

Title: Comparison of three types of drying (supercritical CO₂, air and freeze) on the quality of dried apple – Quality index approach

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1 **Abstract**

2 The aim of this study was to develop a quality index and examine the effects of drying apples using three
3 technologies (supercritical CO₂ drying, air drying and freeze drying) during a period of six months in
4 ambient conditions. Based on nine quality parameters (textural, colour and sensory properties), a
5 mathematical model for calculating a single total quality index (TQI) of dried apples packed in different
6 types of packaging in normal and modified atmosphere has been introduced.

7 At the beginning of the study, apples dried in supercritical CO₂ had the best scores. After six months,
8 samples dried in supercritical CO₂ and freeze dried apples, both packed in polyethylene coated
9 aluminium with 100% N₂, scored similarly. The six month shelf-life research revealed that measurable
10 changes occur during the second half of the shelf-life when it is possible to clearly distinguish differences
11 in the overall quality index of different dried apple slices.

12

13 **Key words:** supercritical drying; air-drying; freeze-drying; total quality index; apples

14

15 **1. Introduction**

16 **1.1 Food drying**

17 One of the oldest fresh fruit preservation techniques is air-drying (Mujumdar, 2014). Adequate
18 understanding of the heat/mass transfer mechanism and its correlation with drying parameters such as
19 temperature, velocity and relative humidity of the air used for drying is required for ideal quality dried
20 product (Unal & Sacilik, 2011). Dried foods should maintain qualities such as flavour, texture,
21 convenience, and functionality and high nutritional content (M. Shafiur Rahman, 2005). This is supported
22 by a review of literature showing that the majority of research analysed physical and mechanical
23 properties (Sette, Salvatori, & Schebor, 2016), colour (Ceballos, Giraldo, & Orrego, 2012) texture profile
24 analysis (Rizzolo et al., 2014) and sensory evaluation (Wojdyło et al., 2016). Besides physical quality
25 characteristics, several authors focused on chemical changes of dried food including total antioxidant
26 activity (Nindo, Sun, Wang, Tang, & Powers, 2003), the content of phenolics (Nayak, Berrios, Powers,
27 Tang, & Ji, 2011) and organic acids (Dupas de Matos et al., 2017; Michalska, Wojdyło, Honke, Ciska, &
28 Andlauer, 2018). As a result of inadequate drying of fresh fruit, various quality degradation processes
29 occur that reduce shelf-life and can cause food spoilage and food safety risks (Bonazzi & Dumoulin,
30 2011).

31 Currently, the most widely used drying techniques are air-drying and freeze-drying. Raghavi, Moses, and
32 Anandharamakrishnan (2018), in their latest review, assume that over 85% of industrial dryers are
33 convective, with hot air or combustion gases used as heat transfer media. Use of elevated air-drying
34 temperatures implies quality degradation of the fruit (Adiletta, Russo, Senadeera, & Di Matteo, 2016;
35 Sette et al., 2016). Indeed, processing fruits at elevated temperatures carries a risk that visual
36 appearance suffers and valuable and thermo-labile nutrients, such as vitamins or carotenoids, might be
37 degraded, and consequently, the fruits will lose their nutritional and health benefits (Polydera, Stoforos,
38 & Taoukis, 2005; Suvarnakuta, Devahastin, & Mujumdar, 2005). Freeze-drying ensures high quality
39 dehydration of fruit but can produce porous, brittle, amorphous and hygroscopic structures (de Santana
40 et al., 2015). Bonazzi and Dumoulin (2011) highlighted various aspects of dried product quality, such as
41 appearance in terms of colour and shape, taste as well as rehydration or dissolving rate, stability over
42 time and type of packaging.

43 Supercritical drying was recently introduced as an alternative process to conventional drying techniques
44 and is assisted by the use of supercritical fluids, usually CO₂ (García-González, Camino-Rey, Alnaief, Zetzi,
45 & Smirnova, 2012). In this process, supercritical CO₂ (scCO₂) is used to dry the product, but
46 simultaneously an inactivation of micro-organisms is achieved due to the antimicrobial activity of the
47 scCO₂. This type of drying is considered as an attractive preservation technology, meeting consumers'
48 demands for a product with high nutritional and sensory qualities (Ferrentino, Balzan, & Spilimbergo,
49 2013). Its main advantages are the relatively low temperature that avoids the thermal effects of
50 traditional heat preservation, retaining the food freshness, in combination with its decontaminating
51 effect (Spilimbergo, Komes, Vojvodic, Levaj, & Ferrentino, 2013).

52 Modified atmosphere packaging (MAP) is often used to maintain characteristics of fruits and vegetables
53 after harvest (Kader, Zagory, Kerbel, & Wang, 1989). This technology relies on modification of the
54 atmosphere by the respiration of the commodity and the permeability of the packaging material (Dash,
55 2014). As a result of using MAP, food keeps its quality characteristics during the extended shelf-life
56 (Farber, 1991).

57

58 **1.2 Food quality**

59 Food quality is considered as a complex concept measured using objective indices (Araujo et al., 2014).
60 Constraints in developing a single total quality score are use of different units for measuring various

61 quality parameters, and no consensus about the weight of each parameter (Finotti, Bersani, & Bersani,
62 2007). Various quality index methodologies were developed for different types of food such as extra-
63 virgin olive oil (Finotti et al., 2007), farmed tambaqui (*Colossoma macropomum*) (Araújo, De Lima, Joele,
64 & Lourenço, 2017) and mushrooms (Djekic et al., 2017b). M. Shafiur Rahman (2005) proposed a quality
65 index model based on nutrition, safety and sensory attributes of dried food. However, quality models for
66 innovative drying technologies such as supercritical drying processes have not been proposed.

67 Quality function deployment (QFD) is a tool developed to design quality aimed at satisfying the customer
68 and transforming the customer's demands into quality targets (Akao, 1990; ReVelle, 2004). The very first
69 step in applying QFD is to develop a house of quality (HOQ) and translate customer requirements to
70 quality characteristics (Park, Ham, & Lee, 2012). Such a HOQ enables the weight importance of each
71 quality characteristic to be calculated. Literature review reveals the use of QFD for chocolates (Viaene &
72 Januszewska, 1999), extra virgin olive oil, (Bevilacqua, Ciarapica, & Marchetti, 2012), Bulgogi bovine
73 meat, (Park et al., 2012) and organic products (Cardoso, Casarotto Filho, & Cauchick Miguel, 2015). No
74 QFD application to any food drying technology has been reported.

75 The aim of this study was to develop a quality index and examine the effects of drying apples using three
76 drying technologies (scCO₂ drying as novel technology compared with classical air drying and freeze
77 drying) stored for six months in ambient conditions. For the purpose of this study, based on nine quality
78 parameters, a mathematical model for calculating a single total quality index (TOI) of dried apples packed
79 in different types of packaging in modified atmosphere has been introduced.

80

81 **2. Material and methods**

82 Two independent research trials were performed in order to develop the quality model. The first trial
83 was designed to identify consumer preferences towards quality characteristics of dried apples. The
84 second trial included the changes of selected quality characteristics of dried apple slices during six
85 months of storage.

86

87 **2.1 Field research**

88 A survey on consumers' perception of quality of dried apples was conducted at the end of 2016. A total
89 of 85 respondents from Belgrade, which has the biggest and most developed food markets in Serbia,

90 were interviewed. The questionnaire consisted of two sections. The first section included general
91 demographic information about the respondents. The second section gave the respondents the
92 opportunity to rank eight sensory/quality characteristics of dried apples (skin colour, flesh colour, odour,
93 overall flavour, sourness, sweetness, hardness, and crispiness) from 1 = the least important for the
94 sensory quality to 8 = the most important for the sensory quality. These characteristics were chosen in
95 line with the research of Tomic, Radivojevic, Milivojevic, Djekic, and Smigic (2016) and M. Shafiur
96 Rahman (2005).

97

98 **2.2 Drying of apple samples**

99 Physiologically mature fruit of the 'Elstar' cultivar were harvested during the 2016 harvest season in a
100 commercial orchard in the Netherlands and stored for around one month in normal atmosphere at $1 \pm$
101 0.5 °C and 90–95% relative humidity before processing. Apples of approximately uniform size and
102 without obvious sunburn were cut into semi-circular slices ca. 50-55 mm in length and 2.2–2.5 mm thick
103 (Defraeye, 2017), without removing the skin, and dried using three different drying methods. Air drying
104 was performed using an Arcen air dryer type 7508 in a stagnant belt dryer (temperature 60 °C, drying
105 time 8 h). Freeze drying was performed in a 20L SuperModulyo freeze dryer (pressure: 0.2 mbar during
106 sublimation and 0.05 mbar during desorption: temperature of sublimation was maintained at -25 °C and
107 gradually increased to 40 °C during desorption; drying time 24 h). Supercritical drying was performed in a
108 patented pilot scale machine using scCO₂ (pressure 125 bar; temperature 50 °C; drying time 16 h),
109 (FeyeCon, 2010).

110

111 **2.3 Storage of dried apple**

112 Dried apple slices were packed under air or modified atmosphere (100% nitrogen) using different
113 packaging materials, as follows: single-layer polyethylene film (PE), multi-layer film consisting of
114 ethylene-vinyl alcohol film placed between two polyethylene films (EVOH-PE), and multi-layer film
115 consisting of aluminium film placed between two polyethylene films (Alu-PE). The packing combinations
116 are shown in Table 1. Each package contained cca. 100 g of dried fruit. Packed dried apple slices were
117 stored **in dark** at ambient temperature (≈ 22 °C) during 6 months and were sampled for analysis after 0
118 months (10 - 15 days after packing), 3 months and 6 months of storage.

119

120 **2.4 Colour changes**

121 To average big differences among slices within each type of sample, visual colour of 10 dried apples slices
122 was measured on both cut surfaces of each slice using a colour analyser (RGB-1002, Lutron Electronic).
123 Data were further expressed in CIELAB coordinates (L^* , a^* and b^*). Total colour difference (ΔE) was
124 determined by using equation 1 (Hunter & Harold, 1987):

125
$$\Delta E = \sqrt{(L^* - L_o^*)^2 + (a^* - a_o^*)^2 + (b^* - b_o^*)^2} \quad /1/$$

126 Values for a_o , b_o , L_o were obtained from the apples dried in scCO₂ just after drying to analyse changes
127 within the subset of scCO₂ apples obtained using the novel technology as well as changes related to the
128 common drying techniques, air and freeze drying.

129 Browning index (BI) of dried apples was calculated using equation 2 (Maskan, 2001; Oliveira, Sousa-
130 Gallagher, Mahajan, & Teixeira, 2012).

131
$$BI = \frac{(a^* - b^*)}{L^*} \quad \text{where} \quad a^* = \frac{100}{L^*} \sqrt{\frac{1}{2} [(L^* - a^*)^2 + (L^* - b^*)^2]} \quad /2/$$

132

133 **2.5 Texture profile analyses**

134 Texture profile analysis of the dried apples was conducted using a texture analyser (Brookfield CT3
135 Texture analyser). The trigger was set at 10 g. Dried apple slices were compressed with a sphere of 12.7
136 mm in diameter with a set deformation depth of 1.0 mm. The speed of the probe was 0.1 mm/s during
137 the penetration. The left and right sides of each slice were used for measurements. Hardness (as peak
138 loads of the compression cycles), cohesiveness (as ratio of energies expanded in compression) and
139 springiness (rate at which a deformed slice returns to its original size and shape) as quality parameters
140 were recorded and analysed. Measurements were performed under ambient conditions within 15
141 minutes after opening the packaging on eight dried apple slices in two replicates for each treatment.

142

143 **2.6 Sensory analysis**

144 Sensory quality rating was conducted by a trained 8-member panel consisting of researchers from the
145 University of Belgrade who were experienced in fruit quality judging. Over a period of three weeks, five
146 2-hour training sessions were performed using dried apple slices prepared in the laboratory by air drying
147 and scCO₂ drying, and three commercially available dried-apple snack products. The analysis was
148 performed using a 5-level quality scoring method as follows: excellent quality (quality score > 4.5); very

149 good quality ($3.5 < \text{score} \leq 4.5$); good quality ($2.5 < \text{score} \leq 3.5$); poor/unsatisfactory quality ($1.5 < \text{score}$
150 ≤ 2.5); very poor quality ($\text{score} \leq 1.5$). Four initially selected characteristics were evaluated: appearance,
151 odour, oral texture, and flavour. Each of the five integer quality scores (1-5) was divided into quarters, to
152 obtain a category scale with 20 alternative responses. The apple samples were labelled with random 3-
153 digit codes and presented to the panellists monadically in random order, three undamaged pieces per
154 assessor. In each case an additional separate apple sample, kept in a closed glass jar, was used for odour
155 judging. No strict instructions were given to the assessors whether to swallow or expectorate individual
156 bites. Low sodium bottled water was used for palate cleansing. All of the samples (Table 1) were
157 evaluated by the panel in two replications after 0 months (10-15 days after packing), 3 months and 6
158 months of storage. The sensory tests were performed in sensory booths in the sensory testing laboratory
159 at the University of Belgrade.

160

161 **2.7 Statistical analysis**

162 Objective colour and texture data were analysed by applying one-way ANOVA (with 'samples' as fixed
163 factor) or two-way ANOVA models combining 'storage time' with 'drying methods' (only the γ samples
164 included) or with 'packaging' (only scCO_2 -dried samples included) as fixed factors, followed by Tukey's
165 HSD *post-hoc* test. Sensory data were first subjected to 3-way ANOVA with 'samples' as fixed factor, and
166 'assessors' and 'replications' as random factors. The mixed model included main effects and all two-way
167 interactions. Then, in order to assess the influence of drying methods, storage time, and packaging on
168 sensory quality scores, two 4-way ANOVA models were applied (both with 'assessors' and 'replications'
169 as random factors): one included only scCO_2 -dried samples with 'storage time' and 'packaging' as fixed
170 factors; the second one included only the γ samples (Table 1) with 'storage time' and 'drying methods' as
171 fixed factors. Both mixed models took into account only the main effects and all two-way interactions.
172 Tukey's HSD test was used to separate the mean sensory scores.

173 The ranking data based on consumers' attitudes towards sensory quality characteristics of dried apples
174 were analysed using Friedman's test followed by the least significant difference *post-hoc* test (ISO, 2006).
175 The level of statistical significance was set at 0.05. Statistical processing was performed using Microsoft
176 Excel 2010 and SPSS Statistics 17.0.

177

178 **2.8 Quality function deployment**

179 The HOQ used in this paper (Fig. 1) consists of three elements: A: demanded quality (WHATs); B: quality
 180 characteristics (HOWs); C: relationship matrix (WHAT vs. HOW). This HOQ was modified according to
 181 Chan and Wu (2005), Park et al. (2012) and Djekic et al. (2017a). Consumer rankings of predetermined
 182 sensory attributes (skin colour, flesh colour, odour, overall flavour, sourness, sweetness, hardness, and
 183 crispiness), obtained within the field research, were used as inputs for defining the weight importance of
 184 defined quality characteristics. W_i is the weight importance of the 'i' demanded quality characteristics
 185 identified by the consumers. Relative weight is the percentage of the weight importance divided by the
 186 sum of all weight importance, equation 3.

$$= \frac{W_i}{\sum W_i} * 100 [\%]$$

187 /3/

188 The nine quality characteristics (HOWs) used in the matrix were the characteristics identified as colour
 189 parameters (ΔE and BI), sensory properties (appearance, odour, oral texture and flavour) and texture
 190 parameters (hardness, cohesiveness and springiness). Relationships between the WHATs and HOWs in
 191 order to identify weight importance were calculated using the following scale of relationships: 9 – very
 192 strong, 3 – strong, 1 – weak, and 0 – no relationship (Cardoso et al., 2015; Park et al., 2012). Absolute
 193 weight importance was calculated using equation 4:

$$= \sum W_i * RS_{ij} \quad /4/$$

195 where:

196 RW_i is the relative weight (WHATs) of 'i' demanded quality characteristic (n – number of demanded
 197 quality characteristics).

198 RS_{ij} is the relationship score (WHATs vs. HOWs) between demanded quality characteristic 'i' and product
 199 quality characteristics 'j' (m – number of product quality characteristics). Based on the absolute
 200 importance, the relative absolute weight importance (RAW) was finally calculated (Park et al., 2012).

201

202 **2.9 Total quality index**

203 The quality parameters were divided into three groups, in line with the work of Finotti et al. (2007).
 204 Parameters of the first kind are those with a target value. The following rule applies – 'the nearer to the
 205 target value the parameter is, the better the quality is', equation 5:

$$= \frac{2 * (-)}{-}$$

206 /5/

207 where:

208 QI – quality index for a parameter; x_i – measured value in the subset of values; T - target value; x_{max} –
 209 maximal value in the subset of values; x_{min} – minimal value in the subset of values. Four sensory
 210 attributes were included in this rule (target values = 5). Parameters of the second kind have the following
 211 rule: 'the smaller the value is, the better the quality is'. For this type of parameter, QI is calculated based
 212 on equation 6:

213
$$= \text{---} \quad /6/$$

214 where:

215 QI is the quality index for a specific quality parameter. Colour parameters were included in this group (ΔE
 216 and BI). Parameters of the third kind have the following rule: 'the higher its value, the better the quality
 217 is'. For this type of parameter, QI is calculated based on equation 7:

218
$$= \text{---} \quad ; x_i \leq x_{max} \quad /7/$$

219 where:

220 QI is the quality index for a specific quality parameter. Texture quality parameters were included in this
 221 group.

222 Upon calculation of all QIs, we can assume that in the new Euclidean space R^m (m is the number of
 223 quality parameters) quality indexes are considered as vectors $QI = (QI_1, QI_2, \dots, QI_m) \in R^m$ (Horn &
 224 Johnson, 1985). The Euclidean norm of the vector, whose components are the indexes QI, multiplied by
 225 weighting factors (RAW) will represent the overall total quality index (TQI) equation 8 (Finotti et al.,
 226 2007).

$$= \text{---} *$$

227 /8/

228 As a conclusion, the rule of thumb is that the further from the origin the vector, the worse its TQI is, and
 229 the nearer to the origin the vector, the better the TQI (Finotti et al., 2007).

230

231 **3. Results and discussion**

232 **3.1 Field research**

233 The results of the survey on consumer attitudes towards the eight selected sensory quality
234 characteristics of dried apples as a snack product are shown in Fig. 2. According to the consumers'
235 opinion, 'flavour' was the attribute ranked as the most important for the sensory quality of the product
236 in question, while 'crispiness' was the least important ($\alpha = 0.05$). The differences among the rest of the
237 included characteristics (skin or flesh colour, odour, hardness, sweetness, and sourness), related to their
238 influence on the sensory quality, were not statistically significant ($p > 0.05$), which implies that they are
239 equally important to the consumers. This information was included within the 'demanded quality'
240 element (WHATs) in QFD.

241

242 **3.2 Colour changes**

243 In this study, different drying technologies initially induced colour changes (Table 2). The colour of air
244 dried and freeze dried apples were statistically different compared to the colour of apples dried in scCO₂
245 ($p < 0.05$) in all three measurement periods. After six months, all dried apple slices (except A- γ) showed
246 significant differences compared to the beginning of the study. Depending on the value of ΔE , when this
247 value is below 2.0, trained observers would notice the difference, while when this values is over 3.5, a
248 clear difference in colour is noticed even by average observers (Mokrzycki & Tatol, 2011). The largest
249 colour differences were for A- γ dried apple slices. Colour changes can occur due to degradation of
250 pigments or non-enzymatic Maillard browning (Dadali, Demirhan, & Özbek, 2007). Colour retention of
251 dried fruits and vegetables can indicate retention of the pigments and nutrients such as carotenoids,
252 flavonoids, phenols, chlorophyll and betalains (Devahastin & Niamnuy, 2010). Soliva-Fortuny, Grigelmo-
253 Miguel, Odriozola-Serrano, Gorinstein, and Martín-Belloso (2001) showed that the faster the initial
254 polyphenol oxidase activity decays, the less the colour changes on apples in MAP occur. When the colour
255 of the dried product is considered as a quality indicator, the colour parameters can be used to optimise
256 the drying process and minimise the degradation of important compounds (Aral & Beşe, 2016).

257 The browning index (BI) is used to characterise the overall changes in browning colour and is one of the
258 most common indicators of browning in food products containing sugar (Quitão-Teixeira, Aguiló-Aguayo,

259 Ramos, & Martín-Belloso, 2008). Browning of apples results from both enzymatic or non-enzymatic
260 reactions and can differ depending on the apple cultivar (Putnik et al., 2017). The formation of browning
261 in dried fruits is often associated with the non-enzymatic Maillard reaction (Baini & Langrish, 2009;
262 Persic, Mikulic-Petkovsek, Slatnar, & Veberic, 2017). Assessing the formation of browning in dried food
263 helps in the selection of an appropriate drying technique, which minimises the degradation of quality in
264 terms of colour (Pathare, Opara, & Al-Said, 2013). Our results show the BI increased over time and was
265 initially the largest for air dried apple slices compared to other dried apple slices ($p < 0.05$). The colour
266 changes in dried apple slices C- α , C- β and A- γ , reflected through the BI, were statistically significant
267 ($p < 0.05$) after six months of storage.

268 Two way ANOVA confirmed statistically significant interactions between different 'drying technologies'
269 and 'storage time', both in colour differences and BI ($p < 0.05$).

270

271 **3.3 Sensory analysis**

272 The results of sensory quality judging are shown in Table 3. Different ANOVA models applied to the
273 quality scores of the evaluated sensory characteristics showed significant changes in sensory quality as
274 affected by 'drying method', 'storage time' and 'packaging'. The most affected were the dried apple
275 slices packed in PE/Air (C- α) and EVOH-PE/N₂ (C- β), followed by the air-dried apple (A- γ). At the end of
276 the observed period, the quality scores of C- α and C- β dried apple slices were within the ranges of 'poor'
277 and 'very poor' quality, while the A- γ dried apple slices retained their initial sensory quality to a greater
278 extent than the former two. Air-drying of apple slices led to product characterised by pronounced shape
279 deformation (incurved and twisted shapes), well preserved skin colour, yellowish-brown colour of flesh,
280 as well as pronounced hardness, brittleness, and apple odour and flavour.

281 The effects of different types of packaging on the sensory quality of dried apple were assessed by
282 observing only the three scCO₂-dried apple slices (Table 1). The effect of scCO₂ drying was reflected in
283 partly deformed shape of the apple slices, the appearance of reddish/pinkish discolorations in flesh
284 originating from the skin colour, the appearance of cracks on the flesh surface, moderate crispiness,
285 good chewiness, and pleasant apple flavour. Supercritical fluids have been used in the food industry as
286 extraction solvents, and actually, the drying process in scCO₂ drying is an extraction process in which
287 water is not removed by vaporisation or sublimation but is dissolved in the scCO₂ (Bourdoux, Li, Rajkovic,
288 Devlieghere, & Uyttendaele, 2016). Water is not the only matter that dissolves during this extraction

289 process. The reddish/pinkish discoloration of flesh obviously occurs as a result of dissolving of the apple
290 skin pigments in the supercritical fluid and their diffusion from skin into the flesh, where the scCO₂
291 serves as a carrier. According to the ANOVA results, it seems that after a relatively short storage period
292 for dry fruits (3 months), 'type of packaging' did not affect the evaluated sensory characteristics. A
293 statistically significant decrease was found only in the texture quality of γ -packaged apples compared
294 with apple slices in α and β packaging (Table 4). Decreases in quality of practical significance were
295 observed only in C- α and C- β apple slices after 6 months of storage (the scores were within the ranges of
296 'poor' and 'very poor' quality). After six months of storage, C- α and C- β apple slices became darker
297 yellow-brown to greyish-brown in colour, typical apple odour and flavour were lost and replaced by hay-
298 like odour and empty dried-fruit flavour, crispiness had completely disappeared and they became more
299 cohesive and flexible, more adhesive on first bite and chew, and also had increased chewiness. These
300 intense changes in sensory characteristics could primarily be attributed to the changes in water activity
301 (a_w) during the storage time, which promotes changes in both physical characteristics of the product
302 matrix and the oxidation rate. In air-packed apple slices (C- α), oxidation changes could further be
303 supported by the presence of oxygen. Starting from 0.19 on average after drying (Table 1), the a_w of the
304 apple slices packed in both PE and EVOH-PE (C- α and C- β) significantly increased after 6 months of
305 storage ($p < 0.01$), in contrast to the Alu-PE packed apple slices in which the increase was not statistically
306 significant at the same level of significance (data not shown). The final average a_w values for C- α and C- β
307 apple slices were 0.46 and 0.48, respectively. Oxidation processes and browning can occur rapidly in
308 dehydrated fruit products depending on the a_w (Lavelli & Corti, 2011). With the exception of lipid
309 oxidation, an increase in a_w can enhance the rate of oxidation by increasing mobility of reactants and
310 bringing existing catalysts into solution (M. Shafiur Rahman, 1995). Investigating the effect of drying and
311 long-term storage on phytochemical contents of apple pomace, Lavelli and Corti (2011) found that air-
312 drying at 60 °C was better than vacuum-drying at 40 °C in terms of anthocyanin and flavanol retention,
313 which could be due to a longer exposure to unfavourable moisture levels during drying. They also found
314 that stability of these antioxidants during storage was greatly affected by the a_w level that ranged from
315 0.11 to 0.75, reporting that the maximum stability was accomplished for all investigated apple
316 phytochemicals at the lowest a_w level. C- α sample resulted in lower score values as compared to C- β
317 after six months of storage, since some of the panellists noticed musty flavour and pointed in their
318 comments traces of mould on some apple slices. The presence of musty sensory properties and moulds

319 are probably correlated with each other. Packages had no traces of damage prior to opening and storage
320 conditions were the same for all samples. Plastics are relatively permeable to small molecules such as
321 gases and water vapour, ranging from excellent to low barrier values, which are important in the case of
322 food products (Siracusa, 2012). Compared to e.g. polyvinylidene dichloride and polyester, PE has the
323 lowest water vapour transmission rate (Kropf, 2004). Aluminium foil is considered to be impermeable to
324 light, water vapour and gases, while EVOH is a good barrier when embedded within waterproof layers,
325 such as PE (Lamberti & Escher, 2007).

326 Taking only γ apple samples into account (the apple slices packed in Alu-PE/N₂, Table 1), the results
327 showed that 'drying method' significantly affected appearance and texture over the observed storage
328 time. The best-preserved sensory characteristics were found in the freeze-dried apple sample (F- γ). After
329 six months of storage, all of the evaluated characteristics of F- γ apple slices retained their initial level of
330 sensory quality ('excellent' or 'very good'). The F- γ apple sample was characterised by apple slices of
331 regular shape (not deformed), pale yellow colour of flesh without red discolorations, typical apple
332 flavour with pleasant sourness, crispiness (at some level even after six months of storage), low hardness,
333 and also good chewiness. In comparing the effects of scCO₂ drying and freeze-drying, the results showed
334 no statistically significant differences in quality scores between C- γ and F- γ apple slices (with the
335 exception of 'appearance') over the period of storage. All of the quality scores of C- γ apple slices were in
336 the range of 'very good' quality. Unlike F- γ , the C- γ apple slices were characterised by reddish/pinkish
337 discolorations of flesh, shape deformations of apple slices (at low level), the presence of cracks on the
338 flesh surface, as well as lower intensity of apple flavour. These results reveal the potential of supercritical
339 drying, as an emerging drying technology. This type of drying can retain dried apple slices for at least six
340 months with similar sensory quality level as obtained by freeze-drying, provided the product is packed in
341 non-permeable materials under inert atmosphere, such as Alu-PE/ N₂. However, disadvantage may lie in
342 the fact that scCO₂ affects the migration of pigments from the peel into the flesh during drying.

343

344 **3.4 Texture profile analysis**

345 Hardness of dried apples showed a gradual decrease for all samples over the storage period, with the
346 greatest decreases found in C- β and F- γ (Table 4). Results of Kutyla-Olesiuk, Nowacka, Wesoly, and
347 Ciosek (2013) confirm that drying methods influence mechanical properties of dried apples. This is

348 mainly since water content has an impact on the loss of the fragility of dried products (Labuza et al.,
349 2004).

350 Comparison of hardness of different dried apple slices during the same storage period showed no
351 statistical differences ($p > 0.05$). Taking the storage time as a factor, significant decreases in hardness
352 were found in dried apple slices C- α , C- β and F- γ , mainly after the period of six months (Table 4). In
353 addition, 2-way ANOVA showed no statistically significant interactions in hardness ($p > 0.05$), neither
354 between 'storage time' and 'drying methods' as factors (only the γ apple slices included), nor between
355 'storage time' and 'packaging' as factors (only scCO₂-dried apple slices included). The reason the greatest
356 decrease in hardness was found in C- α and C- β dried apple slices could be related to the significant
357 changes in a_w observed in these samples after both 3 and 6 months of storage. After 3 months of
358 storage, the a_w levels increased on average to 0.33 and 0.35, and after 6 months to 0.46 and 0.48 in C- α
359 and C- β apple slices, respectively (starting from 0.19). In contrast, in the γ dried apple slices, the increase
360 in a_w was not statistically significant ($p > 0.01$). A negative correlation between moisture content and
361 hardness of dried fruit is expected, as was presented in the work of Ansari, Maftoon-Azad, Farahnaky,
362 Hosseini, and Badii (2014), stating that this behaviour is related to the transition of the glassy dried fruit
363 (tough to deform) into rubbery rehydrated fruit (easy to deform). Rahman and Al-Farsi (2005) and Seow
364 and Thevamalar (1988) confirm hardness as a function of moisture content attributing this behaviour to
365 this transition.

366 Cohesiveness was the texture characteristic that showed significant changes in values ($p < 0.05$), taking
367 into account both the period of storage and drying methods (Table 4). Also, two way ANOVA confirmed
368 that there was a statistically significant interaction in cohesiveness between different drying
369 technologies and storage time ($p < 0.05$).

370 Freeze-drying of apple slices resulted in a lower level of the product springiness ($p < 0.05$) as compared to
371 scCO₂-drying and air-drying methods (Table 4). However, at the end of the shelf-life, dried apple slices
372 showed no statistically significant differences in springiness ($p > 0.05$). Results show that during the shelf-
373 life, C- γ and F- γ dried apple slices expressed statistically significant differences ($p < 0.05$). Two way ANOVA
374 confirmed that there was no statistically significant interaction in springiness ($p > 0.05$) between different
375 'drying technologies' and 'storage time' as factors.

376

377 3.5 Quality function deployment

378 Upon completion of the field research and laboratory testing of dried apples during the six-month
379 period, the next step was to complete the HOQ and establish the absolute and relative importance of
380 each quality characteristic. Fig. 3 reports the relative and absolute importance of the quality
381 characteristics for dried apples packed in modified atmosphere. The three most important characteristics
382 are flavour with 21.5% of RAW, followed by total colour difference (20.1%) and odour (15.8%). These
383 results prioritise customer requirements, which would help in tailoring quality characteristics of the dried
384 apple slices. On the other hand, cohesiveness is among the characteristics that have a low impact on the
385 overall quality of the dried products.

386

387 **3.6 Total quality index**

388 Fig. 4 shows the final TQI scores of the dried apples. At the beginning of the study, apples dried in scCO₂
389 (regardless of the packaging) had the best TQI scores. After three months similar results were obtained
390 for apple slices dried in scCO₂ and freeze dried apples (scores between 0.39-0.44). Air dried apple slices
391 had the worst score at three months. However, after six months, C- α and C- β dried apple slices
392 expressed the worst scores while C- γ and F- γ had similar scores.

393 This method of calculating a unique TQI is capable of comparing and evaluating apples dried in different
394 drying technologies and packed in different MAPs in a quantitative way. It is sensitive to any
395 displacement of QI from their optimal and/or target values (Finotti et al., 2007). Also, this model can
396 enable a large-scale comparison of various products packed in MAPs and was found a reliable, precise,
397 and simple tool for monitoring TQI during shelf-life (Djekic et al., 2017b).

398

399 **4. Conclusion**

400 This research indicates the potential of QFD and the case of a novel TQI in analysing the shelf-life of dried
401 apples using different drying technologies. QFD enabled the merging of consumer research on the most
402 important sensory attributes of dried apples, and made it possible to transfer these demanded quality
403 characteristics to measurable product characteristics. As an outcome, QFD calculated the importance of
404 quality characteristics typical for dried apples and identified the most important attributes that play a
405 significant role in consumer preference. This study established a mathematical index of TQI in order to
406 evaluate the total quality of apples dried using three different drying technologies during their shelf-life.
407 This model enables the evaluation and comparison of different drying techniques.

408 Results revealed two phases in quality deterioration of dried apples during six months of storage. TQI
409 showed that measurable changes occur during the second half of the observed period, when it is
410 possible to clearly distinguish differences in the overall TQI. Although at the end of the examined period
411 C-γ and F-γ dried apple samples had similar scores, further research should deploy investigation of
412 additional quality parameters. The limitation of this research is the fact that various microbiological and
413 chemical parameters have not been included and that one variety of apple fruit was used.

414

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419

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572

573

574 Table 1. Dried apple samples used in the research

Drying method	Initial water activity ¹	Packaging material ²	Atmosphere	Sample abbreviation
scCO ₂ -drying	0.19 ± 0.00	PE	Air	C-α
		EVOH-PE	100% Nitrogen	C-β
		Alu-PE	100% Nitrogen	C-γ
Air-drying	0.18 ± 0.01	Alu-PE	100% Nitrogen	A-γ
Freeze-drying	0.14 ± 0.00	Alu-PE	100% Nitrogen	F-γ

¹ Mean ± standard deviation (n = 3), immediately after drying.

² PE=polyethylene; EVOH-PE=ethylene vinyl alcohol/polyethylene copolymer; Alu-PE = polyethylene coated aluminum.

575

576

577 **Table 2.** The effects of different atmospheres and storage time on the colour properties of dried apples

	Dried apple samples ^{1, 2}				
	C- α	C- β	C- γ	A- γ	F- γ
Total colour difference (ΔE)					
0 months	4.17 \pm 2.08 ^{a, A}	5.94 \pm 3.17 ^{a, A}	4.06 \pm 1.55 ^{a, A}	21.11 \pm 9.09 ^{a, C}	11.37 \pm 1.23 ^{a, B}
3 months	7.76 \pm 5.70 ^{ab, A}	11.04 \pm 8.85 ^{a, A}	7.83 \pm 6.16 ^{a, A}	24.37 \pm 7.16 ^{a, B}	8.39 \pm 2.01 ^{a, A}
6 months	12.65 \pm 7.37 ^{b, A}	17.30 \pm 5.60 ^{b, AB}	14.88 \pm 9.13 ^{b, A}	28.02 \pm 9.24 ^{a, C}	22.53 \pm 6.51 ^{b, BC}
Browning index (BI)					
0 months	34.31 \pm 4.27 ^{a, A}	39.13 \pm 12.55 ^{a, A}	33.44 \pm 6.53 ^{a, A}	77.94 \pm 18.19 ^{a, B}	35.55 \pm 5.49 ^{ab, A}
3 months	36.18 \pm 6.03 ^{a, A}	39.22 \pm 6.92 ^{a, A}	35.15 \pm 6.32 ^{a, A}	76.84 \pm 13.73 ^{a, B}	31.96 \pm 2.81 ^{a, A}
6 months	50.78 \pm 17.66 ^{b, A}	48.37 \pm 5.22 ^{b, AB}	36.42 \pm 4.72 ^{a, C}	82.47 \pm 17.78 ^{a, D}	38.27 \pm 7.42 ^{b, BC}

¹ Sample abbreviations are given in Table 1.

² Values are the arithmetic mean \pm standard deviation (N = 10 samples on both cut surfaces). Values marked with the same small letter within the same column are not statistically different ($\alpha = 0.05$). Values marked with the same capital letter within the same row are not stat. different ($\alpha = 0.05$).

578

579

580 **Table 3.** Effects of different storage conditions on changes in sensory quality characteristics of dried
 581 apples during six months of storage.

	Dried apple samples ^{1,2}				
	C-α	C-β	C-γ	A-γ	F-γ
Appearance					
0 months	3.9±0.6 ^{a, B}	4.1±0.5 ^{a, B}	4.1±0.7 ^{a, B}	3.0±0.8 ^{a, C}	4.8±0.2 ^{a, A}
3 months	4.3±0.5 ^{a, B}	4.3±0.5 ^{a, B}	4.2±0.6 ^{a, B}	3.2±1.1 ^{a, C}	4.9±0.2 ^{a, A}
6 months	1.3±1.3 ^{b, D}	1.7±1.2 ^{b, CD}	3.5±0.7 ^{b, B}	2.1±1.2 ^{b, C}	4.6±0.4 ^{b, A}
Odour					
0 months	3.8±0.6 ^{a, A}	4.0±0.5 ^{a, A}	4.0±0.6 ^{a, A}	4.3±0.8 ^{a, A}	4.1±0.7 ^{a, A}
3 months	3.9±0.8 ^{a, A}	4.3±0.7 ^{a, A}	3.8±1.0 ^{a, A}	4.1±1.0 ^{a, A}	4.5±0.4 ^{a, A}
6 months	2.0±1.0 ^{b, B}	2.3±0.9 ^{b, B}	3.6±0.9 ^{a, A}	3.4±1.1 ^{a, A}	3.6±0.9 ^{b, A}
Texture					
0 months	4.1±0.5 ^{a, A}	4.1±0.6 ^{a, A}	4.5±0.5 ^{a, A}	4.4±0.6 ^{a, A}	4.5±0.4 ^{b, A}
3 months	3.8±0.6 ^{a, C}	4.0±0.5 ^{a, C}	4.7±0.4 ^{a, AB}	4.4±0.6 ^{a, B}	4.8±0.3 ^{a, A}
6 months	1.2±0.6 ^{b, C}	1.6±1.0 ^{b, C}	4.1±0.7 ^{b, A}	3.3±1.2 ^{b, B}	4.1±0.6 ^{c, A}
Flavour					
0 months	4.4±0.4 ^{a, A}	4.5±0.4 ^{a, A}	4.4±0.4 ^{a, A}	4.6±0.4 ^{a, A}	4.7±0.3 ^{a, A}
3 months	4.2±0.8 ^{a, A}	4.2±0.8 ^{a, A}	4.4±0.5 ^{a, A}	4.7±0.4 ^{a, A}	4.6±0.5 ^{a, A}
6 months	0.9±0.9 ^{b, C}	2.3±1.1 ^{b, B}	3.9±0.9 ^{b, A}	4.1±1.3 ^{b, A}	4.0±0.8 ^{b, A}

¹ Sample abbreviations are given in Table 1.

² Values are the arithmetic mean ± standard deviation (N = 16 = 8 assessors x 2 replications). Values marked with the same small letter within the same column are not statistically different ($\alpha = 0.05$). Values marked with the same capital letter within the same row are not stat. different ($\alpha = 0.05$).

582

583

584

585 **Table 4.** The effects of different atmospheres and storage time on the textural properties of dried apples

	Dried apple samples ^{1,2}				
	C- α	C- β	C- γ	A- γ	F- γ
Hardness [g]					
0 months	302.4 ± 98.8 ^{a, A}	270.9 ± 77.5 ^{a, A}	266.6 ± 267.5 ^{a, A}	264.2 ± 138.3 ^{a, A}	175.8 ± 70.0 ^{a, A}
3 months	208.7 ± 35.2 ^{ab, A}	172.8 ± 64.7 ^{b, A}	177.5 ± 221.6 ^{a, A}	239.3 ± 196.8 ^{a, A}	174.8 ± 75.2 ^{a, A}
6 months	165.1 ± 166.4 ^{b, A}	80.1 ± 88.5 ^{c, A}	158.5 ± 183.3 ^{a, A}	178.7 ± 153.3 ^{a, A}	96.6 ± 48.3 ^{b, A}
Cohesiveness					
0 months	0.63 ± 0.15 ^{ab, B}	0.70 ± 0.13 ^{a, A}	0.62 ± 0.15 ^{a, AB}	0.59 ± 0.23 ^{a, AB}	0.47 ± 0.25 ^{a, B}
3 months	0.78 ± 0.36 ^{a, AB}	0.62 ± 0.28 ^{a, AB}	0.77 ± 0.3 ^{a, AB}	0.49 ± 0.21 ^{a, A}	0.84 ± 0.04 ^{b, B}
6 months	0.42 ± 0.05 ^{b, A}	0.62 ± 0.20 ^{a, AB}	0.80 ± 0.51 ^{a, B}	0.72 ± 0.21 ^{a, AB}	0.67 ± 0.45 ^{ab, AB}
Springiness [mm]					
0 months	0.73 ± 0.04 ^{a, A}	0.78 ± 0.03 ^{a, A}	0.77 ± 0.04 ^{a, A}	0.73 ± 0.11 ^{a, A}	0.61 ± 0.04 ^{ab, B}
3 months	0.74 ± 0.08 ^{a, AB}	0.77 ± 0.05 ^{a, A}	0.71 ± 0.05 ^{b, ABC}	0.57 ± 0.38 ^{a, BC}	0.54 ± 0.05 ^{a, C}
6 months	0.75 ± 0.09 ^{a, A}	0.75 ± 0.17 ^{a, A}	0.76 ± 0.03 ^{a, A}	0.89 ± 0.39 ^{a, A}	0.69 ± 0.16 ^{b, A}

¹ Sample abbreviations are given in Table 1.

² Values are the arithmetic mean ± standard deviation (N = 8 samples in 2 replications). Values marked with the same small letter within the same column are not statistically different ($\alpha = 0.05$). Values marked with the same capital letter within the same row are not stat. different ($\alpha = 0.05$).

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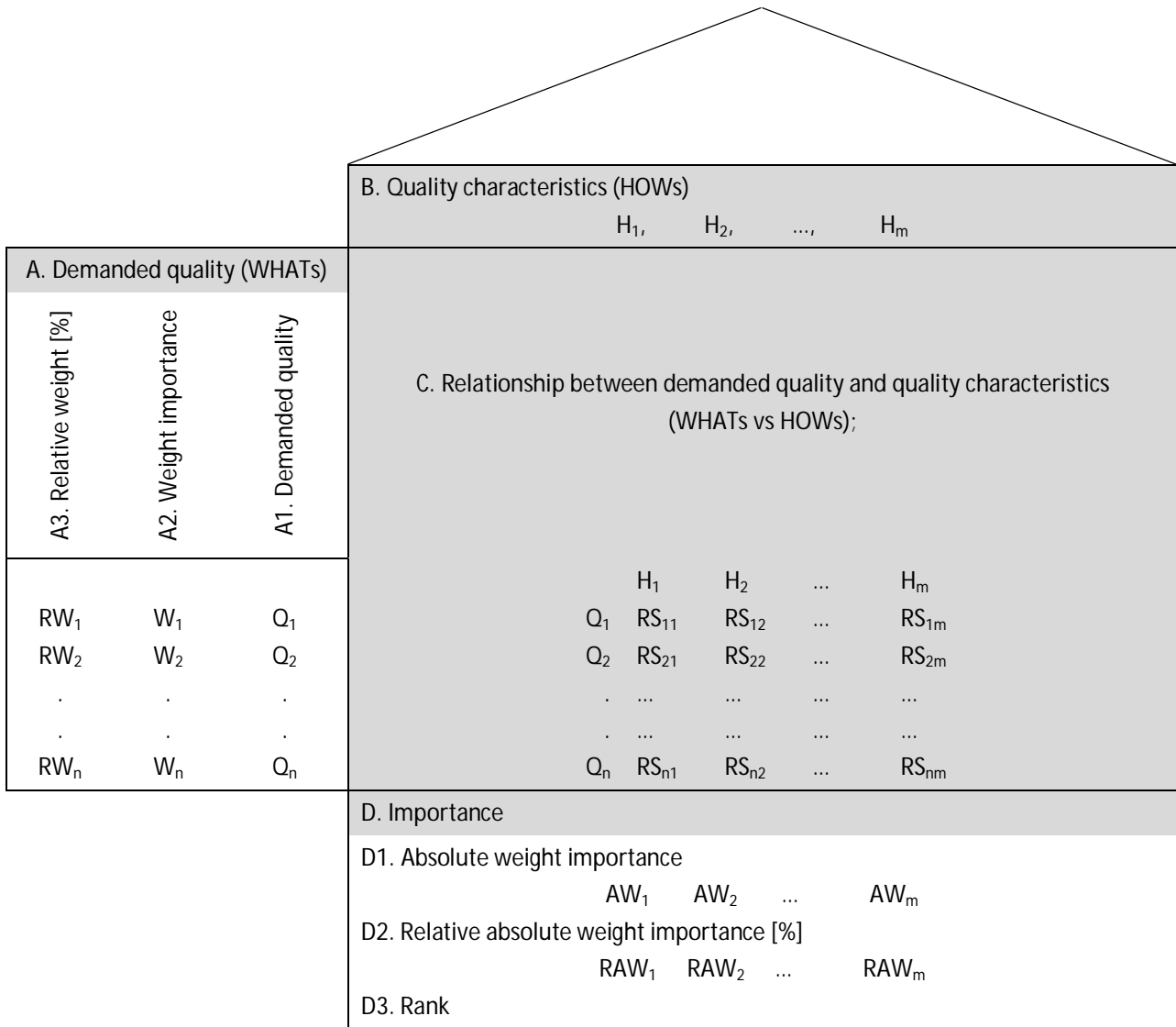


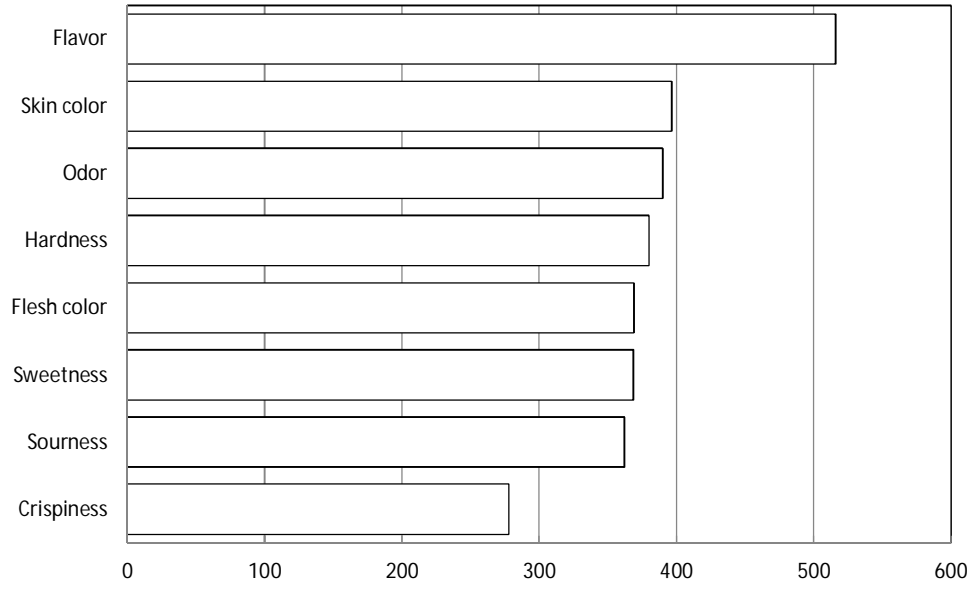
Figure 1. House of quality (HOQ)

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Figure 2. Consumer attitudes towards sensory quality characteristics of dried apples

Legend: Values are the rank sums (N = 85). The characteristics were ranked from 1='the least important' to 8='the most important'.

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Weight		Quality characteristics (HOWs) Demanded quality (WHATs)	Color		Sensory parameters			Other			
Relative weight [%]	Weight importance		Total color difference (ΔE)	Browning Index	Appearance	Odor	Oral texture	Flavor	Hardness	Cohesiveness	Springiness
19.44%	7	Skin color	●	○	○						
11.11%	4	Flesh color	●	○	○						
16.67%	6	Odor				●		○			
22.22%	8	Flavor				○		●			
5.56%	2	Sourness						○			
8.33%	3	Sweetness						○			
13.89%	5	Hardness					●		●	○	○
2.78%	1	Crispiness					●	○	●	○	○
Absolute weight importance			2.75	0.92	0.92	2.17	1.50	2.94	1.50	0.50	0.50
Relative absolute weight importance [%]			20.1%	6.7%	6.7%	15.8%	11.0%	21.5%	11.0%	3.7%	3.7%

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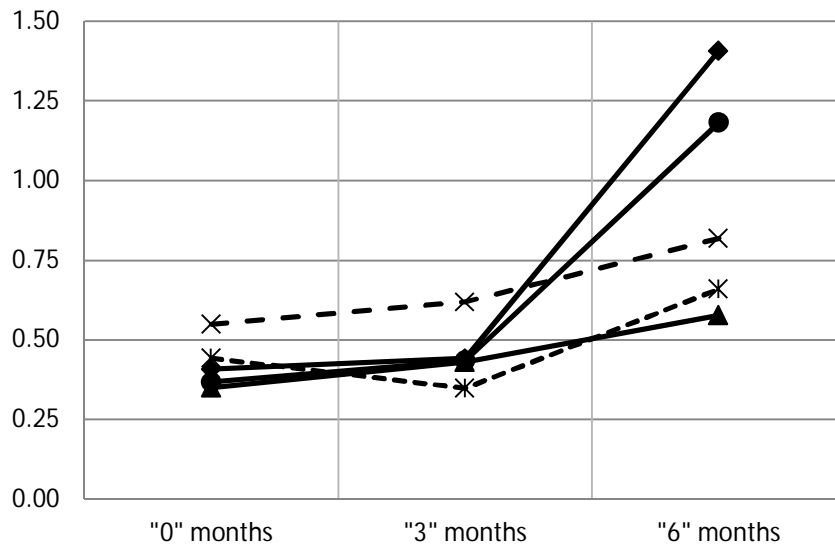
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Figure 3. House of quality for dried apples packed in MAP

Legend: ● 'strong relationship' = 9, ○ 'moderate' = 3, ○ 'weak relationship' = 1 and blank = 'non-existent' or 'zero'

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608 **Figure 4** – Total quality index of dried apples packed in modified atmosphere during shelf-life

609 Legend: CO₂ dried packed in PE with air (◆ C-α); CO₂ dried packed in EVOH-PE with 100% N₂ (● C-β); CO₂ dried packed in AluPE
610 with 100% N₂ (▲ C-γ); Air dried packed in AluPE with 100% N₂ (× A-γ); Freeze dried packed in AluPE with 100% N₂ (* F-γ)

611 Rule of the thumb: the lower the value, the better the total quality index

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