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## Impact of crossflow microfiltration on aroma and sensory profiles of a potential functional citrus-based food

Imane Hammad, Manuel Dornier, Marc Lebrun, Isabelle Maraval, Patrick Poucheret, Claudie Dhuique-Mayer

### ► To cite this version:

Imane Hammad, Manuel Dornier, Marc Lebrun, Isabelle Maraval, Patrick Poucheret, et al.. Impact of crossflow microfiltration on aroma and sensory profiles of a potential functional citrus-based food. *Journal of the Science of Food and Agriculture*, 2022, 102 (13), pp.5768-5777. 10.1002/jsfa.11926 . hal-03816518

**HAL Id: hal-03816518**

**<https://hal.science/hal-03816518>**

Submitted on 9 Feb 2023

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13 **Abstract**

14 **BACKGROUND** – Citrus juices can be cold-concentrated by crossflow microfiltration (CMF)  
15 in order to obtain functional foods enriched in carotenoids, flavonoids and pectins. The work  
16 aimed to characterize the organoleptic quality of this type of micronutrient-dense foods through  
17 their aroma profile and sensory analysis. Two citrus concentrates with and without a  
18 diafiltration step were compared. **RESULTS** – Both citrus products were very different linked  
19 to aroma compound, sugar and organic acid contents. Due to its sugar/acidity balance and its  
20 better aromatic profile responsible for the citrus-floral flavor, the concentrate without  
21 diafiltration was preferred by the sensory panel. Thanks to a simple transfer model, we showed  
22 that retention of volatiles clearly varied from an aroma compound to another. The terpene  
23 hydrocarbons were the most retained by the membrane during CMF probably because they  
24 were strongly associated to insoluble solids by adsorption. **CONCLUSION** – Even the process  
25 modified their organoleptic profiles, both citrus-based products were well rated and can be  
26 consumed directly as pleasant functional drinks.

27

28 **Keywords**

29 Citrus aroma, membrane technology, carotenoid concentrate, volatile compounds, sensory  
30 analysis.

## 31           **1. Introduction**

32 Citrus fruit juices and derived products are widely consumed in the world and are considered  
33 as micronutrient-dense food with attractive taste and aroma. Their nutritional benefits are  
34 mainly attributed to vitamins (C, B, pro-A), dietary fibers (pectins), minerals as well as  
35 phytomicronutrients such as carotenoids and polyphenols<sup>1</sup>. Recently, different studies showed  
36 that some fruit juices could be cold-treated by crossflow microfiltration (CMF) in order to  
37 enhance their nutritional quality<sup>2-4</sup>. Indeed, this process allowed fruit juices to be enriched up  
38 to around 10-fold in carotenoids, some flavonoids and pectins. CMF can be carried out at low  
39 temperature. It allows the organoleptic quality and the nutritional potential of the juice to be  
40 preserved while consuming less energy<sup>5,6</sup>.

41  
42 Bioactive insoluble compounds such as carotenoids, some flavonoids or pectins are retained by  
43 the porous CMF membranes. Moreover, solutes can be removed by addition of a diafiltration  
44 step to modulate sugar content making the potential functional citrus-based food healthier. The  
45 interest of these “citrus concentrates” was evaluated mainly through their nutritional quality.  
46 The carotenoid bioaccessibility and uptake by intestinal cells of a clementine concentrate have  
47 already been reported by Gence et al. (2018)<sup>7</sup>. ~~Overall results indicated that citrus concentrates~~  
48 ~~obtained from industrial flash-pasteurized juices contained more bioaccessible carotenoids.~~  
49 Other formulated citrus juices were concentrated in order to optimize carotenoid/flavonoid  
50 profiles<sup>8,9</sup>. Finally, a functional citrus based-food elaborated with *Citrus clementina* juice was  
51 tested *in vivo* and its consumption was shown to prevent metabolic syndrome/diabetes type 2  
52 in rats<sup>10</sup>. So, the concentrates obtained by CMF can be considered as potential new functional  
53 foods. They could meet consumer demands for food offering health benefits and provide an  
54 alternative to regular citrus juice consumption.

55 These functional citrus based-foods had a similar texture to marmalade or coulis and so could  
56 be consumed directly as a healthy product. The majority of citrus fruits and derived products  
57 are preferred by consumers because of their color, flavor and aroma <sup>11</sup>. Therefore, in parallel  
58 with nutritional studies, it appears necessary to better assess the acceptability of the product by  
59 consumers. If the impact of the membrane process has been evaluated on the bioactive  
60 compounds, its impact on the organoleptic quality and especially on aroma profile is not  
61 documented. However, it is probably strong as already mentioned for other citrus juices <sup>12,13</sup>.

62

63 Consequently, the aim of the present work was to evaluate the impact of CMF technology on  
64 the organoleptic quality of a functional citrus-based food, focusing on flavor. From 2  
65 concentrates obtained with and without a diafiltration step, we examined how the processing of  
66 a mix of clementine and grapefruit juices modified the content of volatiles and sensory profile  
67 through a sensory descriptive analysis coupled with a quantitative analysis of aroma  
68 compounds. Finally, retention of the different aroma compounds by the CMF membrane were  
69 assessed from the measured concentration factors using a simple transfer model.

70

## 71 **2. Materials and methods**

72

### 73 *2.1 Preparation of citrus juices*

74 A formulated 60/40% v/v clementine (*Citrus clementina* Hort. Ex Tan.) and pink grapefruit  
75 (*Citrus paradisi* Macf.) juice was chosen as the initial citrus juice. This blend allowed to obtain  
76 interesting carotenoids and flavonoid profiles, bioactive compounds which have a strong  
77 positive effect on health <sup>9</sup>. Because of the difficulties in sourcing fresh fruits (seasonality) and  
78 in extracting the juice in the required quantities (30 L of juice in total), commercial flash-  
79 pasteurized 100% pure juices purchased from a local food supplier (Carrefour, Saint Clément  
80 de Rivière, France) were used in this study. The juices were stored 3 or 4 days at 4°C before

81 being used to formulate 15 L batches of the citrus juice. As mentioned in a previous study <sup>9</sup>,  
82 this citrus juice had 10.7% total soluble solids, 0.9% titratable acidity (pH 3.45) and 0.36%  
83 suspended insoluble solids. It contained a high concentration 8.7 mg·kg<sup>-1</sup> of carotenoids (β-  
84 carotene, β-cryptoxanthin and lycopene) and 570 mg·kg<sup>-1</sup> of flavonoids (hesperidin and  
85 naringin).

86

## 87 *2.2 Crossflow microfiltration process*

88 CMF was performed as described by Hammad et al. (2020) <sup>9</sup> using the same laboratory device  
89 equipped with 0.2 μm average pore diameter alumina tubular membranes with 55 cm<sup>2</sup> effective  
90 surface area (Pall-Exekia, Bazet, France) and using the same operating conditions (temperature  
91 30°C, transmembrane pressure 2.6 bar, crossflow velocity 5 m·s<sup>-1</sup>). Two concentrates were  
92 obtained by recovering the retentate up to a mass reduction ratio MRR (Eq. 1) between 8 and 9  
93 without or after a final purification step by diafiltration. This last step consisted in adding  
94 distilled water to the system at the end of the concentration until a diamass ratio DMR (Eq. 2)  
95 of around 1 was reached.

$$96 \quad \mathbf{MRR} = \mathbf{1} + \frac{\mathbf{m}_p}{\mathbf{m}_r} \quad \mathbf{(1)}$$

$$97 \quad \mathbf{DMR} = \frac{\mathbf{m}_w}{\mathbf{m}_r} \quad \mathbf{(2)}$$

98 with  $m_p$ ,  $m_r$  and  $m_w$ , mass of permeate, retentate and water added (kg).

99

100 Concentrate obtained without the diafiltration step was labelled DF0 and concentrate with  
101 diafiltration DF1. Both concentrates were stocked in amber sealed bottles under N<sub>2</sub> and kept  
102 frozen (-20°C) until analysis.

103

104        *2.3 Overall sugar and acid composition*

105        Sugar/acid ratio (S/A) was calculated by dividing total soluble solids (TSS) by titratable acidity  
106        (TA). TSS was measured at 25°C with a digital refractometer (Pal3 Pocket, Atago Tokyo,  
107        Japan) and TA was measured by titration with 0.1 mol·L<sup>-1</sup> NaOH.

108

109        *2.4 Analysis of aroma compounds*

110        For aroma compound extraction, 1 g of product with addition of 100 µL of internal standard  
111        (620 mg·L<sup>-1</sup> butanol) were placed in a 10 mL vial and extracted by HS-SPME according to  
112        Corrales et al. (2017) <sup>14</sup>. In the case of the concentrates, the products were diluted 8 times in  
113        order to obtain the same pulp content as in the initial juice. Indeed, the pulp content had a very  
114        strong effect on the release of aroma compounds in the headspace that corroborate the results  
115        already obtained <sup>15</sup>. Extraction was carried out at 50°C with 10 min incubation followed by 45  
116        min trapping, using a 65 µm polydimethylsiloxane/divinylbenzene/carboxen fiber (Supelco,  
117        Bellefonte, USA).

118        Then an Agilent 6890 / MSD 5973 N tandem mass / gas chromatograph (Agilent Technologies,  
119        Palo Alto, USA) and an MPS-2 autosampler (Gerstel GmbH & Co, KG) were used. A polar  
120        capillary column DBWax UI (60 m × 0.25 mm × 0.25 µm), from J & W Scientific (Folsom,  
121        USA) was chosen with a H<sub>2</sub> carrier gas flow rate of 1.5 mL·min<sup>-1</sup>. Injection was realized in  
122        splitless mode at 250°C and the following temperature program was used for elution: 3°C·min<sup>-1</sup>  
123        <sup>1</sup> from 40 to 170°C, then 10°C·min<sup>-1</sup> up to 240°C and held for 10 min. Analyzer and source  
124        temperatures were 150 and 250°C, respectively. Data were analyzed with Mass Hunter version  
125        B.06.00 (Agilent Technologies, Palo Alto, USA). Compound identification was performed by  
126        comparing their mass spectra with those of NIST 2014 (National Institute of Standard  
127        Technology). Co-injection of C8-C20 n-alkane (Sigma-Aldrich, St Louis, USA) was used to

128 calculate the Kovats Retention Index (RI) for comparison with those found on the Flavornet,  
129 Pherobase and NIST websites. Three replicates were carried out for all the samples.  
130 For each aroma compound  $i$ , the concentration factor  $CF_i$  was calculated comparing its contents  
131  $C_i$  in the final concentrate and the initial juice evaluated through semi-quantitative  
132 measurements as butanol equivalent according to Eq. 3.

$$133 \quad C_i = \frac{m_{butanol}}{m_{sample}} \frac{A_i}{A_{butanol}} \quad (3)$$

134 with  $C_i$  content of the compound  $i$  ( $\text{mg}\cdot\text{kg}^{-1}$ );  $A_i$  and  $A_{butanol}$  peak areas of the compound  $i$  and  
135 butanol (internal standard);  $m_{butanol}$  mass of butanol added (mg);  $m_{sample}$  mass of the sample  
136 (kg).

137  $CF_i$  can be used to evaluate the losses of the aroma compound  $i$  (%) in the final retentate through  
138 the Eq. 4.

$$139 \quad \text{Losses of } i = 100 \left(1 - \frac{CF_i}{MRR}\right) \quad (4)$$

140 Standard addition calibration was performed on a selection of aroma compounds for a more  
141 precise quantification. Five points from 0.25 to 8.00  $\text{mg}\cdot\text{kg}^{-1}$  were chosen and the obtained  
142 linear regression coefficients ( $R^2$ ) ranged from 0.90 to 0.97. These compounds were selected  
143 through their well-known relevant contribution to the citrus aromas and their availability by  
144 chemical suppliers.

145

### 146 *2.5 Odor activity values*

147 Odor activity values (OAV) were performed to better appreciate the contribution of each  
148 volatile aroma compound in citrus juice and concentrates. OAV were calculated dividing the  
149 content of the aroma compound by its odor threshold value in water. Thresholds used to  
150 calculate the OAV were obtained from literature<sup>16–18</sup>.

151

### 152 *2.6 Descriptive sensory analysis*

153 A quantitative descriptive analysis was carried out in the CIRAD - UMR Qualisud (Montpellier,  
154 France) sensory analysis laboratory with individual booths, according to the ISO 13299  
155 standard (2016) with 12 panelists (8 women and 4 men between 21 and 60 years old). They  
156 were trained according to the ISO 8586 standard (2012) and were familiarized with sensory  
157 characteristics of citrus concentrates in a single session. These panelists were invited for the  
158 second time to evaluate the citrus DF0 and DF1 concentrates. The samples were presented at  
159 24°C to the panelists in 25 mL transparent glass bottles bearing a random 3-letter code, in a  
160 monadic manner under red lighting. The room temperature and relative humidity were  
161 21.6±0.8°C and 24±3%. Twenty attributes were defined (Table 1) and evaluated on a scale of  
162 0 (weak intensity) to 10 (strong intensity). Samples were evaluated in duplicate and the results  
163 were noted as the average values.

164

### 165 *2.7 Evaluation of the retention of aroma compounds during microfiltration*

166 In order to evaluate the retention rate  $R_i$  for each aroma compound  $i$ , the simple model already  
167 suggested by Servent et al. (2020) <sup>4</sup> was used (Eq. 4). This model allowed the theoretical  
168 concentration factor  $\widehat{CF}_i$  as a function of MRR and DMR to be calculated assuming that there  
169 were no losses,  $R_i$  was constant and the system behave like a perfect stirred reactor.

$$170 \quad \widehat{CF}_i = MRR^{R_i} e^{DMR (R_i - 1)} \quad (4)$$

171 For each compound  $i$ ,  $R_i$  was evaluated thanks to the solver tool of MS-Excel (non-linear GRG  
172 method) by minimizing the sum of squared deviations between experimental and calculated  
173 concentration factors for both DF0 and DF1 trials.

174

### 175 *2.8 Statistical analysis*

176 All statistical analyses were performed using XLSTAT software version 2019.4.1 (Addinsoft,  
177 Paris, France). Data were analyzed using one-way analysis of variance in order to determine

178 significant differences ( $p < 0.05$ ). Tukey's multiple comparison method was used to further  
179 examine any significant difference between results. Principal Component Analysis (PCA) was  
180 performed to distinguish the two citrus products on the quality and sensory attributes as well as  
181 the aroma profile, using volatile compound concentration factor and sensory evaluation scores.  
182

### 183 **3. Results and discussion**

184

#### 185 *3.1 Sensory evaluation of the citrus-based food*

186 The sensory profiles of the two concentrates obtained by CMF without and with the diafiltration  
187 step DF0 and DF1 respectively were graphically illustrated by a radar chart diagram in Fig.1.  
188 The day of tasting, the 20 attributes described in Table 1 were analyzed and evaluated by the  
189 trained sensory panel. Sensory evaluation showed that the two concentrates were significantly  
190 different ( $p < 0.05$ ) for the majority of the attributes (15/20). The scores for citrus and  
191 clementine odor, citrus aroma (clementine and grapefruit), the sweet and sour taste, overall  
192 quality and viscosity mouthfeel, remained the highest for DF0 compared with DF1.

193 The difference between the scores of sweet and sour taste in DF0 (3.55 and 6.25) and in DF1  
194 (1.30 and 3.22) can be explained by the removal of a large percentage of water-soluble  
195 compounds such as sugars and acids in DF1 during the diafiltration step. Conversely,  
196 undesirable sensory attributes including chemical, and oxidized, fermented impression were  
197 significantly ( $p < 0.05$ ) more intense in DF1 than in DF0. This observation could be due to the  
198 high concentration of the compounds 6-methyl-5-hepten-2-one,  $\alpha$ -copaene and octyl acetate in  
199 DF1. According to the literature, it has been shown that these compounds are responsible for  
200 mushroom woody, earthy, spicy, sulfurous and pungent odors<sup>17,19-22</sup>.

201 The high scores of citrus aroma and citrus clementine odor observed in DF0 compared to DF1  
202 could be explained by the fact that the volatile compounds responsible for these attributes were  
203 positively correlated with the amount of sugars in the sample<sup>23</sup>. Indeed, the CMF process also

204 contributed to concentrating the monoterpenes responsible for the citrus flavor<sup>18</sup>. The high  
205 scores of overall quality for DF0 (6.59) which are better than DF1 (4.28), could be related to  
206 monoterpenes which were more concentrated in DF0 than in DF1. In addition, according to the  
207 comments provided by the sensory panel and to better understand this difference between the  
208 two concentrates, we then carried out identification and quantification of the aromatic  
209 compounds described as follows.

210

### 211 *3.2 Profile of aroma compounds in the initial juice*

212 For the more precise quantification through standard addition calibration, the 9 selected aroma  
213 compounds in the initial juice were 4 monoterpene hydrocarbons, D-limonene,  $\beta$ -myrcene,  $\gamma$ -  
214 terpinene, terpinolene; 2 sesquiterpene hydrocarbons, caryophyllene, humulene, and 3  
215 monoterpene alcohols, linalool, terpinen-4-ol et  $\alpha$ -terpineol (Figure 2). As expected for a citrus  
216 juice, D-limonene was widely dominant ( $16.5 \text{ mg}\cdot\text{kg}^{-1}$ ) and the contents of all the other  
217 compounds were below  $1 \text{ mg}\cdot\text{kg}^{-1}$ . These results were in accordance with the values presented  
218 in the literature. For instance, Shui et al. (2019) quantified in sweet orange juices  $24 \text{ mg}\cdot\text{kg}^{-1}$  of  
219 limonene,  $1.4 \text{ mg}\cdot\text{kg}^{-1}$  of myrcene,  $0.06 \text{ mg}\cdot\text{kg}^{-1}$  of  $\gamma$ -terpinene,  $0.20 \text{ mg}\cdot\text{kg}^{-1}$  of linalool,  $0.02$   
220  $\text{mg}\cdot\text{kg}^{-1}$  of caryophyllene<sup>17</sup> and Özkaya et al. (2019) quantified in satsuma mandarin  $0.07 \text{ mg}\cdot$   
221  $\text{kg}^{-1}$  of caryophyllene,  $0.12 \text{ mg}\cdot\text{kg}^{-1}$  of terpinen-4-ol,  $3.8 \text{ mg}\cdot\text{kg}^{-1}$  of  $\alpha$ -terpineol<sup>24</sup>. The major  
222 part of our juice was represented by clementine juice (*Citrus clementina* Hort ex Tan) which is  
223 a hybrid between orange (*Citrus sinensis*) and mandarin (*Citrus deliciosa* Ten) and thus  
224 displaying aroma compounds similar to their parents<sup>25</sup>. Furthermore, no specific volatile  
225 compounds such as nootkatone were identified in grapefruit juice, but we did find some aroma  
226 compounds typical of pink grapefruit such as limonene and linalool ( $30$  and  $0.33 \text{ mg}\cdot\text{kg}^{-1}$   
227 respectively)<sup>26</sup>.

228

229 The OAV results (Table 2) showed that D-limonene, terpinolene, linalool, caryophyllene, and  
230 terpinen-4-ol had olfactory activity values that were higher than 1, which means that these  
231 compounds clearly contributed to the typical aroma of the citrus juice. In this study, particularly  
232 terpinen-4-ol (130), but also linalool (6.9) and terpinolene (5.6), had the highest OAV and were  
233 therefore powerful aromatic compounds in the product. As already mentioned, monoterpene  
234 alcohols played a significant role in the odor/aroma activity of citrus fruit <sup>18</sup>. They included  
235 terpinen-4-ol and linalool that contributed significantly to the citrus and floral odors <sup>27</sup>.

236

### 237 *3.3 Effect of the process on the profile of the aroma compounds*

238 During the microfiltration process of citrus juice, the majority of aromatic compounds were  
239 concentrated by the membrane along with the carotenoids and flavonoids identified in the  
240 previous study <sup>9</sup>. However, the concentration factors ranged over a large interval (Table 3)  
241 which showed that losses in the final retentate varied from one compound to another. For DF0,  
242 among the 36 identified aroma compounds, losses were below 20% for 5 compounds, between  
243 20 and 50% for 7 compounds, between 50 and 80% for 13 compounds and, above 80% for 11  
244 compounds. Globally, terpene hydrocarbons were more concentrated, i.e. were less lost, than  
245 oxygenated compounds.

246

247 The incomplete retention by the membrane of the aroma compounds that passed through the  
248 membrane in the permeate mainly explained these losses. According to Chaparro et al. (2017),  
249 volatile compounds could have different degrees of association with the solid fraction of the  
250 juice <sup>28</sup>. Thus, the compounds most strongly bound to the pulp could be concentrated in the  
251 retentate more easily. Bali (1991) showed that some aroma compounds could be adsorbed on  
252 the vegetal cell walls present in the pulp and would therefore be retained by the membrane <sup>29</sup>.  
253 As mentioned by Cisse et al. (2005), terpene hydrocarbons were highly apolar and a significant

254 portion of these compounds was probably associated through weak bonds to the insoluble  
255 fraction which contains hydrophobic sites <sup>12</sup>.

256 Although when using membranes with a large pore diameter, physico-chemical interactions  
257 between solutes and membrane material are often considered as negligible, they could still be  
258 involved in the differential retention of the aroma compounds. In this study, the membrane  
259 material is particularly hydrophilic ( $\alpha$ -Al<sub>2</sub>O<sub>3</sub>) which may have disfavored the transmembrane  
260 transfer of the more hydrophobic compounds. Moreover, some aroma compounds may have  
261 also been lost by volatilization during microfiltration or undergone changes in their molecular  
262 structure through oxidation reaction for instance. This assumption was the only one that could  
263 explain the concentration factors much lower than 1 that were obtained for carvone, linalool,  
264 decanol and trans-linalool oxide.

265 Guliani et al (2021) have shown that carvone and linalool are highly volatile compounds <sup>30</sup>.  
266 Furthermore, they are unstable and have a short half-life. Linalool is sensitive to chemical  
267 modification (mainly oxidation) because of the double bonds and hydroxyl group present in its  
268 structure <sup>31</sup>. According to the results obtained with DF1, the diafiltration step led to a slight  
269 diminution of the concentration factors for all compounds except for 5-hepten-2one, (-)  
270 carvone, humulene, caryophyllene,  $\alpha$ -copaene, octanal and decanal. This difference could be  
271 explained by the water added during the diafiltration step that leached out the compounds not  
272 totally retained in the permeate and possibly also by the longer duration of the trial that needed  
273 4 more hours.

274 These results were confirmed by the quantification of the 9 main volatile compounds obtained  
275 by standard addition calibration (Figure 2). The amount of terpene hydrocarbons, D-limonene,  
276  $\gamma$ -terpinene, caryophyllene, humulene and terpinolene were similarly 4 to 5-fold concentrated  
277 by CMF (as an example 64.7; 0.59; 0.67 0.41 and 0.99 mg·kg<sup>-1</sup> in the DF0 retentate  
278 respectively). On the contrary, terpinen-4-ol and  $\alpha$ -terpineol contents were increased only by

279 around 20% in the DF0 concentrate but were close to the initial juice for the DF1 concentrate.  
280 Finally, the concentration of linalool markedly decreased in both concentrates (-35 to 47%).  
281 Comparing the DF0 and DF1 concentrates, the diafiltration step led to a decrease of aroma  
282 content between 18 and 33% for most of the compounds and up to 57% for  $\alpha$ -terpineol.  
283 Surprisingly, the concentration of sesquiterpene hydrocarbons increased in the DF1 retentate.  
284 These results could be related to a modification of the solid-liquid adsorption equilibrium  
285 during the diafiltration step<sup>32</sup>.  
286 These trends were reflected in OAV calculation (Table 2). By associating the total OAV to the  
287 odor and aroma perception intensity, CMF globally reinforced the aroma power of the final  
288 product especially in the case of DF0. But at the same time, the process considerably modified  
289 the aroma profile by making it evolve towards the terpene hydrocarbons which could be  
290 considered as less fine than the oxygenated compounds. So, CMF produced final products that  
291 are of interest in terms of flavor quality but the consequences of the aroma distortions induced  
292 by the process on the sensorial perception needs in depth evaluations.

293

#### 294 *3.4 Correlation between the volatiles compounds and sensory attributes of citrus concentrates*

295 PCA based on concentration factors and sensory attributes as well as some physico-chemical  
296 parameters was used to distinguish the two citrus concentrates (Fig. 3). The citrus concentrates  
297 were clearly differentiated and explained by axis 1. DF0 concentrate was characterized by a  
298 better aromatic profile responsible for the citrus-floral odor and the judges gave it a higher  
299 overall quality score. Favorable sensory attributes such as citrus odor and aroma, overall  
300 quality, mouthfeel persistence, and the sweet and sour taste were strongly correlated (from 0.97  
301 to 0.99 Pearson matrix of correlations) with some aroma compounds: p-menth-1-ene, cis-p-  
302 Menth-8-ene,  $\gamma$ -terpinene,  $\alpha$ -terpineol, 1,2-dihydrolinalool. These volatile compounds had a  
303 major impact on the perceived intensity of the sensory quality of DF0 citrus concentrate as

304 confirmed by the sensory panel. We can also notice that favorable sensory attributes were  
305 strongly correlated to titratable acidity (TA) and sugar/acidity ratio (S/A).

306 Opposite and negative correlations (from -0.95 to -0.99) were observed for some aromatic  
307 compounds (humulene, octyl acetate, 6-methyl-5-hepten-2-one,  $\alpha$ -copaene and caryophyllene).  
308 Conversely, they were positively correlated with undesirable attributes (astringency, chemical  
309 oxidized and fermented impression) corresponding to DF1, that were less appreciated by the  
310 sensory panel.

311 Other strong correlations (from 0.86 to 0.98) obtained from Pearson matrix correlations  
312 between sensory attributes (Viscosity M and V; granulometry M and V) and physico-chemical  
313 parameters (particle diameter and limit viscosity) were observed and confirmed the scores  
314 obtained from the sensory panel and the difference between the two concentrates. DF1 was  
315 grainier and less viscous than DF0.

316

### 317 *3.5 Retention evaluation and modelling*

318 Except for the 4 aroma compounds that were obviously lost during the process (carvone,  
319 linalool, linalool oxide and decanol), apparent retentions were calculated thanks to the simple  
320 model described in the material and method section. As shown in Figure 4 that compares the  
321 experimental and calculated concentration factors values, the model provided a satisfactory  
322 representation of the behavior of the great majority of the aroma compounds during the process.

323 Although it slightly overestimated concentration factors for DF0 and underestimated them for  
324 DF1, the calculated points were very close to the first bisector. The model was less efficient for  
325 the 3 measured sesquiterpene hydrocarbons ( $\alpha$ -copaene, caryophyllene, humulene) and for 6-  
326 methyl-5-hepten-2-one. However, these results showed that the model could effectively be used  
327 to evaluate the volatile compounds retention during CMF processing. So volatilization or  
328 chemical change were not the predominant phenomena involved in the loss of aroma

329 compounds. The modifications of the aroma profile during processing could mainly be  
330 explained by simple physical phenomena, retention being related to a more or less strong  
331 association with the insoluble fraction.

332 Comparing the retentions ( $R_i$ ) for the 32 identified aroma compounds (Figure 5), we showed  
333 that almost all terpene hydrocarbons were highly retained by the membrane with  $R_i$  ranging  
334 from 1.00 to 0.63. Two monoterpene hydrocarbons appeared as exceptions with lower  
335 retention: myrcene ( $R_i = 0.50$ ) and ocimene ( $R_i = 0.16$ ). On the contrary, the oxygenated  
336 compounds were less retained with  $R_i$  from 0.62 to 0.18. Terpene hydrocarbons were very  
337 apolar and, these results confirmed that a significant portion of them could be adsorbed on the  
338 pulp, explaining their high retention.

339

#### 340 **4. Conclusion**

341

342 Globally, this study investigated the organoleptic characteristics of a potential functional citrus  
343 based food obtained by membrane technology. The impact of CMF with or without the  
344 diafiltration step led to a very different perception of citrus products linked to aroma compounds  
345 and the sugar/acidity ratio. The concentrate obtained without diafiltration had a better aromatic  
346 profile responsible of citrus-floral odor than the diafiltered concentrate and obtained a higher  
347 overall quality score. However, both citrus products were well noted by the trained panelists  
348 and can be consumed directly as a pleasant healthy drink as well as a nutrient-dense beverage.  
349 Even if it is less appreciated, the diafiltered concentrate is still interesting because the  
350 diafiltration allowed to decrease the solute content of the product (sugars, organic acids) and  
351 thus to increase the purity of carotenoids and hesperidin compared to dry matter. Thanks to a  
352 simple transfer model, the impact of the membrane process on the aroma profile has been  
353 evaluated by the experimental and calculated concentration factors. Finally, although the low  
354 retention or the losses of 4 oxygenated compounds were observed, the main apolar terpene were

355 highly retained by the membrane that could be related to a more or less strong association with  
356 the insoluble fraction. Further investigations are needed to better understand the differential  
357 affinity of the aroma compounds for the pulp and the involved phenomena. For an industrial  
358 use, it would also be necessary to complete the evaluation of the process by applying it to a  
359 fresh fruit juice. Finally, *in vivo* studies have to be performed in order to validate the health-  
360 functionality of this innovative citrus-based food.

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### 365 **Acknowledgements**

366 The authors thank the Algerian government and CIRAD for their financial contribution.

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- 495

496 **Table 1.** Definition of the attributes used to describe sensory characteristics of the citrus  
 497 concentrates.  
 498

Attributes	Definition
<b>Odor</b>	
Citrus	Intensity of the characteristic odor of citrus fruits such as orange, lemon, etc.
Clementine	Intensity of the characteristic odor of clementine.
Grapefruit	Intensity of the characteristic odor of grapefruit.
Artificial	Intensity of the odor of sweet candy, chemical, etc.
<b>Visual</b>	
Brightness	A shiny or glossy appearance resulting from the tendency of a surface to reflect light at one particular angle more than at another.
Granulometry	Texture property related to the perception of the size, shape, and number of particles in the product.
Viscosity	Texture property related to flow resistance.
<b>Flavor</b>	
Sweet taste	Key flavor (sucrose, candy).
Sour taste	Fundamental flavor (citric, tartaric acid).
Bitter taste	Fundamental flavor (quinine solution, caffeine, etc.).
<b>Impression</b>	
Astringency	Contraction of the surface of the mucous of the mouth resulting in dryness in the middle and back of the mouth.
Oxidized, fermented Chemical	Impression of oil degraded by contact with oxygen, aged juice. Which is not natural (detergent, soap, solvent, etc.).
<b>Mouthfeel</b>	
Granulometry	Texture property related to the perception of the size, shape, and number of particles in the product.
Viscosity	Texture property related to flow resistance.
<b>Aroma</b>	
Citrus	Characteristic aroma of citrus fruits such as orange, lemon, etc.
Clementine	Characteristic clementine aroma.
Grapefruit	Characteristic grapefruit aroma.
<b>Persistence in the mouth</b>	Evaluation of the intensity of the time during which the flavor or aroma remains in the mouth after swallowing the sample.
<b>Overall quality</b>	Represents the overall judgment on the juice. It takes into account all the sensory elements, the presence of atypical flavors and their importance as well as the particular characteristics of the fruit juice.

499

500 **Table 2.** Odour Activity Value (OAV) of the aroma compounds quantified in the initial juice  
501 and the concentrates.

	Threshold in pure water (mg·kg <sup>-1</sup> ) <sup>33,34,35</sup>	OAV (concentration/threshold)		
		Initial juice	Concentrate DF0	Concentrate DF1
β-myrcene	0.50	0.30	0.71	0.72
D-limonene	13.30	1.24	4.87	4.69
γ-terpinene	2.10	0.08	0.28	0.26
terpinolene	0.04	5.60	24.15	21.15
linalool	0.10	6.86	4.08	3.98
caryophyllene	0.06	2.36	10.41	15.74
terpinen-4-ol	0.01	130.47	174.87	137.53
humulene	0.12	0.82	3.43	4.94
α-terpineol	9.10	0.03	0.08	0.05

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503 **Table 3.** Aroma compounds identified by SPME/GC-MS analysis and concentration factors in both DF0  
504 and DF1 citrus juice concentrates (mean and standard deviation estimated from 3 repetitions).

Compounds	Concentration factors <sup>A</sup>		Kovats retention index		Odor description <sup>C</sup>
	In concentrate DF0	In concentrate DF1	calculated	Literature <sup>B</sup>	
<b>Monoterpene hydrocarbons</b>	9.94 (1.00)				
m-menthane	7.75 (0.98)	8.90 (1.73)	1018	nd	Peppermint <sup>40</sup>
$\alpha$ -pinene	9.53 (0.74)	6.78 (0.40)	1022	1022	Resin, pine tree, ethereal <sup>33</sup>
p-menthane	5.77 (0.05)	8.35 (1.30)	1034	1122	Pine, grapefruit
3-p-menthene	5.83 (0.48)	5.10 (0.33)	1054	1078	Peppermint, Grapefruit <sup>38</sup>
2-menthene	7.14 (0.42)	5.54 (0.55)	1060	1091	Herbal, grapefruit
cis-p-Menth-8-ene	4.90 (0.45)	6.39 (0.94)	1064	nd	nd
2-methyl-6-methylene 2-octene	4.73 (0.43)	4.61 (0.19)	1071	nd	nd
cis-2,6-dimethyl-2,6-octadiene	4.12 (0.46)	4.34 (0.01)	1074	nd	nd
2,6-dimethyl-2-trans-6-octadiene	8.30 (0.57)	3.85 (0.16)	1091	nd	nd
p-menth-1-ene	2.43 (0.28)	7.57 (1.12)	1113	1150	Citrus, grapefruit
$\beta$ -myrcene	4.05 (0.24)	2.43 (0.11)	1145	1145	Floral, musty, grape, geranium <sup>33,35,37</sup>
D-limonene	3.50 (0.14)	3.87 (0.11)	1188	1199	Lemon, citrus, minty <sup>21,35</sup>
$\gamma$ -terpinene	1.14 (0.13)	3.17 (0.12)	1229	1246	Woody, spearmint, sweet, citrus <sup>33-35,37</sup>
$\beta$ -ocimene	8.74 (0.45)	1.06 (0.05)	1239	1240	Herbaceous, tropical, sweet, warm <sup>21</sup>
p-cymene	4.53 (0.68)	7.75 (0.73)	1257	1272	Citrus, spicy, herbal <sup>37</sup>
terpinolene	2.37 (0.05)	3.92 (0.79)	1270	1283	Plastic <sup>37</sup>
6-methyl-5-hepten-2-one	6.12 (0.78)	4.37 (0.52)	1332	1338	Mushroom <sup>35</sup>
p-cymenene	0.17 (0.06)	5.74 (0.35)	1432	1432	Terpenic, fresh citrus, woody, spice
(-) carvone		0.51 (0.08)	1728	1715	Fresh, minty, herbal <sup>39</sup>
<b>Sesquiterpene hydrocarbons</b>					
$\alpha$ -copaene	6.24 (0.41)	10.22 (1.75)	1492	1492	Woody, earthy, spicy, sulfur, rubbery <sup>20,21</sup>
caryophyllene	4.57 (0.35)	6.90 (1.11)	1666	1667	Musty, green <sup>35</sup>
humulene	4.29 (0.09)	6.18 (1.07)	1597	1595	Sweet, green <sup>34</sup>
<b>Aldehydes</b>					
nonanal	1.99 (0.11)	1.31 (0.09)	1390	1391	Floral, citrus, soapy <sup>21,33,35,37</sup>
decanal	1.98 (0.21)	2.95 (0.68)	1505	1500	Soapy, green <sup>22</sup>
octanal	0.94 (0.11)	1.96 (0.17)	1279	1289	Spicy, herbal, floral, citrus, green <sup>33-35,37</sup>
<b>Alcohols</b>					
1,2-dihydrolinalool	2.55 (0.04)	2.25 (0.11)	1550	1537	Citrus, floral
linalool	0.61 (0.07)	0.59 (0.05)	1563	1547	Floral, fruity, sweet <sup>33,35,36,37</sup>
1-octanol	2.39 (1.04)	1.74 (0.16)	1575	1573	Citrus, sweet, oily
trans-linalool oxide (furanoid)	0.92 (0.22)	0.35 (0.19)	1448	1452	Floral, tea-tike, sweet <sup>36</sup>
terpinen-4-ol	1.38 (0.04)	1.08 (0.07)	1611	1602	Woody, floral, lilac <sup>35</sup>
1-decanol	0.36 (0.08)	1.71 (1.24)	1774	1771	Fresh, floral <sup>35</sup>
$\alpha$ -terpineol	1.62 (0.02)	1.25 (0.03)	1704	1697	Woody, spearmint, lilac <sup>37</sup>
thymol	1.66 (0.09)	1.30 (0.95)	2171	2189	Herbal
3,7-dimethyloctan-3-ol	3.55 (0.27)	2.89 (0.52)	1446	1400	Irritation, stink <sup>35</sup>
<b>Esters</b>					
ethyl acetate	1.76 (0.77)	1.50 (0.26)	848	854	Pear, fruity, solvent <sup>33,35</sup>
octyl acetate	0.97 (0.12)	1.53 (0.11)	1484	1483	Pungent, sour <sup>22</sup>

505 nd data not found; <sup>A</sup>calculated from the concentrations expressed as butanol equivalent; <sup>B</sup>Flavornet; <sup>C</sup>PubChem.

1 **Figure captions**

2 **Figure 1.** Sensory profile of the citrus concentrates DF0 and DF1. Descriptors preceded with  
3 the capital letters O, V, T, I, M and Ar refer to “Odor”, “Visual”, “Taste”, “Impression”, “in  
4 Mouth” and “Aroma” respectively. Significant difference \* < 0.05, \*\* < 0.01 between the DF0  
5 and DF1 concentrates by ANOVA Tukey test.

6 **Figure 2.** Quantification of the major aroma compounds in initial juice and concentrates (based  
7 on standard addition method).

8 **Figure 3.** Principal Component Analysis of sensory analysis results and concentration factors  
9 of the aroma compounds identified in the DF0 and DF1 citrus concentrates (concentration  
10 factors calculated from concentrations expressed as butanol equivalent).

11 **Figure 4.** Comparison between experimental and calculated concentration factors for the  
12 different aroma compounds detected in the DF0 and DF1 citrus concentrates (experimental  
13 concentration factors calculated from concentrations expressed as butanol equivalent).

14 **Figure 5.** Retention of the aroma compounds evaluated through the model (hydrocarbons in  
15 blue and oxygenated compounds in green).

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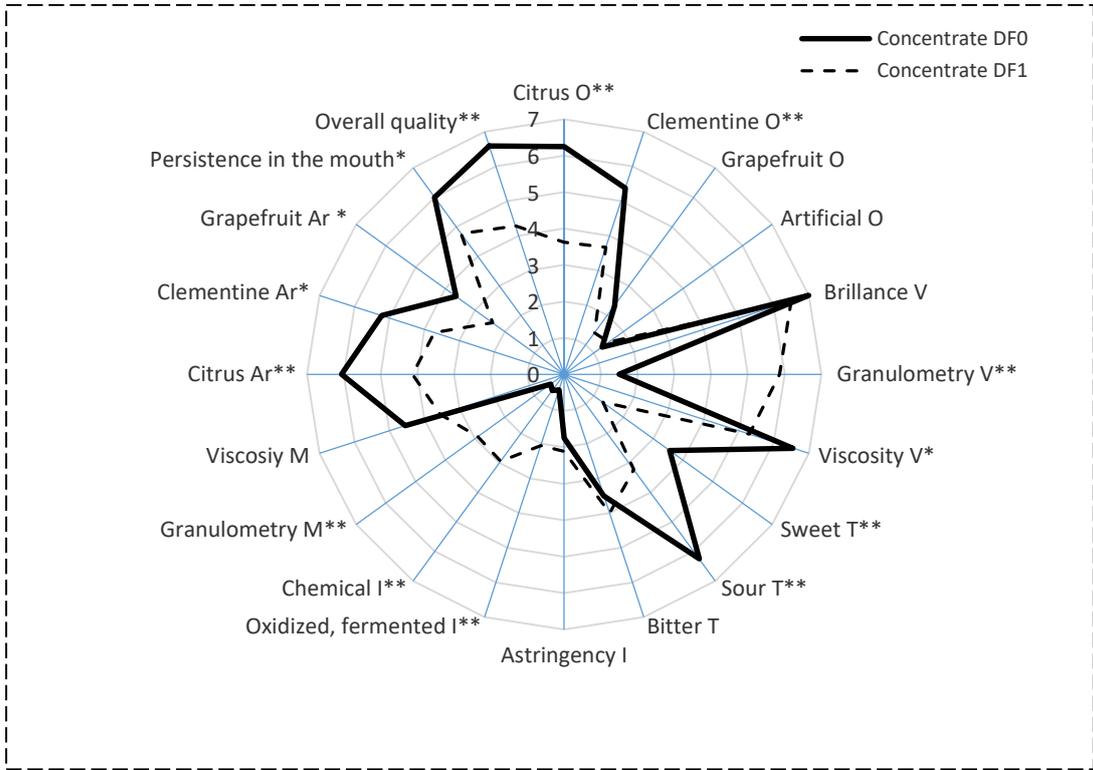
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27 **Figure 1.**

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46 **Figure 2.**

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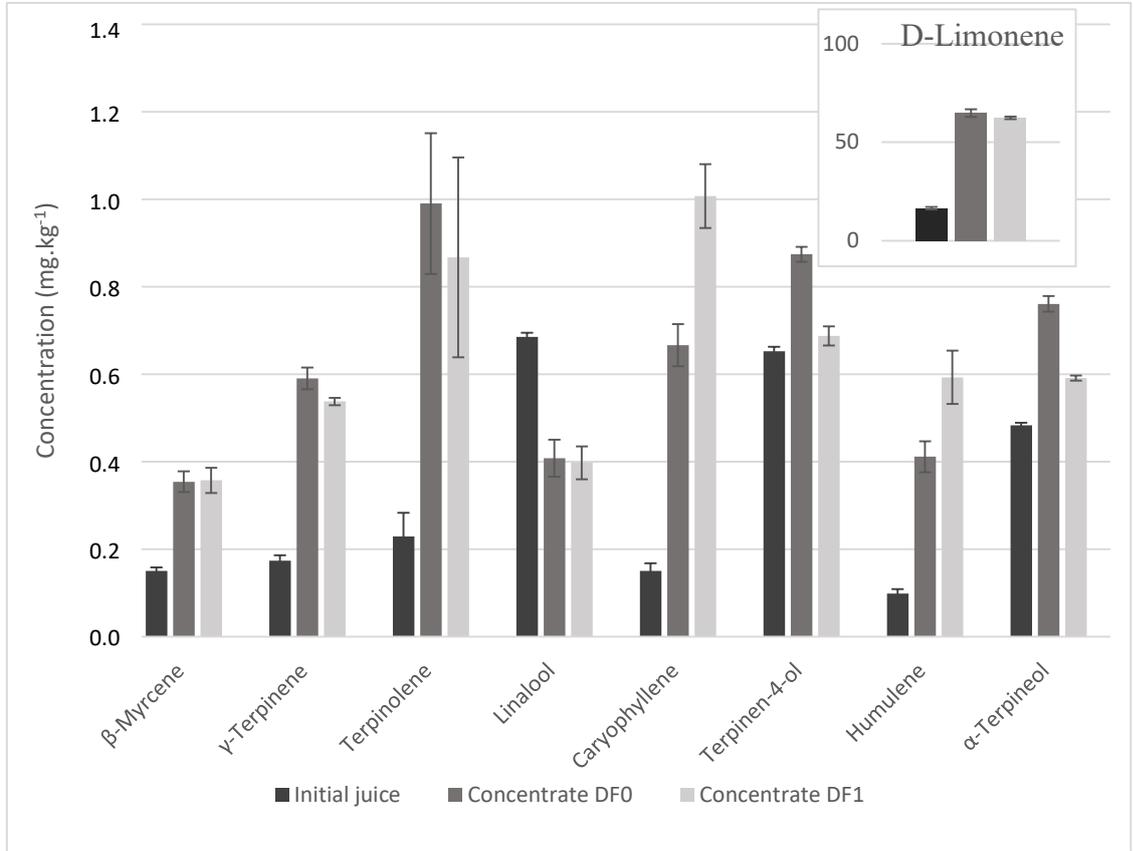
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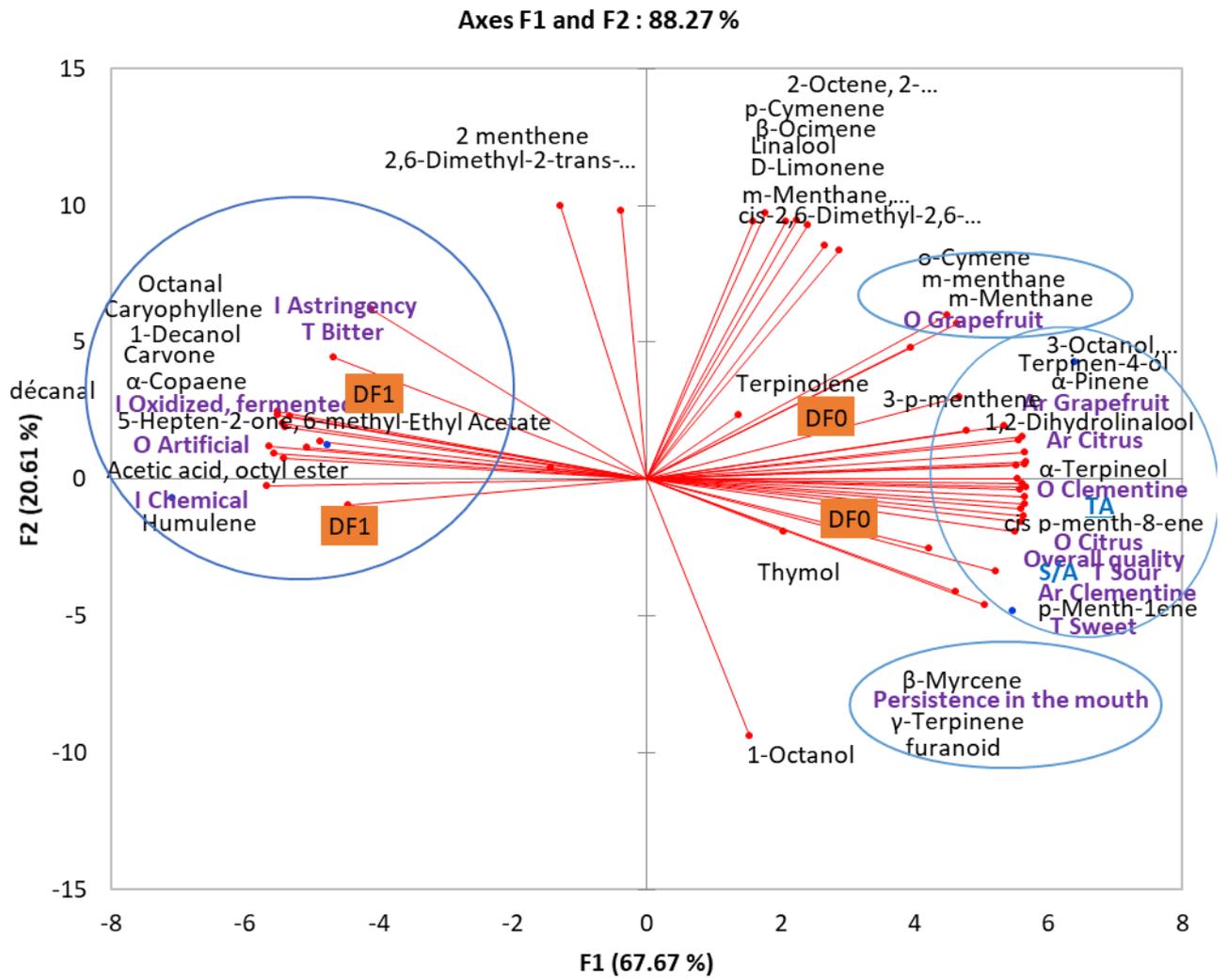
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66 **Figure 3.**

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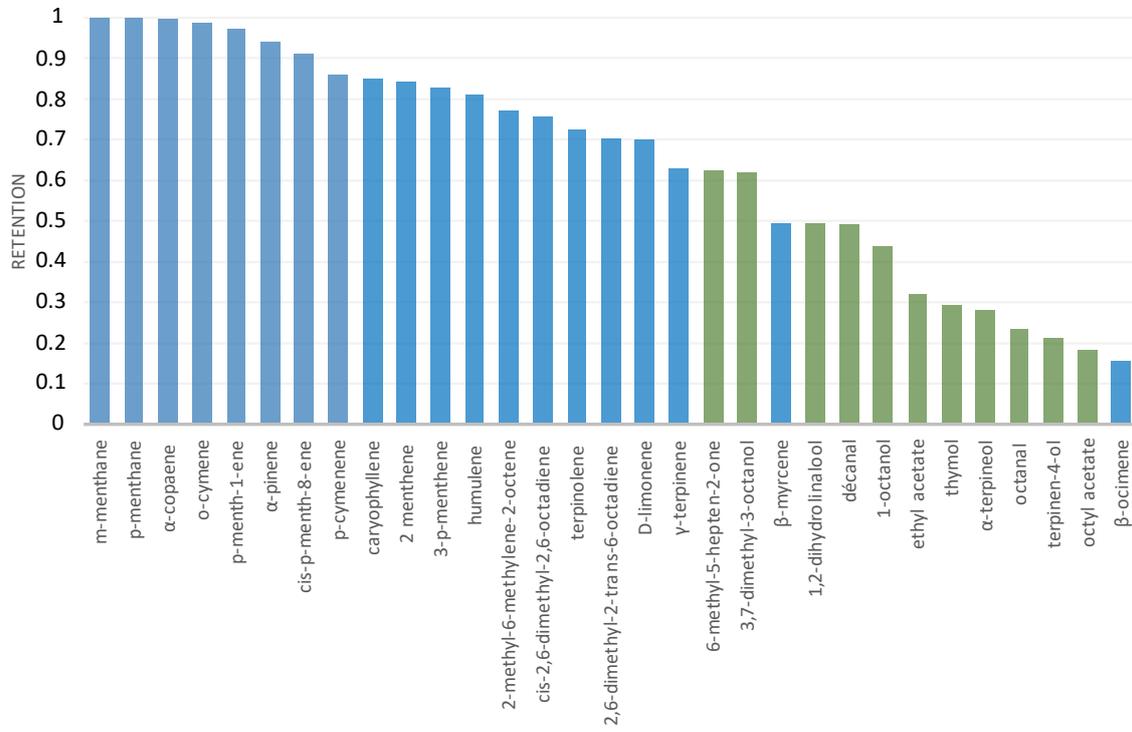


80 **Figure 5.**

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