**American Journal of Applied and Industrial Chemistry**
2021; 5(1): 17-21
http://www.sciencepublishinggroup.com/j/ajaic
doi: 10.11648/j.ajaic.20210501.13

# The Effect of Air Drying and Extraction Methods on the Yield and Chemical Composition of Geranium (*Pelargonium graveolens* L.'Hér) Essential Oils

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**To cite this article:**
Ahmed Mohamed Abouelatta, Attia Youssef Keratum, Sahar Ibrahim Ahmed, Hisham Moustafa El-Zun. The Effect of Air Drying and Extraction Methods on the Yield and Chemical Composition of Geranium (*Pelargonium graveolens* L.'Hér) Essential Oils. *American Journal of Applied and Industrial Chemistry*. Vol. 5, No. 1, 2021, pp. 17-21. doi: 10.11648/j.ajaic.20210501.13

**Received**: March 6, 2021; **Accepted**: March 18, 2021; **Published**: April 7, 2021

**Abstract:** Geranium (*Pelargonium graveolens*) is an economical plant in Egypt, Three methods of extraction (steam distillation, solvent extraction, and hydrodistillation, respectively) were used to extract geranium essential oils from geranium herb. Three types of geranium essential oils (geranium oil, geranium absolute and geranium stripping oil) were extracted with the three different methods. The oil extracted with direct steam distillation is geranium oil was light green with a density of 0.889 g/ml, oil obtained from hydro distillation was geranium sreipping oil which was brownish with a density of 0.903 g/ml. while geranium stripping oil obtained with solvent extraction method was geranium absolute which was dark green with a density of 0.911 g/ml. Geranium herb were air dried for (0, 12, 24 and 48 h). All essential oils obtained were analyzed by Gas Chromatography/mass Spectrometry (GC-MS). Chromatographic analysis of the geranium essential oils demonstrated that citronellol is the main constituent, accounting 29.70%, 31.80% and 18.30% of geranium oil, geranium stripping oil and geranium absolute, respectively. Air drying had a significant effect on geranium oil and geranium stripping oil, for example for geranium oil citronellol percentages were 31.22, 21.20, 28.66 and 35.40 at 0, 12, 24 and 48 h. while geraniol percentages were 9.65, 5.28, 7.85 and 12.43 at same times. For geranium absolute oil air drying had no significant effect on the chemical composition of geranium absolute oil. Air drying had a significant effect on oil obtained from hydrodistillatin method, for example citronellol percentages were 26.44, 28.15, 31.85 and 29.92.

**Keywords:** Geranium Oil, Geranium Absolute, Geranium Stripping Oil, Chemical Composition, Essential Oils, Air Drying

## 1. Introduction

Geranium (*Pelargonium graveolens*, Fam. Geraniaceae) is an economic plant in Egypt, especially in the Beni-Sweif governorate. Egypt is the country with the largest scale of geranium production, followed by China and Morocco [4]. Essential oils are products of secondary metabolism in plants. These oils have strong aromatic components that give a plant its distinctive odor, flavor, or scent [13]. EOs and their major chemical constituents have become the subject of various investigations aiming to evaluate the potential of these compounds for insecticidal properties because they supposedly pose little threat to the environment or to human health [18, 10]. Moreover, their local availability, rapid degradation in the environment and low mammalian toxicity are a few advantages of the use of essential oils as cost-effective control agents [11, 14, 9]. Many aromatic plant species are indigenous to Egypt; however, the insecticidal activities of their essential oils have rarely been studied. Most essential oil chemicals are relatively non-toxic to mammals and fish in toxicological tests, and meet the criteria for “reduced risk” pesticides. Some of these oils and their constituent chemicals are widely used as flavoring agents in foods and beverages and are even exempt from pesticide registration [13]. The essential oil of *Geranium sylvaticum* L.

(Geraniaceae) was isolated by hydrodistillation (HD) and microwave distillation (MD). The composition of the volatile oils was characterized by GC-FID and GC-MS. A total of 73 and 70 compounds were identified, constituting over 90.3% and 98.7% of oil composition of G. sylvaticum, respectively. Sesquiterpene hydrocarbons were shown to be the main group of constituents (HD: 31.7% and MD: 39.8%). The major component of the oils of *G. sylvaticum* was γ - muurolene (HD: 10.9% and MD: 19.6%). The comparative study showed that the amount of total volatiles (98.7%) and the major constituent (39.8%) were better in the MD of G. sylvaticum [12]. *Pelargonium graveolens* essential oil was extracted from dried leaves by a clevenger apparatus and the composition of this essential oil was analyzed by GC-MS. The main results showed that *P. graveolens* essential oil was characterized by the predominance of two compounds: citronellol and geraniol with respective amounts of 27.53 and 25.85%. In addition to that, *P. graveolens* essential oil was characterized by the predominance of oxygenated monoterpenes (70.64%) which were followed by sesquiterpenes (11.88%), acids (1.76%) and hydrocarbon monoterpenes (2.02%). Other compounds were detected with an amount of 11.83% [15]. The extraction method had an effect on the chemical composition of essential oils obtained from *P. graveolens* herb in Egypt, [2]. Air drying had an effect on the yield and chemical composition of *P. graveolens* essential oil, zero time directly after harvesting, after 1, 2, 3, 4, 5 and 6 days from harvesting were studied [8].

# 2. Materials and Methods

### *2.1. Plant Materials*

Geranium herb, *Pelergonium graveolens* L'Hér, (Fam. Geraniacae), grown in Regwa (62 km far from Cairo, Egypt) was harvested after 180 days from plantation within the summer of 2017 and air-dried.

### *2.2. Extraction Techniques*

Geranium herb, *P. graveolens* L'Hér, (Fam. Geraniacae), grown in Regwa (62 km far from Cairo) Egypt were harvested after 180 days from plantation within the summer of 2017 and air-dried. The extraction of crude geranium essential oils was carried out in Hashem Brothers Company for Essential Oils and Aromatic Products (69 Abdel Moneim Riad St., Giza, Egypt). The obtained oil was filtrated twice and maintained in the refrigerator until use.

### *2.3. The Effect of Air Drying on Geranium Oils*

According to Hamouda (2013) the herb of *P. graveolens* L. Herit. was harvested from the field in Regwa (62 km far from Cairo, Egypt) in summer of 2017 [8]. Samples were taken at the mature stage (after 180 days from plantation) for distillation. The distillation period was 1.5 h for all treatments. Four interval periods between harvesting and distillation were studied; Fresh (directly after harvesting), 12, 24 and 48 h from harvesting. Oils obtained from each treatment filtered twice and kept in the fridge till analyzation.

#### *2.3.1. Geranium Oil*

Geranium oil obtained from the geranium herb (*P. graveolens*) which was dried in air for 24 h., 1.5 ton were loaded in a still (3000 $L_3$ capacity), direct water steam passed through the herb inside the still at 120 $C_o$ and under pressure 1.5 bar for 1.5 h to a condenser that using cold water to condense the steam which carrying oil inside according to [7]. After distillation oil separated from water by a glass separator, the oil obtained was filtered twice and kept in the fridge.

#### *2.3.2. Geranium Absolute Oil*

Geranium absolute oil obtained from the geranium herb which was dried in the air for 24 h, one-ton were loaded in a still (5000 $L_3$ capacity), extraction was carried out using solvent (hexane), 3000 L of hexane added to the herb inside the still and covered all the herb with hexane for two hours, then hexane withdrawn for concentration step till 50 L with greenish color. The previous process was repeated two times with different soaking times 2 and 12 h, respectively. All used hexane pumped to primary concentration apparatus. Concentrated hexane (150 L) left for 12 hours to be cooled then filtered on filter paper. The filtered concentrated solvent pumped to the final concentration to remove all the solvent and have the concrete (which contains absolute and wax) under temperature 50 C° and vacuum -1 bar. After 24 h the concrete dissolve in ethyl alcohol 99% purity undercooling and filtered for 5 times to make sure that all the wax had been completely removed then all the solvent (ethyl alcohol which contains absolute) sent to the final concentration apparatus at 70 $C_o$ and vacuum -1 bar alcohol will be removed to have only absolute, [7]. Absolute obtained was filtered twice using filter paper and kept in the fridge.

#### *2.3.3. Geranium Stripping Oil*

Geranium stripping oil isolated from the wastewater obtained during the distillation of geranium herbs by direct steam distillation (during producing geranium oil). Instead of throwing this water to waste water, water pumped to stripping column (which is an especial apparatus of Hashem Brothers Company), with hydrodistillation of this water and after one hour, oil separated from water with a glass separator, the oil obtained filtered twice using filter paper and kept in the fridge.

### *2.4. GC-MS Analysis*

According to (Swigar and Silverstein, 1981) and (Adams, 1995) the constituents of essential oils for each test plant were analyzed by gas chromatography-mass spectrometry (GC/MS) using the model (HP5890) made in the USA system with an HP column (60-meter x 0.25 millimeter, 0.25 µm film thickness) (HP-5ms). The initial temperature was 60 °C and the maximum temperature was 250°C for 65.3 min. The injector temperature was 240°C. Relative percentage amounts were calculated from peaks total area by apparatus software. The compounds were identified by matching the mass spectra data with those held in a computer library

(Wiley 275. L), [20, 3]. All steps of Sample preparation, extraction, and analysis procedure were carried out in the Analytical Laboratory of Hashem Brothers Company for Essential Oils and Aromatic Products.

# 3. Results

*Essential oil, chemical composition*

Data in Table 1 showed that geranium essential oil was light green with a density of 0.889 g/ml. The compounds were listed based on their RT. After 24 h of drying the main components of geranium essential oil were citronellol (28.66%) and geraniol (7.85%), followed by citronellyl formate (6.73%) while the minor compounds were geranyl formate (2.50%) and limonene (2.77%). A total of 14 components were identified, accounting for 85.03% of the total oil.

***Table 1.*** *Main components of geranium oil produced from one ton of plants dried for different periods of time.*

| Component | R. T. (min) | Fresh | Dried | | |
|---|---|---|---|---|---|
| | | | 12 h | 24 h | 48 h |
| Limonene | 9.55 | 0.53%±0.17 | 19.13%±0.20 | 2.77%±0.16 | 1.17%±0.26 |
| Linalool | 12.66 | 5.00%±0.29 | 2.52%±0.01 | 4.27%±0.04 | 4.74%±0.14 |
| Isomenthone | 17.64 | 5.80%±0.35 | 4.36%±0.03 | 5.89%±0.23 | 5.85%±0.03 |
| Citronellol | 19.03 | 31.22%±0.13 | 21.20%±0.11 | 28.66%±0.38 | 35.40%±0.23 |
| Geraniol | 20.22 | 9.65%±0.29 | 5.28%±0.16 | 7.85%±0.32 | 12.43%±0.25 |
| Citronellyl Formate | 20.79 | 6.52%±0.3 | 5.15%±0.09 | 6.73%±0.18 | 7.29%±0.17 |
| Geranyl Formate | 21.90 | 2.20%±0.11 | 1.71%±0.18 | 2.50%±0.28 | 2.62%±0.13 |
| Epi-γ-Eudesmol | 35.45 | 6.17%±0.1 | 3.99%±0.27 | 4.99%±0.26 | 5.82%±0.18 |
| Geranium oil yield/ton herb | | 0.07%±0.01 | 0.10%±0.01 | 0.125%±0.00 | 0.16%±0.01 |

Yield calculated according to the weight of geranium plants after the recorded period (directly before distillation)
aRT, Retention time

Yields of 0.07, 0.1, 0.125 and 0.16% (w/w) were obtained from *P. graveolens* at 0, 12, 24 and 48 h of drying, respectively. The chemical analysis showed that eight major volatile compounds were identified in the essential oil of *P. graveolens*.

Table 1 shows that the chemical composition of geranium essential oil was influenced by sunlight and heat and that the percentages fluctuated among the four treatments. For example, the limonene percentages were 0.53, 19.13, 2.77 and 1.17% for 0, 12, 24 and 48 h of drying, respectively.

Data in table 2 showed that geranium stripping oil was brownish with a density of 0.903 g/ml. A total of 13 components were detected in the essential oil of geranium stripping oil accounting for 79.26% of the total constituents. After 24 h the major compounds were citronellol (31.85%) and geraniol (22.47%) while the minor component was limonene (0.13%). Yields of 0.022, 0.02, 0.021 and 0.016% (w/w) were obtained from *P. graveolens* at 0, 12, 24 and 48 h of drying time, respectively.

***Table 2.*** *Main components of geranium stripping oil following different drying times.*

| Component | R. T. (min) | Fresh | Dried | | |
|---|---|---|---|---|---|
| | | | 12 h | 24 h | 48 h |
| Limonene | 9.50 | 0.17±0.01 | 0.44±0.02 | 0.13±0.01 | 0.23±0.01 |
| Cis oxyderose | 13.04 | 0.87±0.04 | 0.95±0.03 | 0.96±0.03 | 0.98±0.01 |
| Trans oxyderose | 12.04 | 0.27±0.04 | 0.28±0.01 | 0.31±0.02 | 0.31±0.00 |
| Menthone | 15.51 | 1.16±0.09 | 0.38±0.02 | 0.88±0.05 | 0.93±0.02 |
| Isomenthone | 6.72 | 5.75±0.14 | 5.31±0.18 | 5.35±0.04 | 5.41±0.11 |
| Linalool | 4.78 | 13.79±0.11 | 10.56±0.19 | 10.09±0.05 | 10.32±0.18 |
| Guaiadiene | | 0.12±0.01 | 0.14±0.01 | 0.20±0.01 | 0.21±0.00 |
| Citronellyl Formate | 20.54 | 1.18±0.1 | 1.38±0.07 | 1.19±0.10 | 1.29±0.05 |
| Geranyl Formate | 21.90 | 2.29±0.02 | 2.79±0.05 | 2.19±0.02 | 2.46±0.01 |
| Citronellol | 19.03 | 26.44±0.02 | 28.15±0.09 | 31.85±0.17 | 29.92±0.01 |
| Geraniol | 20.22 | 28.69±0.05 | 21.19±0.43 | 22.47±0.04 | 21.62±0.13 |
| Geranyl butyrate | 39.08 | 0.47±0.04 | 0.68±0.02 | 0.49±0.02 | 0.58±0.01 |
| Epi-γ-Eudesmol | 35.61 | 2.65±0.03 | 3.26±0.01 | 3.24±0.14 | 3.26±0.15 |
| Geranium stripping oil yield/ton herb | | 0.022%±0.01 | 0.020%±0.00 | 0.021%±0.00 | 0.016%±0.01 |

Yield calculated according to the weight of geranium plants after the recorded period (directly before hydrodistillation).
aRT, Retention time

Table 2 shows that the chemical composition of geranium stripping oil was affected by the sunlight and heat conditions and that the percentages changed among the four treatments. For example, the limonene percentages were 0.17, 0.44, 0.13 and 0.23 at 0, 24 and 48 h, respectively.

***Table 3.*** *Main components of geranium absolute after different time periods.*

| Component | R. T. (min) | Fresh | Dried | | |
|---|---|---|---|---|---|
| | | | 12 h | 24 h | 48 h |
| Pentanal | 1.71 | 7.58%±0.01 | 7.50%±0.29 | 7.60%±0.11 | 7.57%±0.04 |
| Linalool | 5.10 | 1.35%±0.2 | 1.36%±0.05 | 1.34%±0.02 | 1.35%±0.05 |
| Decadienal | 9.32 | 5.09%±0.05 | 5.10%±0.05 | 5.08%±0.04 | 5.07%±0.04 |
| Citronellol | 8.60 | 18.32%±0.01 | 18.30%±0.17 | 18.33%±0.02 | 18.30%±0.2 |
| Geraniol | 9.24 | 11.09%±0.05 | 11.07%±0.04 | 11.08%±0.04 | 11.10%±0.05 |
| Geraniol Formate | 10.01 | 2.46%±0.03 | 2.45%±0.02 | 2.47%±0.13 | 2.44%±0.25 |
| 2,6 Octadiene | 15.19 | 1.44%±0.02 | 1.43%±0.01 | 1.42%±0.01 | 1.41%±0.00 |
| Geranyl acetate | 15.93 | 1.31%±0.00 | 1.30%±0.20 | 1.32%±0.01 | 1.30%±0.00 |
| Naphthalenemeth-anol | 17.64 | 9.28%±0.16 | 9.27%±0.15 | 9.25%±0.14 | 9.26%±0.15 |
| Longifolene | 18.07 | 1.36%±0.20 | 1.35%±0.20 | 1.36%±0.20 | 1.34%±0.19 |
| 1H-cyclopropa (a) naphthalene | 18.27 | 2.39%±0.05 | 2.38%±0.04 | 2.37%±0.04 | 2.36%±0.03 |
| Geranyl tiglate | 19.12 | 2.36%±0.03 | 2.34%±0.02 | 2.35%±0.20 | 2.33%±0.19 |
| Cyclohexanone | 6.99 | 3.17%±0.10 | 3.16%±0.09 | 3.15%±0.09 | 3.14%±0.08 |
| Phenylethyl tiglate | 16.70 | 2.39%±0.03 | 2.37%±0.04 | 2.36%±0.03 | 2.34%±0.19 |
| Geranium absolute yield/ton herb | | 0.13%±0.03 | 0.15%±0.05 | 0.16%±0.03 | 0.16%±0.03 |

Yield calculated according to the weight of geranium plants after the recorded period (directly before extraction)
aRT, Retention time

As shown in Table 3, there were no significant differences for a single component among the different experimental time periods. Geranium absolute oil was dark green with a yield of 0.16% (w/w) and a density 0.911 g/ml. The retention indices and the percentage of the individual components of geranium absolute are summarized in Table 3. The principal compounds were citronellol (18.33%), geraniol (11.08%) and naphthalenemethanol (9.25%). A total of 15 compounds were identified accounting for 69.53%.

# 4. Discussion

Essential oils (EOs) are generally products of rather complex compositions used contemporaneously in aromatherapy, and for centuries as aromatic medicinal plant species in traditional systems of medicine. Aromatic formulas are used for the treatment of a variety of illnesses, including those that affect the central nervous system [21]. Volatile compounds presenting sedative or stimulatory properties have been identified in EOs from aromatic medicinal species spread across different families and genera. The majority of these substances have small structures with less than 12 carbons and present low polarity chemical functions, being therefore quite volatile. Since most natural EOs are formed by complex mixtures, their bioactivities are obviously dependent on the contribution of their various components [2]. Several studies have indicated that the essential oils obtained via the steam distillation, solvent extraction and hydrodistillation of geranium plants (*P. graveolens*) are often used as fragrances in the perfume industry and more recently for aromatherapy and as herbal medicines, e.g., a study carried out by Džamić *et al*. (2014 on *P. graveolens* oils [6]. Earlier attempts to explore the toxicity of geranium essential oils against the cowpea beetle, *Callosobruchus maculatus*, showed that essential oils affect insects through repellency, inhibition of oviposition, fumigant activities, inhibition of progeny production, and disruption of metabolic pathways [1, 5]. The results of indicate that the method of extraction has a significant effect on the chemical composition of essential oils. Therefore, the air drying of geranium plants had a significant effect on the chemical composition and the percentages of the different components of the geranium oil and geranium stripping oil, while there were no significant effects on the chemical composition or component percentages in the geranium absolute. In agreement with our findings, a study conducted by Hamouda (2013) indicated that the air drying of *P. graveolens* caused a decrease in the plant weight, an increase in the oil percentage and yield, and an improvement in the recovery of the oil components. He also found that the best drying period for the distillation of geranium plants was three days after harvesting. This treatment gave the best results in terms of oil yield and quality, but these results do not agree with those from the current study. We found that the best quality was obtained after 48 h of drying [8]. This difference may have occurred because the other experiments were performed in the desert. Mousavi *et al*. (2014) analysed oil from *P. graveolens* and investigated the following main constituents: citronellol (48.44%), 0-octen-1-01 (18.61%), geraniol (9.70%), p menthone (6.96%), β-caryophyllene (3.13%), germacrene-D (2.92%), caryophyllene oxide (2.52%), geraniol (2.00%), cyclofencheh (1.99%), phenyl ethyl tiglate (1.90%), and geranyl tiglate (1.84%) [16]. Džamić *et al*. (2014) described the chemical composition of *P. graveolens* essential oil. The main compounds were citronellol (24.54%), geraniol (15.33%), citronellyl formate (10.66%) and linalool (9.80%) [6]. Sharopov *et al.* (2014) studied the composition of the essential oil of *P. graveolens* growing in Tajikistan and identified seventy-nine components representing 95.1% of the total oil. The main components of the essential oil were citronellol (37.5%), geraniol (6.0%), caryophyllene oxide (3.7%), menthone (3.1%), linalool (3.0%), β-bourbonene (2.7%), isomenthone (2.1%) and geranyl formate (2.0%) [19]. Nizio *et al*. (2018) studied the effects of different methods of distillation on the chemical composition of *Varronia curassavica* essential oil and found significant effects, which is in agreement with the current study [17]. Mnif *et al*. (2011) studied the chemical composition and the

biological activities of citronellol and geraniol and found that citronellol and geraniol were the main components of the oil, composing 27.53 and 25.85% of the oil, respectively [15].

## 5. Conclusion

*P. graveolens* is an economic plant in Egypt, many farmers and several Companies produce geranium herb and its essential oil.

Current study indicated that the air drying of *P. graveolens* caused a decrease in the plant weight and increased the oil percentage and essential oil yield, and an improvement in the recovery of the oil components. The best drying period for the distillation of geranium plants in the desert is 48 h after harvesting. This treatment gave the best results in terms of oil yield and quality.

Extraction method has a significant effect on the chemical composition of obtained essential oil from geranium herb. According to obtained results Farmers and Essential oils Companies can produce three types of geranium essential oils from the same plant using different extraction methods, which increase the national income.

## Conflict of Interest

The authors declare that they have no conflict of interest.

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