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EFFECT OF STORAGE ON THE VOLATILE OIL CONTENT OF CLOVE BUDS (*Syzygium aromaticum*)

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ABSTRACT

This study has sought to know the effect of conventional storage of dried clove buds (*Syzigium aromaticum*) on their volatile oil content. Volatile oil was extracted from the three differently stored samples using the steam distillation method. The volatile oil content was estimated and the chemical constituents of the volatile oil were fractionated and identified in accordance with AOCA method by using Gas Chromatography/ Mass spectrometry technique (GC/MS). The findings indicated that storage for less than two months caused very slight changes concerning the volatile oil content and the percentages of its active constituents such as eugenol, eugenyl acetate and β-caryophyllene. However, these changes, were not statistically significant.

***Key words**: storage, Syzygium aromaticum, volatile oils.*

1. INTRODUCTION

Spices and herbs were strongly present early in history. They were used in cooking for their distinctive flavor and their medinical and curative properties. Trading in spices was one of the most active and important commercial activities since the dawn of history. Most spices grow in Asia because of its tropical climate nature. Spices are known for their volatile oils and oily resins that account for their distinctive flavor and medicinal properties (Prabuseenivasan *et al*., 2006).

Volatile oils are unstable oils, ethereal oils, aetherolea or a mixture of the above compounds. They are produced naturally in plants. The volatile oil can be extracted using various methods, the most important of which is the mixing of the plant that contains the oil with water, or using dry or steam distillation (Husnu *et al.*, 2007). These may be of a simple chemical structure such as clove (*Syzigium aromaticum*), coriander and vanilla or they may have a complex structure such as nutmeg or laurel (Politeo *et al*., 2007).

Syzigium aromaticum is one of the spices that are rich in volatile oils and acts as natural antioxidants. Musenga *et al.* (2006) mentioned that the essential oil of clove buds contains eugenol, eugenyl acetate, vanillin, B-caryophyllene and crategolic acid. Wenqiang *et al.* (2007) indicated that the main constituent of clove (*Syzigium aromaticum*) buds oil is eugenol, which amounts to 58.77% of the total oil when extracted at a high percentage.

Alma *et al*. (2007) confirmed that the oil extracted from dried clove buds contains eugenol (49-87%,) β-caryophyllene (4-21%,) eugenyl acetate (0.5-21%) and a slight percentage of α-humulene.

These volatile oils are highly beneficial to health. However, they are quickly oxidized by heat, light and air (Djilani and Dicko, 2012). Hence, preservation of these properties requires special handling for storing and grinding spices. Al Shihri and Al-Tameemi (2012) found that most Saudi families grind *Syzigium aromaticum* and keep it in containers for use when required.

Thus, the objective of this study was to determine the effect of storage by two different methods; on *Syzygium aromaticum* spice in terms of its content and components of the volatile oil.

2. MATERIALS AND METHODS

Dried Clove (*Syzigium aromaticum*) buds were purchased from the local markets in Riyadh and samples were prepared as follows:

Sample A: representing the control. A sample of dried clove buds was finely ground immediately upon purchase and analyzed in the laboratory.

Sample B: a sample of whole dried clove buds was stored in glass jars with narrow neck closed tightly for one month at room temperature (20-23˚ C), then finely ground and analyzed immediately.

Sample C: a sample of whole dried clove buds was stored in glass jars with narrow neck closed tightly for one month at room temperature (20-23˚ C), finely ground and stored in glass jars for an additional month at room temperature before being analyzed.

The research adopted the experimental analytical method (Obaidat *et al*., 2000). Steam distillation was used for estimating the oil quantities in the samples. Gas Chromatography / Mass Spectometry (GC/MS, GCMS, QP-2010 Auto Injector-AOC-5000, Vacuum Pump, Column Specification: Rtx.5MS (Crossbond:5% diphenyl L 95% dimethyl polysiloxane), 30 meter, 0.25 mm ID, 0.25um df, cat # 12623 Serial # 882023, Max. Programmable Temp. 350˚C. Minimal bleed at 330 ˚C) was used for quantitative and qualitative estimation of volatile oils in the samples in accordance with the Association of Official Analytical Chemists, method (AOAC, 2000).

The results were summarized in tables and diagrams for easy interpretation. SPSS, V15 was used for statistical analysis of the data. One-way ANOVA was used for determining the statistical differences between the three samples (Steel and Torri 1980).

3. RESULTS AND DISCUSSION

Table (1) and Fig. (1) show that the quantity of the oil extracted from recently purchased and ground clove; the control was 15.34 gm/100 gm of the powder, whereas in the sample B which represents the whole clove buds stored complete for a whole month at room temperature and then ground, the extracted oil was 15.52 gm/100gm and in the whole cloves stored at room temperature (20-23 ˚C) then ground and stored for an additional month at room temperature, representing sample C, the extracted oil quantity was 13.67gm/100gm.

Silano and Anton (2001) stated that the quantity of oil in clove buds amounted to 20% or more of its weight. On the other hand, Peter (2004) stated that ground or complete clove buds contain about 15-20% of their weight as volatile oil. The slight differences in the findings are due to various reasons, key among which are the method used for extracting the oil, precision of the used equipment and the type of solvent (Wenqiang *et al*., 2007). The findings also indicate that storage affects clove total content of oil. This is remarkable when storage is performed after grinding. The difference in this study is, however, not statistically significant because of the short storage period. This is confirmed by Peter (2004) who mentioned that storage of clove buds leads to losing oil through evaporation. The loss ratio depends on several factors most important of which are physical condition, moisture content and the temperature and moisture of the storage location. The total oil percentage in the buds is slightly reduced after storage for a period of one month.

Table (2) and Fig. (2) show the quantity and type of oils present in the studied samples. It is clear that the main constituent of clove buds oil is eugenol, followed by eugenyl acetate and β-caryophyllene. Srivastava *et al*. (2005) revealed that when oil is extracted from Madagascar clove buds by steam distillation the oil will contain eugenol at 82.6% of the total oil with 6% eugenyl acetate, 7.2% of β-caryophyllene, slight percentages of MethlySalisylate estimated at 0.05% and 1.5% Alpha humulene.

Alma *et al*. (2007) extracted oil from Turkish clove buds using steam distillation for three hours. The oil content in the extracted oil was analyzed. They found that the oil extracted from Turkish clove buds contained 18 constituents with eugenol constituting the highest percentage of these constituents at about 87% of the total oil, followed by engenol acetate at about 8.01%

Table (1): Effect of grinding and storage on the yield of the clove buds volatile oil (on dry wt. gm/100gm), mean± SD.

Samples	Sample A (Control)	Sample B	Sample C
Quantity of the extracted oil	15.34±0.99	15.52±0.70	13.67± 0.72

Control Sample A: clove recently purchased, ground and oil immediately extracted from it.
B: Whole Clove buds were stored for one month at room temperature (20-23˚ C), then ground and oil extracted.
C: Whole Clove buds were stored for one month at room temperature (20-23˚ C), then ground and stored for an additional month at room temperature.

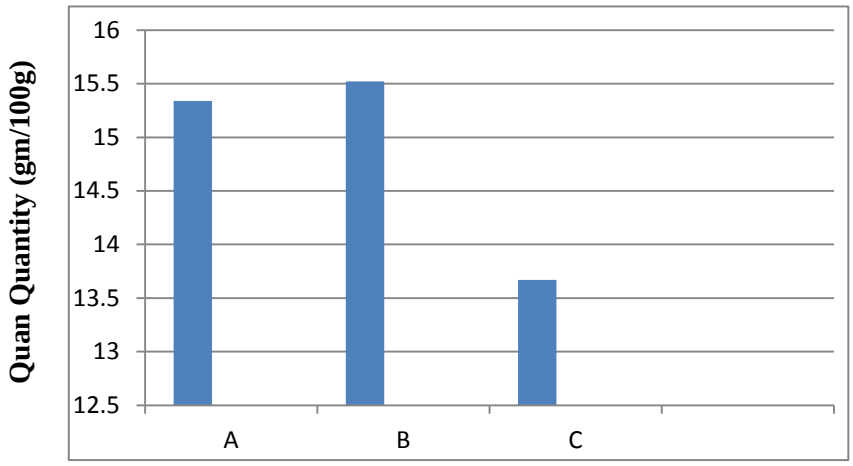

Fig. (1): Quantity of the volatile oil extracted from the three samples (gm/100g).

Table (2): Effect of grinding and storage on the chemical constituents of the clove buds volatile oil by using GC/MS technique.

Chemical constituents	Control Sample A		Sample B		Sample C	
	gm	%	gm	%	gm	%
Eugenol	0.86±10.16	66.24	0.69±10.57	68.11	0.71±8.98	65.68
Eugenyl Acetate	0.12±2.54	16.56	0.00±2.76	17.77	0.01±2.67	19.44
β-Caryophyllene	0.01±2.18	14.21	0.01±1.75	11.26	0.05±1.67	12.23
α-Caryophyllene	0.00±0.25	1.61	0.00±0. 20	1.22	0.00±0.18	1.35
Methyl Salisylate	0.00±0.01	0.07	0.00±0.02	0.11	0.00±0.04	0.31
Caryophyllene Oxide	0.00±0.06	0.38	0.00±0.08	0.50	0.00±0.06	0.46
Copaene	0.00±0.05	0.3	0.00±0.03	0.20	0.00±0.02	0.17
Alpha-Farnesene	0.00±0.04	0.26	0.00±0.03	0.20	0.00±0.01	0.10
Cavicol	0.00±0.02	0.13	0.00±0.02	0.11	0.00±0.02	0.12
α-Cubebene	0.00±0.01	0.09	--	---	----	----
Aromadendrene Oxide-(2)	0.00±0.01	0.07	0.00±0.02	0.11	0.00±0.02	0.11
Cadina-1(10),4—diene	0.00±0.01	0.07	0.00±0.01	0.05	Effects	0.02
Germacrene D	Effects	0.07	Effects	0.01	---	---
α-Terpineol Acetate	---	---	0.00±0.06	0.35	Effects	0.01
Total	15.34	100	15.52	100	13.67	100

Control Sample A: clove recently purchased, ground and oil immediately extracted from it.
B: Whole Clove buds were stored for one month at room temperature (20-23˚ C), then ground and oil extracted.
C: Whole Clove buds were stored for one month at room temperature (20-23˚ C), then ground and stored for an additional month at room temperature.

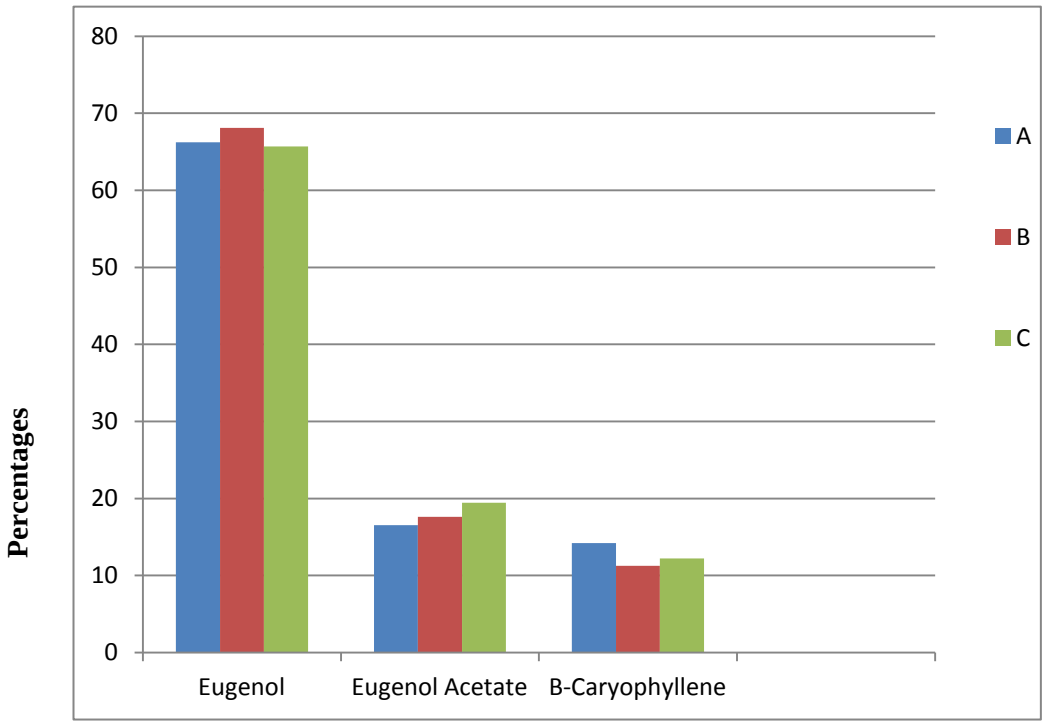

Fig. (2): Percentages of the major constituents of the volatile oil in the three samples.

and then β-caryophyllene with a percentage of 3.56%. The remaining constituents represented only a very small percentage of the total oil. Nassar *et al*. (2007), revealed that eugenol percentage in clove buds oil reached 71.56% and that eugenol acetate reached a percentage of 8.99% of the total oil. The difference in the findings of these studies is due to the different methods used for extracting the total oil form dry clove buds, the equipment used and its precision and the type of solvent (Wenqianq *et al*., 2007). "MMWD" (2008) attributed the variations in the percentages of the constituents of clove bud oil to several causes, key among which are plant hereditary strain, climate, type of soil, agricultural technologies, method of harvesting and the harvested part of the plant. Misharina and Samusenko, (2008) indicated that the quantity and kind of antioxidants in spices and herb essential oils depend on the method used for extracting these oils and the kinds of solutions used in the extraction process. Peter (2004) pointed out that the quantity of eugenol in the extracted oil depends on the time taken by distillation, with fast distillation yielding oil with a high percentage of eugenol. Table (2) shows that the percentages of volatile oil constituents varied very slightly without any significant differences because of the short storage period. But, the Table also shows that the eugenol percentages increased very slightly in sample B compared to control sample A. This may be due to the loss of moisture through evaporation during storage which has led to its concentration. In sample C the eugenol level decreased because of its grinding and storage. The level of Beta caryophyllene decreased in the two stored samples compared to the control sample.

Peter (2004) referred to the effect of storage and grinding on dried clove buds oil, stating that eugenol acetate decreased slightly after storage whereas eugenol increased. These changes usually take place after storage for six months if adequate storage conditions are taken into consideration. Grinding produces an oil with a low content of eugenol because of the loss of a quantity of volatile oil during grinding, hence distillation of ground clove buds is performed immediately after the grinding.

These findings indicate that despite the short period during which the dried clove buds were stored, slight statistically insignificant changes occurred in the quantity and constituents of the volatile oil. Thus, the study recommends conducting further research on the effect of storage on spices volatile oils, storing the materials for longer periods.

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تأثير التخزين على محتوى براعم القرنفل من الزيوت الطيارة

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ملخص

هدفت هذه الدراسة الى التعرف علي تأثير التخزين بالطرق التقليدية على محتوى براعم القرنفل المجففة من الزيت الطيار. حيث تم استخلاص الزيت الطيار من العينات الثلاثة المختلفة بطريقة تخزينها بطريقة التقطير بالبخار وقدرت كميته ثم حللت مكونات الزيت الطيار تبعا لطريقة AOCA وباستخدام جهاز الكروماتوجرافي الغازي وطيف الكتلة. كشفت النتائج ان التخزين لمدة أقل من شهرين احدث تغيرات بسيطة جداً من حيث نسبة الزيت الطيار ونسب مكوناته الفعالة كالايوجينول، خلات الايوجينيل والبيتا- كاريوفيلين وان كانت هذه التغيرات غير معنوية احصائياً.