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1 **Linking sensory and proton transfer reaction-mass spectrometry analyses for the assessment of**
2 **melon fruit (*Cucumis melo* L.) quality traits**

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32 **Abstract**

33 Sixty-seven samples of ten melon types (*Cucumis melo* L.) were evaluated to determine the relationship
34 between their quality traits: sensory attributes, pH, soluble solids, and volatile organic compounds. Fruits
35 from the *cantalupensis*, *conomon*, *dudaim*, *inodorus*, and *momordica* cultivar groups were analyzed. The
36 sensory profiles were assessed using ten attributes covering odor, flavor, and taste characteristics,
37 whereas the volatile profiles were derived by proton transfer reaction-mass spectrometry. Fruits from the
38 *cantalupensis* and *inodorus* cultivars showed an opposite pattern for several quality traits. Fruits from the
39 *dudaim* cultivar were more related to the *cantalupensis*, whereas *conomon* and *momordica* showed an
40 intermediate behavior between *inodorus* and *cantalupensis*. The attributes of odor and flavor intensity,
41 ripe fruit odor, fermentative odor, and fermentative flavor correlated positively to C₃–C₉ esters ($r = 0.43$ –
42 0.73 ; $p \leq 0.01$). Positive correlations were also observed for several alcohols ($r = 0.36$ – 0.82 ; $p \leq 0.05$),
43 including methanol, ethanol, and diol alcohols, as well as for several aldehydes ($r = 0.43$ – 0.85 ; $p \leq 0.01$),
44 such as acetaldehyde, butanal, methyl butanal, heptanal, and decanal. The attributes mentioned above
45 were negatively correlated with two C₉ aldehydes, 2,6-nonadienal and nonenal ($r = -0.45$ to -0.62 ; $p \leq$
46 0.01), whereas sweetness was negatively correlated with two C₆ green leaf volatiles, hexenal and 3-
47 hexenol ($r = -0.50$; -0.67 ; $p \leq 0.001$). The melon fruits presented distinct differences in the quality traits
48 evaluated. These results provide information for the development of new cultivars with characteristic
49 taste combinations without compromising other desirable fruit quality traits.

50

51

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

53 1. Introduction

54 Melon (*Cucumis melo* L.) is a species with high genetic variation, the fruits of which show a wide
55 diversity in morphological, physical-chemical and sensory traits. The sweet melons of the *inodorus* group
56 or the highly aromatic melons of the *cantalupensis* one are generally consumed as fresh fruits. In contrast,
57 while the exotic cultivars of the *conomon*, *dudaim*, or *momordica* groups are either inedible or consumed
58 as fresh, cooked, or pickled vegetables [1, 2]. Melon has an exceptional ripening pattern as it comprises
59 both climacteric and non-climacteric cultivars within a single species, i.e., cultivars with a rise in the
60 respiration rate and ethylene production at the onset of fruit ripening (e.g., *cantalupensis*), and cultivars
61 with little or no ethylene production (e.g., *inodorus*). However, it has also been reported that melon
62 ripening behavior follows a continuous spectrum between the climacteric and non-climacteric references
63 rather than just two ripening patterns [3]. In addition, ethylene-dependent and ethylene-independent
64 pathways can coexist during the ripening process of climacteric melon fruits [4-6]. However, the
65 relationships between fruit quality traits and the biochemical pathways involved in ethylene-dependent
66 and ethylene-independent ripening processes are not entirely understood [7]. The fruits of the
67 *cantalupensis* cultivars are generally more aromatic but show a faster loss of firmness and a shorter shelf-
68 life than the ones of *inodorus* cultivars [8]. These differences are reflected in the sensory attributes and
69 consumer acceptance of commercial cultivars [9, 10], but little is known about the odor and flavor
70 profiles of the *conomon*, *dudaim*, or *momordica* exotic cultivars.

71 Odor and flavor are among the properties that most influence the sensory perception of fruit. Melon odor
72 perception depends on the presence and concentration of volatile organic compounds (VOCs), which
73 comprise a profile typically associated with each cultivar [9, 11]. In addition, flavor perception relies not
74 only on volatile but also non-volatile compounds such as soluble sugars and organic acids. Sweetness is
75 considered a determinant attribute for the eating quality of melon fruit, but a moderate acidity is also able
76 to drive consumers' liking of this fruit species [10]. Interactions between volatile and non-volatile
77 compounds should also be considered as VOCs are known to enhance the perception of several flavor
78 attributes [12-15].

79 Gas chromatography is the most common technique for the assessment of melon VOCs profile, which
80 comprises esters, alcohols, aldehydes, some sulfur-containing compounds, and minor quantities of
81 ketones, terpenes and hydrocarbons [8, 11, 16-22]. An alternative is the use of proton transfer reaction-
82 mass spectrometry (PTR-MS), which allows the headspace VOCs to be drawn from the samples at room
83 temperature (25 °C), simulating the conditions of consumer perception of the fruits. Headspace PTR-MS

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

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

94

95 **2. Material and methods**

96 2.1 Material

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

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

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

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

111 ^a Number of samples for each melon fruit type: Amarillo (n = 3); Cantaloupe (n = 3); Galia (n = 6); Piel de
 112 Sapo (n = 7); Calcuta (n = 10); Dulce (n = 6); Irak (n = 10); Songwhan charmi (n = 8); Piel de Sapo T111 (n =
 113 6); Védrantais (n = 8).

114

115 2.2 Methods

116 2.2.1 Common quality indices

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

123

124 2.2.2 Sensory analysis

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

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

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

140

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

Attributes	Description
<i>Odor</i>	
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.
<i>Flavor</i>	
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.
<i>Taste</i>	
Acidity	Amount of acid perceived during chewing.
Sweetness	Amount of sugar perceived during chewing.

142

143 2.2.3 PTR-MS profiling of VOCs

144 The frozen flesh of each melon fruit was cut into pieces, immersed in liquid nitrogen, and immediately
 145 ground for 15 s at 10,000 rpm using a Grindomix GM 200 (Retsch, Düsseldorf, Germany). Ground
 146 samples were stored (-20 °C) and analyzed within 24 h. For each sample, 1.0 g of ground powder was
 147 weighted in screw cap glass flasks of 250 mL. Before the analyses, the flasks were equilibrated in a water
 148 bath at 25 °C for 30 min. The temperature was selected to match the volatile emission in the headspace of
 149 the flasks and the conditions of the consumer perception of the fruits. The flasks were attached to the inlet
 150 of the PTR-MS system (Ionicon GmbH, Innsbruck, Austria), and the headspace was extracted at a 60
 151 mL/min flow rate. The temperature of both the inlet and the drift chamber was kept at 60 °C. Mass
 152 spectral data in a range between 20 and 160 atomic mass units (amu) was collected with a dwell time of
 153 200 ms. Blank measurements were run between samples to monitor background air, and these values

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

159

160 2.2.4 PTR-ToF-MS tentative identification of VOCs

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

172

173 2.2.5 Data analysis

174 The sensory data was evaluated using a one-way Analysis of Variance (ANOVA) on the mean values per
175 melon across panelists, considering the type of melon as a fixed factor. A Tukey's HSD post hoc test ($p \leq$
176 0.05) was performed to examine significant statistical differences between the melon types. Due to the
177 lack of normality, the PTR-MS data was evaluated using a non-parametric ANOVA (Kruskal-Wallis test),
178 followed by Dunn's multiple comparison test and Bonferroni correction ($p \leq 0.05$).

179 The data of the sensory scores and headspace VOCs measured on the same samples were used to evaluate
180 the relationship between both methods. Principal Component Analysis (PCA) and Pearson's correlation
181 analysis were performed over the 10 sensory attributes and the 40 significantly different VOCs obtained
182 from the ANOVA results. The PCA was performed on the correlation matrix to normalize the different
183 datasets. All the statistical analyses were performed with XLSTAT 2018 software (Addinsoft, Paris,
184 France).

185 3. Results and discussion

186 3.1 Sensory characteristics

187 The significant differences observed for the ten sensory attributes among the melon types are shown in
188 Table 4. ‘Védrantais’ and ‘Dulce’ cultivars showed higher scores for the intensity, ripe fruit, and
189 fermentative odor attributes, whereas the lower were observed for the ‘T111’ line of the Piel de Sapo
190 cultivar followed by the ‘Calcuta’. The same was observed between the commercial *cantalupensis*
191 (Cantaloupe and Galia) and *inodorus* (Amarillo and Piel de Sapo). Other authors observed higher fruity
192 odor for climacteric fruits belonging to the *cantalupensis* cultivar group but smaller differences between
193 these and non-climacteric *inodorus* ones [10]. The slight differences between ‘Irak’, Cantaloupe, and
194 Galia were consistent with previous results for the odor scores of *dudaim* and *cantalupensis* fruits [30].
195 These authors also reported higher odor scores for fruits of both cultivar groups than *inodorus*.
196 ‘Védrantais’ and ‘Dulce’ cultivars were also significantly higher scored for the intensity and fermentative
197 flavor attributes. The lowest scores of these attributes were observed for ‘Calcuta’ and ‘T111’ fruits,
198 respectively. The higher score for fermentative flavor of *cantalupensis* than *inodorus* fruits was consistent
199 with the results of the odor attributes.

200 The sweeter fruits belonged to the commercial varieties and ‘Védrantais’ cultivar. These were followed
201 by ‘Dulce’ and ‘T111’ cultivars, while ‘Calcuta’ was the least sweet. No sweetness differences between
202 *inodorus* and *cantalupensis* fruits were previously observed [10], although changes may occur depending
203 on the type of cultivar studied [11]. *Cantalupensis* fruits were observed to be sweeter than *inodorus*, and
204 both sweeter than *dudaim* fruits [30]. This was consistent with our results for ‘Irak’ cultivar. The highest
205 acidity scores were observed for ‘Védrantais’ and ‘Songwhan charmi’ cultivars. Except for the lowest
206 scores of Amarillo, no significant differences were observed for the rest of the fruits. Other authors
207 reported minimal [10] or not significant acidity differences [11] between *inodorus* and *cantalupensis*
208 cultivars. With the exception of ‘Calcuta’ and ‘Irak’ melons, perceived acidity was substantially lower
209 than sweetness. This reflects the predominance of sweet varieties among the fruits analyzed, as several
210 melon types showed a high SSC level together with near-neutral pH values (Table 2, Appendix). The
211 sweet/acid ratio is an important quality index for other fruit species, but the sweet melon varieties lack
212 acid taste, and their eating quality is mainly determined by sweetness [2]. At these levels, the interaction
213 between acid and sweet tastes has a suppressive effect of sweetness over acidity [31].

214 Small differences were observed for cucumber odor and flavor attributes. ‘Songwhan charmi’ fruits had
215 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) ^a

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

^a Values with different letters in the same row indicate significant differences by Tukey’s HSD post hoc test (p ≤ 0.05).^b Number of samples of each melon type: ‘Dulce’ (n = 4), ‘Védraçais’ (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).

225 3.3 VOCs profile

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, C₄ – C₉ 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].

251

252 *Alcohols*

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

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

261

262 *Aldehydes*

263 Acetaldehyde was the major aldehyde for all the melon types. It was present at significantly higher
264 concentrations in the headspace of the 'Védrantais' cultivar, while lower concentrations were observed
265 for the 'Calcuta' cultivar and commercial *inodorus*. Other authors reported prominent levels of
266 acetaldehyde among melon fruits [16, 34]. Hexenal was determined at significantly higher concentrations
267 for 'Irak' and 'Calcuta' cultivars in comparison to 'Songwhan charmi' and 'T111' cultivars. The lower
268 concentrations of hexenal observed for *inodorus* fruits are consistent with previous works [22, 35].
269 Heptanal was significantly higher for 'Védrantais' cultivar along with commercial *cantalupensis* and
270 'Songwhan charmi', while lower for *inodorus* fruits. Nonenal was significantly higher for 'T111' cultivar
271 along with the commercial *inodorus* fruits. 2,6-nonadienal was significantly higher for 'Irak' cultivar and
272 Amarillo. Lower concentrations of both C₉ aldehydes were observed among the commercial
273 *cantalupensis* fruits. Higher concentrations of nonenal and 2,6-nonadienal among *inodorus* than
274 *cantalupensis* were previously reported [11, 36].

275

276 *Esters*

277 The higher headspace concentrations of esters were observed for the 'Védrantais' cultivar, followed by
278 'Dulce' and the commercial *cantalupensis*. Intermediate concentrations were observed for 'Irak' and
279 'Songwhan charmi', whereas the ester pattern of 'Calcuta' and 'T111' cultivars was more similar to the
280 one of commercial *inodorus*. Within the latter, Piel de Sapo had the lowest ester concentration and
281 showed small differences when compared to the 'T111' cultivar. The C₃ and C₄ esters at *m/z* 75.044 and
282 89.059 were within the most abundant ester related masses, while lower concentrations were observed for
283 C₅–C₉ esters. The differential ester profile of *cantalupensis* and *inodorus* fruits is well documented in the
284 literature [11, 22, 32, 36].

285 *Terpenes*

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

290

291 *Other compounds*

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

296

297 Globally, volatile emission was more pronounced for *cantalupensis* fruits. ‘Védrantais’ cultivar was
298 different from the other *cantalupensis* fruits due to the higher concentrations of most of the VOCs, while
299 the VOC profile of ‘Dulce’ cultivar was more related to the commercial *cantalupensis* fruits, Galia and
300 Cantaloupe. The significantly higher concentrations of two C_9 aldehydes, 2,6-nonadienal and nonenal, the
301 C_5 , C_6 , and C_8 esters, and limonene, found for ‘Dulce’ cultivar, or the higher concentrations of a fragment
302 at m/z 57.069 and methyl butanal found for Galia and Cantaloupe, were on the basis of the differences
303 between the VOC profiles of these melon types. The lower volatile emission was observed for *inodorus*
304 fruits, although some differences were detected between ‘T111’ cultivar and the commercial *inodorus*
305 fruits, Amarillo and Piel de Sapo. The ‘T111’ and Amarillo fruits showed significantly higher
306 concentrations of methanol, 1,2-propanediol, acetic acid, isoprene, methyl acetate, and $C_5 - C_7$ esters than
307 Piel de Sapo. In contrast, higher concentrations of acetaldehyde, a nitrile compound, 1,2-ethanediol, and
308 ethyl acetate/ methyl propanoate were found for ‘T111’ than the other *inodorus* fruits. Regarding the
309 exotic cultivars, the VOCs pattern of the ‘Irak’ cultivar was more similar to that of ‘Dulce’ and the
310 commercial *cantalupensis* fruits for several alcohols, aldehydes, esters, and terpenes, except for methanol,
311 butanal, hexenal, 2,6-nonadienal, $C_4 - C_5$ esters, isoprene, and acetone. The VOC profile of ‘Calcuta’ and
312 ‘Songwhan charmi’ cultivars showed intermediate profiles between the ones of *cantalupensis* and
313 *inodorus* fruits. Additionally, both exotic cultivars had significantly lower concentrations of methanol,
314 whereas ‘Calcuta’ showed a higher concentration of 3-hexenol and hexenal, two C_6 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 LSD test (two right-side columns) and their standard deviations in brackets ^a

Mass (<i>m/z</i>) <i>Tentative identification</i> ^b	Sum formula	Reference	Cultivars (<i>cv.group</i>)					
			Climacteric				Non-climacteric	
			'Dulce' ^c (<i>cantalupensis</i>)	'Védrantais' (<i>cantalupensis</i>)	'Irak' (<i>dudaim</i>)	'Calcuta' (<i>momordica</i>)	'Songwhan charmi' (<i>conomon</i>)	Piel de Sapo 'T111' (<i>inodorus</i>)
29.037 <i>Fragment (alcohol)</i>	C ₂ H ₅ ⁺		16.2 (7.0) ^{ab}	217.4 (112.8) ^a	10.1 (9.3) ^{ab}	3.3 (2.7) ^b	4.8 (4.8) ^b	9.2 (13.6) ^b
31.017 <i>Formaldehyde</i>	CH ₃ O ⁺	[37]	17.5 (5.3) ^{ab}	41.1 (9.0) ^a	10.1 (4.3) ^{ab}	6.4 (2.3) ^b	5.1 (2.6) ^b	5.7 (5.4) ^b
33.034 <i>Methanol</i>	CH ₅ O ⁺	[18, 33]	5385 (2068.8) ^{ab}	6106.8 (1245.3) ^a	1343.7 (579.3) ^{abc}	849.1 (357.1) ^c	564.8 (329.2) ^c	1074.4 (613.4) ^{abc}
41.038 <i>Fragment (alcohol, ester)</i>	C ₃ H ₅ ⁺	[37, 38]	72.1 (36.0) ^{abc}	127.8 (30.6) ^a	53.8 (22.9) ^{abc}	33.4 (12.2) ^{abc}	23.4 (11.5) ^{bc}	17 (12.2) ^c
42.034 <i>Acetonitrile, nitrile fragment</i>	C ₂ H ₄ N ⁺		16.9 (15.6) ^{ab}	36.3 (34.2) ^a	11.7 (7.3) ^{ab}	5.2 (3.4) ^b	14.5 (24.2) ^{ab}	6.5 (6.9) ^{ab}
43.018 <i>Fragment (ester)</i>	C ₂ H ₃ O ⁺	[37, 38]	416 (295.5) ^{ab}	1949.9 (1252.4) ^a	209.1 (143.3) ^{ab}	116 (49.1) ^b	302.7 (235.4) ^{ab}	242.3 (352.2) ^b
43.053 <i>Fragment (alcohol, ester, acetate)</i>	C ₃ H ₇ ⁺	[37, 38]						
45.033 <i>Acetaldehyde</i>	C ₂ H ₅ O ⁺	[4, 34]	3961.2 (1184.9) ^{ab}	12084.9 (4842.8) ^a	4163.5 (4136.2) ^{ab}	1511.3 (1055.5) ^b	5968.6 (3827.4) ^{ab}	3794 (1676.4) ^{ab}
47.049 <i>Ethanol</i>	C ₂ H ₇ O ⁺	[4, 34]	329.7 (117.6) ^{ab}	3737.5 (187.3) ^a	233.5 (164.2) ^{ab}	114.5 (62.3) ^b	146.9 (91.2) ^b	209.3 (241.4) ^b
55.054 <i>Fragment</i>	C ₄ H ₇ ⁺	[37]	28.5 (14.5) ^{ab}	19.7 (7.2) ^{ab}	38.4 (20.9) ^a	29.6 (9.1) ^a	13.3 (4.7) ^{ab}	6.5 (1.2) ^b
57.069 <i>Fragment (alcohol, ester)</i>	C ₄ H ₉ ⁺	[37, 38]	76.3 (43.2) ^{ab}	98.6 (47.3) ^a	35.6 (16.1) ^{abc}	23.8 (7.2) ^{abc}	21 (12.8) ^{abc}	11.3 (12.5) ^{bc}
59.049 <i>Acetone</i>	C ₃ H ₇ O ⁺	[4, 17]	19.3 (3.0) ^{ab}	26.2 (6.7) ^{ab}	32 (8.7) ^a	25.4 (11.4) ^{ab}	26.6 (8.3) ^{ab}	14.8 (6.1) ^b
61.028 <i>Acetic acid</i>	C ₂ H ₅ O ₂ ⁺	[17, 35]	341.3 (287.2) ^{ab}	1539.3 (1324.8) ^a	114.3 (84.9) ^{abc}	72.7 (47.1) ^{bc}	257.3 (230.9) ^{ab}	211.9 (366.4) ^{abc}
63.044 <i>1,2-Ethandiol</i>	C ₂ H ₇ O ₂ ⁺		7.8 (2.1) ^{ab}	24.9 (11.7) ^a	7.2 (5.7) ^{ab}	3.8 (1.2) ^b	10 (5.8) ^{ab}	7 (4.2) ^{ab}
67.054 <i>Fragment (ester)</i>	C ₅ H ₇ ⁺	[37]	1 (0.5) ^{ab}	0.8 (0.4) ^{ab}	1.9 (0.7) ^a	0.8 (0.3) ^{ab}	1.3 (0.5) ^{ab}	2.3 (0.7) ^a

<i>Fragment (terpene, sesquiterpene, aldehyde)</i>			(7.9) ^{ab}	(0.9) ^b	(26.4) ^a	(19.7) ^a	(8.6) ^b	(8.6) ^{ab}	(1.3) ^{ab}
83.086	C ₆ H ₁₁ ⁺	[37]	12.2	6.4	18.1	13.7	2.8	1.4	3.0
<i>Fragment (alcohol, aldehyde, sesquiterpene)</i>			(7.9) ^{ab}	(2.3) ^{abc}	(11.6) ^a	(4.7) ^a	(3.1) ^{bc}	(3.1) ^{bc}	(1.3) ^{ab}
85.099	C ₆ H ₁₃ ⁺	[37]	1.9	4.9	3.7	3.1	0.9	0.6	3.0
<i>Fragment (alcohol)</i>			(1.3) ^{ab}	(1.5) ^a	(1.2) ^{ab}	(1.7) ^{ab}	(0.7) ^b	(0.2) ^b	(2.0) ^{ab}
87.08	C ₅ H ₁₁ O ⁺	[4, 34]	0.5	2.9	0.7	0.6	0.6	0.5	1.0
<i>Methyl butanal</i>			(0.2) ^b	(1.3) ^a	(0.2) ^{ab}	(0.2) ^b	(0.3) ^b	(0.3) ^b	(0.2) ^b
89.059	C ₄ H ₉ O ₂ ⁺	[4, 34]	63.5	354.1	16	7.3	33.3	30	80
<i>Ethyl acetate/ Methyl propanoate</i>			(53.4) ^{ab}	(228.6) ^a	(16.1) ^{abc}	(6.5) ^{bc}	(31.3) ^{abc}	(52.1) ^{abc}	(1.3) ^{ab}
91.074	C ₄ H ₁₁ O ₂ ⁺		6.6	5.6	1.4	2.6	0.9	0.4	2.0
<i>2,3-Butanediol</i>			(5.4) ^{ab}	(2.5) ^a	(0.7) ^{abc}	(1.3) ^{ab}	(0.5) ^{bc}	(0.4) ^{bc}	(1.3) ^{ab}
95.085	C ₇ H ₁₁ ⁺	[37]	0.6	0.5	1.2	0.6	1.8	1	0.5
<i>Fragment (monoterpene)</i>			(0.2) ^{abc}	(0.2) ^{bc}	(0.4) ^{ab}	(0.2) ^{bc}	(0.9) ^a	(0.3) ^{abc}	(0.2) ^b
99.08	C ₆ H ₁₁ O ⁺	[34, 35]	1.3	0.5	10.4	10	0.9	0.3	0.5
<i>Hexenal</i>			(1.4) ^{ab}	(0.2) ^{ab}	(5.1) ^a	(3.9) ^a	(1.9) ^b	(0.3) ^b	(0.2) ^b
101.095	C ₆ H ₁₃ O ⁺	[34, 36]	0.3	0.4	1	0.8	0.2	0.1	0.5
<i>3-Hexenol</i>			(0.2) ^{ab}	(0.1) ^{ab}	(0.7) ^a	(0.3) ^a	(0.2) ^b	(0.1) ^b	(0.2) ^b
103.075	C ₅ H ₁₁ O ₂ ⁺	[34, 36]	14.5	13.1	1.6	1.1	0.5	1.5	1.0
<i>Ester (Ethyl propanoate, Isopropyl acetate, Methyl butanoate, Methyl isobutyrate, Propyl acetate)</i>			(10.6) ^a	(9.2) ^a	(1.4) ^{abc}	(1.0) ^{abc}	(0.4) ^{bc}	(2.6) ^{abc}	(9.2) ^a
115.111	C ₇ H ₁₅ O ⁺	[34-36]	0.3	2.1	0.2	0.2	0.4	0.1	0.5
<i>Heptanal</i>			(0.2) ^{abc}	(2.8) ^a	(0.1) ^{abc}	(0.1) ^{abc}	(0.4) ^{ab}	(0.2) ^{bc}	(0.2) ^b
117.091	C ₆ H ₁₃ O ₂ ⁺	[4, 36]	18.3	18.1	1.4	1.2	0.9	1	1.0
<i>Ester (Butyl acetate, Ethyl butanoate, Isobutyl acetate, Methyl n-methylbutanoate)</i>			(19.0) ^{ab}	(14.7) ^a	(0.9) ^{abcd}	(0.9) ^{abcd}	(0.9) ^{bcd}	(0.9) ^{cd}	(9.0) ^{ab}
123.117	C ₉ H ₁₅ ⁺	[37]	0.9	0.5	1.1	0.4	1.4	2.8	0.5
<i>Fragment (farnesene)</i>			(0.6) ^{ab}	(0.4) ^{ab}	(0.9) ^{ab}	(0.2) ^b	(0.6) ^{ab}	(0.8) ^a	(0.6) ^{ab}
131.107	C ₇ H ₁₅ O ₂ ⁺	[34, 36]	2.5	13.4	0.7	0.2	0.3	0.6	3.0
<i>Ester (Ethyl methylbutanoate, Ethyl pentanoate, Methyl hexanoate, Pentyl cetate)</i>			(1.9) ^{ab}	(9.1) ^a	(0.7) ^{ab}	(0.2) ^{bc}	(0.3) ^{bc}	(0.3) ^{bc}	(3.0) ^{ab}
137.132	C ₁₀ H ₁₇ ⁺	[11, 34]	0.9	0.4	0.2	0.3	0.1	0.1	0.5
<i>Limonene</i>			(0.5) ^a	(0.2) ^a	(0.1) ^{abc}	(0.1) ^{ab}	(0.04) ^{bc}	(0.04) ^c	(0.2) ^b
139.112	C ₉ H ₁₅ O ⁺	[34, 36]	0.4	0.2	1	0.5	0.5	0.8	0.5
<i>2,6-Nonadienal</i>			(0.3) ^{ab}	(0.1) ^b	(0.4) ^a	(0.3) ^{ab}	(0.4) ^{ab}	(0.3) ^{ab}	(0.3) ^{ab}
141.128	C ₉ H ₁₇ O ⁺	[34, 36]	0.5	0.4	0.6	0.3	0.8	1.2	0.5
<i>Nonenal</i>			(0.4) ^{ab}	(0.2) ^{ab}	(0.5) ^{ab}	(0.1) ^b	(0.4) ^{ab}	(0.4) ^a	(0.4) ^{ab}

318 3.4 Correlation between sensory and PTR-MS analyses

319 3.4.1 Principal component analysis

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

337

338 3.4.2 Pearson's correlation analysis

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

343

344 *Alcohols*

345 A positive contribution of alcohols to the attributes of odor ($0.48^{**} \leq r \leq 0.76^{***}$) and flavor intensities
346 ($0.36^* \leq r \leq 0.59^{***}$), ripe fruit odor ($0.42^{**} \leq r \leq 0.80^{***}$), and fermentative odor and flavor ($0.42^{**} \leq r$
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].

349 Methanol and ethanol were reported to be associated with advanced ripening stage [18], whereas the diol
350 alcohols with ester production in melon fruit [17]. Methanol was also negatively correlated with
351 cucumber odor ($r = -0.51^{***}$) and flavor ($r = -0.40^*$).

352

353 *Aldehydes*

354 Several aldehydes were positively correlated with odor ($0.54^{**} \leq r \leq 0.76^{***}$) and flavor intensity
355 ($0.43^{**} \leq r \leq 0.56^{***}$), ripe fruit ($0.49^{***} \leq r \leq 0.76^{***}$) and fermentative odor and flavor attributes
356 ($0.58^{***} \leq r \leq 0.85^{***}$). Most of these aldehydes (acetaldehyde, hexenal, heptanal, and decanal) were
357 associated with a lack of maturity [34], although acetaldehyde was also observed to increase with
358 maturity [18]. Acetaldehyde is particularly important as it increases fruit flavor and contributes to the
359 perception of freshness [39]. Positive contributions of acetaldehyde to the flavor perception of citrus [40],
360 kiwi [41], or tomato fruits [42] have also been observed. Verzera *et al* [22] reported strong correlations
361 between the typical odor and flavor descriptors of melon and several aldehydes, including methyl butanal,
362 2,6-nonadienal and nonenal. The latter two are associated with green or cucumber notes and considered
363 key volatiles in the typical aroma of the non-climacteric fruits belonging to the *inodorus* cultivar [11]. In
364 the present study, no significant correlations were observed between both C₉ aldehydes and cucumber
365 odor or flavor, possibly due to the predominance of climacteric cultivars among the fruits analyzed.
366 However, negative correlations were found between these compounds and odor intensity ($r = -0.53^{**}$; -
367 0.50^{**}), ripe fruit odor ($r = -0.54^{***}$; -0.47^*), and fermentative odor ($r = -0.62^{***}$; -0.52^{***}) or flavor
368 ($r = -0.57^{***}$; -0.45^{**}) attributes. Previous authors observed high negative correlations between 2,6-
369 nonadienal or nonenal with 'fruity', 'sweet-aromatic', and 'chemical' flavor attributes, but also high
370 positive correlations with 'cucurbit' attribute [19].

371

372 *Esters*

373 The correlations found with intensity ($0.48^{**} \leq r \leq 0.69^{***}$), ripe fruit ($0.46^{**} \leq r \leq 0.69^{***}$) or
374 fermentative odor ($0.53^{***} \leq r \leq 0.73^{***}$) and flavor ($0.56^{***} \leq r \leq 0.70^{***}$) are in agreement with
375 previous authors reporting good correlations ($r \geq 0.61$; $p < 0.05$) between ethyl, methyl or acetate esters
376 and melon sensory flavor [18]. High correlations ($r \geq 0.76$) between C₇ – C₉ esters and the fruity odor
377 [20] or between C₅ – C₇ esters and fruity, pineapple-like, and sweet aromas were also observed [21].
378 Additionally, several works pointed out the importance of sulfur-containing esters to the odor and flavor
379 of melon fruits [11, 20, 21], but these were not detected in the present work, possibly due to differences in

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

383

384 *Terpenes*

385 Two terpene related masses, monoterpene (95.085) and farnesene (123.117) fragments, were negatively
386 correlated with odor intensity, ripe fruit or fermentative odor and flavor. The former was positively
387 correlated with cucumber odor ($r = 0.75^{***}$) and flavor ($r = 0.72^{***}$). On the other hand, isoprene and
388 limonene showed positive correlations with odor intensity ($r = 0.50^{***}$; 0.47^{**}), ripe fruit ($r = 0.44^{**}$;
389 0.47^{**}) and fermentative odor ($r = 0.56^{***}$; 0.41^{*}) or flavor ($r = 0.50^{***}$; 0.37^{*}). Limonene was
390 observed to contribute for the odor and flavor of melon [20].

391

392 *Other compounds*

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

398

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

400 The determinations of SSC and pH were satisfactorily correlated with sweetness ($r = 0.67^{***}$; 0.70^{***}).
401 Both parameters were also correlated with flavor intensity ($r = 0.47^{**}$; 0.58^{***}), although this could be
402 due to an indirect effect of the high correlation between sweetness and flavor intensity ($r = 0.77$; $p \leq$
403 0.001). A slight negative correlation was found between SSC and astringency ($r = -0.37^{*}$). This was
404 consistent with the negative correlation between sweetness and astringency attributes ($r = -0.57$; $p \leq$
405 0.001). SSC has a significant positive effect on the sweet and fruity descriptors, as well as a significant
406 negative effect on the green, bitter and astringent descriptors, among fruits, beverages, and flavors [12,
407 46]. A similar pattern was observed for 3-hexenol and hexenal, two C_6 green leaf volatiles, which were
408 negatively correlated with sweetness ($r = -0.50^{***}$; -0.67^{***}) and positively with astringency ($r =$
409 0.50^{**} ; 0.41^{**}). The correlation between hexenal and sweetness was reflected over flavor intensity ($r = -$
410 0.66^{***}), but for 3-hexenol positive correlations with intensity and ripe fruit odor or fermentative odor

411 and flavor attributes ($0.42^{**} \leq r \leq 0.48^{**}$) were observed. The ortho- and retronasal perception of green
412 leaf volatiles was observed to change from 'green' to 'fruity' descriptors due to the interaction of these
413 compounds with sugars and acids [13], but the nature of these interactions can vary with the fruit species.
414 Aprea *et al* [14] observed a negative contribution of 3-hexenol to the sweet perception of apple, whereas
415 Klee and Tieman [47] reported a positive contribution to the 'overall flavor intensity' and 'liking' of
416 tomato. Other authors observed the negative contribution of hexenal to the 'overall flavor intensity' and
417 'liking' of strawberries and blueberries [47], as well as to the sweetness of table grapes [48]. Besides, the
418 interactions between certain VOCs with sugars and acids affect the rate of release and persistence of these
419 volatile compounds in the mouth and, thus, the perceived intensities of aroma and flavor attributes [15].
420 Moreover, a positive correlation between astringency and acidity was also observed ($r = 0.45$; $p \leq 0.01$).
421 This opposed relationship of astringency with sweetness and acidity was observed in other fruits like
422 strawberries [49], apples [50], or kiwifruits [51]. Regarding VOCs, the highest correlation of acidity was
423 with isoprene ($r = 0.54^{***}$), a leaf volatile in the origin of several terpene compounds. Minor correlations
424 were also observed with 1,2-ethanediol ($r = 0.41^{**}$), several aldehydes ($0.39^* \leq r \leq 0.42^{**}$) or acetic acid
425 ($r = 0.37^*$). The interaction between acetaldehyde and sugars or acids is known to enhance the 'fruity' and
426 'tropical flavor' attributes of tomato fruits [42], although in the present study it was only correlated with
427 acidity.

428 pH was positively correlated with SSC ($r = 0.56^{***}$), whereas no significant correlation was observed
429 between pH and acidity. The relationship between pH increase and sugar accumulation was previously
430 observed [1, 2], and both processes are classified as ethylene-independent [5-7]. The melon genotypes
431 with higher sugar levels have pH values closer to the neutral range, whereas the ones with low sugar
432 levels show a broader range of pH values [1]. The characterization of the pH gene, with a major impact
433 on fruit acidity, has contributed to explain the low level of acidity of sweet melon types [52].
434 Additionally, pH was strongly correlated with the majority of the VOCs, as it is a parameter involved in
435 the regulation of several reactions of volatile production [16].

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

<i>m/z</i>	VOCs	Odor attributes ^b				Flavor attributes ^c				Taste attributes ^d	
		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	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***	

^a Significance: *** for $p \leq 0.001$, ** for $p \leq 0.01$ and * for $p \leq 0.05$. Only significant correlation coefficients are shown.

^b Odor attributes: INT: Odor intensity; RPF: Ripe fruit odor; FER: Fermentative odor; CMB: Cucumber odor.

^c Flavor attributes: INT: Flavor intensity; FER: Fermentative flavor; CMB: Cucumber flavor; AST: Astringency.

^d Taste attributes: SWT: Sweetness; ACD: Acidity.

436 **4. Conclusions**

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

446 **Acknowledgments**

447 This research was funded by the Spanish Ministry of Economy and Competitiveness, INIA – Project: RTA2011-
448 00123-00-00, and by the Agency for the Research Centres of Catalonia (CERCA) of the Generalitat de Catalunya.
449 Tiago Bianchi acknowledges the Ph.D. grant from the Spanish National Institute for Agricultural and Food Research
450 and Technology (INIA).

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Table 2 Quality indices determined among melon fruit types (N = 67): mean values and standard deviation in brackets ^a

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

^a Values with different letters in the same row indicate significant differences by Tukey's HSD post hoc test ($p \leq 0.05$).

^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)