

Extraction and Characterization of Chamomile (*Matricaria recutita* L.) Essential Oil Using the Green Technology of Solvent-Free Microwave Extraction [†]

Esteban Herrera ^{1,*}, Claudia Pacheco ² and Luis Olivera ¹

¹ Grupo de Investigación en Bioprocesos y Conversión de la Biomasa, Escuela de Ingeniería Agroindustrial, Universidad San Ignacio de Loyola, Lima 15024, Peru; lolivera@usil.edu.pe

² Esencias Químicas, Lima 15046, Peru; claudiapafe.392@gmail.com

* Correspondence: esteban.herrera@usil.pe; Tel.: +511-992-434-493

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Abstract: This study aimed to assess the benefits of the solvent-free microwave extraction (SFME) on the Chamomile (*Matricaria recutita* L.) essential oil quality and yield compared to the extraction by steam distillation (SD). The oil obtained by SFME and SD presented a blue color, a solubility in 70% ethanol (*v/v*) of 4, a relative density of 0.929–0.925 g/mL, a refractive index of 1.5013–1.4790, and an acidity value of 6.23 and 3.43 respectively. The yields were significantly different between extraction methods, being the highest 0.5 mL (0.083% *v/w*) for SFME and 0.2 mL (0.03%) for SD. The GC-MS analysis showed a markedly difference in sesquiterpenes such as Chamazulene, α -Bisabolol, α -Bisabolol oxide A and α -Bisabolol oxide B. The SFME had a 97% and 20% more content of Chamazulene and α -Bisabolol respectively, whilst SD an 88% and 12% more content of α -Bisabolol oxide A and B respectively. The results suggest that SME is an outstanding alternative for essential oil extraction due to much higher yield and quality compared to the steam distillation.

Keywords: chamazulene; GC-MS; distillation; Bisabolol; sesquiterpenes; yield

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1. Introduction

Chamomile is probably the most widely used medicinal plant [1] due to many health benefits such as anti-inflammatory, antimicrobial, antiparasitic, antioxidant, and cytotoxic properties, among others, attributed mainly to its flavonoids, coumarins and sesquiterpenes content [2,3]. Thus, Chamomile essential oil (EO) and extracts have been increasingly added to food products as a functional ingredient and to increase their shelf life [4,5].

The solvent-free microwave extraction (SFME) method is considered a novel green technology that performs adequately in the extraction of essential oil from herbs or plants [6]. SFME combines the microwave heating and distillation in the absence of externally added solvents [7]. This technique can be completed in minutes instead of hours, presents high reproducibility, low energy consumption and usually higher quality of the final product due to less thermal degradation compounds [8].

The international price of *M. recutita* essential oil range between 850–1500 USD/kg, hence, suitable extraction methods for EO with high yields, strong aromatic profile and functional properties are needed. Therefore, this study aimed to assess the benefits of the solvent-free microwave extraction (SFME) on the Chamomile essential oil quality and yield compared to the extraction by steam distillation (SD), which nowadays is the widely used method.

2. Materials and Methods

2.1. Essential Oil Extraction

For both extraction methods: solvent-free microwave extraction (SFME) and steam distillation (SD), 600 g of fresh chamomile flowers were used (80–82% HR). Fresh plants of chamomile were bought to local companies and flowers were separated manually obtaining 14–15% yield (*w/w*) and immediately processed. Although, usually essential oils are extracted from dried samples, in this study fresh flowers were used due to requirements of the extraction method [7] and due to better quality of final product [5,9]. The extraction of essential oil was performed during autumn with an average condition of 20 °C and 80% HR.

For SFME, flowers were placed in the reactor chamber of the Milestone's Ethos X® (Sorisole, Italy). The setup used for extraction was 1200 W for 45 min and 8 °C of condensation temperature.

For the SD, flowers were placed in a Clevenger system using 2 L of water and the extraction was performed for 2 h.

2.2. Physicochemical Analyses

Density was determined using a pycnometer and the essential oil was taken to 20 °C for analysis. Refractive index was performed using a refractometer Mettler Toledo's RM40 LiquiPhysics® (Greifensee, Switzerland). The EO acidity value was performed following the AOAC Official Method 940.28 [10] and expressed as mg KOH/g EO).

2.3. GC-MS Analyses

The analysis was performed using a GC Agilent 7890 and MS Agilent 5975 (California, United States of America) a fused silica capillary HP-5 column (30 m length × 0.25 mm i.d., 0.25 µm film thickness). Helium was used as the carrier gas at a flow rate of 1.1 mL·min⁻¹. The oven temperature was set for a gradient from 30 °C to 250 °C. The injection temperature was 250 °C and detection temperature 230 °C. The MS was set in TIC mode and EI of 70 eV.

3. Results

3.1. Physicochemical Analyses

The essential oil obtained by SFME and SD presented a deep blue color, a solubility in 70% ethanol (*v/v*) of 4, a relative density of 0.929–0.925 g/mL, a refractive index of 1.5013–1.4790. The only difference was in the acidity index, showing higher values the SFME(6.23) against SD (3.43).

3.2. GC-MS Analyses

The GC-MS results showed that essential oils obtained by SFME (Figure 1) and SD (Figure 2) presented a similar chemical profile. Nevertheless, the quantity of each compound differs from each another (Table 1), in special, Chamazulene content is approximately two times fold in SFME compared to SD. On the other hand, SD presents slightly higher values on α -Bisabolol oxide A and B.

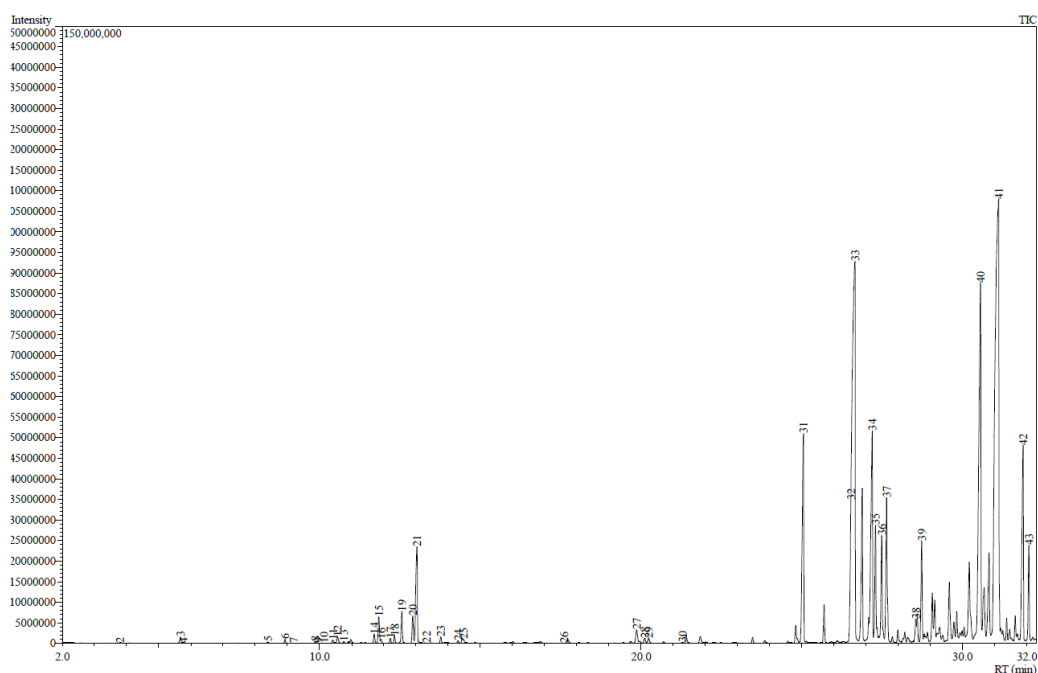


Figure 1. Chromatogram of *M. recutita* essential oil obtained by solvent-free microwave extraction.

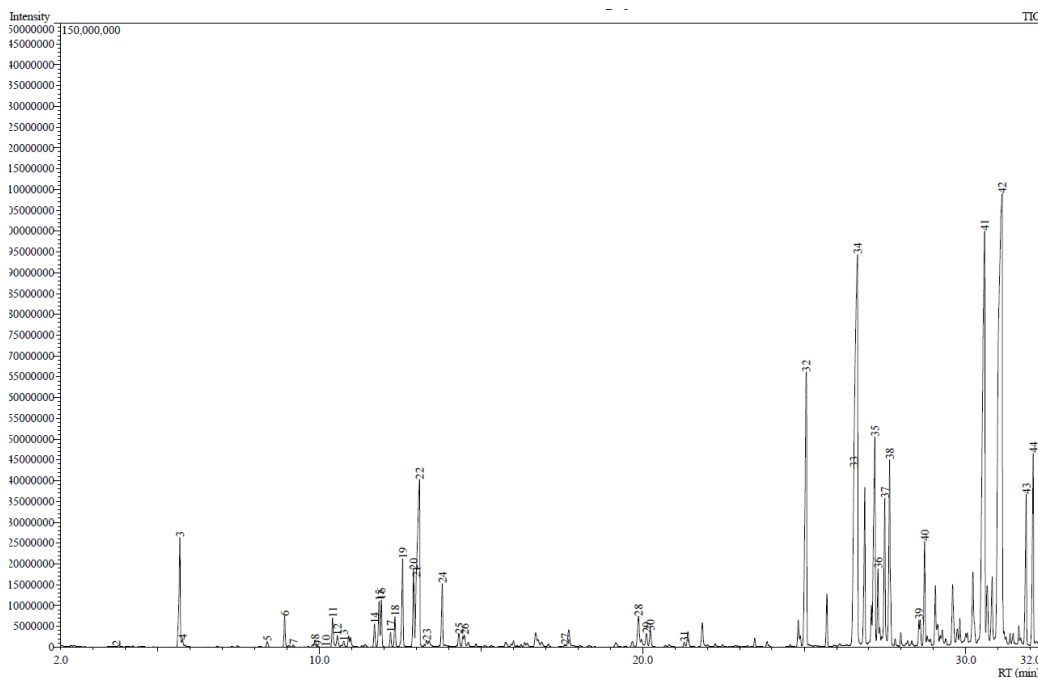


Figure 2. Chromatogram of *M. recutita* essential oil obtained by steam distillation.

Table 1. Physicochemical characterization of chamomile essential oil obtained by solvent-free microwave extraction (SFME) and steam distillation (SD).

Chemical Compound	SFME (%Area)	SD (%Area)
Solvents	13.55	13.53
Ethyl isobutyrate (traces)	0.00	0.00
Ethyl 2-methylbutyrate	0.11	1.74
Ethyl 3-methylbutyrate	0.02	0.04
α -pinene	0.03	0.10
Propyl 2-methylbutyrate	0.08	0.44

Butyl isobutyrate	0.01	0.03
Sabinene	0.04	0.12
β -pinene	0.01	0.01
1-Octen-3-ol (traces)	0.00	0.01
6-methyl-5-hepten-2-one	0.06	0.44
2-pentylfuran	0.15	0.19
Cis- β -Ocimene I	0.03	0.07
p-Cymene	0.18	0.36
Limonene	0.51	0.72
Eucalyptol	0.06	0.60
Trans- β -Ocimene	0.09	0.20
Butyl 2-methylbutyrate	0.13	0.43
Cis- β -Ocimene II	0.58	1.39
γ -Terpinene	0.5	1.10
Artemisia ketone A	2.38	0.45
Trans-2-pentenal	0.00	4.67
Trans-2-Octenol	0.00	0.09
Artemisia alcohol	0.11	0.94
Linalol	0.03	0.10
Isoamyl isovalerate	0.09	0.15
Artemisia ketone B	0.01	0.05
Cis-3-Hexenyl valerate	0.39	0.69
Hexyl isovalerate	0.11	0.21
Trans-2-Hexenyl valerate	0.10	0.27
Methyl trans-2-nonenolate	0.02	0.08
β -Elemene	4.76	5.89
Cis- β -Farnesene	1.75	1.93
β -Caryophyllene	17.47	13.18
Germacrene-D	5.49	4.11
β -Selinene	2.22	1.02
Bicyclogermacrene	2.14	2.14
α -Farnesene	2.99	3.09
Nerolidol (traces)	0.43	0.00
Caryophyllene oxide	1.77	1.47
α -Bisabolol oxide B	10.88	12.18
α -Bisabolol	24.72	20.60
Chamazulene	4.46	2.27
α -Bisabolol oxide A	1.53	2.88

3.3. Yield

The yields were significantly different between extraction methods, being the highest 0.5 mL (0.083% *v/w*) for SFME and 0.2 mL (0.03%) for SD.

4. Discussions

4.1. Physicochemical Analyses

Acidity value of EO obtained by SFME is almost twice of EO from SD. This difference can be attributed to the high microwave power (1200 w) used in the extraction, since increasing microwave power have been associated to more free fatty acid content[11,12].

4.2. GC-MS Analyses

The main difference in chemical profile of essential oils obtained by SFME and SD is the sesquiterpenes content. The SFME showed a 97% more content of Chamazulene and 20% more of α -Bisabolol than SD. These results are remarkable since α -Bisabolol and Chamazulene are considered to be the most valuable components [4], especially due to their contribution to aroma profile and bioactivity [13,14].

Chamazulene derives from matricin [4,5] and has been reported as antioxidant, anti-inflammatory and antispasmodic [15]. On the other hand, α -Bisabolol apparently prevents oxidative stress, inflammatory disorders, infections, neurodegenerative diseases, cancers, and metabolic disorders [15,16].

One study showed that microwave-assisted hydrodistillation achieved 15.08% of chamazulene while hydro-distillation only 1.67% from dried flowers of *M. recutita* [17]. In Damask Rose extracts obtained by SFME, it was found a massive difference in the sesquiterpenes amount, 38.55% against 2.97% obtained by hydrodistillation and 3.37% by steam distillation [8]. Thus, apparently microwave exerts a positive effect in the quantity of sesquiterpenes due to its improved energy transfer compared to traditional heating methods.

Regarding the quality of essential oil, microwaves methods have shown to present better quality than traditional extractions. The lemongrass oil obtained by SFME had a higher amount of citral (74%) in comparison to hydrodistillation (60%) [18]. In other study, although the *T. mastichina* essential oil extracted by SFME did not show significant difference compared to steam distillation or hydrodistillation, the microwave method was quicker and had higher yield [7].

4.3. Yields

The SFME yield was 150% higher than SD and the time for maximum extraction of essential oil was reduced in approximately 60%. This can be due to the effect of microwaves in the cell walls of chamomile, as demonstrated in *C. camphora* fruit peel [19]. Moreover, SMFE has been reported as highly effective in essential oil extraction compared to other methods such as hydrodistillation, salt-assisted extraction, ultrasound-assisted extraction and enzymes-assisted extraction [18,20].

5. Conclusions

Solvent-free microwave extraction is remarkable alternative for Chamomile essential oil extraction due to much higher yield and quality compared to the steam distillation.

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