwave hydro-distillation (MHD) on essential oil extractions of *Mentha arvensis* L. in Lam Dong (LD), Ho Chi Minh City (HCMC), and Ba Ria-Vung Tau (BR) were investigated for the yield and chemical composition of the essential oil. The results showed that the use of MHD (0,075 %) was more effective on oil extraction yield than traditional HD (0.053 %). Optimal Distillation parameters were found distillation times of 20 and 110 minutes and material/water ratio of 1:2,5 and 1:6 g/mL for MHD and HD, respectively. The MHD method was subjected to comparing qualitative chemical compositions from Japanese essential oils in BR, LD, and HCMC, Vietnam. The GC-MS analysis results showed that menthol was a major constituent in the essential oil and was responsible for the distinctive smell and flavor of the Japanese mint. Moreover, the menthol content in oil from LD ound was up to 70.6 % higher than that in HCMC (69.45 %) and BR (67.85 %). In conclusion, the mint essential oil is distilled from ingredients in different regions of Vietnam, such as LD, HCMC, BR-VT, all give essential oil a characteristic scent and a relatively high menthol content of about 60 % - 70 %.

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Keywords
Mentha arvensis L., Lam Dong, Ba Ria - Vung Tau, HCMC, distillation.

1 Introduction

The mint tree is native to Europe and the Mediterranean. In the nineteenth century, the Japanese carried out the distillation of the mint essential oil and became the world's unique commercial producer. At the end of World War II, Brazil started to have several studies on the mint and South America, India, and China. By 1974, the mint was penetrated Vietnam. After the Green Revolution of 1986, mint became an agricultural commodity and thriving up to now [1].

Mint includes many species listed as Ginger mint, Japanese mint, Catmint, Peppermint ... In this study, Japanese mint (*Mentha arvensis L*) was used for essential oil distillation. Throughout the length of Vietnam, mint is grown in various regions such as Bac Ninh, Lao Cai, Ha Giang, Quang Nam, Binh Thuan, Lam Dong, Ba Ria - Vung Tau, Ho Chi Minh City, Tien Giang ...

The main ingredient of Japanese mint is menthol, one of the substances with high antiseptic ability, and mint can be a precious source of natural herbs [2]. Currently, mint leaves have been used widely to make purified tea, flavorings, and spices in foods, candies, sauce… Due to menthol's high antibacterial properties, Japanese mint has been added to personal care cosmetic products such as antiseptic hand sanitizer, antifungal, dandruff shampoo, shower gel, toothpaste... The more prominent use of mint is due to its application in health, such as cough medicine, antibiotics, anti-cancer support… [1].

Various methods have been developed to extract essential oils, such as hydro-distillation (HD), steam-distillation (SD), ultrasonic distillation (UE), microwave-assisted solvent distillation (MASE), microwave hydro-distillation (MHD), microwave hydro-diffusion and gravity (MHG), and solvent-free microwave distillation (SFME)… Dai et al. (2010) [3] showed that the MHD from mint leaves was more efficient than UE. In HD, Japanese mint composition was determined to include menthol 78.21 %, trans-menthone 3.6 %, iso-menthone 2.88 % by Verma et al. (2010) in the Kumaon region in the Western Himalaya [4]. According to Kohari et al. (2020), the yield of Japanese mint essential oil obtained by traditional hydro-distillation was determined at 0.27 %, in which the main component was menthol (60.13 % ± 0.42 % and 66.82 % ± 1.62 % for dry and fresh leaves, respectively) [5]. Another study was conducted by Conde (2017) on several growing methods, distilling essential oils, analyzing the content and composition of compounds in mint [6]. In Dong Nai, the effects of planting density and harvesting time on mint essential oil's growth and performance were studied by Mai Hai Chau (2017) [7].

The chemical compositions of plants are influenced by several external factors, including climate, harvesting times, as well as farming methods. The effects of seasonal variations on some essential oils' chemical and biological characteristics have been reported in some articles [8,9]. Many factors, including extraction methods, influenced the chemical composition of the essential oils. However, there are not many available reports on the chemical composition of mint essential oil from different regions.

Currently, HD is the most common and convenient method of extracting essential oils, but this process is considered time-consuming. Due to its high efficiency, the MHD method's research has become a noticeable area. Microwave extraction is a combination of microwave heating and hydro-distillation. Microwave radiation can disrupt oil-bearing cells while speeding up the movement of aqueous solutions and the diffusion of internal components. Likewise, MHD is an environmentally friendly method. During distillation, the extraction efficiency is affected by several parameters, such as the liquid ratio to the material and extraction time [5,9,10].

The present study was performed due to investigating distillation parameters from HD and MHD methods, evaluating the chemical compositions of essential oils from HD and MHD; and comparing chemical compositions of essential oils from Ba Ria – Vung Tau (BR), Lam Dong (LD) provinces and Ho Chi Minh City (HCMC).

2 Experiments

2.1 Plant materials

Japanese mint (*Mentha arvensis*) was cultivated on the fields of HCMC, BR, and LD. The plant material was harvested about four months of age. The dry leaves were stored in a refrigerator at 6 ^{0}C with the moisture content of about 40 %.

2.2 Distillation of essential oils

The preliminary investigation of mint oils was conducted by two methods, hydro-distillation (HD) and microwave hydro-distillation (MHD). The surveyed parameters, including raw material/water ratio and distillation time, were summarized in Table 1.

Table 1 Investigated parameters of the essential oil distillation procedure

Distillation parameters	Material:water ratio (g/mL)	Distillation time (min)
HD	1/4; 1/5; 1/6; 1/7; 1/8	90; 100; 110;120; 130
MHD	1/1.5; 1/2; ½.5; 1/3; 1/3.5	10; 15; 20; 25 and 30

Hydro-distillation method (HD)

Fresh Japanese peppermint leaves were dried in an oven at 50 ^{0}C for 6hrs until the sample weighs about 40 %. Then, 50 g of dried leaves Japanese mint leaves was separately distilled with water using a Clevenger apparatus. The extracted essential oil–was separated from the distilled water, dried over sodium sulfate, and stored in a refrigerator at 6 ^{0}C for further analysis.

Microwave Hydro-distillation method (MHD)

MHD was performed by microwave irradiation. The dried Japanese mint leaves were placed in a 500 mL removable flask inside the microwave irradiator cabinet with a Clevenger system. The leaves were irradiated under microwave powers at 540 W for different distillation times at atmospheric pressure. The extracted essential oils were separated from the distilled water, dried over sodium sulfate, and stored in a refrigerator at 6 ^{0}C for further analysis.

Determination of essential oil yield:

$$H = \frac{V_{oil}.d}{m_{dr}}.100 = \frac{V_{oil}.d}{m_{fr}(1-w)}.100 \qquad (1)$$

Whereas: H: Yield of essential oil by weight (%); V_{oil}: extracted essential oil volume (mL); m_{dr}: weight of dried material (g); m_{fr}: the weight of fresh material (g); d: density of essential oil (g/mL); w: material moisture (%).

3 Results and discussion

3.1 Effects of distillation factors on the essential oil extraction yields by HD

In the HD method, the material : water ratio was set up in the range of (1 : 4, 1 : 5, 1 : 6, 1 : 7, and 1 : 8) g/mL at the power of 540 W for 100 minutes. The ratios of (1 : 4 and 1 : 5) g/mL exhibited lower yields of essential oil extraction (0.028 % and 0.034 %, respectively) than that of the ratios of (1 : 6, 1 : 7, and 1 : 8) g/mL. This result indicated that the increase in water volume led to a rise in extraction yield of essential oil. However, the ratio of 1 : 6 g/mL was effective and safe in extracting the essential oil from the mint. When less solvent was used, the amount of water was not enough to penetrate the material and low oil yield. When increasing solvent volume, the amount of water was enough to diffuse the essential oil from the material, increasing oil yield. But if the amount of water was increased further, the yield remained constant because the amount of essential oil in the material had been completely separated [4]. As a result, the ratio of 1 : 6 g/mL was selected for further investigations.

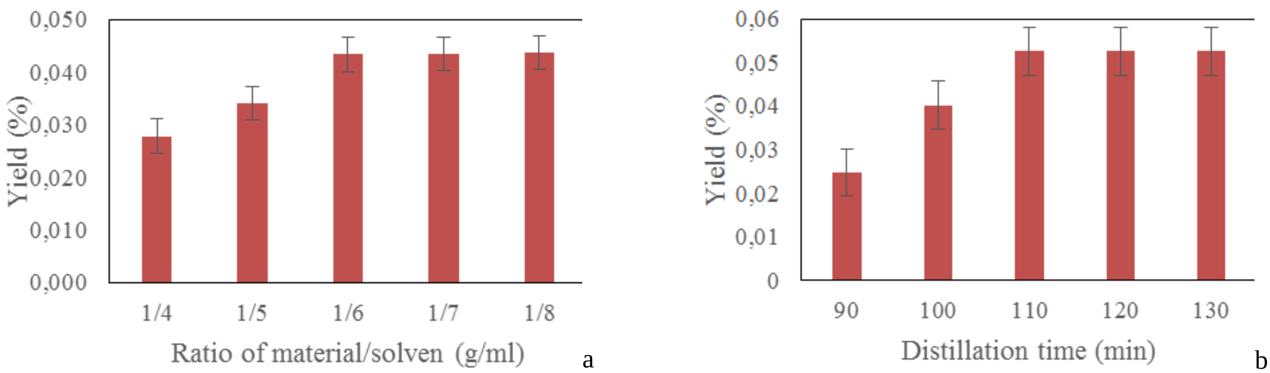

Figure 1 Effects of material/water ratios (a) and distillation time (b) on the essential oil extraction yields by HD

When the distillation time was extended to 130 minutes, the yield reduced to 0,046 %. Indeed, the extension of extraction time allows achieving a high yield of essential oil extraction. However, long extraction time led to the evaporation of oil and the change of oil compositions. As a result, the extraction time of 110 min was selected for further investigation. The results of the experiment had a shorter distillation time compared with Lucchesi's report [10]. Lucchesi et al. reported a yield of 0.610 % for Mentha crispa leaves using HD (270 minutes).

3.2 Effects of distillation factors on the essential oil extraction yields by MHD

In the MHD, the ratios of material : water were investigated in the range of (1 : 1,5; 1 : 2; 1 : 2,5; 1 : 3 and 1 : 3,5) g/mL, at the microwave energy of 540 W for 15 minutes (Figure 2a). Likewise, water volume increased to the highest extraction yield up to 0.068 %, with the ratio of 1 : 2,5 g/mL. However, the extraction yield did not change with the extension of water addition. Accordingly, the ratio of 1 : 2,5 g/mL was selected for further investigation due to the savings and efficiency.

Figure 2b showed that MHD required shorter extraction time than HD at a similar extraction yield. Long distillation period helps to extract the essential oils in the materials completely. However, a prolonged distillation period would evaporate the essential oil into outside environment. Lengthy periods could cause raw materials to burn, reducing the quality of essential oils [9]. The ratio of material : water was used at 1 : 2,5 g/mL in power 540 W in these experiments. With distillation times of 10 to 20 minutes, the amount of essential oil increased continuously and reached the highest value at 20 minutes. But if the distillation time continued to increase to 30 minutes, the essential oil performance remained unchanged. This could be explained by the essential oils removed from the material. The experimental result was similar to that reported by Dai et al. [3], with a yield of 0.091 % for MHD mint (30 minutes). According to this result, the extraction time of 20 min was subjected for further investigation.

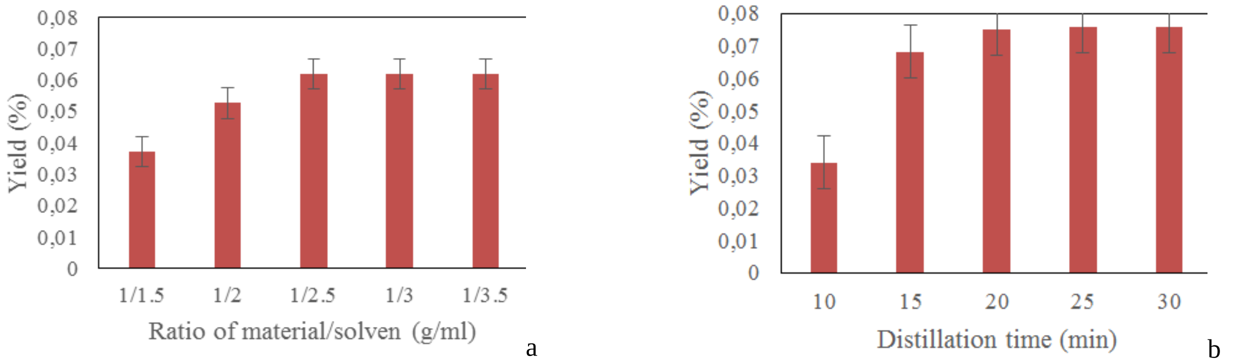

Figure 2 Effects of material/water ratios (a) and extraction time (b) on the essential oil extraction yields by MHD

3.3 Investigation of the composition of Japanese mint oil from different regions

The above results indicated that the highest extraction yield was achieved at the material/water ratio of 1 : 6 g/mL for 110 minutes for the HD method. Meanwhile, the extraction conditions were set up for the MHD method with the material : water ratio of 1 : 2,5 g/mL for 20 min. Samples of essential oils after distillation were refined, which are then carried with GC-MS analysis. The results of the analysis are shown in Table 2.

Table 2 Chemical compositions of mint oils obtained from different regions

Constituents	The present study				Kohari et al. 2020 [5]	
	HD (LD)	MHD			MHD	HD
		LD	BR	HCMC		
α-pinene	0.13	0.06	0.341	0.43	0.18	0.2
sabinene	0.11	0.08	0.637	0.01	0.11	0.11
β-pinene	0.21	0.14	0.139	0.26	0.23	0.24
β-myrcene	0.12	0.09	0.13	0.52	0.14	0.16
3-octanol	0.37	0.57	0.037	0.37	0.58	0.23
o-cymene	0.01	0.02	0.451	0.01	-	-
D-limonene	1.4	0.27	1.194	0.83	0.44	0.25
1,8-cineole	0.1	0.11	0.321	0.03	-	-
trans-β-ocimene	0.2	0.02	0.026	0.03	0.07	0.07
p-mentha-2,4(8)-diene	0.03	0.05	1.8	0.98	-	-
linalool	0.38	0.09	0.31	0.15		-
camphor	0.5	0.46	0.59	0.05	-	-
trans-menthone	20.19	16.3	19.5	12.34	10.54	8.27
cis-methone	4.62	5.01	4.77	4,49	2.97	3.05
(-)-menthol	62.8	70.6	67.85	69.45	59.3	60.13
α-terpineol	0.16	0.25	0.13	0.11	0.18	0.18
cis-3-hexenyl isovalerate	0.14	0.14	0.126	0.26	-	-
pulegone	1.5	0.89	0.127	0.95	-	-
piperitone	3.17	1.23	0.045	1.43	2.11	1.43
methyl acetate	0.64	0.05	0.99	0.01	2.8	2.56
α-copaene	0.01	0.03	0.703	0.03	-	-
(-)-β-bourbonene	0.28	0.17	0.088	0.021	-	-

The results of GC-MS analysis showed that the main content of menthol and cis-menthone of the MHD method (70.6 % and 5.01 %, respectively) was higher than that of HD (62.8 % and 4.62 %, respectively), the content of trans-menthone (20.19 %) in HD method was higher than that of the MHD method (16.3 %). The results showed that essential oil in LD had menthol contents of 62.8 % and 70.6 % from HD and MHD, respectively. These results were higher than when compared with Japanese sources of Kohari (HD: 60,13 % and MHD 59.40 %) [5]. Besides, figure 3 also showed that the contents of trans-menthone and cis-menthone were also higher than the result of

Kohari. The varieties could influence the menthol content in different geographic locations, climatic conditions, and farming methods.

For the HD method, the material should be submerged in water, and the mixture was heated externally by an electric stove. In the MHD, the microwave penetrated the material and heated it from the inside , so the essential oil bags were broken down quickly. Therefore, the distillation time was reduced, and the amount of solvent was much saved when using MHD. According to the influence of distillation parameters on essential oil distillation yield and menthol content, the microwave hydro-distillation method under the extraction conditions of material : water ratio of 1 : 2.5 g/mL for 20 min at 540 W was chosen to perform a material survey from different regions. These obtained results were observed to be similar to the results of Ranitha and colleagues [9].

Mint was directly collected from BR-VT, LD, and HCMC and distilled by MHD under 1 : 2.5 g/mL material : water ratio, 20 minutes duration, and 540 W capacity. The results of the main contents of menthol, trans-menthone, cis-menthone were shown in Figure 3.

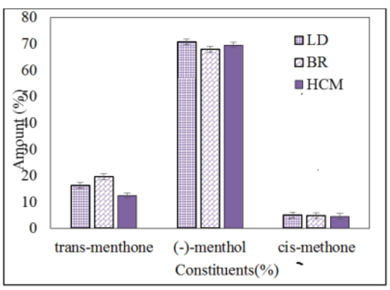

Figure 3 Comparison of the main chemical components of Japanese mint from three regions: Lam Dong (LD), Ba Ria – Vung Tau (BR), and Ho Chi Minh City (HCMC)

Figure 3 showed that mint from LD had some main components such as menthol, cis-menthone, trans-menthone, camphor, piperitone, and pulegone. With mint grown in HCMC, in addition to the main components: menthol, cis-menthone, trans-menthone, and camphor, there were also isoterpinolene, D-limonene, piperitone, and pulegone. In Ba Ria province, menthol's main ingredients include menthol, cis-menthone, trans-menthone, isoterpinolene, piperitone, and pulegone. The menthol content in mint from LD and HCMC showed similar results at 70.6 % and 69.45 %. The result of menthol in BR content was 62.58 % lower than LD and HCMC. The process extracted menthol as a major compound in the *Mentha arvensis* essential oil, which agrees with the results of other research groups [4,5,7].

In three regions, the content of trans-menthone was higher than that of cis-menthone. Results may vary due to cultivation method, time of harvest, and most importantly, geographical location. Japanese mint grown in three different Vietnam regions, including BR, HCMC, and LD, was found to contain a high amount of menthol. Notably, the three regions' quality of essential oils had a strong, spicy, cool, and transparent scent.

4 Conclusion

Research results show that mint essential oil was distilled by the MHD method for higher yield with HD. The method of MHD was more rapid and saving time and energy than the traditional HD method. Distillation time was determined at 20 minutes for MHD and 110 minutes for HD. The ratios of material/solvent were used at 1 : 2.5 g/mL and 1 : 6 g/mL for MHD and HD, respectively. The menthol content in essential oils in LD (70,6 %) was higher than that in HCMC (69.45 %) and BR (67.85 %). The microwave method in essential oil extraction should be paid more attention to because it saves energy and time consuming, economical, and environmentally friendly.

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Khảo sát và đánh giá chất lượng tinh dầu Bạc hà Nhật (*Mentha arvensis*) từ các vùng nguyên liệu khác nhau ở Việt Nam

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Tóm tắt Trong nghiên cứu này, phương pháp chưng cất trực tiếp (HD) và chưng cất có hỗ trợ vi sóng (MHD) từ nguyên liệu Bạc hà Nhật (*Mentha arvensis L.*) được trồng ở Việt Nam đã được khảo sát. Kết quả cho thấy phương pháp chưng cất MHD cho hiệu suất tinh dầu cao hơn (0,075 %) khi so với phương pháp chưng cất HD truyền thống (0,053 %). Các thông số chưng cất cơ bản được tìm thấy như sau: thời gian chưng cất 20 phút và tỉ lệ nguyên liệu : nước 1 : 2,5 g/mL đối với MHD và 110 phút, tỉ lệ nguyên liệu : nước 1 : 6 g/mL đối với HD. Phương pháp MHD được chọn để thực hiện so sánh các thành phần hóa học có trong tinh dầu Bạc hà Nhật từ Bà Rịa - Vũng Tàu, Lâm Đồng và Tp. Hồ Chí Minh. Kết quả phân tích GC-MS cho thấy thành phần hóa học của tinh dầu Bạc hà Nhật từ 3 khu vực là tương tự nhau với sự khác biệt nhỏ về định lượng của một số hợp chất. Menthol là thành phần chính trong tinh dầu bạc hà Nhật và hàm lượng menthol trong tinh dầu nguồn gốc từ Lâm Đồng (70,6 %) cao hơn khi so với hai địa phương còn lại Tp. HCM (69,45 %) và Bà Rịa -Vũng Tàu (67,85 %).

Từ khóa Bạc hà Nhật, Lâm Đồng, chưng cất, Bà Rịa – Vũng Tàu, Tp. HCM.