Menoufia J. Agric. Biotechnology Vol. 6 August (2021): 45–53 https://mjab.journals.ekb.eg

COMPARATIVE STUDY BETWEEN GINGER, CINNAMON AND CHAMOMILE ESSENTIAL OILS (PHYSICAL PROPERTIES, CHEMICAL PROPERTIES AND CHEMICAL COMPOSITION)

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Received: Aug. 1 , 2021 Accepted: Aug. 5 , 2021

ABSTRACT: **Current study designed to compare three important essential oils (Ginger, cinnamon and chamomile) commonly used in Egypt. For this purpose, the essential oils were extracted from ginger rhizomes, cinnamon sticks and chamomile flowers by hydro-distillation; then the physical and chemical properties of these oils were estimated, as well as the identification of the active compounds by using GC-MS. Cinnamon oil recorded the highest values in both specific gravity and refractive index, while ginger oil was the highest in solubility in 80% ethanol. Chamomile oil recorded the highest value in acid number, while the ginger oil was the highest in both saponification number and ester number. GC-MS results identify 12 compounds in ginger oil, 6 compounds in cinnamon oil and 10 compounds in chamomile oil. On the other hand, zingiberene (33.2%) cinnamaldehyde (36.1%) and bisabolol oxide (46.71%) were the main major compounds in ginger oil, cinnamon oil and chamomile oil, respectively. In conclusion the obtained results found that, there are a big difference in physical and chemical properties between these essential oils, they also showed a very different chemical composition which leads to an expected difference in their biological effects.**

Key words: **Antioxidant, Volatile oils, Ginger, Cinnamon, Chamommile**

INTRODUCTION

Plants are a good source of biologically active compounds known as phytochemicals. Phytoconstituents have been found to work as antioxidants by scavenging free radicals and many have curative potential for free radical associated disorders and they also have antimicrobial activity (Molan *et al*., 2012). The World Health Organization (WHO) defines traditional medicinal plants as natural plant materials which are used at least or in the absence industrial processing for the treatment of diseases at a local or regional scale. Traditional herbal medicine has been used in developing and developed countries for thousands of years because it is natural and causes comparatively fewer complications (Liu 2011).

Ginger, the rhizome of *Zingiber officinale* Roscoe, has been used as a spice and food seasoning worldwide for centuries. Several scientific investigations reveal that the pungent flavour and pharmacological activities of fresh ginger are mainly attributed to a group of lipophilic substances, called gingerols (Butt and Sultan, 2011). Several studies have indicated that compounds found in ginger are effective in relief of symptoms from chronic inflammatory diseases and can suppress both the cyclooxygenase and release effect of (ROS) on kidney and can improve renal functions (Tjendraputra *et al.,* 2001).

Gingerol and shogaol in ginger have a health benefits, it can protect heart from blood clotting and ginger can protect liver from oxidative stress and help strongly in improve all liver functions (Polasa and Nirmala, 2003).

The bark of various cinnamon species is one of the most important and popular spices used worldwide not only for cooking but also in traditional and modern medicines. Over-all, approximately 250 species have been identified among the *cinnamon genus*, with trees being scattered all over the world (Sangal, 2011).

Cinnamon is mainly used in the aroma and essence industries due to its fragrance, which can be incorporated into different varieties of foodstuffs, perfumes, and medicinal products, The most important constituents of cinnamon are Cinnamaldehyde and trans-Cinnamaldehyde, which are present in the essential oil, thus contributing to the fragrance and to the various biological activities observed with cinnamon (Yeh *et al.,* 2013). Cinnamon plays a vital role as a spice, but its essential oils and other constituents also have important activities, including antimicrobial, antifungal, antioxidant, and antidiabetic (Prabuseenivasan *et al.,* 2006).

Chamomile is an herb that is widely used and included in the herbal medicine pharmacopoeia of 26 countries. Amino acids, polysaccharides, fatty acids, essential oils, minerals, flavonoids, and phenolic compounds are the main elements of chamomile (Singh *et al*., 2011).

Chamomile had various pharmacological properties including antioxidant, gasteroprotective and hepatoprotective effects and antibacterial and wound healing activities Chamomile flowers contain 1%–2% volatile oil, including α-bisabolol, α-bisabolol oxides A and B, matrisin (which usually turns to kamazolen) and other flavonoids with anti-inflammatory and anti-edema properties (Jabri *et al*., 2017).

Researchers indicated that the pharmacological effect of German chamomile is mainly connected with its essential oil for its spasmolytic, antimicrobial, and disinfective properties and the constituents contain α-bisabolol, bisabolol oxides, chamazulene, and enyn-dicycloethers (Grgesina *et al*., 1995). The current study designed to extract essential oils from ginger rhizomes, cinnamon sticks and chamomile flowers and estimate physical and chemical properties, as well as, identify the main components of each oil by GC-MS.

MATERIALS AND METHODS

1- Extraction of the essential oils

The dried flowers, rhizomes and sticks of chamomile, ginger and cinnamon were gridded or reduced to coarse powder using mortar and pestle. The powder material was homogenized, and 500 g of the powdered material was subjected to hydro-distillation using Clevenger-type apparatus for 3 hours and the essential oil was dried over anhydrous sodium sulphate. Extraction was done five times. Then it was placed in amber bottles and was stored in refrigerator at 4^{o}C until further investigation (Zenebe *et al*., 2017).

2- Physical properties of essential

2.1- Solubility in 80 % ethanol

The solubility in 80 % ethanol was determined by titrating a known volume (1 ml) of essential oil with ethanol 80 % to the point of homogeneity and is calculated as volume /volume.

2.2- Specific gravity

The specific gravity of the oil was determined using a pycnometer (1 ml capacity) as described by Guenther (1960).

2.3- Refractive index

The refractive index was determined using Abbe refractometer model 60. According to the procedure in A.O.A.C (2000)

2.4- Optical rotation

The optical rotation of the oils was determined by a polarimeter at room temperature using sodium lamp. The specific optical rotation of the oils was calculated according to the following equation as stated by Guenther (1960).

3- Chemical characteristics

3.1- Acid number

The acid number was determined according to the method described in A.O.A.C (2000). A known weight of the oil (1g) was dissolved in a neutral ethanol (10 ml) and directly titrated by ethanolic potassium hydroxide (0.1 N) using phenolphthalein as an indicator.

$$Acidnumber = \frac{V \times N \times 56.1}{W}$$

Where V= volume in millileters of KOH solution

N= normality of KOH solution

W= weight of the oil in grams

3.2- The saponification number and ester number

The saponification number of the various substances encountered in the present investigation was determined according to the following procedure which was essentially similar to the standard procedure previously reported by Guenther (1960), used phenolphthalein as an indicator which proved to be a about 1.5 gram of the oil was accurately weighed in a 150ml flask. The oil was treated with a known volume (10 ml or 20 ml) of ethanolic potassium hydroxide (about 0.5 N). The mixture was heated in a water –bath at 100 c for one hour, using an air-cooled condenser. At the end of this period, the excess potassium hydroxide was back titrated with hydrochloric acid (0.15 N) using the indicator. A blank determination was carried out using the same quantity of ethanolic potassium hydroxide. The saponification number was calculated from the following equation:

Saponification number= (A-B) X N X 56.1 /W

A= HCl for blank, ml   B= HCl for sample, ml

N= Normality of HCl

W= weight of sample

The saponification number represents the ester number of the essential oil when the number of the sample is small.

3.3- Ester number

The ester number represents the difference between the saponification number and acid number.

Ester number = saponification number – acid number

4- GC-Ms analysis

The GC-Ms analysis of the essential oil samples was carried out using gas chromatography – mass spectrometry instrument stands at the Laboratory of medicinal and aromatic plants, National Research Center. The chromatographic procedure was carried out using a Finnegan Mat SSQ 7000-GC-MS with auto sampler. A methyl polysiloxane capillary column (DB-5, 50 m X 0.32 mm) was used. Helium was used as the carrier gas. The oven temperature used was maintained at 50ºC for 8 min. The temperature was then gradually raised at a rate of 3°C per min to 180°C per min and maintained at 180°C for 5 min. The temperature at the injection port was 250°C. Quantitative data were obtained from the electronic integration of the FID peak areas. The components of the essential oils were identified by comparison of their mass spectra and retention indices with those published in the literature Adams, (1995) and presented in the MS computer library (WILEY275.L).

RESULTS AND DISCUSSION

1- Physical and chemical properties of ginger, cinnamon and chamomile essential oils

1.1- Physical properties of essential oils

Data in Table (1) showed physical properties of ginger, cinnamon and chamomile essential oils. Ginger oil solubility in 80 % ethanol was 1-4 volume of ethanol, and for cinnamon oil was 1-2.5 volume of ethanol, while for chamomile oil was 1-3 volume of ethanol. Data also showed that, specific gravity was 0.87 for ginger oil, and was 1.02 for cinnamon oil, while it was 0.89 for chamomile oil. Refractive index was 1.47 for ginger oil, and 1.51 for cinnamon oil, while it was 1.41 for chamomile oil. On the other hand, optical rotation of ginger oil is -32 and -1.142 for cinnamon oil, while in chamomile oil is -2.4. These results are in the accordance of many authors (Weeratunga *et al*., 2014; Lopez and Blazquez, 2016).

1-2. Chemical properties of essential oils

Data in Table (2) showed chemical characteristics of three tested essential oils, the results were as the following, acid number for ginger oil was 2.89, and 1.34 for cinnamon oil, while acid number recorded 22 for chamomile oil. Saponification number was 234.1, 182 and 215 for ginger, cinnamon and chamomile oils, respectively, while ester number was 231.21, 180.6 and 193 for ginger, cinnamon and chamomile oils, respectively. These results were in the accordance with Peter (1997) and Salah (2008).

2. The Gc/Ms analysis of ginger and chamomile and cinnamon essential oils

Essential oils are very complex natural mixtures which can contain about 20–60 components at quite different concentrations. They are characterized by two or three major components at high concentrations (20–70%) compared to other components present in trace amounts. For example, carvacrol (30%) and thymol (27%) are the major components of the Origanum compactum essential oil, while menthol (59%) and menthone (19%) of Mentha piperita essential oil. Generally, these major components determine the biological properties of the essential oils (Bakkali et al., 2008). The components include two groups of distinct bio-synthetical origin (Betts, 2001 and Pichersky et al., 2006). The main group is composed of terpenes and terpenoids and the other of aromatic and aliphatic constituents, all characterized by low molecular weight.

Table (1): Physical properties of essential oils

Essential oils / Parameters	Ginger oil	Cinnamon oil	Chamomile oil
Solubility in 80 % ethanol	1-4 volume	1-2.5 volume	1-3 volume
Specific gravity	0.87	1.02	0.89
Refractive index	1.47	1.51	1.41
Optical rotation	-32	-1.142	-2.4

Table (2): Chemical properties of essential oils

Essential oils / Parameters	Ginger oil	Cinnamon oil	Chamomile oil
Acid number	2.89	1.34	22
Saponification number	234.1	182	215
Ester number	231.21	180.6	193

In the same plant variety, the chemical composition of essential oils varies depending on several factors that include the part of the plant used, age of trees, growing season and location, and extraction methods (Rajeswara *et al*., 2007 and Chakraborty *et al*., 2015).

In Table (3), it was recorded the results obtained by coupling chromatography with mass spectrometry (GC/MS) of Ginger, Cinnamon and Chamomile essential oils.

The GC–MS analysis of the ginger Essential oil led to the identification of 12 different components, representing 92.25% of the total oil constituents (Table 5). A total of 19 constituents representing 92.25% of the oil were identified; zingiberene (33.2%), benzene,1-(1,5-dimethyl-4-hexenyl) - 4 - methyl (13.64%), α-sesquiphellandrene (12.99%), α-bisabolene (9.86%), 1,8-cineole (5.75%), camphene (5.63%) and farnesene (5.14%) were the main components comprising 86.21% of the oil. These results are in agreements with those obtained by Gupta et al., (2011); Abhay et al. (2013); Ernest et al., (2016) and Ibrahim, (2019).

Table (3): Main compounds in GC/MS analysis for ginger, cinnamon and chamomile essential oils

Essential oil	RT	Area %	Compounds identified
Ginger	4.77	1.92	α-Pinene
	5.22	5.63	Camphene
	7.53	0.84	D-Limonene
	7.67	5.75	1,8-Cineole
	13.16	1.18	Borneol
	21.99	0.95	α-Elemene
	25.89	13.64	Benzene,1-(1,5-dimethyl-4-hexenyl)-4-methyl
	26.43	33.2	Zingiberene
	26.81	5.14	Farnesene
	26.91	9.86	α –Bisabolene
	27.29	1.15	Cadinene
	27.58	12.99	α-Sesquiphellandrene
Cinnamon	8.02	33.67	Benzyl alcohol
	9.55	0.5	Formic acid, phenylmethyl ester
	17.82	36.1	Cinnamaldehyde
	20.83	4.69	Eugenol
	36.45	0.32	α-Hexyl-cinnamic aldehyde
	36.13	22.95	α -Hexyl-cinnamaldehyde
Chamomile	24.68	20.67	(Z) α -Farnesene
	25.77	2.06	Germacrene D
	26.34	1.36	Ç-Elemene
	27.17	1.18	Naphthalene
	32.61	7.7	Acetic acid,10,11-dihydroxy3,7,11-trimethyl-dodeca-2,6-dienyl ester
	33.69	6.81	3-Cyclohexene-1-propanal, α,4-dimethyl-à-methylene
	33.87	1.64	α-Bisabolol
	35.51	2.94	1,4,5,8-Tetramethylnaphthalene
	36.19	46.71	Bisabolol oxide
	41.04	2.48	2-Methyl-5-styrylthiophene

While in cinnamon essential oil a total of 6 constituents representing 98.23% of the oil were identified; cinnamaldehyde (36.1%), benzyl alcohol (33.67%) and α -hexyl-cinnamaldehyde (22.95%) were the main components comprising 92.72% of the oil. Our obtained results are in the accordance of many author He *et al*., (2005); Leela (2008); Jayaprakasha and Rao, (2011).

On the other hand, in chamomile essential oil a total of 10 constituents representing 93.55% of the oil were identified; bisabolol oxide (46.71), (Z) α -farnesene (20.67%), acetic acid,10,11-dihydroxy 3,7,11-trimethyl-dodeca-2,6-dienyl ester (7.7%) and 3-cyclohexene-1-propanal, α,4-dimethyl-à-methylene (6.81%) were the main components comprising 81.89% of the oil. These data are in the accordance with Costescu et al., (2008); Radulovi et al., (2009); Raal et al., (2011); Lopez and Blazquez, (2016) and Ljiljana et al., (2016).

Conclusion

In conclusion the current study found that, there are a big difference in physical and chemical properties between ginger, cinnamon and chamomile essential oils, they also showed a very different chemical composition which leads to an expected difference in their biological effects. GC-MS results identify 12 compounds in ginger oil, 6 compounds in cinnamon oil and 10 compounds in chamomile oil. On the other hand, zingiberene (33.2%) cinnamaldehyde (36.1%) and bisabolol oxide (46.71%) were the main major compounds in ginger oil, cinnamon oil and chamomile oil, respectively. These variation in physical, and chemical properties, as well as, chemical composition (GC-MS results) leads to an expected difference in their biological effects.

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دراسة مقارنة بين الزيوت الطيارة للزنجبيل والقرفة والبابونج
(الخواص الفيزيائية والخواص الكيميائية والتركيب الكيميائي)

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الملخص العربى

صممت هذه الدراسة للمقارنة بين ثلاثة من الزيوت الطيارة الهامة (الزنجبيل والقرفة والبابونج) الشائعة الاستخدام في مصر. ولهذا الغرض تم استخلاص الزيوت الطيارة من ريزومات الزنجبيل وعيدان القرفة وزهور البابونج بطريقة التقطير في بخار الماء، ثم تم تقدير الخواص الفيزيائية والكيميائية لهذه الزيوت الطيارة كما تم التعرف علي المركبات الفعالة بواسطة جهاز التحليل الغازي الكروماتوجرافي الغازي – مطياف الكتلة (GC–MS). وقد سجل الزيت الطيار للقرفة أعلي قيمة في الوزن النوعي ومعامل الانكسار، في حين سجل الزيت الطيار للزنجبيل أعلي قيمة في الذوبان في الايثانول 80%. أما زيت البابونج فقد سجل أعلي قيمة في رقم الحامض، في حين كان زيت الزنجبيل الأعلي في قيم كلا من رقم التصبن ورقم الاستر.

وأظهرت نتائج التحليل بواسطة (GC–MS) التعرف علي 12 مركب في زيت الزنجبيل، والتعرف علي 6 مركبات في زيت القرفة، في حين تم التعرف علي 10 مركبات في زيت البابونج. ومن ناحية أخري كان مركب الزنجبيرين (33.2%) هو المركب الرئيسي في زيت الزنجبيل، ومركب السينامالدهيد (36.1%) هو المركب الرئيسي في زيت القرفة، أما بالنسبة لزيت البابونج فقد كان مركب البيسابولول (46.71%) هو المركب الرئيسي.

ويمكن تلخيص مادلت عليه نتائج الدراسة في وجود اختلاف كبير في الخواص الفيزيائية والكيميائية للزيوت الطيارة المختبرة (الزنجبيل والقرفة والبابونج)، كما أظهروا اختلافا كبيرا في التركيب الكيميائي مما يؤدي إلي توقع أن يكون هناك اختلاف كبير بينهما في التأثيرات البيولوجية.

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