Food Chemistry 205 (2016) 112–121

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Determination of sugars, organic acids, aroma components, and carotenoids in grapefruit pulps

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ARTICLE INFO

Article history:
Received 15 October 2015
Received in revised form 1 February 2016
Accepted 2 March 2016
Available online 3 March 2016

Keywords:
Grapefruit
Sugar
Organic acid
Volatiles
Carotenoids
HPLC
GC–MS

ABSTRACT

The composition and content of sugars, organic acids, volatiles and carotenoids, in the pulps of six grapefruit cultivars, were examined by HPLC and GC–MS. The results showed that sucrose was the dominant sugar in grapefruit, making up 40.08–59.68% of the total sugars, and the ratio of fructose to glucose was almost 1:1. Citric acid was the major organic acid and represented 39.10–63.55% of the total organic acids, followed by quininic acid. The ratios of individual sugars and organic acids play an important role in grapefruit taste determination. Monoterpenes and sesquiterpenes were the predominant volatiles in grapefruit, in particular D-limonene and caryophyllene. Caryophyllene, α-humulene, humulen-(v1), β-linalool and tert-butyl 2-methylpropanoate are the characteristic aroma compounds of grapefruit. Although β-carotene is the primary carotenoid in grapefruit, the pulp color is mainly determined by the ratios of zeaxanthin, β-cryptoxanthin and lycopene. Our results provide the first complete chemical characterization of the taste, aroma and color of grapefruit.

1. Introduction

Citrus is a large botanical family in which the dominant members are the sweet orange (*Citrus sinensis*), mandarin or tangerine orange (*Citrus reticulata*), grapefruit (*Citrus paradisi*), lemon (*Citrus limon*), and lime (*Citrus aurantifolia*). Grapefruit is one of the major commercial citrus crops, for both the fresh market and for processing (Chebrolu, Jayaprakasha, Jifon, & Patil, 2012). Grapefruits have a unique shape, flavor, color and a long shelf life, all qualities that are attractive to consumers. In addition, they are also an excellent source of many nutrients and phytochemicals that contribute to a healthy diet. Currently, there is an increasing interest in grapefruit because consumption appears to be associated with a reduced risk of certain chronic diseases, such as obesity, diabetes, cancers and cardiovascular disease (Kelebek, 2010).

Taste, aroma and color are important fruit quality factors that determine consumer preference. These traits also provide important information or sensory cues about the nutritional makeup of plant products (Goff & Klee, 2006; Kader, 2008). Grapefruit has a unique, special flavor and a colorful flesh. Its flavor is derived from a combination of its taste and aroma. The taste of grapefruit primarily depends on sugars and organic acids, whereas its aroma depends on a large number of volatile organic compounds (VOCs). Many studies have shown that the primary organic acids of citrus are citric and malic acid, and sucrose is present in large amounts in citrus fruit (Karadeniz, 2004). Previous studies have addressed how thermal treatment, storage (Igual, García-Martínez, Camacho, & Martínez-Navarrete, 2010) and hot air treatment influence the organic acid and sugar metabolism (Chen et al., 2012), sugar, organic acid, and phenolic composition of grapefruit (*C. paradisi* cvs. Rio Red, Star Ruby, Ruby Red and Henderson) (Kelebek, 2010), and the taste-related chemicals in *Ziziphus mauritiana* fruit (Muchuweti, Zenda, Ndhlala, & Kasiyamhuru, 2005). There is no doubt that volatile components play a determinant role in the grapefruit flavor quality. Many studies have also investigated the volatile components in pummelo peel (Cheong et al., 2011; Chung et al., 2012; Shao et al., 2014), essential oil (Sun et al., 2014) and juice (Cheong, Liu, Zhou, Curran, & Yu, 2012). Few researchers have investigated the volatile composition of grapefruit. Ren et al. (2015) characterized the free and bound volatile compounds from pink grapefruit and white grapefruit. Njoroge, Koaze, Karanja, and Sawamura (2005) analyzed the volatile constituents of Red Blush grapefruit (*C. paradisi*) peel essential oils from Kenya. The external color of citrus fruits is one of their most important quality traits, and it is a decisive factor for consumers.

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http://dx.doi.org/10.1016/j.foodchem.2016.03.007
0308-8146/

Grapefruit is characterized by its white, pink and red colors; the coloration of the pulp is primarily influenced by the presence of carotenoids (Rodrigo, Alquézar, Alós, Lado, & Zacarías, 2013). Alquezar, Rodrigo, Lado, and Zacarías (2013) analyzed the carotenoid biosynthetic differences between white and red grapefruit (*C. paradisi* Macf.). Xu, Fraser, Wang, and Bramley (2006) investigated the carotenoid content differences between ordinary citrus and mutant fruits. Alquezar et al. (2013) conducted a comparative physiological and transcriptional study of carotenoid biosynthesis in white and red grapefruit (*C. paradisi* Macf.). Despite previous studies, there is still much to be learned about grapefruit taste, aroma, and color composition.

The objectives of the current study were to identify the composition and content of soluble sugars, organic acids, volatile components and carotenoids in grapefruit pulps, and to create a comprehensive chemical characterization on the taste, aroma and color of grapefruit.

2. Materials and methods

2.1. Chemicals

Sugars (fructose, sorbitol, glucose and sucrose) and organic acids (oxalic acid, tartaric acid, quininic acid, malic acid, citric acid and aconitic acid) were all obtained from Shanghai Sangon Biological Reagent Company (Shanghai, China). *n*-Hexanol, methyl myristate, lutein, zeaxanthin, β-cryptoxanthin lycopene, α-carotene, and β-carotene were obtained from Sigma (St. Louis, MO, USA). All other reagents were of analytical grade and were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

2.2. Fruit materials

Six grapefruit (*C. paradisi* Macf.) cultivars were grown at the National Citrus Germplasm Repository in the Citrus Research Institute at the Chinese Academy of Agricultural Sciences, Chongqing, China (Table 1). All experimental trees were planted in 2001, in rows, in a north–south orientation, with a distance of 3–4 m between rows. Fertilization management and pest control were carried out according to standard practices of the germplasm repositories. During the 2014 harvest season (from the 12th to 30th of January), a total of 240 fruits were picked, from ten trees at the commercial maturity stage, on the basis of external color and size uniformity for each cultivar (Fig. 1). After harvest, fruits were randomly divided into three replicates and manually peeled. Only the pulp was used as the experimental material. Each replicate included 80 fruits. Among these fruits, 20 were used to determine the titratable acid (TA) and soluble solids content (SSC). Sixty grapefruits were ground into a fine powder in liquid nitrogen using a freezer-mill (6750) apparatus (Glen Creston), and then the powder was stored at -80 °C until analysis. In the study, three replicates were performed for all chemical analyses.

2.3. Soluble solids content and titratable acidity determination

SSC and TA were determined according to the method described by Ramful, Tarnus, Aruoma, Bourdon, and Bahorun (2011). Firstly, the pulp juice of peeled fruit was dropped on a digital refractometer (Atago PR-101R, Tokyo, Japan) and the value was read. Each replicate contained 20 fruits and all determinations were performed in triplicate. The temperature of the sample at the time of measurement was also recorded. The degree (°) Brix of the juice was then calculated and a temperature correction was applied. After measuring the SSC, the pulps of all 20 fruits were homogenized in a Waring blender and filtered with muslin cloth. Ten ml of the juice was diluted to 100 ml with distilled water and transferred into a 250 ml beaker, which was placed over a magnetic stirrer to provide continuous stirring of the sample solution. A pH meter probe was then immersed in the solution, and 0.1 N NaOH was added until the pH of the sample exceeded 8.1. TA was expressed as percentage of citric acid (%) and three replicates were used.

Table 1
Grapefruit cultivars used in the present study and their quality index values of pulps.[a,b,c,d]

No.	Repository number	Cultivars	Abbreviation	SSC (%)	TA (%)
1	LG0093	*C. paradis* cv. Marsh	MG	9.10 ± 0.01[d]	1.87 ± 0.09[b]
2	LG0120	*C. paradis* cv. Oroblanco	OR	11.53 ± 0.2[c]	0.90 ± 0.04[c]
3	LG0245	*C. paradis* cv. Cock Tail	CT	12.37 ± 0.31[b]	0.69 ± 0.03[d]
4	LG0094	*C. paradis* cv. Thompson	TG	11.77 ± 0.45[c]	1.92 ± 0.06[b]
5	LG0243	*C. paradis* cv. Red Blush	RB	13.27 ± 0.38[a]	2.14 ± 0.09[a]
6	LG0248	*C. parades* cv. Rio Red	RR	13.13 ± 1.02[a]	1.87 ± 0.03[b]

[a] SSC, soluble solid content.
[b] TA, titratable acidity.
[c] Data are expressed as means ± standard deviation of triplicate samples.
[d] Different lowercase letters between columns represent significant differences between cultivars ($p < 0.05$).

2.4. Determination of sugars and organic acids

Sugars and organic acids were extracted as described by Zhang et al. (2005). Two grams of pulp powder was homogenized by using 5.0 ml of cold ethanol (80%). The solution was then incubated for 20 min in a 35 °C water bath and centrifuged at 10,000×*g* for 10 min. This extraction procedure was repeated three times and the supernatants were combined. The total volume was then adjusted to 25 ml with 80% ethanol. From this mixture, 1 ml was dried under a vacuum (Eppendorf Concentrate Plus, Germany) at 45 °C, and the residue was resuspended in 0.5 ml of distilled water and filtered through a 0.22 μm, 13 mm diameter syringe filter (Shanghai Xingya Purification Material Factory, China). The filtered solution was then used for the sugar and organic acid analysis.

Sugars were analyzed as described previously with some modifications (Gancedo & Luh, 1986). A chromatographic separation of sugars involved acetonitrile: water (80:20, v/v) as the mobile phase at a flow rate of 1.4 ml/min with an Agilent ZORBAX Carbohydrate (4.5 μm, 4.6 mm × 250 mm) column (GL Sciences Inc., Torrance, CA, USA). Eluted peaks were detected with a SHODEX RI101 refractive index detector (JASCO International Co. Ltd, Tokyo, Japan). The data were analyzed with a Chromeleon® 6.8 chromatography data system (Thermo Fisher Scientific Inc., USA).

Organic acids were analyzed by HPLC, as described previously with some modifications (López-Hernández, Oruña-Concha, Simal-Lozano, Vázquez-Blanco, & González-Castro, 1996). The chromatographic separation used for organic acid detection employed $(NH_4)_2HPO_4$ (50 mM, pH 2.7) as the mobile phase, with a flow rate of 0.5 ml/min, and the samples were injected into an ODS C_{18} (4.6 mm × 250 mm) column (Beckman Coulter Inc., Brea, CA, USA). Organic acids were detected with a 2996 diode array detector (Waters Beckman Coulter Inc., Brea, CA, USA). The data were analyzed with a Waters Empower system.

Sugars and organic acids were detected at a wavelength of 210 nm. A calibration curve was prepared using commercial

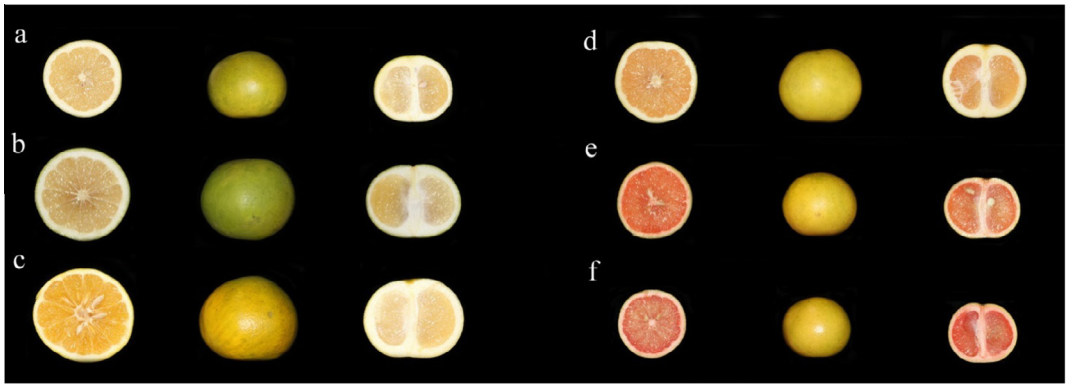

Fig. 1. Grapefruit used in the present study. a, Marsh Grapefruit; b, Oroblanco; c, Cock Tail; d, Thompson Grapefruit; e, Red Blush; f, Rio Red.

standards to determine the relationship between the peak area and concentration. The sugar and organic acid concentrations were expressed as mg/g fresh weight (FW). Three replicates were used for all samples.

2.5. Determination of aromatic volatiles

The concentrations of volatiles were determined according to a previously reported method with some modifications (Eduardo, Chietera, Bassi, Rossini, & Vecchietti, 2010; Xi et al., 2014). 1.5 g of pulp powder was homogenized with 3 ml saturated sodium chloride solution, and then 20 μl authentic *n*-hexanol and methyl myristate were added as the internal standards, to quantify the volatile compounds. The solution was held at 40 °C for 30 min. A solid-phase microextraction (SPME) needle with a 1 cm long fiber coated with 65 μm of divinylbenzene/carboxen/polydimethylsiloxane (DVB/CAR/PDMS) fibers (Supelco Co., Bellefonte PA, USA) was used for volatile extraction.

A GCMS-QP2010 gas chromatograph-mass spectrometer system (Shimadzu Corporation, Kyoto, Japan) with an Rtx-5MS (Restek)-fused silica capillary column (5% diphenyl, 95% dimethyl polysiloxane) (0.32 mm, 30 m, 0.5 μm, J&W Scientific, Folsom CA, USA) was used for compound confirmation. The injection port temperature was 240 °C. The injection volume was 1 μl. Helium was used as the carrier gas at a rate of 1.0 ml/min. The GC oven temperature was held at 40 °C for 3 min, increased by 4 °C/min to 250 °C, and then held for 5 min. Mass spectra were obtained by electron ionization (EI) at 70 eV and a scan range of 40–500 mass units. The detector, ion source and transfer line temperature were set to 150, 200 and 250 °C, respectively.

The chromatograms and mass spectra were evaluated with the GC–MS Postrun Analysis software (SHIMADZU, GC-MS-QP2010, Japan). The compounds were tentatively identified by comparing their mass spectra with those in the data system library (NIST08). Quantitative analysis was carried out using the internal standard method as described by Lan-Phi, Shimamura, Ukeda, and Sawamura (2009). The concentrations of volatile components were expressed as μg/g FW. Three replicates were used in each sample.

2.6. Determination of carotenoids

Carotenoid extraction was conducted as described by Lee and Castle (2001) with some modification. Pulp powder (10 g) was homogenized in 25 ml of extracting solvent (hexane/acetone/ethanol, 50:25:25, v/v) in screw-top tubes. The mixtures were allowed to stand for 30 min in the dark at room temperature. Samples were then centrifuged for 5 min at 6500 rpm at 5 °C (AllegraTM64R Centrifuge, Beckman CoulterTM, USA). The top colored layer of hexane was recovered and transferred to a round-bottom flask, then dried under nitrogen gas with a bath-type nitrogen blowing instrument (FerrenTechnology Co. Ltd).

For saponification, the residue was redissolved with 2 ml of methyl tert-butyl ether (MTBE) and 2 ml of 10% methanolic KOH. The round flask was wrapped with aluminum foil to protect the solution from light, and was shaken overnight at room temperature. The samples were transferred to a separator funnel, and 5 ml of water and 2 ml of 0.1% BHT (butylated hydroxytoluene)/MTBE was added. The colored layer was collected and dried under nitrogen. The residue was dissolved in 2 ml methanol/acetone (2:1, v/v). After being filtered through a Millipore 0.5 μm filter, 1.5 ml solution was injected into the HPLC apparatus for analysis. Analyses were performed under red light to avoid carotenoid degradation during extraction and saponification.

The carotenoids were determined with a Waters High Performance Liquid Chromatographic Acquity system (HPLC) (Milford, MA, USA) equipped with a 2996 PDA detector, an autosampler cooler, a binary solvent delivery module, version 4.1 firmware, a degassing system, a column heater and a C_{30} column (250 × 4.6 mm i.d., 5 μm) (YMC, Wilmington, NC, USA). The system was operated by the Waters Empower software (from Waters Corporation). HPLC conditions were conducted according to the method of Mouly, Gaydou, Lapierre, and Corsetti (1999). The gradient profile and the mobile phase composition are given in Table S1. The flow rate was fixed at 1 ml/min, and the column temperature was set at 25 °C. The absorbances were recorded at 278, 350, 430 and 486 nm. The carotenoids were identified by comparing their retention time and the UV–visible spectra of their peaks with those of the standards. The carotenoids were quantified by a calibration curve of commercial standards and the results were expressed as μg/g. Three replicates were used for all samples.

2.7. Statistical analysis

All data are expressed as the means ± standard deviation of three replicates. A statistical analysis was performed using SPSS v19.0 software (SPSS Inc., Chicago, IL, USA). Significant differences among the samples were calculated using one-way ANOVA followed by Duncan's multiple-range test at the 5% level ($p \leqslant 0.05$).

3. Results and discussion

3.1. Soluble solids content and titratable acidity

The TA and SSC of the grapefruit pulp are shown in Table 1. The SSC of the grapefruit cultivars tested ranged from 9.10% to 13.27%. Cocktail grapefruits had the highest measured SSC value (13.27%),

followed by Rio Red (13.13%). Except for Marsh grapefruits, the SSC of all tested grapefruit cultivars was more than 10%. The TA of all fruits tested ranged from 0.69% to 2.14%. Cocktail grapefruits were the least acidic with a TA value of 0.69% and Red Blush was the most acidic with a TA value of 2.14%. The TA values of Oroblanco and Cocktail grapefruits were less than 1%, but the fruit pulps of the other cultivars were characterized by high TA (>1%).

3.2. Sugar composition and content

Among the four sugars tested (Fig. S1), sucrose, fructose and glucose made up 40.08–59.68%, 23.31–30.99% and 19.01–28.92% of total sugars, respectively (Table 2). The three sugar contents ranged from 26.80 to 71.47 mg/g FW, 17.07–28.75 mg/g FW and 13.77–26.83 mg/g FW in the pulps of the tested grapefruits, respectively. The total sugar content ranged from 57.63 to 120 mg/g FW; Cocktail had the highest level of total sugar and Marsh had the lowest level. Over 100 mg/g FW of total sugar contents were observed in the pulps of Oroblanco, Cocktail, Red Blush and Rio Red grapefruits. No sorbitol was detected in the tested grapefruit.

The citrus fruit pulp primarily contained the following three major sugars: sucrose, fructose and glucose (Albertini et al., 2006). It is well known that fruit sweetness depends not only on the content of each sugar, but also on the ratios of the main individual sugars. The present study showed that sucrose, fructose and glucose are the main sugars in grapefruit and make up over 80% of the total soluble solids. This finding is consistent with previous reports (Kelebek, 2010; Rouseff & Martin, 1985; White & Widmer, 1990). In this study, although similar levels of total sugars were found in grapefruit and sweet lemons (Yu et al., 2012), the sweetness level of the two fruits is significantly different. This discrepancy is primarily a result of the composition ratio of the three major sugars across citrus species/cultivars. In the present study, we found that sucrose is generally the dominant sugar in grapefruit and the ratio of sucrose, fructose and glucose was approximately 2:1:1. However, the ratios of these three sugars are almost equal in sweet orange (Yu et al., 2012). Kelebek and Selli (2011) also found that orange fruits contain glucose and fructose in nearly equal quantities or that fructose is present in a slightly higher amount. Albertini et al. (2006) found that fructose, glucose, and sucrose were the major sugars in lemon, lime and orange, but fructose was the only sugar for which a significant difference could be observed between all acidic and acidless varieties. Fructose concentrations were significantly higher in the two acidless varieties, which is consistent with the fact that fructose is the sweetest of the three main sugars.

3.3. Organic acid composition and content

In the present study, six organic acids, including oxalic acid, tartaric acid, quininic acid, malic acid, citric acid and aconitic acid, were identified in the tested grapefruits (Fig. S2 and Table 3). Citric acid was the dominant organic acid in these pulps, and occupied 39.10–63.55% of total organic acid content, ranging from 12.71 to 27.17 mg/g FW. Quininic acid was the second largest amount of organic acid in the tested grapefruit, with 15.70–33.43% of the total organic acid, ranging from 5.97 to 11.97 mg/g FW. Tartaric acid and malic acid were the third-most common organic acids, and occupied 9.12–13.68% and 5.31–14.43% of total organic acids, respectively. Their contents ranged from 3.63 mg/g FW to 5.89 mg/g FW, and 2.21 mg/g FW to 4.15 mg/g FW in the tested grapefruit, respectively. Oxalic acid was detected in the tested grapefruit, but only represents 1.61–4.26% of the total organic acid, and its content ranged from 0.51 mg/g FW to 1.38 mg/g FW. Trace aconitic acid was detected at less than 3%, with contents below 0.1 mg/g FW in the tested grapefruit. The total organic acid content ranged from 28.75 mg/g FW to 46.59 mg/g FW, where Cocktail grapefruits had the lowest level of total organic acid and Red Blush had the highest level. The total organic acid in Oroblanco, Cocktail, and Red Blush grapefruit was less than 35 mg/g FW.

Many previous studies have shown that citrus fruits primarily contain malic acid, citric acid, quininic acid and oxalic acid. Together, these organic acids influence the citrus fruit acidity. Among these organic acids, citric acid was the largest contributor to the acidity taste, and accounted for 79%, 71%, and 45% of total organic acids in acidic lemon, lime, and orange, respectively (Albertini et al., 2006). In the present study, we found the ratio of citric acid to total organic acids was similar to that of orange. However, the organic acid composition was significantly different across citrus species/cultivars, which is another factor that contributes to the taste difference in different species/cultivars of citrus fruits. The present results suggested that citric acid, quininic acid, tartaric acid and malic acid are the predominant organic acids in grapefruit, as they presented more than 83% of the total organic acids. The ratio of citric acid, quininic acid, tartaric acid and malic acid was about 4:2:1:1, which may contribute to the characteristic acidity taste of grapefruit. Albertini et al. (2006) found that in lemons, limes and oranges, citric acid dominated in acidic varieties, but malic acid exceeded citric acid in acidless varieties. In sweet oranges, the citric acid (1 mg/g FW) and malic acid (1 mg/g FW) contents were two times higher than quinic acid (Yu et al., 2012). However, in the present study, we found that the citric acid and malic acid contents in grapefruit were over three times higher than in sweet orange. Additionally, the quininic acid content was over ten times higher than that of sweet orange. As with the effect of sugars on the sweetness of fruit, besides the absolute content of individual organic acids, the differences in acidity taste across the citrus species/cultivars may also depend on the ratio of organic acids.

3.4. Aromatic volatile composition and content

Table 4 lists the VOCs detected using a HS-SPME-GC–MS platform and the real total ionic chromatogram (TIC) of the volatiles for grapefruit pulp is presented in Fig. S3. A total of 170 volatile compounds have been identified as follows: 20 monoterpenes, 48 sesquiterpenes, 21 terpenic alcohols and aldehydes (18 terpenic alcohols, 3 terpene aldehydes), 18 aliphatic alcohols and aldehydes, 9 ketones, 21 esters (13 aliphatic and 8 monoterpenic acetates), 21 alkanes and 12 others, some of which have been identified only tentatively. Monoterpenes (43.56–72.11%) and sesquiterpenes (11.16–45.39%) were the predominant volatile components of the tested grapefruit pulps, followed by aliphatic alcohols and aldehydes (0.73–12.33%), and then terpenic alcohols and aldehydes (1.19–5.63%). Esters and alkanes were responsible for 0.71–3.16% and 0.67–2.98% of the total VOCs in the tested grapefruit, respectively. 0.25–1.39% of ketones and 0.35–3.46% of other compounds (such as terpenic oxide and aromatic hydrocarbons) were found in these grapefruit.

With regards to the chemical-specific composition, D-limonene was the major VOC in grapefruit pulps, and occupied 30.07–67.55% of total volatiles which was approximately 47.48–95.77% of the total monoterpenes. β-Pinene (2.29–48.09%) presented the second-largest amount of monoterpenes, and D-limonene, β-pinene and 1R-α-pinene were detected in all the tested grapefruit pulps. α-Phellandrene and trans-4,5-epoxy-carane were only detected in Rio Red grapefruits, whereas o-cylene was only detected in Thompson grapefruits. In the sesquiterpene group, caryophyllene was the most common compound in all grapefruit pulps tested, and the ratio to total sesquiterpenes was 28.35–88.75%. α-Humulene was the second-most frequent sesquiterpene in the grapefruit pulps, followed by isocaryophillene. Caryophyllene (9.90–25.69%) was the second-most common compound of all VOCs identified in

Table 2
Sugar content (mg/g FW) in pulp of the grapefruit.[a,b,c]

No.	Cultivars	Fructose	Sorbitol	Glucose	Sucrose	Total
1	MG	17.07 ± 1.33[c]	nd	13.77 ± 0.60[d]	26.80 ± 0.85[e]	57.63 ± 2.78[d]
2	OR	25.59 ± 0.40[b]	nd	22.82 ± 0.40[c]	59.48 ± 0.97[c]	108 ± 1.77[b]
3	CT	25.52 ± 0.47[b]	nd	22.76 ± 0.99[c]	71.47 ± 1.71[a]	120 ± 3.17[a]
4	TG	28.75 ± 0.50[a]	nd	26.83 ± 1.05[a]	37.19 ± 0.95[d]	92.76 ± 2.51[c]
5	RB	24.57 ± 0.45[b]	nd	21.13 ± 0.32[c]	63.14 ± 2.18[b]	109 ± 2.95[b]
6	RR	28.51 ± 1.17[a]	nd	24.73 ± 0.79[b]	55.43 ± 1.14[c]	109 ± 3.11[b]

[a] nd represents not detectable.
[b] Data are expressed as means ± standard deviation of triplicate samples.
[c] Different lowercase letters between columns represent significant differences between cultivars ($p < 0.05$).

Table 3
Organic acid content (mg/g FW) in pulp of the grapefruit.[a,b]

No.	Cultivars	Oxalic acid	Tartaric acid	Quininic acid	Malic acid	Citric acid	Aconitic acid	Total
1	MG	1.14 ± 0.07[b]	3.63 ± 0.47[c]	6.37 ± 0.60 d	3.30 ± 0.33[b]	25.29 ± 0.08[c]	0.08 ± 0.03[a]	39.81 ± 1.56[c]
2	OR	1.38 ± 0.30[a]	4.58 ± 0.94[b]	10.86 ± 0.48[b]	2.89 ± 0.67[b]	12.71 ± 0.08 d	0.08 ± 0.01[a]	32.50 ± 2.48 d
3	CT	0.71 ± 0.16 d	3.67 ± 0.92[c]	7.34 ± 0.54[c]	4.15 ± 0.85[a]	12.79 ± 0.08 d	0.08 ± 0.01[a]	28.75 ± 2.55 e
4	TG	1.05 ± 0.10[b]	4.10 ± 0.42[b]	7.72 ± 0.56[c]	2.21 ± 0.19[c]	26.43 ± 0.09[b]	0.09 ± 0.01[a]	41.59 ± 1.35[c]
5	RB	0.51 ± 0.04 e	4.32 ± 0.63[b]	11.97 ± 0.85[a]	3.23 ± 0.39[b]	26.47 ± 0.09[b]	0.09 ± 0.01[a]	46.59 ± 1.99[a]
6	RR	0.93 ± 0.11[c]	5.89 ± 0.53[a]	6.82 ± 0.57 d	2.52 ± 0.29[c]	27.17 ± 0.10[a]	0.10 ± 0.01[a]	43.42 ± 1.60[b]

[a] Data are expressed as means ± standard deviation of triplicate samples.
[b] Different lowercase letters between columns represent significant differences between cultivars ($p < 0.05$).

the tested grapefruit pulps. Caryophyllene, β-elemene, γ-selinene, α-panasinsen, and δ-cadinene were detected in all the grapefruit pulps. γ-Elemene and α-caryophyllene were only detected in Rio Red; (-)-Aristolene and germacrene B were only found in Oroblanco; β-humulene, α-bergamotene, thujopsene and α-bulnesene were only detected in Marsh; β-panasinsen was only found in Thompson; and α-farnesene was only identified in Red Blush. With respect to terpenic alcohols and aldehydes, β-linalool, L-4-terpineol and α-terpieol were the predominant components, and were detected in all the tested grapefruit pulps. Elemol, tau.-Cadinol and L-perillaldehyde were only detected in Thompson. (-)-Citronellol and (−)-β-citronellol were only detected in Rio Red. Hexenal was the basis for the major aliphatic alcohols and aldehydes, and (*E*)-2-hexenal, heptanal, nonanal and (*E*)-carveol were all detected in all the tested grapefruit pulps. 3-Hexenal was only detected in Rio Red. Ketones were found as trace compounds in the tested grapefruit pulps. Nootkatone and D-dihydrocarvone were detected in all grapefruit pulps, and geranyl acetone was only found in Cocktail. Neryl acetate was the major ester in the tested grapefruit pulps. Acetic acid, butyl ester and butanoic acid, hexyl ester, neryl acetate and tert-butyl 2-methylpropanoate were detected in all the grapefruit pulps. Heptyl ester and octyl propanoate were only detected in Rio Red grapefruit, with neryl acetate found only in Thompson grapefruit. Only a few alkanes were detected in the majority of grapefruit pulps tested here, and their contents were found in trace amounts. 11 alkanes were found in Marsh grapefruit. Some terpenic oxides, benzene and two furans were detected in the grapefruit pulps.

Currently, over 300 VOCs have been identified from *Citrus* fruits (Azam, Jiang, Zhang, Xu, & Chen, 2013). In the present study, more than 60% of these VOCs were found in grapefruit pulp, which showed that grapefruit pulp is rich in VOCs. Although many previous studies have extensively shown that monoterpenes and sesquiterpenes are the predominant VOCs, limonene was the most abundant component in citrus fruits, representing up to 97% in orange (*C. sinensis*) fruit peel (Rodríguez et al., 2011), 88% in mandarins (Tietel, Plotto, Fallik, Lewinsohn, & Porat, 2011), and 80% of the total volatiles in *C. reticulata* Blanco (ponkan) essential oil (Sawamura et al., 2004). However, each species has its own VOCs profile (Rambla et al., 2014). The present study showed that monoterpenes and sesquiterpenes (79.58–91.41%) were also the predominant VOCs in grapefruit; D-limonene (30.07–67.55%), caryophyllene (9.90–25.69%), β-pinene (2.29–48.09%), and α-humulene (0–3.73%) were the dominant VOCs in these grapefruit pulps, which was consistent with main volatile constituents of grapefruit juice (Buettner & Schieberle, 1999), and was similar with the VOCs profile of Shiikuwasha fruits (*Citrus depressa* Hayata) (Asikin et al., 2012). However, although pommelo peels contain substantial quantities of olefins, such as limonene, β-myrcene, α-pinene, β-pinene, α-phellandrene and terpinolene, there are obvious differences in the VOCs profile obtained from pommelos under the same test conditions (Shao et al., 2014; Sun, Ni, Yang, Chen, 2014). Limonene (91.1%) was the most abundant compounds in Red Blush grapefruit oil, followed by R-terpinene (1.3%), R-pinene (0.5%), and sabinene (0.4%). The ratios of R-thujene, R-pinene, γ-terpinene, and terpinolene in the grapefruit oil were less than 0.05% (Njoroge et al., 2005). Limonene and γ-terpinene accounted for approximately 88% of the total volatile components in mandarins, with linalool, α-terpineol, terpinen-4-ol, nonanal, decanal, carvone, limonene, α-pinene and myrcene making up the rest (Tietel et al., 2011). Limonene and myrcene were dominant in both parts of Dangyooja, although more volatile components were detected in Dangyooja peel (Chung et al., 2012). D-Limonene was the most abundant compound, representing 40.6–82.5% of the total aromatic volatiles. Among the 22 identified monoterpene hydrocarbons, four monoterpenes were also present in relatively high amounts in most samples, namely β-myrcene (0.5–4.9% of the relative peak area), *p*-cymene (0.3–7.0%), β-phellandrene (0–3.5%) and dehydro-*p*-cymene (0.5–9.5%) in tangerines and their hybrids (Miyazaki, Plotto, Goodner, & Gmitter, 2011). These results suggested that the differences in the volatile profiles of citrus fruits are primarily quantitative, and only a few compounds are variety-specific.

As mentioned above, monoterpenes are the major volatiles of citrus fruits, with D-limonene usually being the dominant compound. However, citrus aroma quality is mainly determined by its characteristic VOCs. Citrus fruits include not only high concentration volatiles, but also low concentration volatiles with low odor thresholds. As a result, the absence of even a few volatiles might be important for the aroma. Liu et al. (2012) found that monoterpene

Table 4
Volatiles content (μg/g FW) in pulp of the grapefruit.[a,b,c]

No.	Volatiles	1	2	3	4	5	6
		MG	OR	CT	TG	RB	RR
Monoterpene							
1	α-Phellandrene	nd	nd	nd	nd	nd	27.34 ± 1.06[a]
2	β-Phellandrene	36.33 ± 4.12[c]	51.24 ± 2.12[b]	18.25 ± 0.36[d]	15.4 ± 1.43[d]	nd	204 ± 3.22[a]
3	α-Pinene	159 ± 3.45[c]	69.31 ± 3.31[d]	72.52 ± 5.03[d]	144 ± 6.22[c]	249 ± 3.17[a]	192 ± 5.28[b]
4	Camphene	nd	nd	nd	nd	3.21 ± 0.44[a]	nd
5	β-Pinene	792 ± 21.41[c]	327 ± 2.56[d]	339 ± 2.78[d]	711 ± 21.65[c]	13142 ± 15.22[a]	1488 ± 110[b]
6	D-Limonene	11412 ± 108[b]	11130 ± 369[b]	14154 ± 609[a]	10698 ± 309[c]	12975 ± 397[b]	8388 ± 87.23[d]
7	Isolimonene	nd	nd	nd	nd	nd	894 ± 28.23[a]
8	α-Terpinene	93.61 ± 6.12[b]	21.43 ± 4.75[e]	15.86 ± 0.84[d]	72.52 ± 5.23[e]	138 ± 5.55[a]	57.22 ± 2.77[d]
9	Trans-Ocimene	nd	nd	nd	48.66 ± 3.12[a]	nd	24.05 ± 0.57[b]
10	Cis-Ocimene	nd	12.13 ± 1.04[b]	48.43 ± 0.98[a]	nd	nd	nd
11	2-Carene	nd	nd	nd	nd	408 ± 33.17[a]	nd
12	3-Carene	54.23 ± 2.32[d]	12.15 ± 1.65[e]	48.54 ± 3.89[d]	72.32 ± 3.19[c]	111 ± 4.33[b]	174 ± 3.18[a]
13	(+)-4-Carene	nd	nd	9.82 ± 0.33[c]	45.56 ± 0.61[b]	nd	420 ± 19.36[a]
14	γ-Terpinene	81.27 ± 2.23[c]	39.65 ± 3.14[d]	nd	84.55 ± 5.14[c]	126 ± 3.75[b]	192 ± 17.36[a]
15	α-Terpinene	15.35 ± 3.16[b]	nd	nd	12.34 ± 3.76[b]	45.93 ± 0.92[a]	6.78 ± 0.45[c]
16	Trans-4,5-epoxy-Carane	nd	nd	nd	nd	nd	30.15 ± 3.19[a]
17	o-Xylene	nd	nd	nd	15.07 ± 0.35[a]	nd	nd
18	p-Xylene	87.67 ± 6.12[b]	252 ± 3.52[a]	72.8 ± 4.15[b]	nd	54.32 ± 3.85[c]	51.54 ± 3.17[c]
19	Bornylene	30.29 ± 2.52[c]	96.53 ± 5.15[a]	nd	nd	78.33 ± 1.25[b]	nd
20	α-Thujene	nd	6.59 ± 5.22[a]	nd	nd	nd	nd
	Sum	12762 ± 160[c]	12018 ± 402[c]	14779 ± 627[b]	11919 ± 360[c]	27328 ± 468[a]	12149 ± 285[c]
	Relative	51.14%	50.08%	70.54%	43.94%	72.11%	43.56%
Sesquiterpene							
21	Longifolene-(V4)	nd	nd	nd	18.73 ± 0.75[a]	nd	9.24 ± 1.22[b]
22	γ-Elemene	nd	nd	nd	nd	nd	36.35 ± 1.45[a]
23	Elixene	nd	183 ± 6.35[a]	nd	nd	nd	42.43 ± 2.56[b]
24	α-Cubebene	108 ± 3.22[c]	638 ± 7.15[a]	nd	252 ± 11.87[b]	nd	240 ± 16.56[b]
25	α-Bisabolene	21.34 ± 3.15[a]	nd	nd	18.63 ± 0.67[a]	nd	12.16 ± 1.09[b]
26	Copaene	210 ± 18.36[d]	750 ± 7.17[a]	nd	570 ± 5.66[b]	153 ± 2.59[e]	468 ± 8.18[c]
27	β-lemene	54.12 ± 3.82[d]	249 ± 5.89[a]	12.1 ± 0.95[e]	135 ± 3.54[b]	66.5 ± 2.23[d]	105 ± 4.91[c]
28	δ-Elemene	nd	552 ± 7.45[a]	12.7 ± 0.72[b]	nd	nd	nd
29	α-Elemene	nd	24.32 ± 3.22[b]	72.65 ± 6.63[a]	18.32 ± 0.69[b]	nd	nd
30	Isocaryophillene	438 ± 26.92[a]	273 ± 5.45[c]	nd	348 ± 19.24[b]	327 ± 24.71[b]	249 ± 9.65[c]
31	Caryophyllene	6411 ± 39.17[a]	2745 ± 9.56[c]	2075 ± 92.36[d]	6684 ± 84.13[a]	4581 ± 42.18[b]	6426 ± 24.68[a]
32	α-Caryophyllene	nd	nd	nd	nd	nd	916 ± 4.43[a]
33	Caryophyleine-(I3)	24.09 ± 0.82[b]	nd	nd	30.61 ± 1.27[a]	21.42 ± 1.23[b]	18.56 ± 0.52[b]
34	α-Guaiene	nd	18.58 ± 5.87[d]	nd	87.42 ± 0.54[a]	39.45 ± 2.92[c]	66.35 ± 3.47[b]
35	Humulen-(v1)	120 ± 2.88[a]	48.52 ± 5.71[d]	nd	129 ± 17.86[a]	81.25 ± 29 ± 15[c]	105 ± 8.36[b]
36	(+)-Epi-bicyclosesquiphellandrene	102 ± 6.33b	105 ± 9.52[b]	nd	216 ± 7.19[a]	66.3 ± 5.52[c]	114 ± 5.31[b]
37	γ-himachalene	nd	nd	nd	30.48 ± 4.21[a]	nd	24.92 ± 3.13[b]
38	β-Cadinene	nd	nd	nd	18.79 ± 1.25[a]	nd	15.9 ± 1.16[a]
39	γ-Muurolene	nd	183 ± 2.53[a]	nd	nd	nd	30.82 ± 1.71[b]
40	α-Muurolene	6.86 ± 0.37[c]	nd	nd	1263 ± 9.73[a]	15.73 ± 1.56[c]	108 ± 13.19[b]
41	γ-Selinene	36.32 ± 1.15[c]	231 ± 11.47[a]	24.66 ± 2.21[d]	45.85 ± 3.39[c]	117 ± 2.33[b]	24.92 ± 0.97[d]
42	α-Selinene	nd	nd	nd	99.45 ± 4.16[a]	nd	75.15 ± 2.33[b]
43	β-Eudesmene	15.65 ± 2.6[b]	30.57 ± 2.78[a]	nd	30.92 ± 1.17[a]	18.92 ± 1.25[b]	27.65 ± 2.16[a]
44	β-Panasinsen	12.17 ± 1.46[c]	nd	nd	150 ± 8.12[a]	nd	99.66 ± 3.45[b]
45	α-Panasinsen	30.59 ± 1.63[d]	2076 ± 15.57[a]	18.85 ± 0.62[d]	42.33 ± 3.76[c]	84.12 ± 1.76[b]	24.26 ± 1.26[d]
46	γ-Cadinene	nd	54.33 ± 3.22[b]	15.24 ± 1.16[c]	96.48 ± 6.12[a]	15.44 ± 4.12[c]	66.55 ± 5.12[b]
47	δ-Cadinene	150 ± 2.33[d]	237 ± 7.81[c]	12.14 ± 0.82[e]	573 ± 8.37[a]	168 ± 6.55[d]	447 ± 3.22[b]
48	δ-Guaiene	nd	nd	nd	5.51 ± 0.28[b]	nd	42.35 ± 2.64[a]
49	α-Cedrene	33.53 ± 5.42[b]	nd	nd	84.52 ± 2.13[a]	nd	75.69 ± 3.44[a]
50	α-Calacorene	18.91 ± 9.72[b]	nd	nd	27.35 ± 3.13[a]	nd	18.65 ± 6.17[b]
51	γ-Gurjunene	nd	42.29 ± 1.92[a]	nd	15.35 ± 0.39[c]	33.22 ± 1.68[b]	12.73 ± 0.86[c]
52	(+)-Ledene	36.39 ± 3.22[a]	nd	nd	nd	nd	24.45 ± 1.24[b]
53	β-Caryophyllene	nd	87.62 ± 2.91[b]	nd	nd	nd	105 ± 2.56[a]
54	α-Caryophyllene	nd	nd	18.24 ± 2.11[a]	nd	nd	9.32 ± 0.88[b]
55	Caryophyllene	129 ± 3.22[a]	nd	6.52 ± 0.59[c]	135 ± 3.91[a]	114 ± 2.54[b]	6.08 ± 0.15[c]
56	α-Copaene	nd	nd	nd	nd	99.76 ± 7.22[a]	nd
57	(-)-Aristolene	nd	21.29 ± 6.85[a]	nd	nd	nd	nd
58	(+)-Valencene	129 ± 4.38[c]	705 ± 5.67[a]	69.42 ± 3.17[d]	309 ± 6.45[b]	nd	
60	Germacrene B	nd	21.32 ± 3.12[a]	nd	nd	nd	nd
61	α-Humulene	894 ± 32.18[c]	390 ± 6.49[d]	nd	1011 ± 98.25[b]	1278 ± 27.16[a]	nd
62	β-Humulene	39.09 ± 8.45[a]	nd	nd	nd	nd	nd
63	α-Cubebene	nd	15.35 ± 0.98[a]	nd	3.16 ± 0.21[b]	nd	nd
64	β-Panasinsen	nd	nd	nd	15.45 ± 0.29[a]	nd	nd
65	α-Bergamotene	18.86 ± 0.86[a]	nd	nd	nd	nd	nd
66	Thujopsene	18.46 ± 1.19[a]	nd	nd	nd	nd	nd
67	α-Bulnesene	9.7 ± 7.26[a]	nd	nd	nd	nd	nd
68	α-Farnesene	nd	nd	nd	nd	30.5 ± 5.32[a]	
	Sum	9067 ± 190[d]	9681 ± 145[c]	2338 ± 111[f]	12314 ± 311[a]	7311 ± 143[e]	10047 ± 148[b]
	Relative	36.33%	40.34%	11.16%	45.39%	19.29%	36.02%

(*continued on next page*)

Table 4 (*continued*)

No.	Volatiles	1	2	3	4	5	6
		MG	OR	CT	TG	RB	RR
Terpenic alcohols and aldehydes							
69	β-Linalool	72.89 ± 4.53^{d}	108 ± 3.22^{c}	153 ± 5.26^{b}	168 ± 9.25^{b}	99.1 ± 1.27^{c}	420 ± 11.23^{a}
70	Carveol	96.53 ± 5.12^{a}	24.38 ± 3.27^{c}	nd	nd	nd	48.25 ± 2.34^{b}
71	Trans-2,8-menthadienol	nd	nd	nd	81.46 ± 3.56^{a}	nd	54.17 ± 2.33^{b}
72	L-Isopulegol	nd	nd	nd	nd	nd	42.27 ± 2.12^{a}
73	Cis-Verbenol	nd	nd	nd	nd	nd	33.45 ± 1.65^{a}
74	L-4-terpineol	57.26 ± 2.64^{c}	42.86 ± 2.63^{d}	33.52 ± 1.65^{e}	63.92 ± 4.53^{c}	102 ± 3.27^{b}	297 ± 5.76^{a}
75	α-Terpieol	51.24 ± 3.35^{c}	66.35 ± 6.17^{c}	48.23 ± 2.43^{d}	81.16 ± 3.92^{b}	84.23 ± 2.35^{b}	180 ± 2.45^{a}
76	β-Terpineol	33.62 ± 1.65^{b}	nd	nd	3.56 ± 0.47^{c}	48.26 ± 2.35^{a}	nd
77	Cis-Carveol	102 ± 3.45^{a}	45.32 ± 3.56^{d}	nd	84.71 ± 7.46^{b}	114 ± 3.45^{a}	63.82 ± 3.78^{c}
78	Bulnesol	nd	15.09 ± 1.02^{a}	nd	nd	nd	9.42 ± 4.35^{b}
79	Caryophyllenyl alcohol	15.23 ± 1.14^{a}	nd	nd	15.53 ± 1.11^{a}	12.67 ± 0.62^{b}	12.85 ± 1.03^{b}
80	Geranylgeraniol	nd	nd	15.25 ± 2.36^{a}	nd	nd	9.12 ± 0.58^{b}
81	Elemol	nd	nd	nd	12.33 ± 1.59^{a}	nd	nd
82	tau.-Cadinol	nd	nd	nd	21.45 ± 1.87^{a}	nd	nd
83	α-Phellandren-8-ol	30.55 ± 2.11^{a}	nd	nd	nd	nd	nd
84	Trans-Nerolidol	9.73 ± 0.80^{b}	nd	nd	nd	12.24 ± 1.23^{a}	nd
85	β-Citral	nd	21.94 ± 1.62^{b}	nd	21.25 ± 1.14^{b}	12.25 ± 0.63^{c}	147 ± 7.55^{a}
86	α-Citral	12.53 ± 5.13^{c}	30.66 ± 3.35^{b}	nd	3.91 ± 0.22^{d}	27.35 ± 3.87^{b}	159 ± 4.19^{a}
87	L-perillaldehyde	nd	nd	nd	18.42 ± 1.85^{a}	nd	nd
88	(-)-Citronellol	nd	nd	nd	nd	nd	87.15 ± 2.36^{a}
89	(−)-β-Citronellol	nd	nd	nd	nd	nd	27.47 ± 1.32^{a}
	Sum	482 ± 29.92^{d}	355 ± 24.84^{e}	250 ± 11.7^{f}	576 ± 36.97^{b}	512 ± 19.04^{c}	1571 ± 53.04^{a}
	Relative	1.93%	1.48%	1.19%	2.12%	1.35%	5.63%
Aliphatic alcohols and aldehydes							
90	3-Hexenal	nd	nd	nd	nd	nd	144 ± 16.38^{a}
91	Hexanal	279 ± 31.26^{c}	468 ± 41.55^{b}	1173 ± 47.42^{a}	nd	258 ± 11.66^{c}	nd
92	(E)-2-Hexenal	21.56 ± 5.23^{c}	75.72 ± 5.21^{b}	372 ± 3.18^{a}	27.56 ± 4.17^{c}	60.45 ± 3.52^{b}	24.17 ± 1.55^{c}
93	Heptanal	54.27 ± 2.66^{b}	48.29 ± 2.36^{b}	154 ± 7.68^{a}	9.45 ± 2.12^{e}	33.47 ± 1.76^{c}	15.24 ± 1.35^{d}
94	(E)-2-Hepten-1-al	9.43 ± 0.46^{b}	12.98 ± 1.22^{b}	39.35 ± 0.91^{a}	nd	12.65 ± 0.78^{b}	3.43 ± 0.25^{c}
95	Octanal	225 ± 4.67^{b}	nd	nd	nd	nd	1200 ± 13.36^{a}
96	1-Octanol	nd	nd	120 ± 6.23^{b}	nd	nd	546 ± 20.68^{a}
97	Nonanal	141 ± 4.36^{c}	93.66 ± 6.27^{d}	339 ± 5.87^{a}	60.82 ± 2.65^{e}	99.35 ± 6.41^{d}	189 ± 9.25^{b}
98	(E)-Carveol	117 ± 3.64^{b}	18.29 ± 2.12^{d}	15.23 ± 2.44^{d}	5.91 ± 0.52^{e}	180 ± 3.68^{a}	42.32 ± 1.09^{c}
99	1-Nonanol	24.25 ± 2.07^{a}	12.45 ± 1.66^{b}	nd	nd	nd	24.37 ± 1.19^{a}
100	Decanal	69.52 ± 2.15^{d}	69.43 ± 1.76^{d}	102 ± 2.65^{b}	57.05 ± 2.54^{e}	78.77 ± 3.25^{c}	630 ± 8.03^{a}
101	(-)-Citronellol	nd	nd	nd	nd	nd	87.15 ± 2.36^{a}
102	1-Hexanol	nd	24.87 ± 3.13^{c}	99.47 ± 1.52^{a}	24.24 ± 1.32^{c}	54.66 ± 4.12^{b}	nd
103	1-Octen-3-ol	30.23 ± 2.17^{c}	33.82 ± 3.85^{c}	69.41 ± 4.51^{a}	2.54 ± 2.18^{d}	45.32 ± 3.13^{b}	nd
104	α-hellandren-8-ol	nd	nd	nd	nd	63.24 ± 2.02^{a}	nd
105	2-Methyl-2-butenal	15.35 ± 0.57^{b}	nd	15.25 ± 0.31^{b}	9.25 ± 0.82^{c}	18.16 ± 1.87^{a}	nd
106	trans-2-Decenol	18.23 ± 0.87^{a}	6.32 ± 1.92^{b}	nd	nd	nd	nd
107	5-(Hydroxymethyl)spiro[2.4]heptan-5-ol	nd	nd	57.24 ± 2.35^{a}	nd	nd	nd
	Sum	1039 ± 61.35^{c}	864 ± 71.05^{e}	2583 ± 86.59^{b}	197 ± 16.32^{f}	935 ± 44.42^{d}	2864 ± 75.65^{a}
	Relative	4.16%	3.60%	12.33%	0.73%	2.47%	10.26%
Ketones							
108	Methylheptenone	33.45 ± 1.07^{b}	nd	nd	30.56 ± 2.07^{b}	300 ± 11.2^{a}	30.2 ± 1.64^{b}
109	Nootkatone	30.75 ± 0.94^{d}	171 ± 2.36^{a}	30.38 ± 3.32^{d}	51.53 ± 1.47^{c}	102 ± 3.45^{b}	18.23 ± 1.09^{e}
110	D-Dihydrocarvone	36.44 ± 1.47^{a}	12.56 ± 0.32^{e}	27.25 ± 3.25^{c}	30.72 ± 2.17^{b}	30.53 ± 1.24^{b}	21.44 ± 3.17^{d}
111	1-Octen-3-one	24.45 ± 1.16^{a}	nd	nd	18.56 ± 2.15^{b}	nd	nd
112	6-Camphenone	42.28 ± 2.66^{a}	nd	nd	24.53 ± 1.74^{b}	27.63 ± 6.75^{b}	nd
113	(+)-Carvone	24.15 ± 1.32^{a}	nd	15.27 ± 1.36^{b}	18.25 ± 1.31^{b}	3.23 ± 0.19^{c}	nd
114	Juniper camphor	nd	69.23 ± 2.16^{a}	9.25 ± 0.85^{c}	54.52 ± 1.66^{b}	63.43 ± 2.58^{a}	nd
115	(+)-Camphor	30.22 ± 0.67^{a}	nd	15.87 ± 0.45^{b}	nd	nd	nd
116	Geranyl acetone	nd	nd	27.46 ± 1.53^{a}	nd	nd	nd
	Sum	222 ± 9.29^{c}	253 ± 4.84^{b}	125 ± 10.76^{d}	229 ± 12.57^{c}	527 ± 25.41^{a}	69.87 ± 5.90^{e}
	Relative	0.89%	1.05%	0.60%	0.84%	1.39%	0.25%
Esters							
117	Acetic acid, butylester	12.43 ± 1.25^{d}	51.65 ± 3.12^{b}	84.35 ± 2.42^{a}	12.31 ± 3.41^{d}	21.44 ± 4.46^{c}	9.82 ± 0.63^{d}
118	Butanoic acid, hexylester	21.45 ± 1.24^{a}	9.98 ± 1.54^{c}	15.63 ± 0.65^{b}	21.92 ± 0.93^{a}	21.8 ± 0.85^{a}	12.54 ± 0.82^{b}
119	Heptyl acetate	nd	nd	nd	nd	nd	21.32 ± 0.42^{a}
120	Ethyl octanoate	nd	nd	nd	117 ± 9.86^{b}	153 ± 8.43^{a}	54.67 ± 2.65^{c}
121	Octyl acetate	81.53 ± 1.58^{b}	nd	nd	57.64 ± 4.23^{c}	81.61 ± 3.75^{b}	288 ± 7.82^{a}
122	L-Bornyl acetate	21.56 ± 2.22^{c}	33.17 ± 2.11^{a}	27.23 ± 2.57^{b}	nd	27.32 ± 3.55^{b}	18.56 ± 1.67^{c}
123	Octyl propanoate	nd	nd	nd	nd	nd	15.29 ± 2.17^{a}
124	Nonyl acetate	15.32 ± 0.47^{b}	nd	nd	18.29 ± 0.86^{b}	18.82 ± 1.37^{b}	27.45 ± 0.47^{a}
125	Citronellol acetate	54.56 ± 2.54^{b}	18.37 ± 0.82^{c}	nd	48.54 ± 3.19^{b}	78.29 ± 5.54^{a}	78.26 ± 3.45^{a}
126	Neryl acetate	60.17 ± 1.26^{c}	18.34 ± 1.22^{d}	24.32 ± 1.59^{d}	69.62 ± 2.22^{b}	69.71 ± 2.93^{b}	81.26 ± 2.53^{a}
127	Acetic acid, geraniol ester	54.87 ± 4.33^{c}	nd	nd	nd	105 ± 3.25^{a}	93.43 ± 3.56^{b}
128	Decyl acetate	nd	nd	nd	33.2 ± 1.23^{b}	27.3 ± 3.22^{b}	84.5 ± 5.38^{a}
129	Perilla acetate	12.2 ± 3.1^{b}	nd	nd	36.54 ± 5.15^{a}	30.44 ± 43.26^{a}	33.62 ± 2.48^{a}
130	tert-butyl 2-methylpropanoate***	9.42 ± 0.83^{c}	27.67 ± 0.64^{b}	30.92 ± 0.91^{a}	24.52 ± 0.92^{b}	9.64 ± 0.76^{c}	6.82 ± 0.85^{c}

Table 4 (*continued*)

No.	Volatiles	1	2	3	4	5	6
		MG	OR	CT	TG	RB	RR
131	Farnesyl acetate	6.49 ± 0.43[c]	nd	nd	12.34 ± 0.75[b]	nd	57.19 ± 1.72[a]
132	Neryl acetate	nd	nd	nd	6.28 ± 0.45[a]	nd	nd
133	3-heptan-4-yl 2-methylpropanoate	15.89 ± 1.15[b]	nd	15.73 ± 0.53[b]	18.28 ± 0.63[a]	15.67 ± 3.67[b]	nd
134	cis-Carvyl acetate	36.72 ± 7.16[b]	nd	nd	6.55 ± 0.47[c]	54.23 ± 2.29[a]	nd
135	Methyl octanoate	15.39 ± 0.60[a]	nd	nd	15.89 ± 0.40[a]	nd	nd
136	2-ethylhexyl acetate	18.69 ± 0.63[a]	12.26 ± 0.35[b]	6.72 ± 0.75[c]	nd	18.92 ± 1.25[a]	nd
137	(3Z)-3-Hexenyl formate	nd	nd	303 ± 23.19[a]	nd	nd	nd
	Sum	437 ± 28.79[d]	171 ± 9.80[e]	508 ± 32.61[c]	499 ± 34.70[c]	733 ± 88.58[b]	883 ± 36.62[a]
	Relative	1.75%	0.71%	2.42%	1.84%	1.93%	3.16%
Alkanes							
138	Octadecane	nd	nd	24.99 ± 0.97[a]	nd	nd	nd
139	Tridecane	nd	nd	12.45 ± 1.22[c]	15.76 ± 0.63[b]	18.69 ± 0.66[a]	nd
140	Hexadecane	15.34 ± 0.89[a]	nd	12.92 ± 0.93[b]	nd	15.45 ± 1.22[a]	nd
141	Guaiazulene	nd	6.32 ± 0.67[a]	nd	nd	nd	nd
142	Tetradecane	24.34 ± 0.35[a]	nd	nd	nd	nd	nd
143	Pentadecane	nd	39.29 ± 2.11[a]	nd	nd	nd	nd
144	Eicosane	nd	6.65 ± 0.43[a]	nd	nd	nd	nd
145	10,10-Dimethyl-2,6-dimethylenebicyclo[7.2.0]undecane	129 ± 7.21[b]	nd	nd	156 ± 3.16[a]	96.33 ± 1.22[c]	126 ± 7.46[b]
146	2-ethenyl-1,1-dimethyl-3-methylidenecyclohexane	nd	258 ± 12.58[a]	nd	nd	nd	nd
147	4-ethenyl-4,9,9-trimethyl-6-methylidenebicyclo[5.2.0] nonane	63.46 ± 4.33[a]	nd	nd	nd	63.91 ± 2.12[a]	nd
148	(E)-3-Heptene	120 ± 3.11[a]	nd	nd	132 ± 1.76[a]	nd	nd
149	Cyclooctatetraene	72.81 ± 8.14[a]	nd	nd	nd	36.69 ± 6.15[c]	60.89 ± 3.53[b]
150	1,3,8-p-Menthatriene	204 ± 13.82[a]	nd	nd	nd	99.69 ± 6.81[b]	nd
151	4-Acetyl-1-methylcyclohexene	45.69 ± 2.11[a]	15.83 ± 1.32[c]	21.45 ± 0.82[b]	nd	nd	nd
152	3,9-Epoxy-p-mentha-1,8(10)-diene	nd	nd	nd	24.94 ± 2.14[a]	nd	nd
153	Eudesma-3,7(11)-diene	nd	33.66 ± 6.55[a]	nd	nd	nd	nd
154	1S,2S,5R-1,4,4-Trimethyltricyclo[6.3.1.0(2,5)]dodec-8(9)-ene	nd	nd	nd	63.59 ± 1.65[a]	nd	nd
155	1,7,7-Trimethylnorbornene	nd	nd	93.54 ± 5.11[a]	nd	48.98 ± 4.53[b]	nd
156	3-acetoxy-4-(1-hydroxy-1-methylethyl)-1-methyl-Cyclohexene	33.52 ± 1.49[b]	nd	nd	63.45 ± 3.23[a]	nd	nd
157	4-methylidene-1-propan-2-ylbicyclo[3.1.0]hex-2-ene	3.88 ± 0.22[a]	nd	nd	nd	nd	nd
158	2-ethenyl-1,3,3-trimethylcyclohexene	30.91 ± 1.81[b]	nd	12.46 ± 0.35[c]	nd	39.61 ± 3.19[a]	nd
	Sum	743 ± 35.34[a]	360 ± 23.66[c]	178 ± 9.4[d]	456 ± 12.57[b]	419 ± 25.90[b]	187 ± 10.99[d]
	Relative	2.98%	1.50%	0.85%	1.68%	1.11%	0.67%
Others							
159	Ethylbenzene	33.56 ± 5.17[b]	87.27 ± 3.69[a]	nd	24.25 ± 1.61[c]	39.13 ± 1.65[b]	21.45 ± 1.18[c]
160	trans-Limonene oxide	15.12 ± 1.86[a]	9.29 ± 0.56[b]	nd	nd	15.43 ± 4.93[a]	9.21 ± 1.13[b]
161	2,5-ditert-butylphenol	nd	nd	nd	nd	nd	33.45 ± 1.66[a]
162	1-Isopropyl-4,7-dimethyl-1,2,4a,5,6,8a-hexahydronaphthalene	nd	nd	nd	nd	nd	15.28 ± 0.56[a]
163	Caryophyllene oxide	69.45 ± 4.32[a]	21.71 ± 1.17[c]	nd	60.26 ± 2.74[a]	6.83 ± 3.84[d]	42.56 ± 7.69[b]
164	trans-Linalool oxide	nd	nd	nd	795 ± 5.27[a]	nd	nd
165	Naphthalene	18.26 ± 2.17[a]	12.93 ± 1.54[b]	18.94 ± 0.69[a]	nd	nd	
166	2,4-di-tert-butylphenol	45.28 ± 3.93[d]	129 ± 5.88[b]	150 ± 4.35[a]	54.82 ± 0.85[c]	39.27 ± 2.34[e]	nd
167	Isonox 132	nd	18.32 ± 3.12[b]	21.08 ± 0.87[a]	nd	nd	nd
168	Toluene	nd	18.63 ± 1.86[a]	nd	3.28 ± 0.64[b]	3.29 ± 0.51[b]	nd
169	Propylbenzene	nd	nd	nd	nd	3.48 ± 0.52[a]	nd
170	6-dimethyl-2,3,3a,4,5,7a-hexahydro-1-benzofuran	24.58 ± 2.31[a]	nd	nd	nd	24.56 ± 5.34[a]	nd
	Sum	206 ± 19.76[c]	297 ± 17.82[b]	190 ± 5.91[c]	938 ± 30.50[a]	132 ± 11.23[d]	122 ± 20.51[d]
	Relative	0.83%	1.24%	0.91%	3.46%	0.35%	0.44%
	Total	24956 ± 534[c]	23999 ± 399[d]	20952 ± 096[e]	27127 ± 795[b]	37896 ± 834[a]	27892 ± 789[b]

[a] nd, not detectable.
[b] Data are expressed as means ± standard deviation of triplicate samples.
[c] Different lowercase letters between rows represent significant differences between cultivars ($p < 0.05$).

hydrocarbons predominated in Mangshanyegan fruits, in particular D-limonene and β-myrcene, which accounted for 85.75% and 10.89% of the total volatiles, respectively. However, the combined results of GC-O, quantitative analysis, odor activity values (OAVs), and omission tests revealed that β-myrcene and (*Z*)- and (*E*)-linalool oxides were the characteristic aroma compounds of Mangshanyegan, contributing to the balsamic and floral notes of its aroma. Generally, like in other citrus species/cultivars, D-limonene was also important to the background aroma of the grapefruit. Odor activity values (OAVs) were calculated using the concentrations of the aroma active compounds and their respective odor activity values threshold values in water obtained from the literature. An OAV greater than one indicates that the volatile chemical has an effect on the aroma quality of fruit. In the present study, high concentrations of caryophyllene, β-pinene, and α-humulene, with odor thresholds of 1.54 μg/g, 4.16 μg/g and 0.12 μg/g (Boonbumrung et al., 2001; Liu et al., 2012;) presented more than 1347, 69 and 3250 OAVs in grapefruit, respectively, and therefore can have a significant effect on the perceived aroma of grapefruit. Other rich and widely detected grapefruit volatiles, such as α-pinene, α-terpinene, humulen-(v1), β-linalool, α-terpieol, nonanal, nootkatone, neryl acetate and tert-butyl 2-methylpropanoate, also presented high OAVs, wih values of 20, 79, 306, 12167, 137, 17, 106, 9, 406, respectively (Buttery, Seifert, Guadagni, Black, & Ling, 1968; Dharmawan, Kasapis,

Table 5
Carotenoid content (μg/g FW) in pulp of the grapefruit.[a,b,c]

No.	Cultivars	Lutein	Zeaxanthin	β-Cryptoxanthin	Lycopene	α-carotene	β-carotene	Total carotenoid
1	MG	1.09 ± 0.04[a]	nd	nd	0.25 ± 0.03[c]	nd	0.35 ± 0.03[d]	1.68 ± 0.10[e]
2	OR	1.05 ± 0.01[a]	nd	nd	nd	nd	0.24 ± 0.014[d]	1.29 ± 0.02[e]
3	CT	1.11 ± 0.03[a]	0.93 ± 0.04[a]	1.28 ± 0.03[a]	nd	nd	2.58 ± 0.163[c]	5.89 ± 0.25[c]
4	TG	1.06 ± 0.01[a]	0.87 ± 0.05[a]	1.13 ± 0.15[b]	nd	nd	3.77 ± 0.09[c]	4.83 ± 0.09[d]
5	RB	1.05 ± 0.01[a]	nd	nd	4.35 ± 0.26[b]	nd	8.11 ± 0.06[b]	13.51 ± 0.32[b]
6	RR	1.07 ± 0.01[a]	0.46 ± 0.03[b]	1.00 ± 0.01[c]	5.66 ± 0.20[a]	1.34 ± 0.01[a]	64.66 ± 0.21[a]	74.19 ± 0.45[a]

[a] nd represents not detectable.
[b] Data are expressed as means ± standard deviation of triplicate samples.
[c] Different lowercase letters between columns represent significant differences between cultivars ($p < 0.05$).

Sriramula, Lear, & Curran, 2009; Guadagni, Buttery, & Harris, 1966; López et al., 2007; Takeoka, Flath, Mon, Teranishi, & Guentert, 1990). Taken together, this data suggests that caryophyllene, α-humulene, humulen-(v1), β-linalool and tert-butyl 2-methylpropanoate may play very important roles in the perceived aroma of grapefruit.

3.5. Carotenoid composition and content

Six carotenoids, including lutein, zeaxanthin, β-cryptoxanthin, lycopene, α-carotene and β-carotene, were identified in the tested grapefruit cultivars (Fig. S4 and Table 5). β-carotene was the dominant carotenoid in these pulps and represented 18.40–87.15% of the total carotenoids. β-carotene ranged from 0.24 to 64.66 μg/g FW; Rio Red grapefruits had the highest level of β-carotene, and its content was approximately 7.97–186 times that of other cultivars. Lutein made up the second largest amount of carotenoids in the tested grapefruit, and its content made up 1.44–81.60% of total carotenoids, ranging from 1.05 to 1.11 μg/g FW. No differences in lutein were found in these grapefruit. Zeaxanthin and β-cryptoxanthin were only detected in Cocktail and Rio Red grapefruits. Lycopene was detected in Red Blush and Rio Red grapefruits and α-Carotene was only detected in Rio Red.

Various citrus fruits differ in their carotenoid profiles. Mandarin, orange, and clementine are usually rich in β-cryptoxanthin, violaxanthin, lutein, and zeaxanthin. Pummelo accumulates primarily phytoene, phytofluene, ζ-carotene, and β-carotene, while citron, lemon, and lime contain low levels of carotenoids (Kato, 2012; Yuan, Zhang, Nageswaran, & Li, 2015). Violaxanthin is the major carotenoid in oranges, whereas β-cryptoxanthin is the major carotenoid in mandarins (Yuan et al., 2015). The present results showed that β-carotene or/and lycopene were the major carotenoids in grapefruit. However, Alquezar et al. (2013) found that of the nine major carotenoids identified, phytoene was the dominant carotenoid in the white Marsh and red Star Ruby grapefruits and the phytoene content of the white Marsh was five times that of the red Star Ruby. In the present study, significantly higher β-carotene content was observed in the white cultivar (Red Blush and Rio Red) than in the red cultivars (Marsh and Oroblanco). The most important finding is that only rich lycopene was detected in the red pulp grapefruits, Red Blush and Rio Red. It is very interesting that of the orange cultivars, Cocktail and Thompson grapefruits presented significantly higher zaeaxanthin and β-cryptoxanthin than yellow and red grapefruit. Therefore, even different carotenoids may affect the yellow, orange and red colors of grapefruit, differences in grapefruit color between cultivars mainly depends on the ratio of zaeaxanthin, β-cryptoxanthin and lycopene.

4. Conclusion

This study profiles the chemicals that contribute to the taste, aroma and color of grapefruit. Sucrose is the dominant sugar in grapefruit, occupying 40.08–59.68% of the total sugars, where the ratio of fructose to glucose was almost 1:1. Citric acid was the major organic acid, making up 39.1–63.55% of the total organic acid, followed by quininic acid. The ratios of individual sugars and organic acids play an important role in the taste determination of grapefruit. Grapefruits contain rich aromatic volatiles and a total of 170 VOCs were identified from these fruits. Monoterpenes and sesquiterpenes (79.58–91.41) were the major volatiles, in particular D-limonene and caryophyllene, followed by β-pinene and α-humulene. According to their OAVs, caryophyllene, α-humulene, humulen-(v1), β-linalool and tert-butyl 2-methylpropanoate are the characteristic aroma compounds of grapefruit, and might have a significant effect on the perceived aroma of grapefruit. Though β-carotene is the primary carotenoid in grapefruit, followed by lutein and β-cryptoxanthin, the color differences between cultivars are mainly determined by the ratio of zaeaxanthin, β-cryptoxanthin and lycopene. Our findings provide valuable information for grapefruit quality breeding and consumer guidelines.

Acknowledgement

We thank Drs. Rachel Maczis Shahan and Julie Caruana (Department of Cell Biology and Molecular Genetics, University of Maryland) for comments on an earlier version of this manuscript. This work was supported by the National Natural Science Foundation of China (No. 31471855).

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.foodchem.2016.03.007.

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