VARIABILITY IN PEEL COMPOSITION AND QUALITY EVALUATION OF PEEL OILS OF CITRUS VARIETIES

Muhammad Mushtaq Ahmad* , Salim-ur-Rehman**, Tahir Mahmood Qureshi*, Muhammad Nadeem*, Muhammad Asghar***

ABSTRACT

Variation in chemical composition of peels and quality of peel essential oils of Kinnow (Citrus reticulata), Fewtrell’s Early (Citrus reticulata), Malta (Citrus sinensis), Mousami (Citrus sinensis), Grapefruit (Citrus paradise) and Eureka lemon (Citrus limon) were studied at Institute of Food Science and Nutrition, University of Sargodha, Sargodha, Pakistan during 2015-16. The results revealed that the highest fat contents (1.68%), protein (6.24%) and fibre (6.11%) were found in grapefruit peel, ash (4.39%) in Kinnow peel, moisture (75.39%) in Eureka lemon peel and nitrogen free extract (11.68%) in Mousami peel. After extracting the oils from each peel through cold expressing method, yield was estimated. The highest yield (1.21%) was estimated in Malta peel, followed by peels of Eureka lemon (1.12%), Mousami (0.98%), Grapefruit (0.73%), Kinnow (0.32%) and Fewtrell’s Early (0.22%) with regards to chemical composition of peel oils the data displayed the highest aldehyde contents (42.733%) in Kinnow peel oil followed by grapefruit (40.473%), Fewtrell’s Early (30.020%), Mousami (26.760%) and Eureka lemon (11.803%). Similarly, acid number of Kinnow, Fewtrell’s Early, Malta, Mousami, Grapefruit and eureka lemon oils was estimated at 2.213, 1.337, 1.253, 1.323, 1.863 and 1.390, respectively. Malta peel oil showed the lowest ester number (11.747), and the highest (27.120) in grapefruit peel oil. The results conclude that the highest yield was estimated in Malta peel oil, aldehyde contents and acid number in Kinnow peel oil and ester number in Grapefruit peel oil.

KEYWORDS: Citrus peel oils; cold expressing method; yield, quality; aldehyde contents; acid number; ester number.

INTRODUCTION

Citrus fruits are consumed both fresh and in processed form. The peels and membrane residues called primary waste fractions confine to 40-50 percent of wet fruit mass (7). Flavedo a coloured outer layer and albedo the white inner layer are the two main functional peel components (17). The flavedo

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part contains oblate-shaped oil glands or oil sacs, which extend to different depths into the albedo layer of these fruits. These glands discharge peculiar chemicals through metabolic processes, characterized by each type of citrus aroma and flavor, called essential oils (2), ranging from 0.5 to 3.0 kg per ton of fruit (22). Essential oils are composed of 200 natural compounds (8) which form a complex mixture, classified as oxygenated volatiles and non-volatile residues or secondary metabolites. The volatile compounds consist of aldehydes, ketones, alcohols, acids and esters and constitute 90-95 percent of the whole oil (23). The non-volatile residues consist of paraffin waxes, steroids and triterpenoids, fatty acids, cumarins and psoralen flavones and constitute 1-10 percent of the whole oil and accumulated in balloon like cells or oil glands (15). The aldehyde contents, being the major oxygenated compounds, in particular neral and geranial, ranging from 3-15 percent of the whole oil, are considered to be a quality reference (9, 10).

Covering an area of 194.0 thousand hectares in Pakistan, annual production of citrus fruits in the year 2013-2014 was 2168.0 thousand tons (3), experiencing increasing trend each year. Keeping in view the abundant availability and remarkable differences in colour, flavor, peel thickness, sizes and peel essential oils, a study was planned to examine the variability in composition of peels and yield and quality of peel oils of citrus fruits. So that their quality criteria of these essential oils may be established.

MATERIALS AND METHODS

This study was conducted at the Institute of Food Science and Nutrition, University of Sargodha, Sargodha Pakistan during the year 2015-16 Kinnow, Fewtrell’s Early, Malta, Mousami, Grapefruit and Eureka lemon were collected from local orchards of Sargodha. Research work on peel composition, yield and quality of peel oils of these fruits was carried out through the following steps.

Percent peel portion and proximate analysis

The fruits were peeled manually and percent peel portion was calculated on the basis of 20 fruits of each variety. Similarly, the proximate analysis for moisture, ash, protein, crude fat and nitrogen free extract (NFE) of peel of each variety was carried out using AOAC (4) methods.

Cold expressing and extraction of oils

In the next step of extracting peel oil, peels were shredded to a size of 20x3 cm, using citrus peel shredder (Tunsbull’s Model No. 266069). Keeping in
view the convenience and management of the process, oils yield, time of extraction, quality stability, efficiency and other feasible indicators, cold expressing method was used to extract the oils from peels and expressed at 15000 psi using locally made hydraulic press. The mixture obtained after extraction was centrifuged in refrigerated centrifuge (Model No. ALC-4227R) with 15000 rpm at 28-30°C for 45 minutes followed by separation through separating funnel. Each oil, was then filtered through the powder of sodium sulfate to remove traces of moisture.

Yield of oils and quality evaluation

The yield of each peel was then calculated and quality evaluation for aldehyde contents (Sodium sulfate method), ester number and acid number (5) was determined. The results so obtained were then analyzed statistically (24) to judge the comparative probability and validity of research.

RESULTS AND DISCUSSION

Percent peel portion

The results (Fig. 1) revealed that peel portion of Eureka lemon was the highest ranging from 39.0-48.0 percent, followed by Mousami (38.0-43.0%), Grapefruit (35.0-41.0%), Malta (35.0-40.0%), Kinnow (32.0-33.0%) and Fewtrell’s Early (28.0-32.0%). Citrus peels are the major source of essential oils due to the presence of oil glands in outer flavedo layer. Weiss (27) also found that peel portions of sweet orange, lemon and mandarin were 25.0 percent 40.0 percent and 28.0 percent respectively. Similarly, in author study (4) various fruits such as mandarin, orange, grapefruit and lemon had the peel ranging from 25.0-45.0 percent. It was also observed earlier (6) that peel portions of various citrus fruits such as tangerine, orange, grapefruit and lemon were 25.6-33.0 percent, 21.5-38.1 percent, 33.7-36.4 percent and 32.0-46.6 percent, respectively. The results of present study delineated some sort of difference between the percent peels of citrus fruits of Pakistani origin and similar research studies carried out earlier, which may be attributed to the variation in soil conditions and climate (12).

Yield of essential oils

The results (Fig. 1) indicated that yield of peel oil of malta was the highest (1.21%), among all citrus fruits, followed by that of Eureka lemon, Mousami, grapefruit, Kinnow and Fewtrell’s early i.e. 1.12, 0.98, 0.73, 0.32 and 0.22 percent respectively.

After studying the proximate composition of some citrus fruits, Weiss (27) also reported almost similar results and stated that total oil contents of sweet orange, eureka lemon, mandarin and bergamot orange were 0.80, 0.90, 0.80 and 0.55 percent respectively, depending mainly on climacteric conditions. In another report on the importance of aromatic plants, Ohloff (18) described that yield of cold pressed peel oils of orange, bergamot and petitgrain was 0.5 percent each whereas, mandarin oil had 0.2 percent yield. Similarly, he also mentioned that 850 kg of carefully picked orange flowers yielded one kg of neroli oil after steam distillation Tu et al., (25, 26) studied the physical analysis of citrus peel oils of various varieties and found that yield of citrus essential oil differed with individual plant species ranging from 0.2 to 2.0 percent.

![Graph showing percent peel portion and yield of essential oils of various citrus fruits.](image_url)

**Fig. 1.** Percent peel portion and yield of essential oils of various citrus fruits.

The yield of essential oils is influenced by many factors such as variety, presence of gums in peels, difference in ratio of albedo and flavedo layers, extraction method, maturity and stage of harvesting etc. From the above results and discussion it is inferred that cold expressed method applied in present study was the most favorable extraction method. So the results of peel oil yield of citrus varieties are in conformity with similar scientific studies. However, oil yields of Kinnow and Fewtrell’s Early were low as compared to other fruits which might be due to the presence of some gums in the peels as was experienced during oil extraction phase in this study.
Chemical examination of citrus peels

Chemical examination of these peels for moisture, ash, crude fat, crude fibre, protein and NFE, was carried out. The results (Table 1) delineated that Malta and Eureka lemon had maximum moisture contents, (75.18 and 75.39%) having non-significant difference to each other. However, significant differences in moisture contents of peels were observed (Table 1).

Table 1. Mean values of chemical examination of peels of citrus cultivars.

<table>
<thead>
<tr>
<th>Citrus fruits</th>
<th>Moisture (%)</th>
<th>Ash (%)</th>
<th>Fat (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kinnow</td>
<td>73.35±1.35b</td>
<td>4.38±0.15ab</td>
<td>1.59±0.05ab</td>
</tr>
<tr>
<td>Fewtrell’s early</td>
<td>70.65±1.55e</td>
<td>3.79±0.14c</td>
<td>1.35±0.08c</td>
</tr>
<tr>
<td>Malta</td>
<td>75.18±1.89a</td>
<td>4.24±0.19b</td>
<td>1.51±0.06b</td>
</tr>
<tr>
<td>Mousami</td>
<td>72.42±0.15c</td>
<td>3.51±0.20d</td>
<td>1.35±0.09c</td>
</tr>
<tr>
<td>Grapefruit</td>
<td>71.35±1.33d</td>
<td>3.45±0.09d</td>
<td>1.68±0.05a</td>
</tr>
<tr>
<td>Eureka lemon</td>
<td>75.39±1.11a</td>
<td>4.57±0.30a</td>
<td>1.52±0.05b</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Citrus fruits</th>
<th>Fiber (%)</th>
<th>Protein (%)</th>
<th>NFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kinnow</td>
<td>5.46±0.17bd</td>
<td>5.21±0.15b</td>
<td>10.22±1.11b</td>
</tr>
<tr>
<td>Fewtrell’s early</td>
<td>5.30±0.15c</td>
<td>6.11±0.10a</td>
<td>10.69±1.19b</td>
</tr>
<tr>
<td>Malta</td>
<td>5.25±0.23cd</td>
<td>5.26±0.09b</td>
<td>8.43±1.26c</td>
</tr>
<tr>
<td>Mousami</td>
<td>5.53±0.25b</td>
<td>5.25±0.14b</td>
<td>11.68±1.49a</td>
</tr>
<tr>
<td>Grapefruit</td>
<td>6.11±0.19a</td>
<td>6.24±0.12a</td>
<td>11.06±1.65a</td>
</tr>
<tr>
<td>Eureka lemon</td>
<td>5.15±0.13d</td>
<td>6.09±0.15a</td>
<td>7.04±0.55d</td>
</tr>
</tbody>
</table>

Mean values sharing same letter are non-significant to each other (P> 0.01).

Ash contents of the peels of these fruits were also determined. The results indicated highly significant difference in ash contents of all peels ranging from 3.45-4.57 percent, with minimum in Grapefruit and maximum in Eureka lemon. With regards to fat contents, also significant difference was noted among varieties with the highest mean value in Grapefruit peel (1.68%) and the lowest in mousami peel (1.35%). The results of crude fibre contents of peels showed almost the same pattern and with highly significant difference in peels of these fruits, being the highest in grapefruit (6.11%) followed by Mousami (5.53%), Kinnow (5.46%), Fewtrell’s Early (5.30%), Malta (5.25%) and Eureka lemon (5.15%).

Similarly, citrus varieties showed highly significant difference in crude portion i.e. 5.23, 6.11, 5.375, 5.24 and 6.02 percent in Kinnow, Fewtrell’s Early, Malta, Mousami, Grapefruit and Eureka lemon, respectively. For nitrogen free extracts (NFE), highly significant difference was noted among these peels. The highest mean value of NFE was noted in mousami (11.68%) followed by Grapefruit (11.06%), Kinnow (10.22%), Fewtrell’s early (10.69%) and Eureka lemon (10.89%).

lemon (8.04%). NFE contents displayed its inverse proportion to the oil yield as Malta and Eureka lemon peels which had less NFE and more oil yield.

In a previous study (3) peels of grapefruit and orange were evaluated on dry weight bases and found that grapefruit peel contains 17.90 percent dry matter, 3.90 percent ash, 1.70 percent crude fat, 6.70 percent crude protein, 10.60 percent crude fiber and 77.10 percent carbohydrates. Similarly, the peel of orange contained 16.10 percent dry matter, 3.70 percent ash, 1.90 percent crude fat, 6.80 percent crude protein, 6.20 percent crude fiber and 81.40 percent carbohydrates. While studying varietal effects on the production of glucose through acid hydrolysis of citrus peels, Aftab (1) evaluated the peels of Mousami, Grapefruit and Kinnow for proximate analysis on dry weight basis and found almost similar results. The results on moisture and crude protein of Kinnow peel also agree to those of Raza (21), who reported 78.9 percent the moisture and 7.08 percent protein contents of Kinnow.

The slight variation in moisture, ash, ether extract, fiber and NFE of citrus peels in present and earlier studies may be due to variation in climate, type of soil, cultural practices, season and maturity and ripening conditions of fruit (12, 20). With regard to difference in protein content, it is argued that intercropping is a common practice in Pakistan, where, soils are deficient in N, P, Zn, Mn and Fe (16). So, the deficiency of N might be the reason of reduced amount of protein in the citrus peels.

**Aldehyde contents:** Aldehyde contents of citrus peel oils are the primary indicator of flavor strength and reference of quality. The results (Table 2) exhibited highly significant difference among aldehyde contents peel oils. These results also delineated that Kinnow peel oils had the highest aldehyde contents (42.73%) followed by Grapefruit (40.47%), Fewtrell’s Early (32.62%), Mousambi (26.79%), Malta (17.68%) and Eureka lemon (11.80%).

**Acid number:** Acid number is also one of the parameters to judge the quality of essential oils. The results indicated highly significant difference also in acid number of peel oils of these fruits. Maximum acid number was observed in Kinnow (2.21) followed by Grapefruit (1.88) against minimum in Malta (1.25) comparison of means revealed that the acid number of kinnow, fewtrell’s early, malta, mousami, grapefruit and eureka lemon was 2.213, 1.337, 1.253, 1.323, 1.863 and 1.390, respectively.
Variability in peel composition and quality evaluation

**Ester number:** The peel oils of the citrus fruits were also analysed for ester number. The results indicated highly significant difference in ester number of these oils. Mean values revealed the lowest ester number (11.74) in Malta peel oil whereas grapefruit peel oil, had the highest (27.12) (Table 2).

Table 2. Mean values of quality parameters of essential oils of various citrus fruits.

<table>
<thead>
<tr>
<th>Citrus cultivars</th>
<th>Mean values</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Aldehyde contents (%)</td>
</tr>
<tr>
<td>Kinnow</td>
<td>42.73±0.67a</td>
</tr>
<tr>
<td>Fewtrell's early</td>
<td>32.62±1.37c</td>
</tr>
<tr>
<td>Malta</td>
<td>17.68±0.17e</td>
</tr>
<tr>
<td>Mousami</td>
<td>26.79±2.11d</td>
</tr>
<tr>
<td>Grapefruit</td>
<td>40.47±1.44b</td>
</tr>
<tr>
<td>Eureka lemon</td>
<td>11.80±0.32f</td>
</tr>
</tbody>
</table>

Mean values sharing same letter are non-significant to each other (P> 0.01).

feasibility of cold expressing method. Ojeda de Rodrígue (19) studied the chemical properties of oils of Brazilian mandarin peel, Palestine sweet orange peel and California lemon peel. They observed delineated that Brazilian mandarin oil had 1.40-1.50 percent aldehyde contents, whereas, aldehyde contents of Palestine sweet orange oil and California lemon oil were 1.40 and 2.30 to 3.50, respectively. Similarly, present results are also in compliance with those of Guenther (11) who observed that ester number of mandarin, orange, grapefruit and lemon oils ranged from 18.7 to 20.8, as acetylene and acid number of cold pressed oil of mandarin, grapefruit and eureka lemon was 2.70, 1.70 and 1.50, respectively. The varying results of chemical characteristics of citrus peel essential oils, in previous studies may be due to the variation in climatic conditions, season, fertilizer application, rootstock (14), locality, growth period, time of harvest, type of soil, fruit size, storage conditions of fruits and oils, stage of maturity and method of extraction (13).

CONCLUSION

It is concluded that peel portion of Eureka lemon was the highest followed by Mousami, Grapefruit, Malta, Kinnow and Fewtrell’s early, which might be due to genetic variability of secreting glands and environmental factors. It was also found that NFE contents were inversely related to the oil yield as depicted from results of Malta and Eureka lemon. It has also been concluded that Fewtrell’s Early had the lowest yield, whereas, Malta had the highest. It might be due to the presence of gums, difference in variety and ratio of albedo and flavedo layers etc., as delineated by earlier studies.

In case of chemical examination the study concludes that kinnow fruit peel oil had the highest aldehyde contents and referred to be more valued while comparing with other peel oils as endorsed by earlier studies in this regard. Similarly, the results also revealed that malta peel oil had the lowest acid and ester numbers as compared to other essential oils, which may contribute to stability in quality of peel essential oils. However, some variability in composition and quality of peel oils of Pakistani citrus varieties might be rather due to several environmental and physiological factors such as tissue age (maturity), climate season, soil type, presence of gums and storage conditions.

REFERENCES


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CONTRIBUTION OF AUTHORS:

Muhammad Mushtaq Ahmad : Planned and supervised the study
Salim-ur-Rehman : Assisted in writeup
Tahir Mahmood Qureshi : Assisted in conducting the research
Muhammad Nadeem : Assisted in applying statistical analysis
Muhammad Asghar : Assisted in conducting the research work