Food Chemistry 139 (2013) 171–183

Contents lists available at SciVerse ScienceDirect

Food Chemistry

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

An integrated approach for flavour quality evaluation in muskmelon (*Cucumis melo* L. *reticulatus* group) during ripening

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ARTICLE INFO

Article history:
Received 6 July 2012
Received in revised form 20 December 2012
Accepted 28 December 2012
Available online 7 January 2013

Keywords:
Melon maturity
Ripening
Aroma
Flavour
Volatiles
Sensory descriptive analysis
zNose
Headspace sorptive extraction

ABSTRACT

Numerous and diverse physiological changes occur during fruit ripening and maturity at harvest is one of the key factors influencing the flavour quality of fruits. The effect of ripening on chemical composition, physical parameters and sensory perception of three muskmelon (*Cucumis melo* L. *reticulatus* group) cultivars was evaluated. Significant correlations emerging from this extensive data set are discussed in the context of identifying potential targets for melon sensory quality improvement. A portable ultra-fast gas-chromatograph coupled with a surface acoustic wave sensor (UFGC–SAW) was also used to monitor aroma volatile concentrations during fruit ripening and evaluated for its ability to predict the sensory perception of melon flavour. UFGC–SAW analysis allowed the discrimination of melon maturity stage based on six measured peaks, whose abundance was positively correlated to maturity-specific sensory attributes. Our findings suggest that this technology shows promise for future applications in rapid flavour quality evaluation.

1. Introduction

Netted muskmelon (*Cucumis melo* L., *reticulatus* group), also commonly called cantaloupe, is an orange-fleshed, sweet and aromatic melon that is highly popular in the United States, representing a large share of the produce market. In 2010, the estimated net domestic use of muskmelon totaled over 2.6 billion pounds, and muskmelon ranked fourth in U.S. annual per capita consumption of fresh fruit after bananas, watermelons and apples (USDA-ERS, 2010). Consumer surveys assessing "overall preference" for several muskmelon cultivars highlighted that flavour, sweetness and texture were important factors in determining consumer liking of melons (Lester, 2006). While these attributes are dictated by the specific cultivar, or genetic makeup, of the muskmelon, maturity at harvest has also been shown to have a large impact on the sugar content (related to sweetness), volatile content (related to flavour and aroma) and texture of melon fruit (Beaulieu & Grimm, 2001; Beaulieu, Ingram, Lea, & Bett-Garber, 2004; Beaulieu & Lancaster, 2007; Pratt, 1971).

Harvesting firmer and early mature fruits is a commercial practice commonly adopted in order to maximise post-harvest life during handling, shipping and storage of climacteric fruits (Kader, 2008). However, this practice is detrimental for flavour quality because it does not allow full development of the fruit aroma profile (Beaulieu, 2006; Beaulieu et al., 2004; Wyllie, Leach, & Wang, 1996).

Typically, muskmelon fruit maturity in the field is determined by the extent of the development of an abscission layer (also called "slip" in the trade) between the vine and the fruit. In California, melons are generally harvested at ¾- to full-slip stage for local market distribution. However, genetic, environmental and agronomic factors often complicate maturity assessment by influencing fruit physiology and the development of this abscission zone, resulting in variable postharvest fruit quality. In addition, melons destined for long distance transport are typically harvested earlier, sometimes even before the clear development of an abscission zone.

Due to the interactions of many parameters (e.g., sugar content, aroma profile, colour, texture) in determining fruit sensory characteristics, measuring a single composition parameter such as sugar content is seldom sufficient to reflect an objective assessment of overall fruit flavour quality. From an applicative perspective, a comprehensive assessment of flavour quality is often unfeasible due to the requirement of expensive analytical instrumentation, highly trained personnel and time- and labour-consuming proce-

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http://dx.doi.org/10.1016/j.foodchem.2012.12.042

dures. While the availability of rapid methods for the detection of external, visual quality has allowed the distribution of aesthetically superior fruit, the lack of rapid methods for flavour quality control may hinder the delivery of more flavourful fruit to consumers.

In our study we first evaluated the effect of ripening on sensory perception, chemical composition and physical measurements of melon fruit. Changes in chemical composition and physical properties during the ripening process were then correlated to sensory attributes in an attempt to predict flavour perception by chemical composition analysis. Finally, an ultra-fast gas-chromatograph was evaluated for its ability to monitor changes in melon volatile concentrations during ripening and to predict the sensory perception of flavour.

2. Materials and methods

2.1. Plant material and sample preparation

Muskmelons (*C. melo* L., *reticulatus* group) cv. Navigator, Mas Rico and Thunderbird, were grown in Davis, CA (38.55 N, 121.74 W), and provided by HM. Clause Seed Company (Modesto, CA, USA). Each of the three cultivars was planted on three different dates with approximately 2-week intervals during the spring of 2010 and grown on raised beds using standard commercial cultivation practices with drip irrigation. Each cultivar plot corresponding to one "planting date" was used to supply one of three iterations of fruit materials for sensory testing and physiochemical analysis.

Fruit were harvested at five different maturity stages, ranging from early mature to fully ripe, between July and September 2010. The maturity at harvest was assessed in the field by examining the presence or absence of the abscission zone formed around the peduncle, commonly called "slip". Early mature melons corresponded to fruits that had reached full size, but did not show a visible abscission zone around the peduncle (thereafter named "pre-slip" fruit). Fruits were classified as "full slip" when a fully developed abscission zone (as evidenced by the development of a crack around the peduncle) was visible at harvest time. Under our growing conditions, the abscission zone developed very quickly (within less than a day), making the standard ¼-, ½- and ¾-slip maturity assessment impractical. Therefore, "full-slip" melons were grouped in three classes, "slip A" (Sa), "slip B" (Sb) and "slip C" (Sc), based on increasing force needed to detach the fruit from the plant (force applied to detach the fruit: Sa > Sb > Sc). Six melons were chosen for each maturity stage based on the absence of external and internal defects, and size homogeneity. Fruits were rinsed with tap water in order to remove dirt and dust, cut longitudinally into four wedges, and a further classification of the "pre-slip" fruit was performed visually based on the degree of colour lightness of the flesh: "pre-slip light orange" (PL) and "pre-slip dark orange" (PD).

Seeds and cavity tissue were removed, and two opposite wedges per fruit were selected for physiochemical analysis and the other two opposite wedges were used for sensory analysis on the same day.

2.2. Environmental conditions

Average and maximum daily air temperature and total solar radiation data were retrieved from the web-site of California Information Management System (CIMIS; http://www.cimis.water.ca.gov) weather station, located 5 miles North-West (38.54 N, 121.78 W) of HM. Clause Seed Company fields. Total solar radiation was converted into photosynthetic active radiation (PAR) in 400–700 nm wavebands, according to Thimijan and Heins (1983), and expressed in $\mu mol\ m^{-2}\ s^{-1}$.

2.3. Sensory analysis

2.3.1. Panellist recruiting and training

Students and staff with various backgrounds in sensory testing were recruited from the UC Davis campus to participate in sensory panel training.

Nine panellists underwent six one-hour training sessions, over a period of 3 weeks. During the initial training sessions, panellists generated and agreed upon a list of sensory attributes presented in Table 1. The attributes were rated on a 10 cm unstructured scale, anchored at the ends with "none" and "strong", except for colour intensity, which ranged from "pale orange" to "strong orange", and unevenness of colour, which ranged from "even" to "uneven".

2.3.2. Sensory descriptive analysis

Melon balls (1.3 cm in diameter) carved from melons of the same cultivar and maturity stage were gently mixed in a mixing bowl, and a set of five balls was put into five 162-mL plastic cups with lids labelled with random three digit codes. The samples were served in complete randomised order, as established by the software program CSA, Compusense Five (Version 5.0, Compusense, Guelph, Ontario, Canada, 2008), in individual booths and under normal fluorescent white light, at ambient room temperature (20 °C). Water and unsalted crackers were used as rinsing agents between samples.

A modified quantitative descriptive analysis (QDA) (Stone, Sidel, Oliver, Woolsey, & Singleton, 1974) was used to evaluate the samples. Five samples per cultivar, representing all five maturity stages, were evaluated during each session and each cultivar evaluation was repeated three times (corresponding to the three different planting dates). Panellists evaluated the samples one by one, on a computer screen, according to the order presented in the software program. Appearance of the fruit was evaluated first based on the overall average colour intensity and unevenness of colour for the five melon balls. Panellists then smelled the sample, and evaluated the aroma attributes. Two sample balls were used for evaluation of the texture attributes. Panellists swallowed the samples after completing the evaluation. Flavours, tastes and flavour lasting sensation were evaluated on the remaining three sample balls.

2.4. Physical and chemical analysis

2.4.1. Physical measurements

Colour measurement was performed using a Minolta Colorimeter (CR-300, Minolta, Ramsey, NJ, USA). L^*, a^*, b^* values were recorded on six replicates per sample, from the side of $2 \times 2 \times 2$ cm flesh cubes obtained from the equatorial region of the fruit. Hue angle, h, was calculated as $h = \arctan(a^*/b^*)$.

Puncture and compression assessments were performed to evaluate fruit flesh firmness, using a TA.XT2 Texture Analyzer (Texture Technologies, Scarsdale, NY, USA). Puncture testing was performed with a 100 g force load on the side of a $2 \times 2 \times 2$ cm flesh cube obtained from the equatorial region of the fruit. A 5 mm diameter flat-head stainless steel cylindrical probe travelled at a rate of 1 mm s^{-1} for a total of 6 mm. The area under the curve from 0 to 6 mm was used as the puncture measurement. Compression testing using a 38 mm flat compression probe was performed with a 100 g force load on the side of a $2 \times 2 \times 2$ cm cube. Pretest speed was 10 mm s^{-1} with a test speed of 0.5 mm s^{-1}, followed by a post-test speed of 10 mm s^{-1}. The compression measurement determined from the graph was total force area ($N * s$). Six melon cubes were analysed for puncture and compression for each cultivar, maturity level, and planting date.

Table 1
Sensory attributes and reference standards.

Attribute	Attribute descriptions	References
Appearance		
Colour intensity	The intensity of the most dominant colour of the sample (all five melon balls included), ranging from very pale orange to strong orange	None
Unevenness of colour	The degree of evenness of the colour, ranging from even (no white spots in flesh), to uneven (many white spots) in the sample	None
Aroma		
Overall aroma	The intensity of aroma of any type, ranging from weak overall aroma, to strong aroma present in the sample	None
Fruity aroma	The intensity of fruity aroma like mixed fruit juice, a fresh fruity aroma, ranging from none to strong fruity aroma	Dole mixed fruit juice (orange, peach, mango juice) Scale point: 10
Marshmallow aroma	The intensity of sweet aroma, includes the sweet sensation of sugar, marshmallows, and bubblegum, ranging from none to strong sweet aroma.	Marshmallows, small size Scale point: 10
Cucumber aroma	The intensity of green, fresh aroma, like cucumber, squash, ranging from none to strong, green aroma	English cucumber, peeled and cut into 1 cm slices Scale point 10
Musky aroma	The intensity of the spicy aroma of cedar wood, like cedar mothballs, but also the musky smell of animal, ranging from none to a strong musky aroma	A piece of cedar wood Scale point: 10
Buttery aroma	The intensity of the aroma of butter, ranging from none to a strong buttery aroma	Real butter, ca. 1 cm^3 Scale point: 10
Texture		
Juiciness	The amount of juice released in the sample when biting into it with the front teeth, ranging from no juice released, to lots of juice released	Ripe peach Scale point: 10
Firmness	The force required to compress the sample between the back teeth, ranging from soft to firm	Banana, yellow, ripe Scale point: 0–1 English cucumber with peel Scale point: 10
Crunchiness	The amount of sound generated when chewing the back teeth, ranging from no sound, like a banana, to a lasting crunchy sound	Granny Smith apple wedge Scale point: 10
Fibrousness	The presence of fibres in the flesh, ranging from no fibres to many fibres in the sample	No reference
Taste and flavour		
Sweet taste	The intensity of sweet taste like sugars, but also like candy (marshmallows or bubblegum), ranging from none to a strong sweet taste	Marshmallow, small size Scale point: 10
Sour taste	The intensity of fresh sour taste like Granny Smith apples, ranging from none to a strong sour taste	Granny smith apple Scale point: 10
Fruity flavour	The intensity of the flavour of fruit juice of mixed fruits, ranging from none to a strong fruity flavour	Dole mixed juice (orange, peach, mango): Scale point: 10
Bitter taste	The intensity of bitter taste, like caffeine, but also like the peel of cucumber, or an unripe fruit, ranging from none to a strong bitter taste	English cucumber peel Scale point: 10
After flavour	How long the flavour lasts in the mouth, ranging from a few seconds to longer than 20 s	None

2.4.2. Chemical measurements

Soluble solids content (SSC) was determined by refractometry (Reichert AR6 Series, Depew, NY, USA) using 200 μL of juice obtained from squeezing melon balls (using a garlic press) sampled from blossom end, stem end and equatorial regions of the melons. Titratable acidity (TA) was determined by diluting 4 g of juice (obtained as above) in 20 mL deionised water and titrating to pH 8.2 using an automatic titrator (Radiometer TitraLab Tim850, Radiometer Analytical SAS, Lyon, France). TA is reported as citric acid equivalents. Each measurement was performed in triplicate for each sample. For ethylene and carbon dioxide analysis, 25 g of melon pieces (obtained randomly along melon wedges using an 11 mm i.d. cork borer) were enclosed in air-tight glass vessels (250 mL volume). After equilibration for 15 min, a 10-mL headspace sample was analysed by gas chromatography coupled with flame ionisation detector using a Varian 3800 (Walnut Creek, CA, USA) equipped with a 6 m × 3 mm alumina column held at 50 °C. For carbon dioxide analysis, 10-mL headspace sample was injected into a rapid gas analyzer (VIA510; Horiba, Fukuoka, Japan). Ethylene and CO_2 concentrations were calculated by comparing the sample response to authentic standard curves.

Juice samples obtained as described for SSC and TA measurements were used for sugar and acid analysis. Fructose, glucose, sucrose, citric, malic and glutamic acid concentrations were evaluated by enzymatic assays as described in Vermeir, Nicolai, Jans, Maes, and Lammertyn (2007) using enzyme reagent kits (R-Biopharm, Marshall, MI, USA) according to the manufacturer's instructions. Standard solutions were used to verify the accuracy of the method.

2.4.3. Volatile compound analysis

2.4.3.1. Sample preparation. For a detailed sample preparation, see Vallone, Lloyd, Ebeler, and Zakharov (2012). Briefly, after removing the seeds and seed cavity tissue, melon balls (2.5 cm in diameter) from six fruits were pooled and mixed, and then 200 g of sample were homogenised with 200 mL of saturated $CaCl_2$ solution and 50 μL of a 100 mM solution of 2-methylbutyl isovalerate in methanol, added as an internal standard. Then, 5-mL aliquots of sample without foam were pipetted into 20-mL glass amber vials. The vials, sealed with a steel screw cap fitted with a Teflon/silicon septum, were flash frozen in liquid nitrogen, and stored at −80 °C until analysis. For volatile analysis, samples were thawed for one hour at room temperature immediately prior to analysis.

2.4.3.2. Headspace sorptive extraction and gas chromatography mass spectrometry. Headspace sorptive extraction (HSSE) and gas chromatography mass spectrometry (GC–MS) was performed using an Agilent (Santa Clara, CA, USA) gas chromatograph (7890A) and mass spectrometer detector (5975C), equipped with a thermal desorption unit (TDU) and cryo-cooled injection system (CIS) (Gerstel Inc., Mülheim an der Ruhr, Germany). The headspace sampling was performed with a 10-mm magnetic stir bar (also called

Twister™) coated with 24 μL of polydimethylsiloxane (Gerstel Inc., Germany). The bar was suspended above the sample in the 20-mL amber vial and exposed to the headspace for 30 min at room temperature, while the sample was stirred at 550 rpm. For thermal desorption, the following parameters were used: initial TDU temperature of 30 °C, heated to 40 °C at 720 °C min^{-1}, then to 250 °C at 60 °C min^{-1} and held for 5 min; solvent venting as desorption mode and vent time of 0.01 min. Simultaneously, the analytes were transferred into the PTV-injector where they were cryogenically focused at −80 °C, using liquid nitrogen. The injection mode was set as solvent vent; vent flow of 50 mL min^{-1} and vent pressure of 0 psi until min 0; purge flow to split vent of 5 mL min^{-1} at min 1; gas saver of 20 mL min^{-1} after 2 min. The initial injection temperature was programmed at −80 °C, then heated to 280 °C at 12 °C s^{-1} and held for 5 min. The GC was equipped with a DB-5MS capillary column (30 m × 0.25 mm; film thickness 0.25 μm). The injector temperature was 220 °C and helium carrier gas flow rate was 1.2 mL min^{-1}. MS parameters were as follows: the transferline was set to 230 °C; the source was 230 °C; the quadrupole temperature was 150 °C; and the scan range was from 30 to 300 *m/z* at a rate of 3 scans s^{-1}. Spectral deconvolution was performed with AMDIS software version 2.69 (Stein, 1999) and spectral alignment was performed with Mass Profiler Professional (version B2.01; Agilent, Santa Clara, CA, USA).

Volatile compound identity was confirmed by matching retention time and mass spectra of authentic standards whenever possible. The percent relative response ratio of each volatile compound was calculated using the internal standard peak area.

All samples were analysed in triplicate.

2.4.3.3. Ultra-fast chromatography and surface acoustic wave sensor analysis. An ultra-fast gas chromatograph (GC) coupled with a surface acoustic wave (SAW) sensor called zNose™ (model 4500; Electronic Sensor Technology-EST; Newbury Park, CA, USA), was used as a rapid method for volatile compounds analysis. The UFGC–SAW analysis was carried out in two steps, sampling and injection. A detailed description of the sampling and injection parameters is found in Vallone et al. (2012).

2.5. Statistical analysis

The sensory data was captured by the Compusense software program, which converted each panellist rating to a value between 1.00 and 9.00 (with 1.00 anchored at the low intensity end and 9.00 anchored at the high intensity end of the scale). A four-way analysis of variance (generalised Linear Model) was run with a mixed model (9 judges × 5 ripening stages × 3 cultivars × 3 planting dates). The judges were evaluated as a random effect. The overall panel performance, judge repeatability and discrimination ability were evaluated on a randomly selected set of replicates using the software PanelCheck version 1.4.0 (Tomic et al., 2010).

A three-way analysis of variance (ANOVA) was performed to analyse the effects of maturity at harvest, cultivar and planting date on sensory attributes, physical and chemical measurements. Tukey's test was performed for all pair-wise comparisons to evaluate statistically significant differences among the means. A principal component analysis (PCA) was performed to reduce the dimensionality of the data, and to visualise relations among all the variables (sensory and physiochemical). First, Pearson's correlation coefficients were calculated for each pair-wise combination, using log transformed and mean centered data. Then the new variables (principal components) were obtained from the eigenvectors of the estimated correlation matrix of the original variables (Borgognone, Bussi, & Hough, 2001). Since the variables measured had different units, the correlation matrix was selected to standardise the original variables and obtain a variance equal to 1. The Scree test was used to select the number of principal components to retain. Regression analysis was performed on selected variables for which the assumption of dependence was satisfied, and the coefficient of determination and significance of the slope were calculated. The analysis of variance and Tukey's test were performed using the statistical program SAS (version 9.1.3; SAS Institute Inc., Cary, NC, USA), while JMP (version 10.0.0; SAS Institute Inc., Cary, NC, USA) was used for principal component and regression analysis. Pearson's correlation analysis was performed using the package Hmisc (Harrell & with contributions from many other users, 2012) in the statistical programming language R (R 2.14.0-Development Core Team 2008, Vienna, Austria).

3. Results and discussion

3.1. Physiochemical and sensory evaluation of three melon cultivars at different maturity stages

Maturity at harvest is one of the key factors influencing melon quality, due to the numerous physiological changes that take place during the ripening process. As expected with field-grown fruit, significant differences were observed in some sensory and physiochemical parameters, not only as a function of maturity and cultivar, but also as influenced by environmental factors (i.e., "planting date") (Table 2, Table S3). Changes in natural environment that occurred between the three growing periods may have influenced the composition of the fruits at harvest time. An increase in maximum air temperature as well as a decrease in photosynthetic active radiation (PAR) were observed between the first and last planting date (data not shown). In some muskmelon varieties, a seasonal variation in colour, SSC and sugar content has been observed (Beaulieu, 2005; Beaulieu, Lea, Eggleston, & Peralta-Inga, 2003).

3.1.1. Sensory evaluation

Judge performance quality, based on the evaluation reproducibility on a replicate set of samples, was evaluated using PanelCheck (Tomic et al., 2010). This analysis showed that the panel used the attributes in a similar and consistent way (data not shown).

Significant differences in most sensory attributes were found across maturity stages and among the three cultivars (Table 2). Significant planting date and interactions effects were also observed for various sensory attributes.

When scores for each sensory attribute were averaged across all maturity stages and planting dates, the cultivar Navigator was generally perceived to have the highest "intensity of colour", "overall aroma", "sweet taste", and "fruity flavour", and a lower intensity of "cucumber aroma" and "bitter taste" (Table S1). Mas Rico generally had the lowest perceived "intensity of colour" and "fruity aroma", but was scored high for "cucumber aroma", "firmness" and "crunchiness". The Thunderbird cultivar was the most bitter and had the lowest "sweet taste" and "fruity flavour" intensities. Previous studies comparing sensory attributes in different melon cultivars also found significant differences in taste, aroma and texture perception (Senesi, Di Cesare, Prinzivalli, & Lo Scalzo, 2005; Senesi, Lo Scalzo, Prinzivalli, & Testoni, 2002).

The different cultivars tended to follow similar changes in sensory perception during maturation. The sensory scores for "colour intensity", "overall aroma", "fruity aroma", "marshmallow aroma", "musky aroma", "juiciness", "sweet taste", "fruity flavour" and "after flavour" increased with increasing maturity, while sensory scores for "cucumber aroma", "firmness", "crunchiness", "fibrousness" and "sour taste" attributes generally decreased as maturity increased. The largest increase in perceived intensity was found between "pre-slip light" and "dark" stages for "colour intensity",

Table 2
Analysis of variance: main effects and significance levels for sensory attributes[a]

Attribute	Planting date	Cultivar	Stage	Cultivar × planting date	Maturity × planting date	Maturity × cultivar
Colour intensity	0.1163	**0.0474**	**<.0001**	**0.0182**	0.147	0.3856
Unevenness of colour	0.8228	0.0581	0.409	0.6984	0.9427	0.7645
Overall aroma intensity	0.488	**0.006**	**<.0001**	0.6256	0.1083	0.4969
Fruity aroma	0.6474	**0.0149**	**<.0001**	0.6671	0.731	0.9127
Marshmallow aroma	0.1388	**0.0004**	**<.0001**	0.3935	**0.0126**	0.2461
Cucumber aroma	0.3273	**0.0149**	**<.0001**	0.2738	0.0502	0.9612
Musky aroma	**0.0076**[b]	**<.0001**	**<.0001**	0.5352	0.0565	**0.0052**
Buttery aroma	0.3558	0.6761	**<.0001**	0.1144	0.2091	0.5615
Juiciness	0.7213	**0.0016**	**<.0001**	**0.0121**	0.1354	0.3717
Firmness	0.2941	**0.0014**	**<.0001**	0.0602	0.223	**0.0152**
Crunchiness	0.1526	**0.0049**	**<.0001**	**0.0233**	0.1189	**0.0126**
Fibrousness	0.1029	**0.0079**	**0.0016**	0.3154	0.819	0.474
Sweet taste	0.7308	**0.0203**	**<.0001**	0.4296	0.3061	0.1542
Sour taste	0.2178	0.4213	**0.0243**	0.1667	0.5661	**0.0115**
Fruity taste	0.6636	**0.0008**	**<.0001**	**0.069**	0.3313	0.0584
Bitter taste	**<.0001**	**<.0001**	**0.037**	**0.0163**	0.1093	**0.0145**
After flavour	0.2427	0.2993	**<.0001**	0.1691	0.8615	0.9947

[a] Data calculated for three melon cultivars, five maturity stages and three planting dates.
[b] Bold *p*-values indicate statistical significance ($p < 0.05$).

"sweet taste" and "fruity flavour" attributes. Large changes in the texture attributes, "firmness" and "crunchiness", were observed between "pre-slip dark", "slip A" and "slip B" stages. A larger increase in "sweet taste" and "fruity flavour" attributes was perceived in Mas Rico from the "slip A" to "slip B" stages, compared to the other two varieties. "Musky aroma" perception increased steadily and to a greater extent in Navigator throughout ripening.

Our results are in general agreement with similar studies evaluating sensory attributes of melon at different ripening stages, where the largest differences in firmness, taste and flavour attributes were perceived between the "unripe" and "ripe" stages (which may correspond to "pre-slip" and "slip A/B" stages in our study), and little difference in these attributes was noted between "ripe" and "overripe" stages (similar to "slip B" and "slip C" in our study) (Senesi et al., 2005). Similarly, Beaulieu et al. (2004) observed significant changes in sensory attributes such as "sweet aromatic", "sweet taste", "surface wetness" and "hardness" of muskmelons harvested at stage "1/4 slip" or "1/2 slip", but not in later maturity stages ("3/4 slip" and "full slip"). These observations highlight the importance of maturity assessment of melon to determine the optimal fruit harvest date in the field, given the large variation in sensory scores between "pre-slip" and "slip" stages.

3.1.2. Physical and chemical measurements

The physical and chemical parameters considered in our study were chosen to reflect taste (soluble solids content, titratable acidity, individual sugars and acids), aroma (volatiles), texture (compression and puncture), colour (L^*, a^*, b^*) and physiological stage (ethylene and CO_2) of the fruits sampled. The concentration of sugars (sucrose, glucose, and fructose) was significantly affected by maturity stage and/or cultivar (Table 3). Sucrose levels gradually increased between the "pre-slip light" and "slip C" stages in all cultivars (from an average of 17.8 g L^{-1} to an average of 30.3 g L^{-1}, respectively). The opposite trend was observed for glucose concentration, which decreased from an average (across cultivars) of 25.9 g L^{-1} at the "pre-slip light" stage to an average of 19.4 g L^{-1} at the "slip C" stage. Fructose levels were generally lower in Mas Rico than in the other two cultivars, but did not vary significantly throughout the ripening process for any of the cultivars. Consistent with our results, an increase in sucrose and a decrease in glucose content during ripening have previously been observed in other muskmelons cultivars (Chrost & Schmitz, 1997; Wang, Wyllie, & Leach, 1996). However, in these studies fructose content also decreased as the fruits ripened, suggesting a contribution from the two monosaccharides (fructose and glucose) to support sucrose accumulation. A recent investigation into sugar metabolism in immature and ripe muskmelon cv. Dulce highlighted the complexity of this metabolic network, and suggested that catabolism of the photosynthates raffinose oligosaccharides (raffinose and stachyose) contributed to the sustained accumulation of sucrose in ripe fruits (Dai et al., 2011). Cultivar-specific regulation of soluble carbohydrate metabolism with regards to fructose utilisation for sucrose biosynthesis may explain the differences in fructose and glucose accumulation patterns in the current and previous studies. Interestingly, soluble solids content (SSC), traditionally used as a reflection of sugar content in fruit and to assess maturity in melon, was variable between planting dates and did not change significantly during ripening between the "pre-slip light" and "slip C" stages in any of the three cultivars used in our study. Consistent with our results, a wide variation in SSC (from 6.5% to 12.5%) was observed by Yamaguchi, Hughes, Yabumoto, and Jennings (1977) in melons from a single harvest, and maturity at harvest did not affect SSC. Conversely, more recent reports noted an increase in SSC during ripening in other muskmelon cultivars (Beaulieu & Lea, 2007; Senesi et al., 2005). Differences in cultivars, growing conditions and fruit maturity assessment methods between studies, and variations in soluble solids content within the fruit (Mizuno, Kato, Harada, Miyajima, & Suzuki, 1971) may account for the discrepancies observed.

Organic acid (citrate and malate) content and titratable acidity of the fruit varied between planting dates and cultivars such that no trend could be clearly defined as a function of ripening (Tables 3 and S2). While very few studies have quantified organic acid concentrations in muskmelon during ripening, seasonal and genotypic effects on organic acid levels in ripe fruit have previously been reported (Beaulieu et al., 2003). The levels of glutamate, on the other hand, consistently increased in all three cultivars between the "pre-slip light" (0.3 g L^{-1}) and "pre-slip dark" (0.9 g L^{-1}) stages, and reached a plateau thereafter. Glutamate levels were significantly (1.5- to 2.3-fold) higher in Navigator than in Mas Rico and Thunderbird at the "pre-slip dark" and more mature stages.

In muskmelon, which belongs to the "climacteric" fruit group, the plant hormone ethylene acts as a signal for the onset of ripening, and coordinates numerous physiological and metabolic processes that occur during ripening (Pech, Bouzayen, & Latche, 2008). In climacteric fruits, the onset of ripening is also generally concomitant with a rise in respiration. Therefore, fruit physiologi-

Table 3
Physical and chemical[a] parameters measured in three melon cultivars harvested at five maturity stages from three planting dates.

Cultivar[b]	Maturity[c]	Planting date	Puncture (N)	Compression (N)	L^*	a^*	b^*	Hue	Ethylene ($\mu L\ kg^{-1}\ h^{-1}$)	CO_2 ($mL\ kg^{-1}\ h^{-1}$)	SSC (%)	TA (%)	Glutamic acid ($g\ L^{-1}$)	Citric acid ($g\ L^{-1}$)	Malic acid ($g\ L^{-1}$)	Sucrose ($g\ L^{-1}$)	Glucose ($g\ L^{-1}$)	Fructose ($g\ L^{-1}$)
T	PL	1	18.0	40.2	65.1	11.6	37.0	0. 30	2.7	186.7	8.7	0.08	0.29	3.06	0.34	18.0	28.0	29.0
		2	19.1	36.9	63.5	9.1	34.2	0.26	1.7	102.4	7.8	0.31	0.17	2.98	0.32	14.6	26.7	28.6
		3	18.7	50.6	64.8	7.9	32.6	0.24	6.1	170.7	8.7	0.06	0.22	3.17	0.31	12.1	25.8	25.8
M	PL	1	19.0	46.0	66.2	11.0	37.3	0.29	3.2	193.4	10.7	0.27	0.52	2.96	0.45	37.3	26.1	26.5
		2	18.0	45.7	66.3	9.7	34.7	0.27	3.6	201.8	8.0	0.07	0.21	2.48	0.43	16.3	26.2	25.5
		3	21.8	48.2	66.9	9.8	35.2	0.27	3.4	127.9	6.6	0.08	0.29	2.78	0.38	8.4	24.1	24.2
N	PL	1	16.5	39.9	67.4	10.5	35.5	0.29	3.8	236.0	9.4	0.04	0.73	2.46	0.41	23.3	28.2	28.6
		2	16.7	38.7	66.3	9.9	32.8	0.29	13.9	189.6	7.9	0.95	0.27	2.56	0.34	15.3	26.2	27.0
		3	16.2	41.4	67.9	10.5	35.4	0.29	6.0	249.8	9.0	0.03	0.40	2.68	0.02	14.7	21.8	27.2
T	PD	1	12.6	27.4	65.9	10.7	38.0	0.27	3.3	177.8	9.3	0.06	0.64	2.41	0.40	26.3	29.1	31.2
		2	11.8	23.3	63.9	10.2	35.0	0.28	8.3	126.8	9.9	0.27	0.55	2.54	0.36	30.7	25.9	27.6
		3	18.6	36.8	62.6	10.3	35.0	0.29	2.9	258.3	10.0	0.12	0.35	3.16	0.36	30.6	26.0	27.1
M	PD	1	16.8	41.9	65.6	11.9	38.3	0.30	3.3	203.8	11.2	0.23	0.88	2.73	0.51	43.7	25.4	26.4
		2	17.0	34.0	64.1	10.9	35.4	0.30	16.2	128.9	9.0	0.06	0.55	1.98	0.49	25.1	24.7	24.8
		3	19.4	36.2	67.9	10.5	34.8	0.29	5.3	151.9	7.6	0.06	0.64	2.21	0.48	17.9	22.2	22.4
N	PD	1	15.7	37.1	65.5	11.9	37.6	0.31	7.2	210.2	10.4	0.03	1.36	2.22	0.49	34.2	26.0	28.3
		2	11.6	27.4	62.7	10.2	33.3	0.29	12.1	253.7	10.0	0.79	1.39	1.61	0.45	27.1	26.6	27.8
		3	16.4	35.7	65.8	11.5	36.3	0.31	6.1	252.9	10.6	0.05	1.35	2.17	0.16	21.4	22.4	19.7
T	Sa	1	6.6	14.6	68.6	9.9	35.6	0.27	21.9	240.1	8.5	0.06	0.68	1.94	0.24	21.7	20.8	23.6
		2	7.7	15.8	58.1	10.1	33.0	0.30	13.9	70.2	8.8	0.32	0.84	2.15	0.30	23.4	24.6	28.6
		3	7.6	17.5	63.4	10.0	35.2	0.28	19.0	175.7	8.3	0.07	0.57	2.87	0.23	23.8	25.8	29.8
M	Sa	1	7.8	18.3	62.6	11.9	38.1	0.30	15.7	172.7	10.6	0.24	1.11	2.86	0.37	38.6	26.1	28.1
		2	13.1	32.3	63.0	11.1	35.7	0.30	13.8	160.0	9.4	0.06	0.69	1.67	0.43	28.9	25.4	26.3
		3	11.8	23.6	65.1	10.2	34.4	0.29	13.8	172.4	8.0	0.06	1.20	1.93	0.41	24.5	20.1	20.6
N	Sa	1	8.8	21.8	64.5	11.7	36.7	0.31	19.1	175.8	10.2	0.04	1.49	2.33	0.42	33.4	27.0	28.8
		2	11.2	25.5	65.3	11.7	36.4	0.31	20.5	295.3	9.1	0.68	1.26	0.85	0.49	29.1	23.3	25.3
		3	9.5	20.5	66.1	11.8	36.7	0.31	20.3	263.6	9.6	0.05	1.13	2.17	0.02	16.1	22.1	27.3
T	Sb	1	5.6	10.9	67.1	11.1	37.8	0.28	15.5	213.5	8.0	0.06	0.98	1.93	0.29	22.1	21.3	25.8
		2	9.7	23.4	61.4	10.7	36.1	0.29	13.5	152.4	8.9	0.30	1.22	1.46	0.41	28.3	21.6	26.3
		3	7.7	14.3	61.6	11.1	35.8	0.30	19.2	267.2	8.7	0.06	0.72	2.24	0.30	25.5	20.7	24.9
M	Sb	1	11.7	31.6	62.8	11.3	37.2	0.29	23.9	263.5	10.3	0.24	1.23	2.47	0.44	40.6	22.6	24.8
		2	7.7	14.3	56.2	10.8	33.6	0.31	11.3	138.9	9.6	0.06	0.60	2.18	0.44	36.1	21.0	23.5
		3	9.2	20.8	65.2	11.0	35.5	0.30	19.1	173.0	9.1	0.06	0.96	2.07	0.40	34.3	19.8	22.2
N	Sb	1	7.2	14.1	60.9	11.0	34.1	0.31	16.1	176.8	9.2	0.04	1.53	1.67	0.48	31.4	22.0	26.1
		2	9.2	20.8	62.2	11.0	34.8	0.30	11.0	253.4	8.1	0.76	1.59	0.92	0.49	21.3	21.0	23.7
		3	8.6	22.1	64.6	11.9	36.8	0.31	20.4	208.9	9.4	0.05	1.97	1.78	0.04	29.0	28.0	35.9
T	Sc	1	4.1	11.6	67.6	10.8	37.1	0.28	16.4	201.8	8.2	0.06	0.83	1.77	0.31	27.7	18.9	26.2
		2	6.1	11.6	62.6	10.2	35.2	0.28	12.6	59.7	8.7	0.29	1.24	1.88	0.38	25.7	21.8	26.2
		3	6.1	16.6	61.4	9.6	33.6	0.28	20.4	212.7	8.7	0.06	0.56	1.91	0.33	32.6	18.3	24.3
M	Sc	1	5.4	16.6	63.5	11.5	37.4	0.30	19.8	221.0	9.6	0.30	0.64	2.87	0.29	35.8	21.7	26.1
		2	8.7	22.8	59.6	11.9	36.0	0.32	14.2	167.4	10.3	0.06	0.78	2.24	0.49	42.2	19.6	21.8
		3	9.1	22.0	62.5	12.4	37.2	0.32	13.6	210.6	9.8	0.07	0.68	2.19	0.48	42.8	18.2	20.1
N	Sc	1	6.5	16.8	61.7	11.1	35.8	0.30	15.8	175.6	9.0	0.04	1.73	1.62	0.55	31.8	18.3	24.4
		2	6.4	15.8	59.4	10.9	34.5	0.31	7.1	201.0	8.6	1.21	0.85	1.29	0.53	28.5	18.7	24.0
		3	8.1	18.0	64.4	12.1	37.1	0.31	19.2	217.0	9.0	0.05	1.70	0.79	0.02	5.4	19.0	21.5

Data are reported as mean value of three replicates.
[a] Volatile compounds are reported in Table 4.
[b] Cultivar: T = Thunderbird; M = Mas Rico; N = Navigator.
[c] Maturity stage at harvest: PL = pre-slip light; PD = pre-slip dark; Sa = slip A; Sb = slip B; Sc = slip C.

cal state during ripening was monitored using flesh ethylene production rates and respiration rates. These parameters were highly variable and affected by environmental factors; however, a general trend in ethylene production could be noted, as there was a significant and consistent sharp increase in ethylene production rate between the "pre-slip dark" (7.2 $\mu L\,kg^{-1}\,h^{-1}$) and "slip A" (17.6 $\mu L\,kg^{-1}\,h^{-1}$) stages in all three cultivars. This sharp increase in ethylene production may constitute a signal for the initiation of faster ripening and may explain the sharper decrease in firmness between these two stages, as ethylene has been shown to regulate cell wall breakdown in melon (Nishiyama et al., 2007).

Comprehensive analysis of melon volatile profiles was carried out using headspace sorptive extraction coupled with GC–MS (HSSE-GC–MS), and 82 volatile compounds were detected (Table 4). Consistent with previous reports (Beaulieu, 2006; Beaulieu & Grimm, 2001; Senesi et al., 2005; Wang et al., 1996; Wyllie, Leach, Wang, & Shewfelt, 1995), the total volatile content significantly increased during ripening. Total volatile concentration in "pre-slip" and "slip A" fruit was less than 20% and 50%, respectively, that of fully ripe fruits ("slip B"). Among all the volatiles detected, twenty-eight compounds were significantly affected by fruit maturity at harvest (Table 4).

Volatile esters were the predominant constituents of melon flesh aroma, representing from 48% to 94% of the total volatiles measured in pre-slip and slip B fruits, respectively.

Several volatiles measured in this study were previously identified as important muskmelon aroma compounds (Beaulieu & Grimm, 2001; Homatidou, Karvouni, Dourtoglou, & Poulos, 1992; Jordan, Shaw, & Goodner, 2001; Kourkoutas, Elmore, & Mottram, 2006; Wyllie & Leach, 1992; Wyllie, Leach, Wang, & Shewfelt, 1994). Various esters quantified in our study were previously investigated for their sensory significance in melon aroma and have been identified as odour active compounds by gas chromatography–olfactometry (Jordan et al., 2001; Schieberle, Ofner, & Grosch, 1990; Wyllie et al., 1994). As reported by these authors, some esters measured in our study were found to actively contribute to the fruity and floral notes characteristic of melon aroma, including isobutyl acetate (described as floral), ethyl 3-(methylthio)propanoate (clean/fresh/melon/green), heptyl acetate (clean/fresh/floral), and (Z)-3-hexen-1-yl acetate (green/herbal/banana).

The aldehyde fraction greatly decreased in ripe fruits. Although environmental conditions affected the concentrations of nearly all aldehydes, hexanal, 2-heptenal and 2,4-nonedienal were generally present at higher concentration in early mature fruits compared to fully ripe fruits. Beaulieu and Grimm (2001) also found higher concentration of 2-heptenal in immature muskmelons (*Cucumis melo reticulatus* group cv. Sol Real and Athena). Hexanal and (E)-2-nonenal, both measured in our study, have been reported to be responsible for green and cucumber-like notes in melon and in cucumber (*Cucumis sativus* L.) aroma (Schieberle et al., 1990; Ullrich & Grosch, 1987). The remaining fraction of the total volatile profile was comprised of sulphur-containing esters, alcohols, ketones, terpenoids, norisoprenoids and unidentified compounds. The concentration of sulphur-containing esters, alcohols and terpenoids significantly increased as fruits ripened.

In addition to ripening-associated changes in melon aroma, quantitative differences in volatile concentrations were observed between the three cultivars investigated in this study. Cultivar-specific differences in aroma profiles were also reported in previous studies (Beaulieu & Grimm, 2001; Obando-Ulloa, Ruiz, Monforte, & Fernandez-Trujillo, 2010; Senesi et al., 2005; Wyllie & Leach, 1992; Yamaguchi et al., 1977), highlighting a strong genetic control of the aromatic trait.

On average, the total volatile content was higher in Navigator than in Thunderbird and Mas Rico (68% and 36% of the total volatile content of Navigator, respectively).

Although sulphur-containing esters were measured in all three cultivars, a significant increase in ethyl-(methylthio)acetate and ethyl-3-(methylthio)propionate concentrations was observed during ripening only in Navigator. In a broad screening conducted on 26 melon cultivars with the intent to investigate the incidence of sulphur-containing esters in the aroma profile, ethyl-(methylthio)acetate was frequently detected and its presence in the aroma extract appeared to be cultivar dependent (Wyllie & Leach, 1992).

3.2. Investigating relationships between sensory attributes and physiochemical parameters

To investigate relationships among sensory perception and physiochemical parameters, and the effects of ripening on overall melon flavour, a correlation analysis was performed on all variables, followed by principal component analysis (PCA) on the correlations matrix to visualise the relationships. The first six components explained 70.3% of the total variation, with the first two components accounting for 43.8% and 10.5% of the total variation (Fig. 1). While variation along the second component was likely due to environmental effects, on the first component, separation of the samples was primarily driven by maturity at harvest. Early mature fruits ("pre-slip" light and dark) clustered on the left side of the plot and were negatively correlated with the more mature fruits ("slip B" and "slip C"), which were grouped on the opposite quadrant (Fig. 1).

Indeed, all the variables that had significantly higher values in pre-slip fruits were found on the left quadrant of the plot. Flesh texture ("compression" and "puncture") was positively correlated with sensory firmness ($r = 0.87$ and 0.92) and crunchiness ($r = 0.88$ and 0.92) sensory attributes, which were scored high in "pre-slip" fruits (Fig. 1). Cucumber aroma was significantly correlated to firmness, as measured with both Texture Analyzer and by the sensory panel ($r \geqslant 0.81$). "Cucumber aroma" was also correlated with the volatile compound, 2-heptenal ($r = 0.78$), which is derived from oxidation/degradation of fatty acids and has been previously reported to contribute to the flavour of fresh cucumbers (Grosch & Schwarz, 1971; Ullrich & Grosch, 1987). Significant associations between the sensory attributes "hardness", "cucurbit" (which could correspond to our sensory "firmness" and "cucumber aroma") and total aldehyde levels were also found in the Western Shipper cultivar Sol Real (Beaulieu & Lancaster, 2007).

Ethylene was produced at higher levels in fully ripe fruits and was also negatively correlated to firmness evaluated using both instrumental (compression and puncture: $r = -0.69$ and -0.74) and sensory assessments (crunchiness and firmness: $r = -0.74$ and -0.75). Ethylene has been shown to play an important role in the regulation of many ripening processes including fruit softening in melon (Ayub et al., 1996; Hadfield et al., 2000; Pech et al., 2008). For example, in ripening Charentais melons (*Cucumis melo, cantalupensis* group), fruit softening due to cell wall disassembly was shown to be an ethylene-dependent mechanism (Nishiyama et al., 2007). Ethylene has also been shown to be involved in regulating volatile production and aroma development during melon ripening (Bauchot, Mottram, Dodson, & John, 1998; El-Sharkawy et al., 2005; Flores et al., 2002). Consistently, we observed that in fully ripe fruits, sensory attributes such as "overall aroma intensity", "juiciness", "fruity flavour", "marshmallow aroma" and "fruity aroma" were significantly correlated with ethylene production ($r \geqslant 0.59$). In addition, ethylene production was highly correlated with the concentrations of numerous volatile compounds, particu-

Table 4
Analysis of variance of volatile compounds: significance levels of main effects and interactions.

	Volatile compound	CAS #	RT[a]	RI[b]	RI[c]	Cultivar	Maturity	Planting date	Maturity × cultivar	Maturity × planting date	Cultivar × planting date
1	Ethyl acetate	141-78-6	2.12	674	605/628[d]	**0.045**	**0.005**	0.735	0.532	0.827	0.308
2	Propyl acetate	109-60-4	2.99	722	707/720[d]	**0.023**	**0.015**	0.167	0.087	0.249	0.188
3	Ethyl isobutyrate	97-62-1	3.68	761	751/762[d]	**<.0001**	**<.0001**	0.471	**0.0002**	0.435	0.454
4	Isobutyl acetate	110-19-0	3.94	775	768/776[d]	**0.025**	**<0.001**	0.361	0.829	0.608	0.129
5	Hexanal	66-25-1	4.44	802	801	0.306	**0.009**	**0.002**	0.580	**0.012**	**0.037**
6	Ethyl butanoate	105-54-4	4.50	804	803	**<.0001**	**<.0001**	0.190	**<.0001**	0.277	0.225
7	Butyl acetate	123-86-4	4.81	816	812	**0.002**	**<.0001**	0.094	**0.004**	0.296	0.362
8	2-Ethyl-3-methylbutanal	26254-92-2	5.29	834		0.871	0.154	**0.001**	0.297	**0.009**	**0.023**
9	Ethyl 2-methylbutanoate	7452-79-1	5.69	849	846	**<.0001**	**<.0001**	0.533	**0.0003**	0.660	0.595
10	Methyl thiobutyrate	2432-51-1	5.78	852	869[*]	**<.0001**	**0.0003**	0.176	**0.003**	0.496	0.221
11	2-Methylpropyl propanoate	540-42-1	6.21	868	863	**0.007**	**0.0002**	0.142	0.072	0.505	**0.027**
12	Hexanol	111-27-3	6.32	872	865	**0.0002**	**<.0001**	0.333	**0.002**	0.816	0.441
13	3-Methylbutyl acetate	123-92-2	6.47	878	876	0.144	**0.001**	**0.035**	0.132	**0.005**	0.607
14	2 Methylbutyl acetate	624-41-9	6.53	880	877	**0.001**	**<.0001**	**0.012**	0.187	**0.020**	0.626
	2-Butyl furan	4466-24-4	6.87	892	894[d]	0.082	0.153	0.123	0.119	0.156	0.161
15	Heptanal	111-71-7	7.14	902	902	**<.0001**	**0.015**	**0.001**	0.171	**0.056**	**0.001**
16	Ethyl pentanoate	539-82-2	7.17	903	898[d]	**<.0001**	**0.002**	0.130	**0.003**	0.497	0.085
17	Pentyl acetate	628-63-7	7.61	916	912	**0.006**	**<.0001**	**0.003**	0.055	**0.017**	0.266
18	Butyl propanoate	590-01-2	7.74	920	907	0.236	**0.005**	**0.024**	0.926	0.408	**0.001**
19	Prenyl acetate	1191-16-8	7.89	924	918	**0.006**	**<.0001**	0.411	**0.011**	0.409	0.337
20	Methyl hexanoate	106-70-7	7.93	925	922	**<.0001**	**<.0001**	0.784	**<.0001**	0.582	0.827
21	S-methyl 3-methylbutanethioate	23747-45-7	8.34	938	938	0.061	**<.0001**	0.297	0.678	0.833	0.043
22	Ethyl (E)-2-methyl-2-butenoate	5837-78-5	8.43	940	949[d]	**<.0001**	**<.0001**	0.071	**<.0001**	0.517	0.077
23	2-Heptenal	18829-55-5	8.93	955	955	0.058	**<.0001**	**0.019**	0.174	**0.022**	0.078
24	Isobutyl butanoate	539-90-2	8.96	956	953[d]	**0.004**	**<.0001**	**0.006**	**0.004**	0.077	0.177
25	Benzaldehyde	100-52-7	9.02	958	962	**<0.001**	**<.0001**	0.696	**0.004**	0.847	0.287
26	3-Methylbutyl propanoate	105-68-0	9.52	972	974[d]	**0.001**	**0.002**	0.202	**0.015**	0.122	**0.002**
27	1-Octen-3-one	4312-99-6	9.71	978	975	0.082	**0.002**	**0.048**	0.683	0.817	0.574
28	1-Octen-3-ol	3391-86-4	9.77	980	978	**<.0001**	**0.001**	0.317	0.200	0.299	**0.001**
29	Ethyl (methylthio)acetate	4455-13-4	9.91	984	981	**<0.001**	**<.0001**	0.298	**0.003**	0.614	0.249
30	6-Methyl, 5-hepten-2-one	110-93-0	10.02	987	985[d]	0.223	0.319	**0.001**	0.304	0.132	0.264
31	2-Pentylfuran	3777-69-3	10.14	991	989/992[d]	0.658	0.496	**0.033**	0.650	0.440	0.158
32	2-Octanone	111-13-7	10.16	991	992[d]	**0.005**	**0.001**	**0.010**	0.561	0.971	0.666
33	Ethyl hexanoate	123-66-0	10.50	1001	999	**<.0001**	**<.0001**	0.328	**<.0001**	0.629	0.192
34	Octanal	124-13-0	10.55	1003	1003	**0.001**	0.095	**0.011**	**0.007**	**0.018**	**<.0001**
35	(Z)-3-hexen-1-ol, acetate	3681-71-8	10.75	1008	1004/1008[d]	**0.001**	**<.0001**	0.063	**0.008**	0.373	0.157
36	Hexyl acetate	142-92-7	10.98	1015	1011/1015[d]	**0.023**	**<.0001**	**0.012**	0.120	0.068	0.264
37	(E)-4-hexen-1-yl acetate[e]	42125-17-7	11.27	1023		**0.003**	**<.0001**	**0.037**	0.018	0.102	0.328
38	Methyl heptanoate	106-73-0	11.36	1025	1021	**<.0001**	**<.0001**	0.055	**<.0001**	0.393	**0.035**
39	Limonene	5989-27-5	11.38	1026	1029	**0.002**	0.109	0.495	**0.033**	**0.038**	0.260
40	Eucalyptol	470-82-6	11.47	1028	1032/1030[d]	**0.004**	**<.0001**	**0.008**	**0.041**	0.217	**0.039**
41	2-Ethyl hexan-1-ol	104-76-7	11.51	1029	1029[*]	**0.001**	0.072	0.106	0.364	0.767	0.202
42	Benzyl alcohol	100-51-6	11.73	1035	1033	**0.021**	**0.0003**	0.523	0.075	0.498	0.737
43	2,6-Dimethyl-4-heptanone	68514-40-9	12.16	1047		0.260	0.299	**0.012**	0.386	0.134	**0.0002**
44	(E)-oct-2-enal	2548-87-0	12.50	1057	1060[d]	**0.011**	0.690	0.173	0.906	0.838	0.064
45	2,3-Butanedioldiacetate (I)	1114-92-7	12.77	1064	1064	**0.002**	**<.0001**	0.809	**0.012**	0.964	0.157
46	1-Octanol	111-87-5	13.03	1072	1070	**0.008**	**0.014**	0.273	**0.030**	0.858	0.806
47	2,3-Butanedioldiacetate (II)	1114-92-7	13.24	1077		**0.040**	**<.0001**	0.851	0.115	1.000	0.180
48	2-Nonanone	821-55-6	13.76	1092	1091[d]	**0.004**	**0.007**	**0.003**	**0.008**	0.127	0.057
49	Methyl benzoate	93-58-3	13.80	1093	1091[d]	**0.006**	**0.003**	0.421	0.115	0.980	0.070
	Tetrahydro linalool	78-69-3	13.97	1098	1098[d]	0.367	0.907	0.422	0.403	0.553	0.409
50	Ethyl 3-(methylthio) propionate	13327-56-5	14.02	1099	1098	**<.0001**	**<.0001**	0.076	**<.0001**	0.420	0.109
51	Nonanal	124-19-6	14.18	1104	1104	**0.001**	0.101	0.088	0.270	0.081	**0.002**
	n-Amyl isovalerate[IS]	25415-62-7	14.36	1108	1107						
52	Heptyl acetate	112-06-1	14.53	1113	1111	0.825	**<.0001**	**0.002**	0.614	**0.044**	0.092

53	Methyl octanoate	111-11-5	14.95	1125	1126[d]	**<.0001**	**<.0001**	0.143	**<.0001**	0.446	0.123
	Unknown1		15.68	1146		**<.0001**	0.305	0.246	0.623	0.126	**<.0001**
54	2-Ethylhexyl acetate	103-09-3	15.88	1151	1144[*]	**0.004**	**<.0001**	**<.0001**	0.070	**0.136**	0.287
55	(E)-2-nonenal	18829-56-6	16.14	1159	1162/1155[d]	**0.001**	0.350	0.753	0.941	0.825	0.052
56	Benzyl acetate	101-41-7	16.33	1164	1261[d]	**<.0001**	**<.0001**	0.147	**0.024**	0.340	0.496
57	Ethyl benzoate	93-89-0	16.51	1169	1172/1170[d]	**<.0001**	**<.0001**	0.109	**<.0001**	0.442	0.159
	Unknown2		17.19	1188		**0.014**	**<.0001**	0.766	0.294	0.459	0.653
58	Ethyl octanoate	106-32-1	17.52	1197	1194	**<.0001**	**0.001**	0.062	**0.0003**	0.398	0.062
59	Decanal	112-31-2	17.77	1205	1205	**0.001**	0.347	0.128	0.675	0.259	**0.017**
60	Octyl Acetate	112-14-1	18.02	1212	1213	**0.017**	**0.0001**	**0.013**	0.133	0.093	0.136
61	2,4-Nonadienal	5910-87-2	18.04	1213	1216	0.482	**0.002**	0.511	0.090	0.925	0.183
62	β-Ciclocitral	432-25-7	18.22	1218	1220	**0.012**	0.083	**<.0001**	0.314	0.733	0.056
63	Ethylphenyl acetate	101-97-3	19.12	1244	1243	**<.0001**	**<.0001**	0.433	**<.0001**	0.624	0.332
64	2-Phenylethyl acetate	103-45-7	19.51	1256	1255	0.302	**<.0001**	**0.001**	0.549	0.263	0.105
65	(E)-2-decenal	3913-81-3	19.68	1261	1261[d]	0.118	0.430	0.621	0.798	0.330	**0.025**
	Unknown3		19.90	1267		**0.014**	**0.011**	0.932	0.391	0.081	**<.0001**
66	2-Undecanone	112-12-9	20.78	1293	1296[d]	**0.019**	**0.048**	**0.018**	0.055	0.181	0.107
67	Undecanal	112-44-7	21.21	1306	1306	**0.002**	0.787	0.288	0.621	0.081	**0.025**
68	α-Terpinyl acetate	80-26-2	22.57	1348	1350[d]	**0.001**	**0.0002**	0.373	0.116	0.590	**0.003**
	Unknown4		22.60	1349		0.064	0.452	0.090	0.370	0.159	0.307
69	3-Phenylpropyl acetate	122-72-5	23.26	1369	1373	0.284	**0.001**	**0.012**	0.689	0.116	0.379
	Unknown 5		23.32	1371		**0.025**	0.500	0.052	0.317	0.173	0.317
70	Ethyl decanoate	110-38-3	24.11	1396	1392	**<.0001**	**0.022**	0.098	0.146	0.456	**0.018**
	Tetradecane	629-59-4	24.21	1399	1399[d]	0.065	0.725	0.439	0.681	0.260	0.245
71	Dodecanal	112-54-9	24.49	1408	1407[d]	**0.001**	0.603	0.408	0.263	0.111	**0.020**
72	Geranyl acetone	3796-70-1	25.85	1453	1448/1453[d]	**0.017**	0.150	**0.001**	0.324	0.948	0.083
73	β-Ionone	14901-07-6	26.84	1485	1484	**0.005**	0.237	**<.0001**	0.301	0.934	0.066
74	Dihydroactinidiolide	17092-92-1	27.91	1533	1539[d]	**0.042**	0.442	**0.003**	0.187	0.560	0.147

Bold *p*-values indicate statistical significance (<0.05).

[a] RT: retention time expressed in minutes.

[b] RI: retention indices calculated from C8–C20 *n*-alkanes.

[c] RI: retention indices reported in the literature for DB-5MS capillary GC columns (Beaulieu & Grimm, 2001).

[d] RI: retention index reported in Pherobase, Flavournet and NIST library 8.0 for DB-5MS and HP-5MS capillary GC column.

[e] Tentatively identified.

[*] RI: retention index reported in Pherobase on DB-1 capillary column.

[IS] Internal standard.

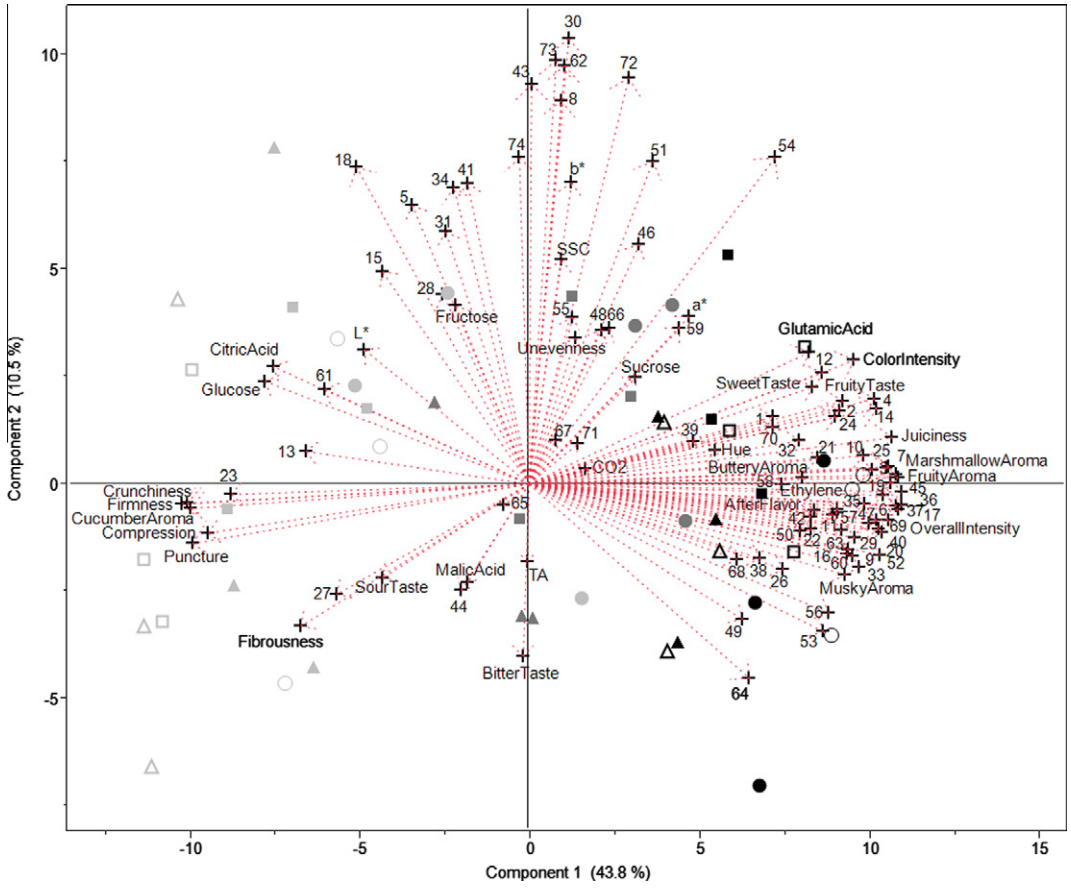

Fig. 1. Biplot from Principal Component Analysis of physical and chemical parameters and sensory attributes. Symbol shape corresponds to different cultivars: square = Thunderbird; triangle = Mas Rico; circle = Navigator. Maturity is represented by different shades of grey: open symbols, light grey = pre-slip light; filled symbols, light grey = pre-slip dark; filled symbols, dark grey = slip A; filled symbols, black = slip B; open symbols, black = slip C. Numbers correspond to identified volatiles reported in Table 4; only the volatiles significantly influenced by maturity at harvest, cultivar and/or planting date effects were included in the analysis. The length of the dotted arrows from the origin of the axis to a particular parameter indicates how well the variation in that parameter is explained by the model.

larly esters, e.g., 2-methylbutyl acetate ($r = 0.73$), pentyl acetate ($r = 0.66$), heptyl acetate ($r = 0.63$), and hexyl acetate ($r = 0.63$).

The "fruity aroma" sensory attribute was significantly and positively correlated with many volatiles, including benzyl acetate, pentyl acetate, hexyl acetate, benzaldehyde, eucalyptol, heptyl acetate, isobutyl butanoate and prenyl acetate (correlation coefficients $r \geqslant 0.76$). Benzyl acetate, hexyl acetate and eucalyptol have previously been described in various melon types as characteristic impact flavour or aroma compounds (CIFAC) with fruity, floral and/or fresh odour characters (Beaulieu, 2005; Jordan et al., 2001; Kourkoutas et al., 2006).

Two isomers of 2,3-butanediol diacetate detected in our samples have also been reported in several melon varieties (Beaulieu & Grimm, 2001; Homatidou et al., 1992; Kourkoutas et al., 2006; Wyllie & Leach, 1990). The racemic mixture was described to have a sweet smell and high odour threshold, and therefore suggested not to significantly impact the aroma of melon cv. Golden Crispy (Wyllie & Leach, 1990). In our samples, however, both isomers were positively correlated to the "marshmallow aroma" sensory attribute ($r = 0.76$).

"Musky aroma" was positively correlated to the presence of ethyl-(methylthio)acetate and ethyl butanoate ($r \geqslant 0.80$). In our study the perception of "musky aroma" was higher in Navigator; the volatile profile for this variety was also significantly richer in sulphur-containing esters compared to the other varieties. It has been reported that trace amounts of some sulphur compounds, such as ethyl-(methylthio)acetate, might contribute to the musky note in muskmelon (*Cucumis melo* cv. Makdimon) (Wyllie et al., 1994).

"Sweet taste" was positively correlated to concentrations of sucrose ($r = 0.53$), as expected, and more surprisingly, to glutamic acid ($r = 0.53$). Human sweet and umami taste receptors, located in taste buds, respond to diverse sweetening components (such as sugars, synthetic sweeteners and sweet-tasting proteins) and glutamate and purine nucleotides (such as 5′-inosine monophosphate and 5′-guanosine monophosphate), respectively. Interestingly, sweet and umami receptor protein complexes have been shown to share a common subunit (Li et al., 2002), and several studies in rats have suggested that glutamate can activate both types of receptors, likely evoking both sweet and umami tastes (Heyer, Taylor-Burds, Tran, & Delay, 2003; Ninomiya et al., 2000; Sako & Yamamoto, 1999; Yamamoto et al., 1991). While the role of glutamate as flavour enhancer is well established in human taste perception, most studies have focused on the effect of glutamate concentration on taste modalities (e.g. saltiness, bitterness and umami) associated with savoury foods (Fuke & Ueda, 1996; Yeomans, Gould, Mobini, & Prescott, 2008). On the other hand, the specific effect – if any – of glutamate on sweetness perception in humans is still unclear (Kemp & Beauchamp, 1994; Maga, 1983). Our results suggest an intriguing role for glutamate in sweetness perception in melons that warrants further investigation.

3.3. Evaluation of melon flavour quality using a portable ultra-fast gas chromatograph (the zNose™)

An analysis of variance performed on a total of 22 peaks measured by UFGC–SAW showed that the abundance of 6 peaks (peak

2, 5, 9, 13, 14 and 17) was significantly influenced by maturity at harvest regardless of the cultivar, although some quantitative differences were also observed between cultivars (Table S3).

To our knowledge, no previous UFGC–SAW studies investigated relationships between peaks detected and sensory attributes. Towards this goal, we selected peaks and sensory attributes that were significantly affected by maturity at harvest, cultivar and/or planting date, and a PCA was performed on the correlation matrix (Fig. 2). The first two components accounted for 62% of the total variance explained. Consistent with aforementioned results of PCA on physiochemical parameters and sensory attributes, the samples separation was driven by a maturity effect on the first component, while a planting date effect was the main driver of the separation on the second component. As shown in the biplot (Fig. 2), peaks 9 and 14 were positively correlated to "cucumber aroma" ($r = 0.83$ and 0.85), "firmness" ($r = 0.78$) and "crunchiness" ($r = 0.78$ and 0.77) sensory attributes. In fully ripe fruits, the highest significant correlations for "fruity aroma" were observed with peak 5 ($r = 0.95$), peak 2 ($r = 0.90$) and peak 17 ($r = 0.93$). Peak 17 was also found to be significantly correlated to "sweet taste" and "fruity flavour" attributes ($r = 0.70$ and 0.73).

To evaluate the prediction ability of these correlations, regression analysis was conducted on aforementioned sensory attributes and peaks. Significant linear relationships were observed between "fruity aroma" and peak 2 ($R^2 = 0.81$; $y = 4.8301x - 0.5669$), peak 5 ($R^2 = 0.78$; $y = 0.5095646 + 0.0773868x$) and peak 17 ($R^2 = 0.54$; $y = 0.3484706 + 0.0919787x$), and between peak 17 and "fruity flavour" ($R^2 = 0.70$; $y = 0.2985612 + 0.1009298x$), and "sweet taste" ($R^2 = 0.67$; $y = 0.3883602 + 0.0964181x$). Significant linear relationships were also observed between "cucumber aroma" and peaks 9 and 14, although they only explained 38% and 41% of the model variation, respectively.

These results indicate that several peaks detected by UFGC–SAW may represent markers for these sensory attributes.

This technology has been used in several studies for rapid volatile analysis of fresh produce (Du, Olmstead, & Rouseff, 2012; Li, Heinemann, & Irudayaraj, 2007; Li, Wang, Raghavan, & Vigneault, 2009; Vallone et al., 2012; Watkins & Wijesundera, 2006). However, to our knowledge, only few studies have focused on fruit maturity assessment using UFGC–SAW.

To evaluate mango (*Mangifera indica* L.) ripeness, Li et al. (2009) selected a peak, whose concentration increased consecutively the respiration climacteric, and used it to predict mango ripeness, achieving an 80% accuracy rate.

UFGC–SAW analysis has also been used in a recent investigation to discriminate among blueberries harvested at three maturity stages (Du et al., 2012). Using 14 selected peaks, a clear separation between fully ripe and less ripe berries was successfully achieved for one of the two cultivars assessed ("Primadonna" and "Jewel"). As the authors suggested, the greater separation of the fully ripe berries of the cultivar "Primadonna" might be ascribed to the higher level of total volatiles produced by this cultivar.

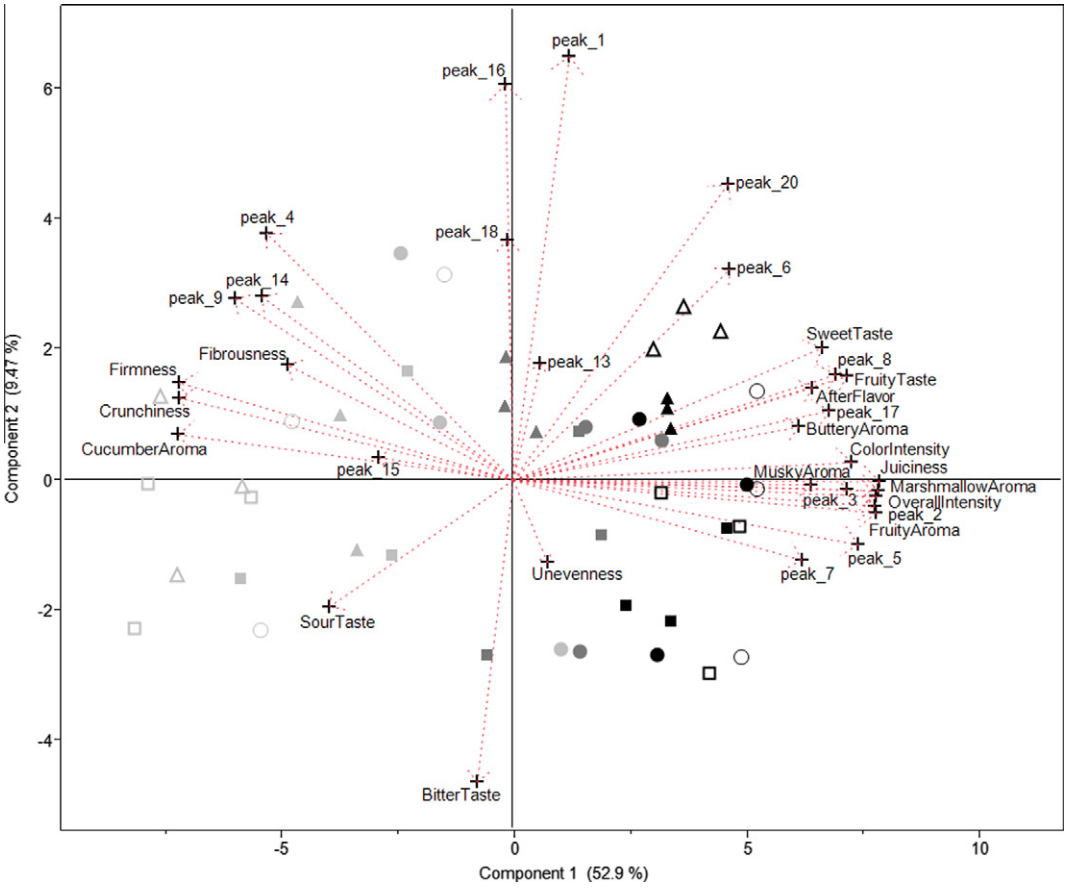

Fig. 2. Biplot from principal component analysis of peaks from UFGC–SAW analysis and sensory attributes. Symbol shape corresponds to different cultivars: square = Thunderbird; triangle = Mas Rico; circle = Navigator. Maturity is represented by different shades of grey: open symbols, light grey = pre-slip light; filled symbols, light grey = pre-slip dark; filled symbols, dark grey = slip A; filled symbols, black = slip B; open symbols, black = slip C. The length of the dotted arrows from the origin of the axis to a particular parameter indicates how well the variation in that parameter is explained by the model.

4. Conclusions

Maturity at harvest, cultivar and planting date qualitatively and quantitatively affected chemical composition and physical characteristics of melon, ultimately impacting sensory perception.

Overall, the perception of sweetness, fruity and musky notes was greater in ripe fruit, while cucumber notes were predominant in less ripe fruit. Instrumental measurements of flesh firmness were generally correlated with sensory texture perception. After esters, aldehydes were the most abundant group of volatiles in pre-slip fruit. Hexanal, typically imparting green notes to fruit aromas, was the predominant aldehyde measured, while 2-heptenal was significantly correlated with cucumber aroma in pre-slip fruit. In riper fruit, esters and sulphur containing compounds predominated and correlated with perceived fruity and musky aromas. The cultivar Navigator was generally perceived to have the highest intensity of colour, aroma, taste, and flavour attributes, while Mas Rico scored higher for cucumber notes and firmness. Identifying all the variables affecting fruit quality and monitoring them during the pre- and post-harvest life of a fruit remains a challenge due to the lack of a single rapid, comprehensive method for physiochemical analysis.

Volatiles play an important role in determining melon flavour, and the significant correlations existing among various volatiles and sensory attributes found in our and previous studies, suggest that this relationship might be potentially exploited to predict sensory perception by measuring volatiles.

Here, a portable volatile sensing system (UFGC–SAW) was able to discriminate melons of different maturity stages based on 6 measured peaks, whose abundances were positively correlated to sensory attributes associated with different maturity stages of melon. Our findings suggest that this technology shows much promise for future applications in rapid aroma measurement. Further investigations are needed to confirm the ability of the UFGC–SAW instrument in predicting fruit sensory properties.

Acknowledgements

This project was supported by the Specialty Crops Research Initiative Competitive Grants Program Grant No. 2009-51181-05783 from the USDA National Institute of Food and Agriculture. We are grateful to Bill Copes (HM. Clause) for providing the melons used in this study, and to Robin Clery (Givaudan) for his kind gift of 2,3-butanediol diacetate. We thank Edward Orgon, Michael Mace, Aly Depsky and Sharon Wei for technical assistance.

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.2012.12.042.

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