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Linking sensory and proton transfer reaction-mass spectrometry analyses for the assessment of melon fruit (Cucumis melo L.) quality traits Tiago Bianchi<sup>1,2</sup>, Luis Guerrero<sup>1</sup>, Yannick Weesepoel<sup>2</sup>, Jason Argyris<sup>3</sup>, Alex Koot<sup>2</sup>, Marta Gratacós-Cubarsí<sup>1</sup>, Jordi Garcia-Mas<sup>3</sup>, Saskia van Ruth<sup>2,4,5</sup>, Maria Hortós<sup>1,\*</sup> <sup>1</sup>IRTA, Food Technology Centre, Finca Camps i Armet, 17121 Monells, Girona, Spain <sup>2</sup>Wageningen Food Safety Research, part of Wageningen University & Research, P.O. box 230, 6700 AE Wageningen, The Netherlands <sup>3</sup>IRTA, Centre for Research in Agricultural Genomics, CSIC-IRTA-UAB-UB, 08193 Bellaterra, Barcelona, Spain <sup>4</sup>Food Quality and Design Group, Wageningen University & Research, P.O. box 17, 6700 AA Wageningen, The Netherlands <sup>5</sup>School of Biological Sciences, Queen's University Belfast, 19 Chlorine Gardens, Belfast, BT9 5DL, Northern Ireland, United Kingdom 16-digit ORCID of the authors: Tiago Bianchi (0000-0002-4327-1092), Luis Guerrero (0000-0001-7062-874X), Yannick Weesepoel, Jason Argyris (0000-0003-2685-9959), Alex Koot, Marta Gratacós-Cubarsí, Jordi Garcia-Mas (0000-0001-7101-9049), Saskia van Ruth (0000-0003-3955-7976), Maria Hortós (0000-0001-7823-9626) \* Corresponding author at: IRTA, Food Technology Centre, Finca Camps i Armet, 17121 Monells, Girona, Spain. E-mail addresses: maria.hortos@irta.cat

#### Abstract

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Sixty-seven samples of ten melon types (Cucumis melo L.) were evaluated to determine the relationship between their quality traits: sensory attributes, pH, soluble solids, and volatile organic compounds. Fruits from the cantalupensis, conomon, dudaim, inodorus, and momordica cultivar groups were analyzed. The sensory profiles were assessed using ten attributes covering odor, flavor, and taste characteristics, whereas the volatile profiles were derived by proton transfer reaction-mass spectrometry. Fruits from the cantalupensis and inodorus cultivars showed an opposite pattern for several quality traits. Fruits from the dudaim cultivar were more related to the cantalupensis, whereas conomon and momordica showed an intermediate behavior between inodorus and cantalupensis. The attributes of odor and flavor intensity, ripe fruit odor, fermentative odor, and fermentative flavor correlated positively to  $C_3$ – $C_9$  esters (r = 0.43– 0.73;  $p \le 0.01$ ). Positive correlations were also observed for several alcohols (r = 0.36 - 0.82;  $p \le 0.05$ ), including methanol, ethanol, and diol alcohols, as well as for several aldehydes (r = 0.43-0.85;  $p \le 0.01$ ), such as acetaldehyde, butanal, methyl butanal, heptanal, and decanal. The attributes mentioned above were negatively correlated with two  $C_9$  aldehydes, 2,6-nonadienal and nonenal (r = -0.45 to -0.62;  $p \le$ 0.01), whereas sweetness was negatively correlated with two C<sub>6</sub> green leaf volatiles, hexenal and 3hexenol  $(r = -0.50; -0.67; p \le 0.001)$ . The melon fruits presented distinct differences in the quality traits evaluated. These results provide information for the development of new cultivars with characteristic taste combinations without compromising other desirable fruit quality traits.

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Keywords: flavor, melon fruit, odor, PTR-MS, sensory analysis, volatile organic compounds

#### 1. Introduction

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Melon (Cucumis melo L.) is a species with high genetic variation, the fruits of which show a wide diversity in morphological, physical-chemical and sensory traits. The sweet melons of the *inodorus* group or the highly aromatic melons of the *cantalupensis* one are generally consumed as fresh fruits. In contrast, while the exotic cultivars of the *conomon*, dudaim, or momordica groups are either inedible or consumed as fresh, cooked, or pickled vegetables [1, 2]. Melon has an exceptional ripening pattern as it comprises both climacteric and non-climacteric cultivars within a single species, i.e., cultivars with a rise in the respiration rate and ethylene production at the onset of fruit ripening (e.g., cantalupensis), and cultivars with little or no ethylene production (e.g., inodorus). However, it has also been reported that melon ripening behavior follows a continuous spectrum between the climacteric and non-climacteric references rather than just two ripening patterns [3]. In addition, ethylene-dependent and ethylene-independent pathways can coexist during the ripening process of climacteric melon fruits [4-6]. However, the relationships between fruit quality traits and the biochemical pathways involved in ethylene-dependent and ethylene-independent ripening processes are not entirely understood [7]. The fruits of the cantalupensis cultivars are generally more aromatic but show a faster loss of firmness and a shorter shelflife than the ones of *inodorus* cultivars [8]. These differences are reflected in the sensory attributes and consumer acceptance of commercial cultivars [9, 10], but little is known about the odor and flavor profiles of the conomon, dudaim, or momordica exotic cultivars. Odor and flavor are among the properties that most influence the sensory perception of fruit. Melon odor perception depends on the presence and concentration of volatile organic compounds (VOCs), which comprise a profile typically associated with each cultivar [9, 11]. In addition, flavor perception relies not only on volatile but also non-volatile compounds such as soluble sugars and organic acids. Sweetness is considered a determinant attribute for the eating quality of melon fruit, but a moderate acidity is also able to drive consumers' liking of this fruit species [10]. Interactions between volatile and non-volatile compounds should also be considered as VOCs are known to enhance the perception of several flavor attributes [12-15]. Gas chromatography is the most common technique for the assessment of melon VOCs profile, which comprises esters, alcohols, aldehydes, some sulfur-containing compounds, and minor quantities of ketones, terpenes and hydrocarbons [8, 11, 16-22]. An alternative is the use of proton transfer reactionmass spectrometry (PTR-MS), which allows the headspace VOCs to be drawn from the samples at room temperature (25 °C), simulating the conditions of consumer perception of the fruits. Headspace PTR-MS

allows a highly sensitive, real-time volatile detection (pptv, parts per trillion by volume detection and less than 1 minute for a complete spectrum acquisition) without any sample pretreatment. The method is based on a soft chemical ionization by protonated water molecules  $(H_3O^+)$ , which perform a non-dissociative proton transfer to most of the common VOCs without reacting with any of the natural components of air [23, 24].

The aim of the present study was to assess the odor and flavor profiles of ten types of melon fruits and evaluate the correlation between their quality traits: i.e., between their sensory attributes, pH, soluble solids, and VOCs. Melon genotypes belonging to the *cantalupensis*, *conomon*, *dudaim*, *inodorus*, and *momordica* cultivar groups, together with commercial reference varieties from *cantalupensis* and *inodorus* cultivars, were selected to represent the variation within the species.

## 2. Material and methods

2.1 Material

Fruits of ten melon (*Cucumis melo* L.) types (Table 1), comprising genotypes from the subspecies *melo* and *agrestis*, together with commercial reference varieties, were analyzed (N = 67). The commercial varieties were obtained from a local market, while the cultivars were grown at the 'IRTA-Torre Marimon' greenhouse (41°36'47.88"N 2°10'10.45"E, Barcelona, Spain) and harvested at physiological maturity. The fruits were harvested at 40-45 days after pollination (dap) for 'Irak' and 'Calcuta' cultivars, 45 dap for 'Védrantais' and 50 dap for 'Dulce', which corresponded to the change in color and abscission of the fruits, or at 50 dap for 'Songwhan charmi' and 55 dap for 'Piel de Sapo-T111' as it was previously determined to be the point at which these cultivars had high sucrose content, and thus optimal fruit quality [3].

Fruits were transversally cut into 2 cm slices, and both stem and blossom-ends discarded. The middle slice was used for pH and soluble solids content (SSC) determinations, while the two contiguous slices were covered with plastic wrap and stored at 4 °C until the sensory analysis. The flesh of the remaining slices was vacuum-packed in double-layer aluminum bags and stored at -80 °C for further analyses, after the removal of the skin plus 1 cm of underlying flesh and the placental tissue.

**Table 1** Melon fruits used in this study  $(N = 67)^a$ 

Melon fruit (accession)	Subspecies Cultivar group		Respiration pattern	Country of origin	
Cultivars					
'Dulce'	melo	cantalupensis	climacteric	USA	
'Védrantais'	melo	cantalupensis	climacteric	France	
'Irak' (C-1012)	melo	dudaim	climacteric	Irak	
'Calcuta' (PI-124112)	agrestis	momordica	climacteric	India	
'Songwhan charmi' (PI-161375)	agrestis	conomon	non climacteric	Korea	
'Piel de Sapo' (T111)	melo	inodorus	non climacteric	Spain	
Commercial varieties					
Galia	melo	cantalupensis	climacteric	Spain	
Cantaloupe	melo	cantalupensis	climacteric	Spain	
Amarillo	melo	inodorus	non climacteric	Spain	
Piel de Sapo	melo	inodorus	non climacteric	Spain	

<sup>111 &</sup>lt;sup>a</sup> Number of samples for each melon fruit type: Amarillo (n = 3); Cantaloupe (n = 3); Galia (n = 6); Piel de

## 2.2 Methods

#### 2.2.1 Common quality indices

The pH measurements were performed in the flesh of the middle slice of each fruit using a puncture electrode pH-meter with temperature correction probe, model 5053-T (Crison Instruments, Barcelona, Spain). Flesh from the same slice was hand-squeezed, and the soluble solids content (SSC) was measured in the juice using a Quick-Brick<sup>TM</sup> 90 digital refractometer (Mettler-Toledo, GmbH, Germany). Both parameters were measured in triplicate, and the values expressed as average results (N = 67) (Table 2, Appendix).

## 2.2.2 Sensory analysis

The sensory analyses were performed by an eight-member panel with extensive experience in quantitative and descriptive methods, selected and trained following ISO 8586-1:1993 [25] and ISO 8586-2:1994 [26]. Ten descriptors of odor, flavor, and taste attributes (Table 3) were chosen during training sessions of open discussion between the panelists. Different commercial melon samples were evaluated during these sessions in order to have a wide range of sensory characteristics frequent in melon fruits, following a procedure previously described [27].

A total of 38 samples obtained from the same fruits used for the chemical determinations was assessed at harvest. Two melon slices (2 cm) of each fruit sample were cut into 8 pieces of similar size, placed in a

<sup>112</sup> Sapo (n = 7); Calcuta (n = 10); Dulce (n = 6); Irak (n = 10); Songwhan charmi (n = 8); Piel de Sapo T111 (n = 10);

<sup>113 6);</sup> Védrantais (n = 8).

plastic dish labeled with a random number of three digits, and given to each one of the 8 assessors. All of them assessed the same number of samples per session in different presentation orders, following a Williams Latin square design to block first-order and carry-over effects. A non-structured 10 cm linear scale was used for the evaluation of each descriptor, in which 0 meant low intensity and 10 meant high intensity. Mineral water was used as a palate cleanser between samples. The analyses were performed in a test room designed following ISO 8589:2007 [28] and the samples evaluated under white lighting (700  $lux \pm 150 lux$ ).

**Table 3** Sensory attributes and description used for sensory analysis

Attributes	Description
Odor	
Odor intensity	Strength of melon overall odor perceived during chewing.
Ripe fruit	Typical fruity odor in a range from under to overripe.
Fermentative	Presence of chemical or solvent-like odor.
Cucumber	Presence of cucumber characteristic odor.
Flavor	
Flavor intensity	Strength of melon overall flavors perceived during chewing.
Fermentative	Presence of chemical or solvent-like flavor.
Cucumber	Presence of cucumber characteristic flavor.
Astringency	Drying out, roughness aftertaste felt in any mouth surface.
Taste	
Acidity	Amount of acid perceived during chewing.
Sweetness	Amount of sugar perceived during chewing.

# 2.2.3 PTR-MS profiling of VOCs

The frozen flesh of each melon fruit was cut into pieces, immersed in liquid nitrogen, and immediately ground for 15 s at 10,000 rpm using a Grindomix GM 200 (Retsch, Düsseldorf, Germany). Ground samples were stored (-20 °C) and analyzed within 24 h. For each sample, 1.0 g of ground powder was weighted in screw cap glass flasks of 250 mL. Before the analyses, the flasks were equilibrated in a water bath at 25 °C for 30 min. The temperature was selected to match the volatile emission in the headspace of the flasks and the conditions of the consumer perception of the fruits. The flasks were attached to the inlet of the PTR-MS system (Ionicon GmbH, Innsbruck, Austria), and the headspace was extracted at a 60 mL/min flow rate. The temperature of both the inlet and the drift chamber was kept at 60 °C. Mass spectral data in a range between 20 and 160 atomic mass units (amu) was collected with a dwell time of 200 ms. Blank measurements were run between samples to monitor background air, and these values

were subtracted from the sample measurements. All values were corrected for transmission, converted to ppbv according to the procedure described by Lindinger *et al* [24] and considering a reaction rate constant of  $k_R = 2 \times 10^{-9}$  cm<sup>3</sup>/s. All the analyses were carried out in independent triplicates, and the average mass spectra calculated. The masses m/z 32 ( $O_2^+$ ) and m/z 37 (water cluster ion) were removed from the dataset, and mass spectral data (m/z 20-160) of the 67 melon fruits were used for data analysis.

## 2.2.4 PTR-Tof-MS tentative identification of VOCs

Volatile organic compounds tentative identification was performed using a PTR-Tof-MS 8000 system (Ionicon GmbH, Innsbruck, Austria). A representative subset of samples (n=6) was selected considering the variability observed in the PTR-MS results. The procedure was identical as in the previous section, except that only 0.25 g of the ground powder was used. The ionization conditions in the reaction chamber were maintained as follows: drift temperature 60 °C, drift voltage 421 V, and drift pressure 3.80 mbar. The instrument was operated at E/N value of 133 Townsend (1Td = $10^{-17}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>). A further description of PTR-Tof-MS is given elsewhere [23]. The sample measurements lasted 60 s with an acquisition rate of 1 spectrum/s. Baseline removal and spectra alignment by internal calibration of the ToF data were performed according to a procedure previously described [29]. The interfering ions ( $O_2^+$ , NO<sup>+</sup>, and water clusters) and their isotopologues were excluded from the dataset. VOCs were tentatively identified based on the PTR-ToF-MS results and the existing literature.

France).

## 173 2.2.5 Data analysis

The sensory data was evaluated using a one-way Analysis of Variance (ANOVA) on the mean values per melon across panelists, considering the type of melon as a fixed factor. A Tukey's HSD post hoc test ( $p \le$ 0.05) was performed to examine significant statistical differences between the melon types. Due to the lack of normality, the PTR-MS data was evaluated using a non-parametric ANOVA (Kruskal-Wallis test), followed by Dunn's multiple comparison test and Bonferroni correction ( $p \le 0.05$ ). The data of the sensory scores and headspace VOCs measured on the same samples were used to evaluate the relationship between both methods. Principal Component Analysis (PCA) and Pearson's correlation analysis were performed over the 10 sensory attributes and the 40 significantly different VOCs obtained from the ANOVA results. The PCA was performed on the correlation matrix to normalize the different datasets. All the statistical analyses were performed with XLSTAT 2018 software (Addinsoft, Paris,

#### 3. Results and discussion

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3.1 Sensory characteristics

The significant differences observed for the ten sensory attributes among the melon types are shown in Table 4. 'Védrantais' and 'Dulce' cultivars showed higher scores for the intensity, ripe fruit, and fermentative odor attributes, whereas the lower were observed for the 'T111' line of the Piel de Sapo cultivar followed by the 'Calcuta'. The same was observed between the commercial cantalupensis (Cantaloupe and Galia) and inodorus (Amarillo and Piel de Sapo). Other authors observed higher fruity odor for climacteric fruits belonging to the cantalupensis cultivar group but smaller differences between these and non-climacteric inodorus ones [10]. The slight differences between 'Irak', Cantaloupe, and Galia were consistent with previous results for the odor scores of dudaim and cantalupensis fruits [30]. These authors also reported higher odor scores for fruits of both cultivar groups than inodorus. 'Védrantais' and 'Dulce' cultivars were also significantly higher scored for the intensity and fermentative flavor attributes. The lowest scores of these attributes were observed for 'Calcuta' and 'T111' fruits, respectively. The higher score for fermentative flavor of cantalupensis than inodorus fruits was consistent with the results of the odor attributes. The sweeter fruits belonged to the commercial varieties and 'Védrantais' cultivar. These were followed by 'Dulce' and 'T111' cultivars, while 'Calcuta' was the least sweet. No sweetness differences between inodorus and cantalupensis fruits were previously observed [10], although changes may occur depending on the type of cultivar studied [11]. Cantalupensis fruits were observed to be sweeter than inodorus, and both sweeter than dudaim fruits [30]. This was consistent with our results for 'Irak' cultivar. The highest acidity scores were observed for 'Védrantais' and 'Songwhan charmi' cultivars. Except for the lowest scores of Amarillo, no significant differences were observed for the rest of the fruits. Other authors reported minimal [10] or not significant acidity differences [11] between inodorus and cantalupensis cultivars. With the exception of 'Calcuta' and 'Irak' melons, perceived acidity was substantially lower than sweetness. This reflects the predominance of sweet varieties among the fruits analyzed, as several melon types showed a high SSC level together with near-neutral pH values (Table 2, Appendix). The sweet/acid ratio is an important quality index for other fruit species, but the sweet melon varieties lack acid taste, and their eating quality is mainly determined by sweetness [2]. At these levels, the interaction between acid and sweet tastes has a suppressive effect of sweetness over acidity [31]. Small differences were observed for cucumber odor and flavor attributes. 'Songwhan charmi' fruits had higher cucumber odor, followed by Amarillo ones, while the rest of the fruits had lower scores for this

216	attribute. This pattern was reflected in cucumber flavor perception for 'Songwhan charmi' fruits but not
217	for Amarillo ones, possibly due to the high sweetness perception observed for Amarillo melons.
218	Regarding astringency, the highest scores were observed for 'Irak' cultivar, while for commercial Piel de
219	Sapo the least. No differences were observed between the rest of the fruits, neither cultivars nor
220	commercial varieties.
221	These results showed that panelists distinguished cantalupensis and inodorus fruits by their odor and
222	flavor, but also perceived small differences between cantalupensis cultivars and their commercial
223	relatives as well as unique traits of the exotic cultivars. Our results provide information for the quality-
224	oriented programs with an aim to produce more aromatic and flavorful melon cultivars.

**Table 4** Sensory panel scores of the odor, flavor, and taste attributes among melon fruit types: mean values and standard deviation in brackets (n = 38) <sup>a</sup>

			Cultiva	ars (cv. group)				Commercial varie	ties (cv. group)	
		Climac	teric		Non-cli	macteric	Clima	cteric	Non-clir	nacteric
Attributes	'Dulce' b	'Védrantais'	'Irak'	'Calcuta'	'Songwhan charmi'	Piel de Sapo 'T111'	Galia	Cantaloupe	Amarillo	Piel de Sapo
	(cantalupensis)	(cantalupensis)	(dudaim)	(momordica)	(conomon)	(inodorus)	(cantalupensis)	(cantalupensis)	(inodorus)	(inodorus)
Odor										
Odor intensity	$7.95 (0.33)^{ab}$	9.03 (0.06) <sup>a</sup>	$6.00 (0.36)^{c}$	4.71 (0.62) <sup>cd</sup>	5.29 (0.67) <sup>c</sup>	$3.31 (0.83)^d$	$6.36 (1.02)^{bc}$	6.61 (1.05) <sup>bc</sup>	$3.79 (0.06)^d$	$3.95 (0.59)^d$
Ripe fruit	7.80 (0.38) <sup>ab</sup>	8.91 (0.36) <sup>a</sup>	5.11 (0.88) <sup>cde</sup>	$3.76 (0.88)^{e}$	4.12 (0.66) <sup>de</sup>	3.33 (0.32) <sup>e</sup>	6.20 (1.38) <sup>bc</sup>	5.95 (0.90) <sup>bcd</sup>	$3.83 (0.38)^{e}$	3.45 (0.74) <sup>e</sup>
Fermentative	4.23 (0.60) <sup>ab</sup>	$6.50 (0.08)^a$	2.38 (0.19) <sup>bc</sup>	1.35 (0.78) <sup>cd</sup>	1.70 (0.65) <sup>cd</sup>	$0.12 (0.07)^{d}$	$3.85 (1.61)^{b}$	3.46 (1.21) <sup>bc</sup>	$0.39 (0.06)^{cd}$	$0.11 (0.14)^{d}$
Cucumber	$0.60 (0.44)^{b}$	$0.02 (0.10)^{b}$	$0.73 (0.59)^{b}$	$0.74 (0.62)^{b}$	3.10 (0.62) <sup>a</sup>	$0.78 (0.50)^{b}$	$0.41 (0.28)^{b}$	$0.51 (0.41)^{b}$	1.53 (0.72) <sup>ab</sup>	$0.89 (0.24)^{b}$
Flavor										
Flavor intensity	6.81 (0.55) <sup>ab</sup>	7.94 (0.39) <sup>a</sup>	5.28 (0.56) <sup>cd</sup>	$3.86 (0.34)^d$	5.97 (0.41) <sup>bc</sup>	5.58 (0.29) <sup>bcd</sup>	6.86 (0.58) <sup>ab</sup>	7.11 (0.31) <sup>ab</sup>	$6.03 (0.25)^{bc}$	6.63 (0.49) <sup>ab</sup>
Fermentative	$3.41 (0.54)^{b}$	5.94 (0.39) <sup>a</sup>	2.46 (0.62) <sup>bc</sup>	$0.99 (1.07)^{cd}$	1.08 (0.63) <sup>cd</sup>	$0.20 (0.38)^d$	2.67 (0.39) <sup>b</sup>	3.23 (0.91) <sup>b</sup>	0.89 (0.11) <sup>cd</sup>	$0.22~(0.27)^d$
Cucumber	$0.59 (0.43)^{b}$	$0.17 (0.31)^{b}$	$1.53 (0.82)^{b}$	1.38 (1.17) <sup>b</sup>	4.05 (0.77) <sup>a</sup>	$0.80 (0.04)^{b}$	$0.59 (0.42)^{b}$	0.73 (0.64) <sup>b</sup>	$0.49 (0.07)^{b}$	$0.31 (0.18)^{b}$
Astringency	1.37 (0.26) <sup>ab</sup>	1.43 (0.55) <sup>ab</sup>	2.27 (0.63) <sup>a</sup>	1.84 (0.11) <sup>ab</sup>	1.65 (0.50) <sup>ab</sup>	$0.99 (0.06)^{ab}$	1.22 (0.37) <sup>ab</sup>	1.29 (0.35) <sup>ab</sup>	$0.87 (0.19)^{ab}$	$0.77(0.39)^{b}$
Taste										
Acidity	1.10 (0.22) <sup>ab</sup>	1.53 (0.03) <sup>a</sup>	1.19 (0.29) <sup>ab</sup>	$1.22 (0.61)^{ab}$	1.46 (0.32) <sup>a</sup>	$0.70 (0.24)^{ab}$	1.16 (0.31) <sup>ab</sup>	1.23 (0.23) <sup>ab</sup>	$0.46 (0.05)^{b}$	$0.67 (0.23)^{ab}$
Sweetness	4.26 (0.87) <sup>ab</sup>	5.22 (0.57) <sup>a</sup>	1.15 (0.50) <sup>cd</sup>	$0.55 (0.12)^d$	2.56 (0.72) <sup>bc</sup>	4.17 (0.15) <sup>ab</sup>	5.77 (0.90) <sup>a</sup>	5.67 (0.33) <sup>a</sup>	5.48 (0.63) <sup>a</sup>	5.90 (0.68) <sup>a</sup>

<sup>&</sup>lt;sup>a</sup> Values with different letters in the same row indicate significant differences by Tukey's HSD post hoc test (p ≤ 0.05).

<sup>b</sup> Number of samples of each melon type: 'Dulce' (n = 4), 'Védrantais' (n = 3), 'Irak' (n = 4), 'Calcuta' (n = 3), 'Songwhan charmi' (n = 6), Piel de Sapo 'T111' (n = 2), Galia (n = 4), Cantaloupe (n = 3), Amarillo (n = 2), Piel de Sapo (n = 7).

## 3.3 VOCs profile

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226 The VOCs profile of melon fruit is influenced by cultivar, maturity stage, harvest conditions, or storage. 227 Moreover, as the pathways involved in the formation of specific compounds (such as esters) are known to 228 depend on the production of ethylene, the climacteric and non-climacteric melon fruits exhibit different 229 volatile profiles. Among the key volatile compounds reported in melon,  $C_4 - C_9$  esters have the highest 230 impact over the aroma of climacteric fruits, considered very aromatic, whereas C6 and C9 alcohols and 231 aldehydes have the highest impact over the aroma of non-climacteric fruits, generally considered as less 232 aromatic [4-8]. 233 In the present study, significant differences were observed for 40 compounds among the ten melon fruit 234 types (Table 5). The VOC profile consisted of 9 alcohols (including 3 alcohol fragments at m/z 29.037, 235 71.058 and 85.099), 9 aldehydes, 1 compound at m/z 143.143 tentatively identified as an alcohol/ 236 aldehyde (Nonanal/ nonenol), 8 esters (including 1 ester fragment at m/z 67.054), 4 terpenes (including a 237 monoterpene fragment at m/z 95.085 and a farnesene fragment at m/z 123.117), and 4 other volatiles or 238 related compounds (acetone, acetic acid, and a nitrile compound at m/z 42.034). Some fragments of 239 several possible origins (alcohols, aldehydes, esters, terpenes) were also observed for m/z 41.038, 43.018/ 240 43.053, 55.054, 57.069, 81.070, and 83.086. The VOC profiles obtained for the different melon types 241 showed that, among the climacteric fruits, the ones belonging to the cantalupensis cultivar group 242 ('Védrantais', 'Dulce', Galia, and Cantaloupe) had a higher concentration of alcohols, aldehydes, and 243 esters. The opposite was observed for the non-climacteric fruits, especially those belonging to the 244 inodorus cultivar group (Piel de Sapo 'T111', Amarillo and Piel de Sapo), which are reported to have a 245 lower volatile concentration. Regarding the exotic cultivars, the fruits of the dudaim cultivar ('Irak') 246 showed several similarities with the VOC profile of the other cantalupensis fruits. In contrast, fruits of the 247 momordica ('Calcuta') and conomon ('Songwhan charmi') cultivars showed an intermediate behavior 248 between cantalupensis and inodorus. This was in agreement with previous works reporting similarities 249 between the VOC profile of several dudaim, conomon, and momordica fruits with either cantalupensis or 250 inodorus regardless of their climacteric or non-climacteric classification [32].

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Alcohols

The abundance of alcohols was significantly higher for 'Védrantais', mostly followed by 'Dulce' and 'Irak' cultivars, and lower for 'Calcuta', 'Songwhan charmi', and 'T111'. A similar alcohol profile was observed for the commercial *cantalupensis* (Cantaloupe and Galia) and the 'Irak' cultivar. Methanol was

the major alcohol observed for all the melon types followed by ethanol. It is a marker of pectin degradation involved in the regulation of ethanol production during ripening [33]. Ethanol is produced by the reduction of acetaldehyde, and the changes in the concentration of both compounds occur in a related pattern [16]. The ratio methanol/ethanol in melon fruit differs between cultivars, ripening stage, and processing [18, 33].

## Aldehydes

Acetaldehyde was the major aldehyde for all the melon types. It was present at significantly higher concentrations in the headspace of the 'Védrantais' cultivar, while lower concentrations were observed for the 'Calcuta' cultivar and commercial *inodorus*. Other authors reported prominent levels of acetaldehyde among melon fruits [16, 34]. Hexenal was determined at significantly higher concentrations for 'Irak' and 'Calcuta' cultivars in comparison to 'Songwhan charmi' and 'T111' cultivars. The lower concentrations of hexenal observed for *inodorus* fruits are consistent with previous works [22, 35]. Heptanal was significantly higher for 'Védrantais' cultivar along with commercial *cantalupensis* and 'Songwhan charmi', while lower for *inodorus* fruits. Nonenal was significantly higher for 'T111' cultivar along with the commercial *inodorus* fruits. 2,6-nonadienal was significantly higher for 'Irak' cultivar and Amarillo. Lower concentrations of both C<sub>9</sub> aldehydes were observed among the commercial *cantalupensis* fruits. Higher concentrations of nonenal and 2,6-nonadienal among *inodorus* than *cantalupensis* were previously reported [11, 36].

## Esters

The higher headspace concentrations of esters were observed for the 'Védrantais' cultivar, followed by 'Dulce' and the commercial *cantalupensis*. Intermediate concentrations were observed for 'Irak' and 'Songwhan charmi', whereas the ester pattern of 'Calcuta' and 'T111' cultivars was more similar to the one of commercial *inodorus*. Within the latter, Piel de Sapo had the lowest ester concentration and showed small differences when compared to the 'T111' cultivar. The  $C_3$  and  $C_4$  esters at m/z 75.044 and 89.059 were within the most abundant ester related masses, while lower concentrations were observed for  $C_5$ — $C_9$  esters. The differential ester profile of *cantalupensis* and *inodorus* fruits is well documented in the literature [11, 22, 32, 36].

285 Terpenes

Isoprene and a monoterpene fragment at m/z 95.085 were responsible for the higher terpene concentration determined for 'Védrantais', 'Songwhan charmi' and 'Irak' cultivars. The lower terpene concentrations were observed for 'Calcuta' cultivar. A farnesene fragment at m/z 123.117 was present at significantly higher concentrations for 'T111' cultivar, along with the commercial *inodorus* fruits.

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Other compounds

Acetic acid and a nitrile compound (m/z 42.034) were present at significantly higher concentrations in the headspace of 'Védrantais' cultivar and lower for 'Calcuta' and commercial *inodorus*. Acetone was determined at a significantly higher concentration for 'Irak' in comparison to 'T111' cultivar and Amarillo fruits. No significant differences were observed for the rest of the fruits.

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Globally, volatile emission was more pronounced for cantalupensis fruits. 'Védrantais' cultivar was different from the other cantalupensis fruits due to the higher concentrations of most of the VOCs, while the VOC profile of 'Dulce' cultivar was more related to the commercial cantalupensis fruits, Galia and Cantaloupe. The significantly higher concentrations of two C<sub>9</sub> aldehydes, 2,6-nonadienal and nonenal, the C<sub>5</sub>, C<sub>6</sub>, and C<sub>8</sub> esters, and limonene, found for 'Dulce' cultivar, or the higher concentrations of a fragment at m/z 57.069 and methyl butanal found for Galia and Cantaloupe, were on the basis of the differences between the VOC profiles of these melon types. The lower volatile emission was observed for *inodorus* fruits, although some differences were detected between 'T111' cultivar and the commercial inodorus fruits, Amarillo and Piel de Sapo. The 'T111' and Amarillo fruits showed significantly higher concentrations of methanol, 1,2-propanediol, acetic acid, isoprene, methyl acetate, and  $C_5 - C_7$  esters than Piel de Sapo. In contrast, higher concentrations of acetaldehyde, a nitrile compound, 1,2-ethanediol, and ethyl acetate/ methyl propanoate were found for 'T111' than the other inodorus fruits. Regarding the exotic cultivars, the VOCs pattern of the 'Irak' cultivar was more similar to that of 'Dulce' and the commercial cantalupensis fruits for several alcohols, aldehydes, esters, and terpenes, except for methanol, butanal, hexenal, 2,6-nonadienal, C<sub>4</sub> - C<sub>5</sub> esters, isoprene, and acetone. The VOC profile of 'Calcuta' and 'Songwhan charmi' cultivars showed intermediate profiles between the ones of cantalupensis and inodorus fruits. Additionally, both exotic cultivars had significantly lower concentrations of methanol, whereas 'Calcuta' showed a higher concentration of 3-hexenol and hexenal, two C<sub>6</sub> green leaf volatiles.

- 315 These results are consistent with the intermediate ripening expression between the climacteric and the
- 316 non-climacteric pattern previously observed for several exotic melon cultivars [3, 32].

**Table 5** Tentative identification by PTR-Tof-MS (three left-side columns) and concentration (ppbv) of the significantly different VOCs among melon fruit types (N = 67) by deviations in brackets <sup>a</sup>

Mass $(m/z)$	Sum			Climac	cteric		Non-cl	imacteric
Tentative identification <sup>b</sup>	formula	Reference	'Dulce' c' (cantalupensis)	'Védrantais' (cantalupensis)			'Songwhan charmi' (conomon)	Piel de Sapo 'T111' (inodorus)
29.037	$C_2H_5^+$		16.2	217.4	10.1	(momordica)  3.3	4.8	9.2
Fragment (alcohol)	_ ,		$(7.0)^{ab}$	$(112.8)^{a}$	$(9.3)^{ab}$	$(2.7)^{b}$	$(4.8)^{b}$	$(13.6)^{b}$
31.017	$CH_3O^+$	[37]	17.5	41.1	10.1	6.4	5.1	5.7
Formaldehyde			$(5.3)^{ab}$	$(9.0)^{a}$	$(4.3)^{ab}$	$(2.3)^{b}$	$(2.6)^{b}$	$(5.4)^{b}$
33.034	CH <sub>5</sub> O <sup>+</sup>	[18, 33]	5385	6106.8	1343.7	849.1	564.8	1074.4
Methanol			$(2068.8)^{ab}$	$(1245.3)^{a}$	$(579.3)^{abc}$	$(357.1)^{c}$	$(329.2)^{c}$	$(613.4)^{abc}$
41.038	$C_{3}H_{5}^{+}$	[37, 38]	72.1	127.8	53.8	33.4	23.4	17
Fragment (alcohol, ester)			$(36.0)^{abc}$	$(30.6)^{a}$	$(22.9)^{abc}$	$(12.2)^{abc}$	$(11.5)^{bc}$	$(12.2)^{c}$
42.034	$C_2H_4N^+$		16.9	36.3	11.7	5.2	14.5	6.5
Acetonitrile, nitrile fragment			$(15.6)^{ab}$	$(34.2)^{a}$	$(7.3)^{ab}$	$(3.4)^{b}$	$(24.2)^{ab}$	$(6.9)^{ab}$
43.018	$C_2H_3O^+$	[37, 38]	416	1949.9	209.1	116	302.7	242.3
Fragment (ester)			$(295.5)^{ab}$	$(1252.4)^{a}$	$(143.3)^{ab}$	$(49.1)^{b}$	$(235.4)^{ab}$	$(352.2)^{b}$
43.053	$C_3H_7^+$	[37, 38]						
Fragment (alcohol, ester, acetate)								
45.033	$C_2H_5O^+$	[4, 34]	3961.2	12084.9	4163.5	1511.3	5968.6	3794
Acetaldehyde			$(1184.9)^{ab}$	$(4842.8)^{a}$	$(4136.2)^{ab}$	$(1055.5)^{b}$	$(3827.4)^{ab}$	$(1676.4)^{ab}$
47.049	$C_2H_7O^+$	[4, 34]	329.7	3737.5	233.5	114.5	146.9	209.3
Ethanol			$(117.6)^{ab}$	$(187.3)^{a}$	$(164.2)^{ab}$	$(62.3)^{b}$	$(91.2)^{b}$	$(241.4)^{b}$
55.054	$C_4H_7^+$	[37]	28.5	19.7	38.4	29.6	13.3	6.5
Fragment			$(14.5)^{ab}$	$(7.2)^{ab}$	$(20.9)^{a}$	$(9.1)^{a}$	$(4.7)^{ab}$	$(1.2)^{b}$
57.069	$C_4H_9^+$	[37, 38]	76.3	98.6	35.6	23.8	21	11.3
Fragment (alcohol, ester)			$(43.2)^{ab}$	$(47.3)^{a}$	$(16.1)^{abc}$	$(7.2)^{abc}$	$(12.8)^{abc}$	$(12.5)^{bc}$
59.049	$C_3H_7O^+$	[4, 17]	19.3	26.2	32	25.4	26.6	14.8
Acetone			$(3.0)^{ab}$	$(6.7)^{ab}$	$(8.7)^{a}$	$(11.4)^{ab}$	$(8.3)^{ab}$	$(6.1)^{b}$
61.028	$C_2H_5O_2^{+}$	[17, 35]	341.3	1539.3	114.3	72.7	257.3	211.9
Acetic acid			$(287.2)^{ab}$	$(1324.8)^{a}$	$(84.9)^{abc}$	$(47.1)^{bc}$	$(230.9)^{ab}$	$(366.4)^{abc}$
63.044	$C_2H_7O_2^{^+}$		7.8	24.9	7.2	3.8	10	7
1,2-Ethanediol			$(2.1)^{ab}$	$(11.7)^{a}$	$(5.7)^{ab}$	$(1.2)^{b}$	$(5.8)^{ab}$	$(4.2)^{ab}$
67.054	$C_5H_7^+$	[37]	1	0.8	1.9	0.8	1.3	2.3
Fragment (ester)			$(0.5)^{ab}$	$(0.4)^{ab}$	$(0.7)^{a}$	$(0.3)^{ab}$	$(0.5)^{ab}$	$(0.7)^a$

Fragment (terpene, sesquiterpe	ne, aldehyde)		$(7.9)^{ab}$	$(0.9)^{b}$	$(26.4)^{a}$	$(19.7)^{a}$	$(8.6)^{b}$	(8.6) <sup>ab</sup>	(1
83.086	$C_6H_{11}^{+}$	[37]	12.2	6.4	18.1	13.7	2.8	1.4	3
Fragment (alcohol, aldehyde, se	esquiterpene)		$(7.9)^{ab}$	$(2.3)^{abc}$	$(11.6)^{a}$	$(4.7)^{a}$	$(3.1)^{bc}$	$(3.1)^{bc}$	(
85.099	$C_6H_{13}^{+}$	[37]	1.9	4.9	3.7	3.1	0.9	0.6	3
Fragment (alcohol)			$(1.3)^{ab}$	$(1.5)^{a}$	$(1.2)^{ab}$	$(1.7)^{ab}$	$(0.7)^{b}$	$(0.2)^{b}$	(2
87.08	$C_5H_{11}O^+$	[4, 34]	0.5	2.9	0.7	0.6	0.6	0.5	1
Methyl butanal			$(0.2)^{b}$	$(1.3)^{a}$	$(0.2)^{ab}$	$(0.2)^{b}$	$(0.3)^{b}$	$(0.3)^{b}$	((
89.059	$C_4 H_9 O_2^{\ +}$	[4, 34]	63.5	354.1	16	7.3	33.3	30	8
Ethyl acetate/ Methyl propanoa	te		$(53.4)^{ab}$	$(228.6)^{a}$	$(16.1)^{abc}$	$(6.5)^{bc}$	$(31.3)^{abc}$	$(52.1)^{abc}$	(
91.074	$C_4H_{11}O_2^{\ +}$		6.6	5.6	1.4	2.6	0.9	0.4	2
2,3-Butanediol			$(5.4)^{ab}$	$(2.5)^{a}$	$(0.7)^{abc}$	$(1.3)^{ab}$	$(0.5)^{bc}$	$(0.4)^{bc}$	(
95.085	$C_7 H_{11}^{+}$	[37]	0.6	0.5	1.2	0.6	1.8	1	0
Fragment (monoterpene)			$(0.2)^{abc}$	$(0.2)^{bc}$	$(0.4)^{ab}$	$(0.2)^{bc}$	$(0.9)^{a}$	$(0.3)^{abc}$	((
99.08	$C_6H_{11}O^{\scriptscriptstyle +}$	[34, 35]	1.3	0.5	10.4	10	0.9	0.3	0
Hexenal			$(1.4)^{ab}$	$(0.2)^{ab}$	$(5.1)^{a}$	$(3.9)^{a}$	$(1.9)^{b}$	$(0.3)^{b}$	((
101.095	$C_6H_{13}O^{\scriptscriptstyle +}$	[34, 36]	0.3	0.4	1	0.8	0.2	0.1	0
3-Hexenol			$(0.2)^{ab}$	$(0.1)^{ab}$	$(0.7)^{a}$	$(0.3)^{a}$	$(0.2)^{b}$	$(0.1)^{b}$	((
103.075	$C_5H_{11}O_2^{\ +}$	[34, 36]	14.5	13.1	1.6	1.1	0.5	1.5	1
Ester (Ethyl propanoate, Isopro	pyl acetate, Methyl bu	tanoate,	$(10.6)^{a}$	$(9.2)^{a}$	$(1.4)^{abc}$	$(1.0)^{abc}$	$(0.4)^{bc}$	$(2.6)^{abc}$	(9
Methyl isobutyrate, Propyl acet	ate)								
115.111	$C_7H_{15}O^+$	[34-36]	0.3	2.1	0.2	0.2	0.4	0.1	0
Heptanal			$(0.2)^{abc}$	$(2.8)^{a}$	$(0.1)^{abc}$	$(0.1)^{abc}$	$(0.4)^{ab}$	$(0.2)^{bc}$	((
117.091	$C_6H_{13}O_2^{\ +}$	[4, 36]	18.3	18.1	1.4	1.2	0.9	1	1
Ester (Butyl acetate, Ethyl butan	noate, Isobutyl acetate	,	$(19.0)^{ab}$	$(14.7)^{a}$	$(0.9)^{abcd}$	$(0.9)^{abcd}$	$(0.9)^{\text{bcd}}$	$(0.9)^{cd}$	(9
Methyl n-methylbutanoate)									
123.117	$C_9H_{15}^{\ +}$	[37]	0.9	0.5	1.1	0.4	1.4	2.8	0
Fragment (farnesene)			$(0.6)^{ab}$	$(0.4)^{ab}$	$(0.9)^{ab}$	$(0.2)^{b}$	$(0.6)^{ab}$	$(0.8)^{a}$	((
131.107	$C_7H_{15}O_2^{\ +}$	[34, 36]	2.5	13.4	0.7	0.2	0.3	0.6	3
Ester (Ethyl methylbutanoate, E	Ethyl pentanoate, Meth	yl hexanoate,	$(1.9)^{ab}$	$(9.1)^{a}$	$(0.7)^{ab}$	$(0.2)^{bc}$	$(0.3)^{bc}$	$(0.3)^{bc}$	(3
Pentyl cetate)									
137.132	$C_{10}H_{17}^{+}$	[11, 34]	0.9	0.4	0.2	0.3	0.1	0.1	0
Limonene			$(0.5)^{a}$	$(0.2)^{a}$	$(0.1)^{abc}$	$(0.1)^{ab}$	$(0.04)^{bc}$	$(0.04)^{c}$	((
139.112	$C_9H_{15}O^+$	[34, 36]	0.4	0.2	1	0.5	0.5	0.8	0
2,6-Nonadienal			$(0.3)^{ab}$	$(0.1)^{b}$	$(0.4)^{a}$	$(0.3)^{ab}$	$(0.4)^{ab}$	$(0.3)^{ab}$	((
141.128	$C_9H_{17}O^+$	[34, 36]	0.5	0.4	0.6	0.3	0.8	1.2	0
Nonenal			$(0.4)^{ab}$	$(0.2)^{ab}$	$(0.5)^{ab}$	$(0.1)^{b}$	$(0.4)^{ab}$	$(0.4)^{a}$	(

3.4 Correlation between sensory and PTR-MS analyses

3.4.1 Principal component analysis

A PCA was performed on the sensory scores and headspace VOCs measured on the same samples, to which the SSC and pH were added as supplementary variables (Fig. 1). The first three principal components (PCs) explained 68% of the variance (46%, 13%, and 9%, respectively). Three main groups can be observed, in the clockwise direction from the third to the fourth quadrant. One of *inodorus* fruits, formed by the 'T111' cultivar along with Amarillo and Piel de Sapo commercial fruits, a second one formed by the exotic cultivars ('Calcuta', 'Irak' and 'Songwhan charmi'), and another of cantalupensis fruits, formed by the 'Védrantais' and 'Dulce' cultivars along with Cantaloupe and Galia commercial fruits. The higher positive loadings of the majority of VOCs and odor intensity, ripe fruit odor, and fermentative odor and flavor attributes contributed to the opposed projection of inodorus and cantalupensis fruits along with the PC 1. 'Védrantais' exhibited a clear differentiation, not only from the rest of the melon types but also from the other cantalupensis fruits. The separation of 'Calcuta' and 'Irak' from the other melon fruits was mainly due to the high positive loadings of the fragments at m/z 81.070 and 83.086, hexenal, and astringency, together with high negative loadings of nonenal, flavor intensity and sweetness. 'Songwhan charmi' melons had intermediate characteristics between inodorus, exotic, and cantalupensis. Cantaloupe, 'Dulce' and Galia were further separated from the rest of the fruits along with PC 3 due to the high positive loadings of an ester (m/z 67.054) and monoterpene (m/z 95.085) fragments, as well as high negative loadings of cucumber odor and flavor attributes.

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3.4.2 Pearson's correlation analysis

The significant correlations found between sensory attributes and VOCs are shown in Table 6. Most of the volatiles showed an impact over odor intensity, ripe fruit odor, and fermentative odor attributes. The same was observed for fermentative flavor but to a lower extent for flavor intensity. Fewer correlations were observed for the attributes of cucumber odor and flavor, sweetness, acidity, and astringency.

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Alcohols

A positive contribution of alcohols to the attributes of odor  $(0.48^{**} \le r \le 0.76^{***})$  and flavor intensities  $(0.36* \le r \le 0.59***)$ , ripe fruit odor  $(0.42** \le r \le 0.80***)$ , and fermentative odor and flavor  $(0.42** \le r \le 0.80***)$ 347  $\leq 0.82^{***}$ ) was observed. Total alcohols were reported to be positively correlated with the overall flavor 348 [18], but that correlation was observed to change during storage for several flavor attributes [19]. Methanol and ethanol were reported to be associated with advanced ripening stage [18], whereas the diol alcohols with ester production in melon fruit [17]. Methanol was also negatively correlated with cucumber odor (r = -0.51\*\*\*) and flavor (r = -0.40\*).

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Aldehydes

Several aldehydes were positively correlated with odor  $(0.54** \le r \le 0.76***)$  and flavor intensity  $(0.43^{**} \le r \le 0.56^{***})$ , ripe fruit  $(0.49^{***} \le r \le 0.76^{***})$  and fermentative odor and flavor attributes  $(0.58*** \le r \le 0.85***)$ . Most of these aldehydes (acetaldehyde, hexenal, heptanal, and decanal) were associated with a lack of maturity [34], although acetaldehyde was also observed to increase with maturity [18]. Acetaldehyde is particularly important as it increases fruit flavor and contributes to the perception of freshness [39]. Positive contributions of acetaldehyde to the flavor perception of citrus [40], kiwi [41], or tomato fruits [42] have also been observed. Verzera et al [22] reported strong correlations between the typical odor and flavor descriptors of melon and several aldehydes, including methyl butanal, 2,6-nonadienal and nonenal. The latter two are associated with green or cucumber notes and considered key volatiles in the typical aroma of the non-climacteric fruits belonging to the *inodorus* cultivar [11]. In the present study, no significant correlations were observed between both C<sub>9</sub> aldehydes and cucumber odor or flavor, possibly due to the predominance of climacteric cultivars among the fruits analyzed. However, negative correlations were found between these compounds and odor intensity (r = -0.53\*\*; – 0.50\*\*), ripe fruit odor (r = -0.54\*\*\*; -0.47\*), and fermentative odor (r = -0.62\*\*\*; -0.52\*\*\*) or flavor (r = -0.57\*\*\*; -0.45\*\*) attributes. Previous authors observed high negative correlations between 2,6nonadienal or nonenal with 'fruity', 'sweet-aromatic', and 'chemical' flavor attributes, but also high positive correlations with 'cucurbit' attribute [19].

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Esters

The correlations found with intensity  $(0.48^{**} \le r \le 0.69^{***})$ , ripe fruit  $(0.46^{**} \le r \le 0.69^{***})$  or fermentative odor  $(0.53^{***} \le r \le 0.73^{***})$  and flavor  $(0.56^{***} \le r \le 0.70^{***})$  are in agreement with previous authors reporting good correlations  $(r \ge 0.61; p < 0.05)$  between ethyl, methyl or acetate esters and melon sensory flavor [18]. High correlations  $(r \ge 0.76)$  between  $C_7 - C_9$  esters and the fruity odor [20] or between  $C_5 - C_7$  esters and fruity, pineapple-like, and sweet aromas were also observed [21]. Additionally, several works pointed out the importance of sulfur-containing esters to the odor and flavor of melon fruits [11, 20, 21], but these were not detected in the present work, possibly due to differences in

the analytical methodology [34, 43]. Esters are particularly related to the fruity notes of climacteric cultivars, but their odor active values and, thus, their contribution to aroma was reported to be substantially lower than that of aldehydes and alcohols [44].

- 384 Terpenes
  - Two terpene related masses, monoterpene (95.085) and farnesene (123.117) fragments, were negatively correlated with odor intensity, ripe fruit or fermentative odor and flavor. The former was positively correlated with cucumber odor (r = 0.75\*\*\*) and flavor (r = 0.72\*\*\*). On the other hand, isoprene and limonene showed positive correlations with odor intensity (r = 0.50\*\*\*; 0.47\*\*), ripe fruit (r = 0.44\*\*; 0.47\*\*) and fermentative odor (r = 0.56\*\*\*; 0.41\*) or flavor (r = 0.50\*\*\*; 0.37\*). Limonene was

- 392 Other compounds
- A nitrile compound at m/z 42.034 and acetic acid were correlated with intensity, ripe fruit, and fermentative odor, as well as fermentative flavor attributes. Acetone was slightly correlated with cucumber odor (r = 0.39\*) and flavor (r = 0.46\*\*). Acetone is associated with solvent or ethereal descriptors, but its aromatic character was reported to change from 'glue/ alcohol' in deionized water, to 'sweet' in ethanol-methanol-water solution, or 'green' in deodorized tomato homogenate [45].

399 Effect of SSC, pH, and volatiles over sweetness, acidity and astringency attributes

observed to contribute for the odor and flavor of melon [20].

The determinations of SSC and pH were satisfactorily correlated with sweetness (r = 0.67\*\*\*; 0.70\*\*\*). Both parameters were also correlated with flavor intensity (r = 0.47\*\*; 0.58\*\*\*), although this could be due to an indirect effect of the high correlation between sweetness and flavor intensity ( $r = 0.77; p \le 0.001$ ). A slight negative correlation was found between SSC and astringency (r = -0.37\*). This was consistent with the negative correlation between sweetness and astringency attributes ( $r = -0.57; p \le 0.001$ ). SSC has a significant positive effect on the sweet and fruity descriptors, as well as a significant negative effect on the green, bitter and astringent descriptors, among fruits, beverages, and flavors [12, 46]. A similar pattern was observed for 3-hexenol and hexenal, two  $C_6$  green leaf volatiles, which were negatively correlated with sweetness (r = -0.50\*\*\*; -0.67\*\*\*) and positively with astringency (r = 0.50\*\*; 0.41\*\*). The correlation between hexenal and sweetness was reflected over flavor intensity (r = -0.66\*\*\*), but for 3-hexenol positive correlations with intensity and ripe fruit odor or fermentative odor

and flavor attributes  $(0.42^{**} \le r \le 0.48^{**})$  were observed. The ortho- and retronasal perception of green leaf volatiles was observed to change from 'green' to 'fruity' descriptors due to the interaction of these compounds with sugars and acids [13], but the nature of these interactions can vary with the fruit species. Aprea et al [14] observed a negative contribution of 3-hexenol to the sweet perception of apple, whereas Klee and Tieman [47] reported a positive contribution to the 'overall flavor intensity' and 'liking' of tomato. Other authors observed the negative contribution of hexenal to the 'overall flavor intensity' and 'liking' of strawberries and blueberries [47], as well as to the sweetness of table grapes [48]. Besides, the interactions between certain VOCs with sugars and acids affect the rate of release and persistence of these volatile compounds in the mouth and, thus, the perceived intensities of aroma and flavor attributes [15]. Moreover, a positive correlation between astringency and acidity was also observed (r = 0.45;  $p \le 0.01$ ). This opposed relationship of astringency with sweetness and acidity was observed in other fruits like strawberries [49], apples [50], or kiwifruits [51]. Regarding VOCs, the highest correlation of acidity was with isoprene (r = 0.54\*\*\*), a leaf volatile in the origin of several terpene compounds. Minor correlations were also observed with 1,2-ethanediol (r = 0.41\*\*), several aldehydes ( $0.39* \le r \le 0.42**$ ) or acetic acid (r = 0.37\*). The interaction between acetaldehyde and sugars or acids is known to enhance the 'fruity' and 'tropical flavor' attributes of tomato fruits [42], although in the present study it was only correlated with acidity. pH was positively correlated with SSC (r = 0.56\*\*\*), whereas no significant correlation was observed between pH and acidity. The relationship between pH increase and sugar accumulation was previously observed [1, 2], and both processes are classified as ethylene-independent [5-7]. The melon genotypes with higher sugar levels have pH values closer to the neutral range, whereas the ones with low sugar levels show a broader range of pH values [1]. The characterization of the pH gene, with a major impact on fruit acidity, has contributed to explain the low level of acidity of sweet melon types [52]. Additionally, pH was strongly correlated with the majority of the VOCs, as it is a parameter involved in the regulation of several reactions of volatile production [16].

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 Table 6 Pearson correlation coefficients between VOCs, sensory attributes, SSC and pH determinations a

m/z VOCs	NOC		Odor a	attributes <sup>b</sup>		Flavor attributes <sup>c</sup>				Taste attributes <sup>d</sup>	
m/z	VOCs	INT	RPF	FER	CMB	INT	FER	CMB	AST	SWT	ACD
29.037	Fragment (alcohol)	0.50***	0.51***	0.56***		0.36*	0.66***				
31.017	Formaldehyde	0.76***	0.76***	0.80***	-0.44**	0.52***	0.85***				0.39*
33.034	Methanol	0.76***	0.80***	0.82***	-0.51***	0.59***	0.78***	-0.40*		0.37*	
41.038	Fragment (alcohol, ester)	0.79***	0.81***	0.84***	-0.41**	0.52***	0.73***				0.36*
42.034	Acetonitrile, nitrile fragment	0.50**	0.49**	0.51***			0.48**				
43.018/ 43.053	Fragment (ester)/ Fragment (alcohol, ester, acetate)	0.61***	0.57***	0.66***		0.44**	0.69***				0.37*
45.033	Acetaldehyde	0.56***	0.49***	0.59***		0.56***	0.60***				0.42**
47.049	Ethanol	0.51**	0.51**	0.57***		0.36*	0.66***				
55.054	Fragment	0.56***	0.48**	0.49***			0.43**		0.47**	-0.48**	0.39*
57.069	Fragment (alcohol, ester)	0.75***	0.76***	0.81***	-0.35*	0.53***	0.69***				0.34*
59.049	Acetone				0.39*			0.46**			
61.028	Acetic acid	0.59***	0.55***	0.64***		0.41**	0.67***				0.37*
63.044	1,2-Ethanediol	0.60***	0.54***	0.65***		0.52***	0.67***				0.41**
67.054	Fragment (ester)	-0.52***	-0.50**	-0.58***			-0.49**				
69.070	Isoprene	0.50***	0.44**	0.56***			0.50***		0.38*		0.54***
71.085	Fragment (alcohol)	0.48**	0.48**	0.52***		0.41**	0.52***				
73.064	Butanal	0.63***	0.60***	0.69***		0.53***	0.66***				0.42**
75.044	Methyl acetate	0.69***	0.67***	0.73***	-0.38*	0.50***	0.70***				
77.059	1,2-Propanediol	0.70***	0.66***	0.74***	-0.35*	0.45**	0.71***				
81.070	Fragment (terpene, sesquiterpene, aldehyde)					-0.70***			0.42**	-0.68***	
83.086	Fragment (alcohol, aldehyde, sesquiterpene)								0.38*	-0.51***	
85.099	Fragment (alcohol)	0.61***	0.59***	0.64***	-0.34*		0.54***		0.35*		
87.080	Methyl butanal	0.59***	0.60***	0.69***		0.43**	0.58***				
89.059	Ethyl acetate/ Methyl propanoate	0.58***	0.56***	0.64***		0.43**	0.67***				0.36*
91.074	2,3-Butanediol	0.65***	0.63***	0.62***			0.59***				
95.085	Fragment (monoterpene)		-0.40*	-0.37*	0.75***		-0.44**	0.72***		-0.39**	
99.080	Hexenal					-0.66***			0.41**	-0.67***	
101.095	3-Hexenol	0.48**	0.42**	0.48**	-0.34*		0.42**		0.50***	-0.50***	
103.075	Ester (Ethyl propanoate, Isopropyl acetate, Methyl butanoate, Methyl isobutyrate, Propyl acetate)	0.68***	0.68***	0.70***	-0.34*	0.48**	0.60***				
115.111	Heptanal Fd 11 to 1 to 1	0.59***	0.52***	0.62***		0.41**	0.60***				0.41*

SSC			0.47**	-0.35* -0.37*	0.67*** -0.45**
pH	0.35*	-0.36*	0.58*** 0.42**	-0.42**	0,70***

<sup>&</sup>lt;sup>a</sup> Significance: \*\*\* for  $p \le 0.001$ , \*\* for  $p \le 0.01$  and \* for  $p \le 0.05$ . Only significant correlation coefficients are shown. <sup>b</sup> Odor attributes: INT: Odor intensity; RPF: Ripe fruit odor; FER: Fermentative odor; CMB: Cucumber odor. <sup>c</sup> Flavor attributes: INT: Flavor intensity; FER: Fermentative flavor; CMB: Cucumber flavor; AST: Astringency. <sup>d</sup> Taste attributes: SWT: Sweetness; ACD: Acidity.

## 4. Conclusions

The sensory and PTR-MS analyses allowed the identification of specific odor and flavor traits associated with the melon cultivars evaluated, regardless of the group formation into *inodorus*, *cantalupensis*, and exotic fruits. These methodologies highlighted the enhanced sweetness of the *inodorus* and *cantalupensis* fruits, both commercial and elite cultivars, and the similar volatile profiles of 'Irak' and *cantalupensis* melons. A reasonable correlation between melon sensory attributes and PTR-MS spectral data was observed. Our results provide new information for the improvement of melon fruit quality. As new cultivars are being developed with high sugar and high acid levels, the results presented herein can be used as a tool to achieve distinct taste combinations without compromising desirable odor and flavor traits. Additional research to explore these correlations on new cultivars with extended shelf life would also be valuable.

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#### 583 Appendix

**Table 2** Quality indices determined among melon fruit types (N = 67): mean values and standard deviation in brackets <sup>a</sup>

			s (cv. group)	Commercial varieties (cv. group)						
		Climac	teric		Non-c	limacteric	Clim	acteric	Non-climacteric	
Parameters	'Dulce' b	'Védrantais'	'Irak'	'Calcuta'	'Songwhan charmi'	Piel de Sapo 'T111'	Galia	Cantaloupe	Amarillo	Piel de Sapo
	(cantalupensis)	(cantalupensis)	(dudaim)	(momordica)	(conomon)	(inodorus)	(cantalupensis)	(cantalupensis)	(inodorus)	(inodorus)
pН	6.3 (0.2) <sup>abc</sup>	6.5 (0.1) <sup>a</sup>	5.2 (0.4) <sup>e</sup>	5.5 (0.2) <sup>de</sup>	5.6 (0.1) <sup>de</sup>	5.8 (0.1) <sup>cde</sup>	6.4 (0.1) <sup>abc</sup>	6.5 (0.1) <sup>ab</sup>	5.9 (0.3) <sup>bcd</sup>	5.7 (0.2) <sup>cde</sup>
SSC (°Brix)	8.2 (1.6) <sup>cd</sup>	9.6 (1.3) <sup>bc</sup>	$4.7(1.0)^{d}$	7.4 (4.7) <sup>cd</sup>	9.3 (1.1) <sup>bc</sup>	9.9 (0.4) <sup>abc</sup>	12.8 (2.6) <sup>ab</sup>	14.0 (2.9) <sup>a</sup>	10.6 (0.6) <sup>abc</sup>	12.6 (1.6) <sup>abc</sup>

a Values with different letters in the same row indicate significant differences by Tukey's HSD post hoc test ( $p \le 0.05$ ).
b Number of samples of each melon type: 'Dulce' (n = 6), 'Védrantais' (n = 8), 'Irak' (n = 10), 'Calcuta' (n = 10), 'Songwhan charmi' (n = 8), Piel de Sapo 'T111' (n = 6), Galia (n = 6), Cantaloupe (n = 3), Amarillo (n = 3), Piel de Sapo (n = 7)