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Title: Pectin-honey coating as novel dehydrating bioactive agent for cut fruit: enhancement of the functional properties of coated dried fruits

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Keywords: Fruit dehydration, pectin-honey edible coating; antioxidant properties; food quality preservation; nutritional upgrading.

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Abstract: In this paper, a novel and sustainable process for the fruit dehydration is described. Specifically, edible coatings based on pectin and honey were prepared and used as dehydrating and antimicrobial agents of cut fruit samples, in this way promoting the fruit preservation from irreversible deteriorative processes.

Pectin-honey coating was tested on apple, cantaloupe melon, mango and pineapple. The analysis were performed also on uncoated dehydrated fruits (control). The coated fruit evidenced enhanced dehydration percentage, enriched polyphenol and vitamin C contents, improved antioxidant activity and volatile molecules profile. Moreover, the antimicrobial activity against *Pseudomonas* and *Escherichia Coli* was assessed. Finally, morphological analysis performed on fruit fractured surface, highlighted the formation of a non-sticky and homogeneous thin layer. These outcomes suggested that the novel fruit dehydration process, performed by using pectin-honey coating, was able to both preserve the safety and quality of dehydrated fruits, and enhance their authenticity and naturalness.

**Cover Letter**

**P. Finglas**

**Editor of Food Chemistry**

**Quadram Institute Bioscience, Norwich, England, UK**

**Dear Professor Finglas,**

Thank you very much for giving us the opportunity to resubmit the paper in a totally revised version. The title of the manuscript was modified, as rightly suggested by one of the referees and following reported: **“Pectin-honey coating as novel dehydrating bioactive agent for cut fruit: enhancements of the functional properties of coated dried fruits”**.

by Gabriella Santagata, Salvatore Mallardo, Gabriella Fasulo, Paola Lavermicocca, Francesca Valerio, Mariaelena Di Biase, Michele Di Stasio, Mario Malinconico, Maria Grazia Volpe for publication in one of the forthcoming issues of *Food Chemistry*.

Dear Editor, following the worthy amendments of the four Reviewers, we totally revised the paper, both in contents and in the form. The introduction was streamlined and the aim and novelty of the paper were better highlighted. All the referee suggestions were considered and accepted: sentences were modified and/or removed if not pertinent to the aim of the paper, English was improved and updated references were included, whereas the results and discussion section was better deepened. We really wish that this new wholly modified version of the paper could satisfy the reviewers demands and, of course, could match your positive response.

We are really glad about your re-consideration of our manuscript and we thank you in advance for your kind reply,  
looking forward to hearing from you soon,

Kind Regards,

Gabriella Santagata and Maria Grazia Volpe on behalf of the co-authors.

Any communication should be sent to the corresponding authors:

## **Highlights**

Novel approach for cut fruit dehydration using pectin-honey bioactive edible coating

The coated fruit (PH) showed improved dehydration percentage with respect the control

PH coating improved polyphenol content, antioxidant and antibacterial properties

Organoleptic properties by volatile molecules were improved in dehydrated PH fruits

Morphological analysis showed the physical stability of the coating on fruit surface

1 **Pectin-honey coating as novel dehydrating bioactive agent for cut fruit:**  
2 **enhancement of the functional properties of coated dried fruits**

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13 **Abstract**

14 In this paper, a novel and sustainable process for the fruit dehydration was described. Specifically,  
15 edible coatings based on pectin and honey were prepared and used as dehydrating and antimicrobial  
16 agents of cut fruit samples, in this way promoting the fruit preservation from irreversible  
17 deteriorative processes.

18 Pectin-honey coating was tested on apple, cantaloupe melon, mango and pineapple. The analysis  
19 were performed also on uncoated dehydrated fruits (control). The coated fruit evidenced enhanced  
20 dehydration percentage, enriched polyphenol and vitamin C contents, improved antioxidant activity  
21 and volatile molecules profile. Moreover, the antimicrobial activity against *Pseudomonas* and  
22 *Escherichia Coli* was assessed. Finally, morphological analysis performed on fruit fractured  
23 surface, highlighted the formation of a non-sticky and homogeneous thin layer. These outcomes  
24 suggested that the novel fruit dehydration process, performed by using pectin-honey coating, was

25 able to both preserve the safety and quality of dehydrated fruits, and enhance their authenticity and  
26 naturalness.

27

## 28 **Keywords**

29 Fruit dehydration, pectin-honey edible coating; antioxidant properties; food quality preservation;  
30 nutritional upgrading.

31

## 32 **1.Introduction**

33 Dehydration of fruit is one of the oldest techniques of food preservation consisting essentially in a  
34 decrease of water content with following increase in solute concentration (Chen & Mujumdar,  
35 2008; Kudra & Mujumdar, 2009).

36 Almost all industrial devices used as fruit dryers are convective type with either hot air or  
37 combustion gases as heat transfer media; anyway, in the last year, novel drying methods exploiting  
38 microwave or ultrasound approaches, high electric field, heat pump drying and refractance window  
39 drying technology are used to improve the dehydration process by reducing the energy consumption  
40 and, at the same time, preserving the quality of the dried products (Zarein, Samadi, Ghobadian,  
41 2015; Sabarez, Gallego-Juarez, Riera 2012).

42 Moreover, the fruit dehydration processes are usually supported by the addition of chemical  
43 additives, such as ascorbic acid, citric acid, glucose oxidase-catalase and sodium bisulfite that, in  
44 the last few decades, has increased to a great extent, since they represent a common approach to  
45 ensure the reduction or elimination of sanitary risks and microbiological contamination, while  
46 preserving, at the same time, the food sensorial characteristics (Bourdoux, Li, Rajkovic,  
47 Devlieghere, and Uyttendaele, 2016).

48 Anyway, the upcoming concept of "green foods" quality is oriented toward a reduced content of  
49 chemical additives since their ascertained healthy toxic effects on human immune system, leading  
50 to various disorders and diseases, not least, to serious risk of cancer (Dar et al., 2017).

51 Finally, since the increasingly concern related to harmful environmental impact of the high  
52 greenhouse gases emission during the food drying methods (Mujumdar, 2010), the applicative  
53 science research and the industrial technologies have been focusing their attention on new eco-  
54 friendly fruit drying methodologies able to drastically reduce the energy consumption while  
55 preserving the safety and quality of dehydrated fruits, by enhancing their authenticity and  
56 naturalness.

57 A novel approach, exploited in the frame of this paper, can be represented by the use of edible  
58 coatings. They are defined as thin layers of edible material applied to the surface of a food, which  
59 provides a barrier against migration of moisture, oxygen, carbon dioxide, aromas, lipids, and other  
60 solutes (Castilho Garcia, Cássia Caetano, de Souza Silva, Aparecida Mauro, 2014; Lago-Vanzela,  
61 Nascimento, Fontes, Mauro, Kimura, 2013).

62 An edible coating must be eaten as part of the whole product. Therefore, the composition of edible  
63 coatings must conform to the regulation applied to the food product (Vargas et al., 2008).

64 Several studies reported that polysaccharides from different sources are promising materials for the  
65 preparation of films and coatings with tailored behavior (Chengcheng, Pollet & Averous, 2017).

66 This is not surprising considering that they are natural, nontoxic, biodegradable polymers. Among  
67 polysaccharides, pectin is of worthy interest as a potential coating component, because of its unique  
68 colloidal properties. This biopolymer is mainly obtained from citrus peel or apples pomace and  
69 contains smooth (linear) regions and hairy, branched regions. Its chemical structure is widely  
70 reported in literature and will not be discussed here (Ström et al., 2007; Nesic et al., 2017).

71 Its efficiency in enhancing food protection has been well recognized by many research groups, as  
72 shown by Laurienzo et al. (2010). In their paper, the authors showed that pectin edible films could

73 preserve some volatile components and free sugar content of dried fruits, controlling the gas  
74 exchange, the moisture transfer and the oxidation processes (Laurienzo et al., 2010).

75 In addition, the development of functional edible coatings with inherent active ingredients  
76 represents an upcoming technological challenge for the food industry. Indeed, several compounds,  
77 such as antioxidants, antimicrobials, flavors and probiotics, can be incorporated into the polymeric  
78 formulations and consumed with the food, by enhancing its safety, nutritional and sensorial  
79 attributes (Rojas-Graü et al., 2009; Robles-Sánchez et al., 2013).

80 Based on the above considerations, and aimed to obtain a dehydrating and functional edible coating,  
81 a natural compound, honey, was included in pectin water formulations.

82 Actually, the high honey hygroscopicity and low pH (pH range 3.2-4.5) are the two key factors for  
83 the hampering of microorganism growth. Honey bactericidal activity against food spoilage and  
84 pathogenic organisms have been widely explored in numerous studies (Moundoi, Padilla-Zakour &  
85 Worobo, 2004; Eteraf-Oskouei, & Najafi, 2013).

86 Hence, the aim of the present work was to use a pectin-honey (PH) based coating as innovative eco-  
87 sustainable method for dehydrating selected kinds of cut fruits and contemporary preserving their  
88 safety and quality by exploiting the antimicrobial activity of honey. The dehydration process was  
89 carried out at low temperature; in this way, the nutritional and sensorial properties of dried fruit, not  
90 treated with sugar and/or chemical additives, could be maintained by providing a final high quality  
91 food. The effects of PH coating on fruits water content (dehydration process), total polyphenols and  
92 vitamin C amount, antioxidant activity, volatile molecules profile, microbiological quality and fruit  
93 surface adhesion were investigated. Uncoated dehydrated fruit slices were used as control reference.

94

## 95 **2. Materials and methods**

### 96 2.1. Materials

97 Fresh fruit samples of apple (Ap), cantaloupe melon (Cm), mango (Ma) and pineapple (Pi) slices

98 were purchased from a local market and immediately stored at 4°C, in order to avoid any  
99 fermentation process. Uniform sized, defect-free fruits were selected.

100 Citrus pectin (low esterification degree) was obtained from Herbstreight & Fox KG, Pektin-  
101 Fabriken (Germany). Honey was collected between May and July 2015 in the South of Italy  
102 (Benevento area) and stored in the dark, at room temperature.

103 2,2'-Diphenil-1-picryl hydrazyl (DPPH), gallic acid, Folin-Ciocalteu and ethanol were obtained  
104 from Sigma-Aldrich. Starch paste and iodine were purchased by Carlo Erba.

105

## 106 2.2. Films and cut fruit coating preparation

107 Low ester citrus pectin (2%wt/v) was dissolved in demineralized water at 100°C, while stirring.  
108 After that, 10% w/w of honey was added in pectin solution and dispersed by magnetic stirring.  
109 After 30 minutes, 50 mL of neat pectin (P) and pectin/honey (PH) solutions were respectively  
110 casted on Petri dishes and allowed to dry at room temperature, under ventilated hood for 48 hours  
111 up to obtain dried detectable films. Prior each measurements, all films were conditioned in climatic  
112 chamber at room temperature and 50% RH.

113 The fruit samples of Ap, Cm, Ma, and Pi were thoroughly washed, peeled, cut into sections of about  
114 2 cm and dipped in PH solution for 30 seconds. The identification codes of coated samples used  
115 were ApPH, CmPH, MaPH and PiPH.

116

## 117 2.3. Dehydration process and determination of free water content

118 The coated samples, ApPH, CmPH, MaPH and PiPH and control samples Ap, CM, Ma, and Pi were  
119 placed on polypropylene grids and allowed to be slowly dehydrated by allocating them in a  
120 ventilated fridge at 4°C and relative humidity of 40% for about 15 days. The free water content of  
121 the samples was evaluated as follows: all the fruit slices were periodically withdraw from  
122 refrigerator, weighted, put in an oven at 90°C and re-weighted. This procedure was repeated up the

123 reaching of dehydrated samples constant weight (%). For each kind of cut fruit, three specimens  
124 were used and the results were expressed as the average value of water losing percentage.

#### 125 2.4. Extraction and determination of total polyphenol

126 The fruit samples were grounded into fine particles with a laboratory mixer (Ultraturrax T8, Ika-  
127 Werke, Germany). Aliquots of 0.5g of selected kind of fruit were treated with 10 ml of ethanol-  
128 water mixture(50:50 v/v) for 30 min at room temperature (Soong & Barlow, 2004). The samples  
129 were centrifuged at 4000 rpm for 30 min by using a centrifuge (MOD. 4235, A.C.L. International  
130 S.r.l., Milano), and the supernatant was collected for the evaluation of total phenolic content and  
131 antioxidant capacity (see 2.6paragraph) acid, Folin-Ciocalteu and ethanol were obtained from  
132 Sigma-Aldrich. Starch paste and iodine were purchased from Carlo Erba.

133 The total phenolic content was determined according to the Foline Ciocalteu procedure (Soong &  
134 Barlow, 2004).The absorbance was measured at 760nm. Phenolic concentration was determined by  
135 comparing the absorbance of the samples with a standard. The results were expressed as milligrams  
136 of gallic acid equivalents in 100 g of dry fruit (mg GAE/100g dry fruit). The measurement was  
137 performed in triplicate and the result was reported as the averaged value.

138

#### 139 2.5. Antioxidant capacity

140 The antioxidant capacity of dehydrated fruit slices was studied according to the procedure described  
141 by Sánchez-Moreno et al., (1998), (Sánchez-Moreno, Larrauri, & Saura-Calixto, 1998).

142 The absorption ofthe samples was measured with a spectrophotometer UV-Vis (Beckman Coulter,  
143 mod. DU 730, Brea, CA, USA)at 515 nm against a blank of methanol without DPPH. The  
144 antiradical activity was expressed as percentage inhibition (%I) of the samples compared to the  
145 initial DPPH absorption ( $A_0$ ), according to the following formula:

$$146 \quad I(\%) = \frac{(A_0 - A_t)}{A_0} \times 100 \quad (2)$$

147 Where  $A_0$  is the absorbance of initial time  $t=0$  min,  $A_t$  is the absorbance of the sample at time  $t= 30$   
148 min. The results were expressed as the average value of three repetitions.

149

## 150 2.6. Iodometrically determination of vitamin C

151 The ascorbic acid content of samples was determined iodometrically. Specifically, the samples were  
152 homogenized, diluted in 100 ml of ultrapure water mQ and filtered. A volume of 10 ml of these  
153 solutions were recovered, added with 1ml of a starch paste (indicator) and titrated with a solution of  
154 iodide (0.00489 M) (A.O.A.C., 1990). The result was expressed as the average value of three  
155 measurements for each kind of sample.

156

## 157 2.7. Determination of volatile molecules profile

158 The extraction of volatile compounds was realized through the SPME technique, combined with gas  
159 chromatography paired to mass spectrometry (SPME-GC / MS) (Komes et al., 2007).

160 2g of sample were weighed in vials of 20 mL with 2g of Sodium Chloride and 3g of distilled water.

161 Volatile compounds (VOCs) were extracted from the samples by a fiber polydimethylsiloxane  
162 (PDMS) with 100  $\mu\text{m}$  film thicknesses (Supelco), kept for 45min in block, heated to 45  $^{\circ}\text{C}$  in the  
163 head space of the sample. The analysis of the volatile compounds was performed after 21 days, by  
164 using a GC / MS Agilent 7890A / 5975C autosampler Gerstel MPS2, INNOWax using a capillary  
165 column (30mm x 0.250mm x 0.5 $\mu\text{m}$ ) at the following temperature program: 40  $^{\circ}\text{C}$  for 2 min,  
166 heating ramp up to 230 $^{\circ}\text{C}$  at 5 $^{\circ}\text{C}/\text{min}$ , and isotherm step at 230 $^{\circ}\text{C}$  for 10 min. Identification of  
167 volatile compounds was achieved by comparing mass spectra with the Wiley library (Wiley7, NIST  
168 05). The amount of the individual compounds was expressed as a peak area percentage, obtained by  
169 automatic integration of the area of the compound with respect to the sum of peak areas of all  
170 compounds identified in the chromatograms. All the analyses were performed in triplicate.

171

## 172 2.8. Microbiological analysis

173 Dried fruit samples (2 g) were homogenized for 2 min in sterile Ringer solution (Oxoid, Milan,  
174 Italy) (1:5 dilution) using a stomacher (Seward, London, UK). After 30 min at room temperature, an  
175 aliquot of the suspension was used to prepare serial decimal dilutions in NaCl 0.85% (p/vol) and  
176 Tween 80 0.25% (vol/vol); 100 µl of each dilution was inoculated on agar plates by spread plating.  
177 Total aerobic bacterial count (TBC) was evaluated using Plate Count Agar (PCA, Difco, Becton  
178 Dickinson Co., Sparks, MD, USA) supplemented with cycloheximide (0.17g/L) incubated at 30°C  
179 for 48 h. *Pseudomonas* spp. (Ps) were counted on *Pseudomonas* agar with CFC supplement  
180 (Oxoid). Lactic acid bacteria (LAB) were counted on de Man-Rogosa-Sharpe agar (MRS, Oxoid) at  
181 30°C for 48 h. Yeast and moulds (YM) were counted on Sabouraud Dextrose Agar (Difco)  
182 supplemented with chloramphenicol and chlortetracycline (both 0.05 g/L) and incubated at 25°C for  
183 5-7 days. *Escherichia coli* β-glucuronidase positive was enumerated by pour plating 1 mL of  
184 decimal dilutions in Tryptone Bile X-Glucuronide Agar plates (TBX, Oxoid) incubated at 30°C for  
185 4 h and thereafter at 44°C for 44 h: typical blue–green coloured colonies were counted. Another  
186 aliquot of the food suspension was subjected to heat treatment (80°C for 10 min) to count spore-  
187 former bacteria. Therefore, the suspension was pour plated (1 mL) and decimally diluted and spread  
188 plated (100 µL) on Brain Heart Infusion (BHI) agar plates incubated at 37°C for 24h. Microbial  
189 counts were expressed as mean of log CFU/g of dehydrated fruit.

190

## 191 2.9. Viscosity of P and PH water solution

192 The viscosity of polysaccharide based solutions was measured at room temperature (23 ±1 °C), by  
193 means of direct reading at the rotary viscometer (Brookfield Co., U.S, model LV). The screw feeder  
194 was initially calibrated to account for the different bulk densities, in this way selecting the suitable  
195 spindle. Three different tests at different angular velocity of 6, 12 and 30 rpm were performed.

196 Evaluations were carried out in triplicate in order to average the final viscosity values. For each  
197 sample and measurement, the viscosity value was monitored and read when it was constant.

198

#### 199 2.10. Scanning Electron Microscopy (SEM)

200 Morphological analysis of films was performed by means of a FEI Quanta 200 FEG Scanning  
201 Electron Microscope (SEM) on cryogenically fractured cross-sections. SEM observations were  
202 performed in low vacuum mode ( $P_{H_2O}$ : 0.7 torr), using a large field detector (LFD) and an  
203 acceleration voltage of 5–20 kV. Prior to the observation, the sample surfaces were coated with a  
204 homogeneous layer ( $18 \pm 0.2$  nm) of Au–Pd alloy by means of a sputtering device (MED 020, Bal-  
205 Tec AG). The micrographs reported showed a magnification of 400x.

206

#### 207 2.11. Statistical analysis

208 The averages and the standard deviations were calculated with Microsoft Office Excel 2016. The  
209 differences in the mean values were determined by multiple comparison procedure (Fisher LSD  
210 Method,  $P < 0.05$ ), applied after an analysis of variance (ANOVA) and a Normality Test (Shapiro-  
211 Wilk) (Tabachnick & Fidell, 2007). Statistical analysis was performed using STATISTICA (data  
212 analysis software system), version 10 (StatSoft, Inc., Tulsa, OK, USA). For each test, data are  
213 expressed as the average of three measurements, and in the graphics the vertical bars indicate  
214 standard deviation. Columns with different letters are significantly different following Fisher's  
215 protected LSD test ( $P < 0.05$ ).

216

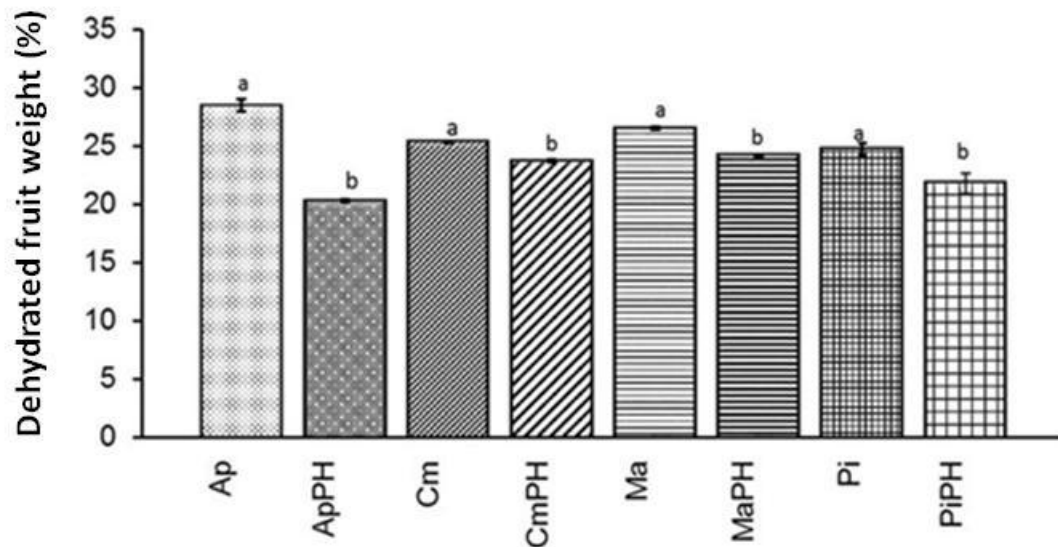
### 217 **3. Results and discussion**

218 Fruit dehydration process required approximately 15 days to be completed, as previously found by  
219 Laurienzo et al., (2010).

220 In Fig 1, the percentage of dehydrated fruit weight of all analysed samples, is reported. It is worthy  
221 to observe that the presence of coating induced higher dehydration percentage in all tested samples.  
222 Generally, the coated samples showed a greater water loss than the untreated sample (control):  
223 indeed, as evidenced in Fig. 1, they show a dehydrated samples weight in the range between 20-  
224 25%, which is lower compared to the control samples, whose dehydrated weight was in the range of  
225 25-30%.

226 The dehydration process, was ruled by the low temperature and relative humidity difference  
227 between the fruit slices and the refrigerator, accounting for the slow releasing of unbound water  
228 molecules from the fruit surface to the external environment (refrigerator); anyway, the internal  
229 ventilation and the presence of honey in PH treated samples, avoided any risk related to the  
230 microbial proliferation on the fruit surface, as antimicrobial assays following discussed, evidenced  
231 (Galus, Turska & Lenart, 2012). Moreover, the presence of the coating induced a higher efficiency  
232 of dehydration process. This outcome could be explained by considering that pectin and honey are  
233 both hydrophilic and hygroscopic since the presence of many polar groups, such as hydroxyl and  
234 carboxylated groups. It was likely that the thin film membrane could drain the surface adsorption of  
235 water molecules from fruit slices, thus inducing the following releasing towards the external  
236 environment by means of permeation process.

237



238

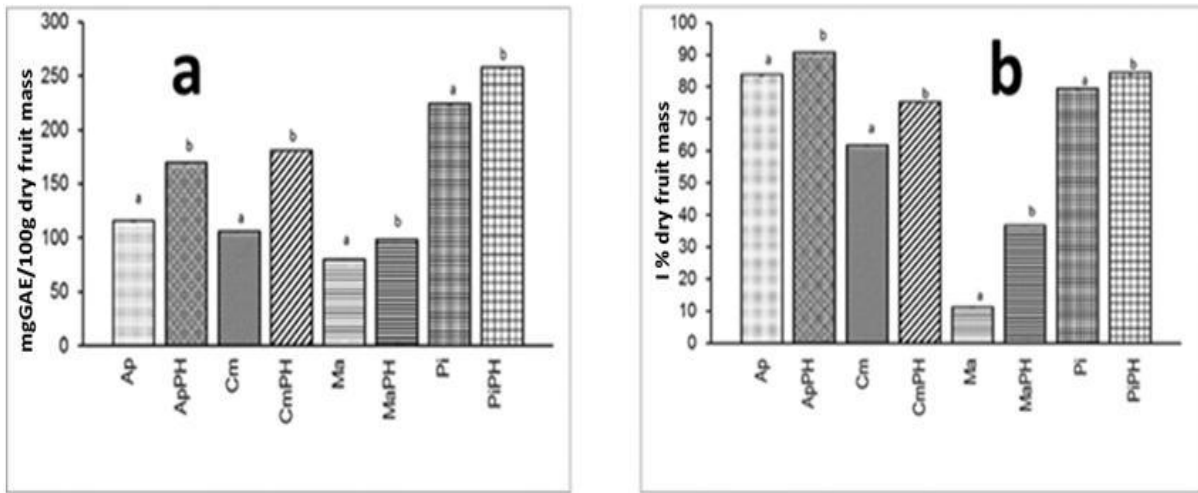
239 **Fig 1.** Moisture content of untreated and PH coated dehydrated fruits. For each fruit, columns with  
 240 different letters are significantly different following Fisher's protected LSD test ( $P < 0.05$ ).

241

242 In Fig 2a, the total polyphenol content of dehydrated fruit samples is reported. Specifically, the  
 243 uncoated fruit specimens (control) evidenced a polyphenol content in a range of 80-225mg GAE  
 244 /100g of dry mass sample, with the following decreasing order: Pi, Cm, Ap and Ma; as concerning  
 245 coated samples, the total polyphenol content followed the same lessening order, even if the values  
 246 were shifted at higher concentration interval, specifically 100 and 270 mg GAE /100g. This result is  
 247 ascribed to the significant PH fruit dehydration (see Fig 1), followed by the increasing of the solutes  
 248 and bioactive molecules contents, (Torreggiani & Bertolo, 2001). Moreover, it is plausible that the  
 249 higher polyphenol content of dehydrated treated samples was supported by the honey contribution,  
 250 since it is well known that as natural dietary antioxidant, honey polyphenols content is very high  
 251 (Al-Mamary, Al-Meerri& Al-Habori, 2002).

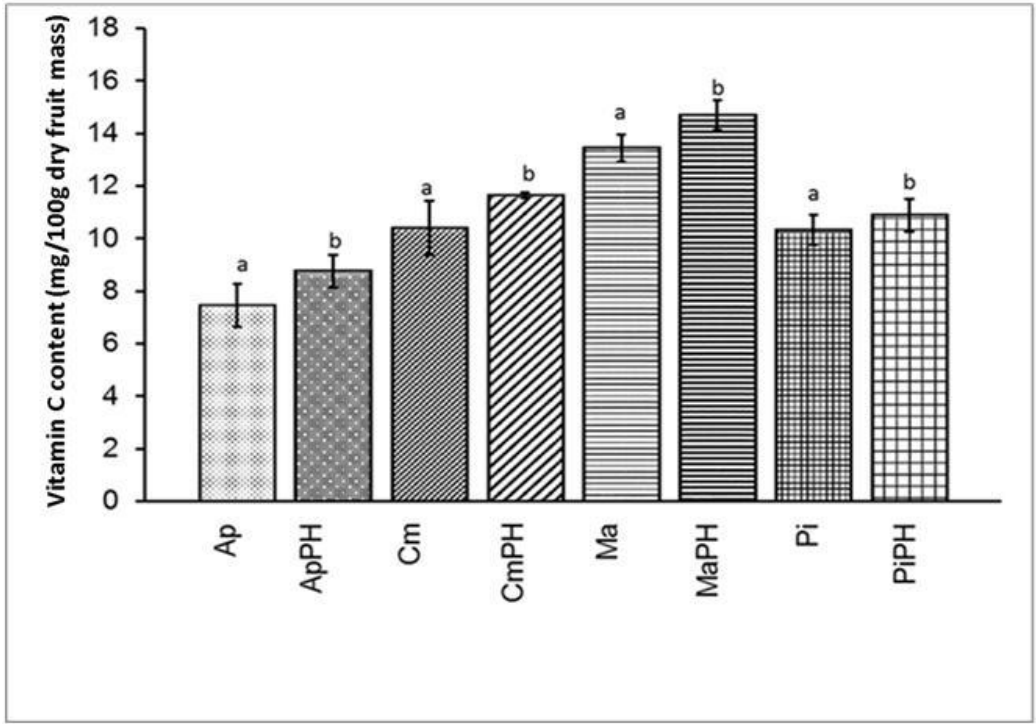
252 The assessment of the free radical scavenging capacity of a substance is an important method to  
 253 determine its antioxidant activity (Pokorný, 2007). In the specific case, DPPH scavenging activity  
 254 assays of coated fruits showed higher percentage inhibition (%I) if compared to the control samples,

255 as evidenced in Fig 2b. Here too, both enhanced dehydration and high honey concentration of PH  
256 coated fruit could be responsible of the enhanced antioxidant effect.



257  
258 **Fig 2a,b.** Total polyphenol content (a) and antioxidant properties (b) of untreated and PH coated  
259 dehydrated fruits. For each fruit, columns with different letters are significantly different following  
260 Fisher's protected LSD test ( $P < 0.05$ ).

261  
262 As far as vitamin C is concerned, Santos and Silva (2008) claimed that vitamin C can be easily  
263 degraded by means of photo-oxidation, thermal-oxidation and enzyme catalyzed reactions. For this  
264 reason, vitamin C is often used as a quality indicator of food processes (Santos & Silva, 2008). In  
265 the specific case, pectin-honey coating efficiently boosted vitamin C retention in quite all treated  
266 fruit slices, as evidenced in Fig 3. The difference between neat and treated dehydrated fruit samples  
267 was statistically significant in all fruit samples ( $P < 0.05$ ).



268 -

269 **Fig 3.** Vitamin C content of untreated and PH coated dehydrated fruits. Data is the average of three  
 270 measurements, and the vertical bars indicate standard deviation. For each fruit, columns with  
 271 different letters are significantly different following Fisher's protected LSD test ( $P < 0.05$ ).

272

273 Since aroma is one of the most appreciated fruit properties, volatile flavor compounds are likely to  
 274 play a key role in determining the perception and acceptability of products by consumers.  
 275 Depending on their presence and amount, volatile compounds affect the organoleptic characteristics  
 276 of dehydrated fruits as well as their sensorial perception being responsible for their unique flavour  
 277 and aroma. (Cheong et al., 2010).

278 The qualitative and quantitative data of cut fruit volatile compounds, grouped as esters and acetates,  
 279 alcohols, aldehydes and ketones and terpenoids, are reported in Table 1.

280 The importance of terpenoids in fruits is due to the appreciated floral notes (Lignou et al 2014). The  
 281 analysis of data in Table 1 evidences that among all volatile compounds, high concentration of  
 282 terpenoids may be found in all untreated fruits, whereas in all coated samples, terpenoids contents  
 283 decreased. This experimental outcome suggests that the polysaccharide based coating induced the

284 terpenoids evolution from dehydrated fruit. Likely, the scarce physical entanglements between the  
 285 polar residues of polysaccharide-honey macromolecular pattern and the hydrophobic groups of  
 286 these molecules, together with their relative small size, fostered the permeation throughout the  
 287 hydrophilic macromolecular pathways, allowing an easy volatilization.  
 288 (De Roos, 2000; Scoottitantawat et al., 2004; Ortega-Toro et al., 2016).  
 289 Moreover, in pineapple coated samples, the concentration of esters, responsible of fruity and floral  
 290 notes, severely increases since the enhanced metabolism of fatty acids induced by pectin-honey  
 291 coating components (Obando-Ulloa, Moreno, Garcí'a-Mas, Nicolai, Lammertyn, Monforte, 2008).  
 292 In Ap and Cm samples, the aldehydes and ketones concentration improved. The unbranched  
 293 aldehydes and ketones represent an important part of the flavor of this fruits, with aromas usually  
 294 defined as green/grassy (Lignou et al., 2014).

295

296 **Table 1.** Relative peak area of volatile compounds (VCRPA%) identified in fruit samples

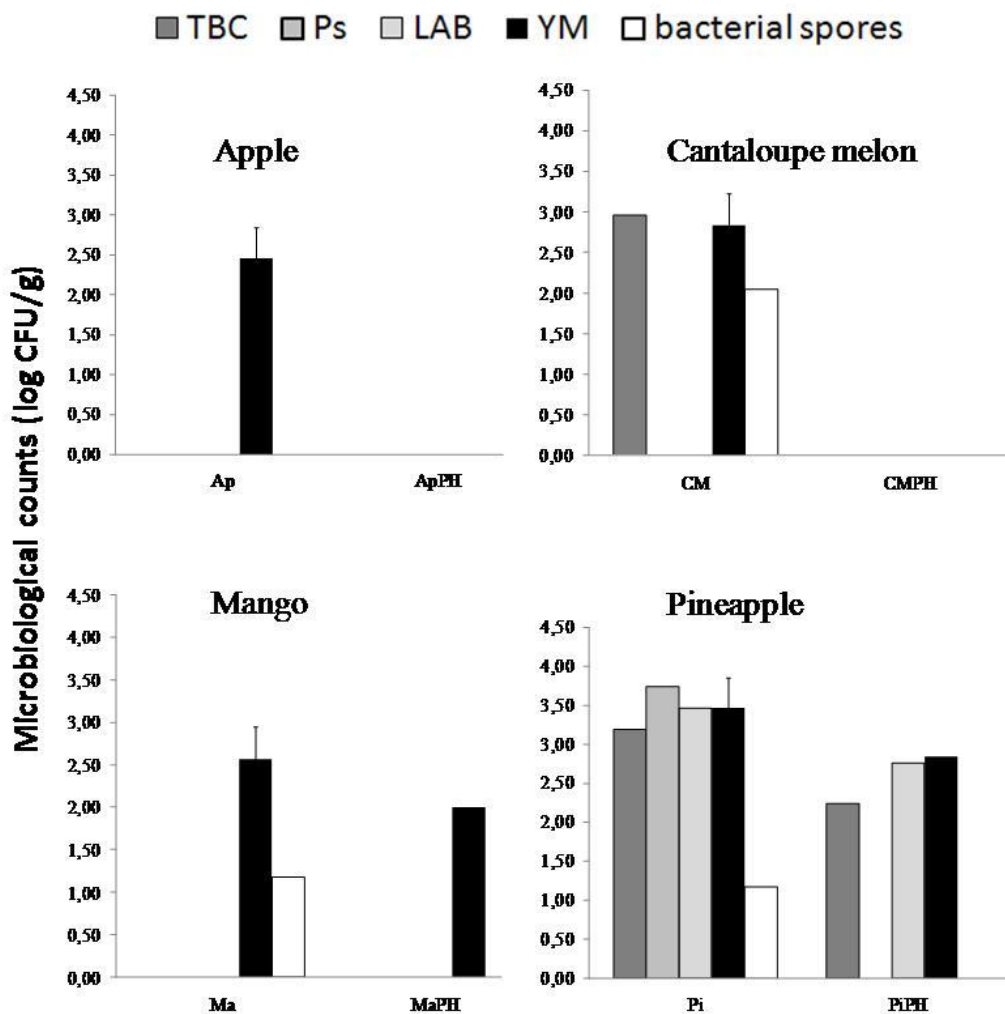
Samples/(VCRPA%)	Pi	PiPH	Ma	MaPH	Ap	ApPH	Cm	CmPH
Esters and Acetates	11,63± 0,02	48,45 ±0,04	4,91±0,02	4,22±0,02	3,26±0,04	2,25±0,04	3,25±0,03	5,60±0,05
Alcohols	1,75±0,02	4,05±0,02	2,02±0,03	5,04±0,03	3,92±0,02	8,98±0,02	6,60±0,02	7,10±0,02
Ald. and ketones	7,28±0,04	8,76±0,03	7,56±0,02	8,36±0,02	15,77±0,02	49,37±0,02	9,92±0,01	12,40±0,07
Terpenoids	71,27±0,01	29,07±0,01	83,19±0,09	73,70±0,07	69,36±0,02	28,15±0,03	67,44±0,04	50,12±0,06
Others	0,89±0,02	1,91±0,02	1,28±0,08	3,91±0,06	1,88±0,04	1,86±0,03	1,92±0,05	1,23±0,05

297 Results are expressed as mean ± standard deviation

298

299 Dried food products are considered safe from a microbiological point of view since they are  
 300 characterized by a low water activity ( $a_w$ ), and usually no growth occurs below  $a_w$  of about 0.62  
 301 (Sagar & Kumar , 2010). As reported in Fig.4, control fruits were mainly contaminated by yeast and  
 302 moulds and by bacterial spores. Pineapple was the most contaminated fruit while apple sample  
 303 showed a low microbial load. *Pseudomonas* spp. were present only Pi sample, while the absence of  
 304 *Escherichia coli*- $\beta$ -glucuronidase positive was ascertained in all samples (data not shown).

305 ApPH and CmPH did not show contamination, while MaPH showed a yeast and mould load lower  
 306 than control. Only in the case of pineapple, microbial counts were generally higher than the  
 307 detection limits in all dehydrated products. The dehydration process allowed to obtain coated fruits  
 308 with an improved microbiological quality. The lower microbial load in coated fruits could be  
 309 partially due to the use of honey, known for its antimicrobial features (Moundoi, Padila-Zakour, &  
 310 Worobo, 2001) in the dehydration process.



311  
 312 **Fig. 4** Microbiological counts (logCFU/g) of untreated and PH coated dehydrated fruits samples.  
 313 Total bacterial count (TBC, dark grey), *Pseudomonas* spp (Ps, grey), Yeast and Moulds (YM,  
 314 black), Lactic acid bacteria (LAB, light grey), Spore formation bacteria (white).  
 315 Values indicated as 0.00 log CFU/g were lower than detection limits (TBC, Ps, LAB and YM.  
 316 Detection limits <2logCFU/g; Spore-forming bacteria: Detection limit <1logCFU/g).

317 Evaluation of viscosity was necessary in order to evaluate the physical stability of the coatings after  
318 their application on fruit substrates. Indeed, the polysaccharide based solutions should both follow  
319 the wrinkle inhomogeneous fruit surface, in this way providing a non-sticky stable sheen, and avoid  
320 a detrimental slippery texture after fruit dipping. Therefore, after drying, a homogeneous glossy  
321 coating should be expected to form, able to preserve dehydrated fruits from any water-induced  
322 oxidative reactions, responsible of structural changes and fruit properties deterioration (Doymaz &  
323 Göl, 2011). This outcome strictly depends on the physical interaction occurring between fruit  
324 surface and polysaccharide based solution. As far as P and PH solutions are concerned, the viscosity  
325 data were  $2631 \pm 12\%$  cps and  $2982 \pm 9\%$  cps respectively, thus evidencing the higher viscosity of  
326 pectin-honey solution, as expected, due to the presence of honey.

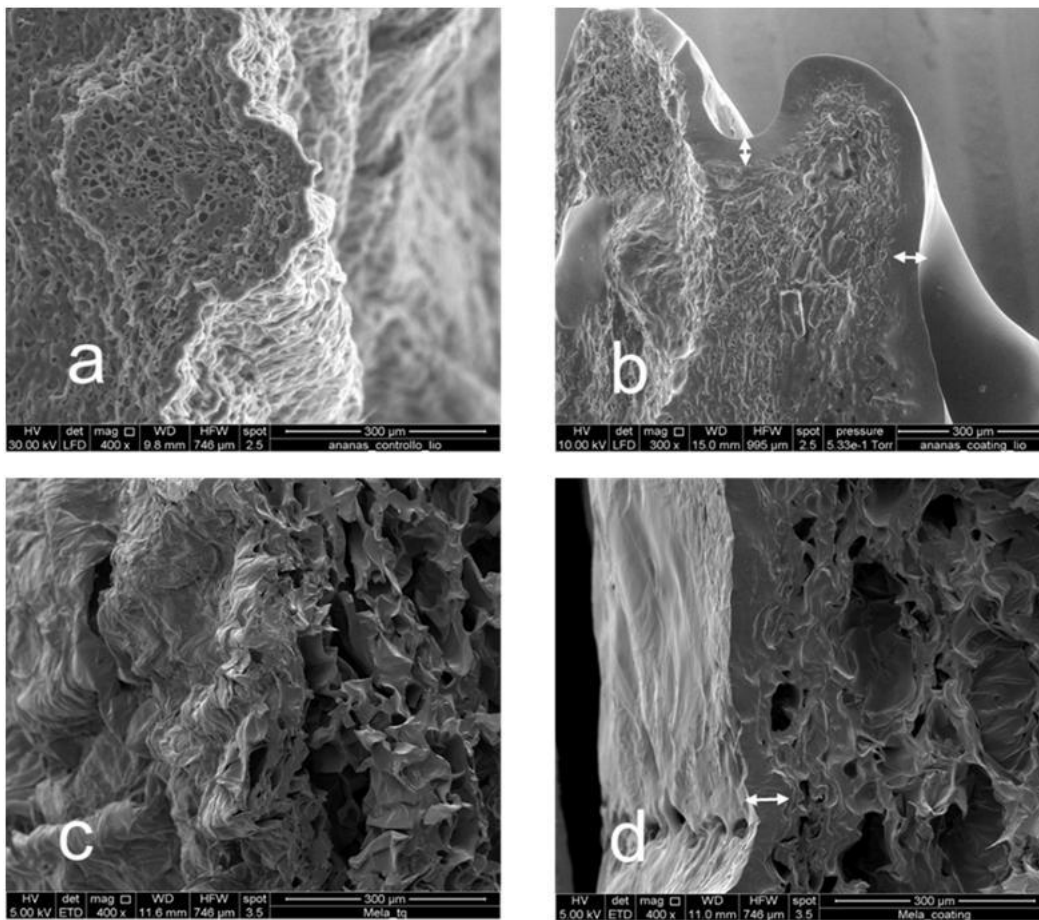
327 In order to evidence the PH coating adhesion on fruit slice surfaces, in Figg. 5a,b and 5c,d, as an  
328 example, the micrographs of cryogenically fractured cross-sections of neat (a,c) and coated (b,d)  
329 pineapple and apple slices respectively, are reported.

330 In both pictures of neat fruits, it is possible to observe similar irregular voids and cell walls  
331 dispersed throughout the inner surface; anyway, apple structure morphology is spongy and  
332 evidences bigger and deeper holes if compared to tight pineapple network (Mchugh & Senesi,  
333 2000).

334 These peculiar structural features are responsible of the different distribution and adhesion of  
335 surface coating. Indeed, in Fig 5b a regular and smoothed distribution of the coating is visible; the  
336 film thickness of about  $80 \mu\text{m}$  and the good interfacial adhesion between pineapple surface and the  
337 polymeric layer, highlights the efficiency of the coating; even if similar results are discernible in  
338 case of apple too (see Fig 5d), the film thickness is somewhat irregular, due to some permeation of  
339 PH water solution inside porous apple structure (Rojas-Grau et al., 2009). Nevertheless, a stable  
340 layer of film forms, well glued on apple surface.

341 The homogeneous coating development is fairly ascribed to the physical interactions occurring  
342 between polar groups of both fruits and pectin-honey solution, able to both improve structural

343 integrity and preserve appearance and texture, thus underlining the actual effectiveness of pectin-  
344 honey blend as a protective coating. (De Britto & Assis, 2010).



345  
346 **Fig 5a,b,c,d.** SEM cross-section micrographs of Pi (a), PiPH (b), Ap (c) and ApPH (d) slices

347

### 348 **Conclusion**

349 In this work, pectin-honey films were prepared and used as novel functional coatings for fruits both  
350 dehydration and quality preservation. The edible coating, tested on apple, cantaloupe melon, mango  
351 and pineapple fruits, evidenced higher dehydration efficiency if compared to control. Moreover, the  
352 polyphenol concentration, antioxidant activity and vitamin C content were particularly heightened  
353 in coated samples, pointing out the pectin-honey coating protection. The analysis of volatile  
354 compounds highlighted a general improvements of volatile compounds and sensorial flavors in all  
355 coated fruit samples, except for terpenoids. The innovative dehydration process allowed to obtain

356 coated fruits with an improved microbiological quality respect to the uncoated controls. Last but not  
357 least, morphological analysis underlined the formation of a non-sticky, stable and continuous  
358 coating on fruit substrates. All these outcomes emphasized the efficiency of pectin-honey films as  
359 eco-friendly system for fruit dehydration process, able to both preserve the safety and quality of  
360 dehydrated fruits, and enhance their authenticity and naturalness.

361

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368

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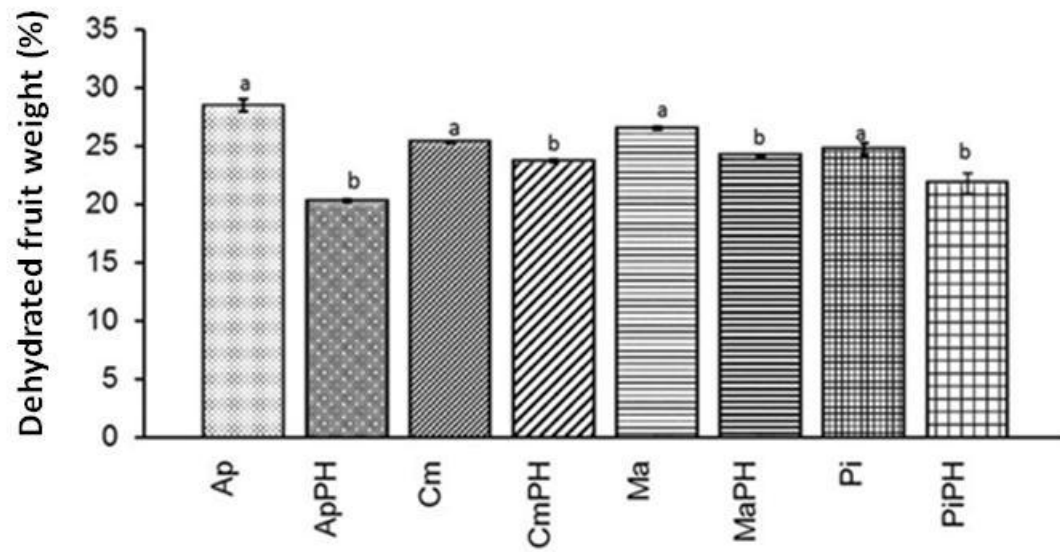
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## Tables

**Table 1.** Relative peak area of volatile compounds (VCRPA%) identified in fruit samples

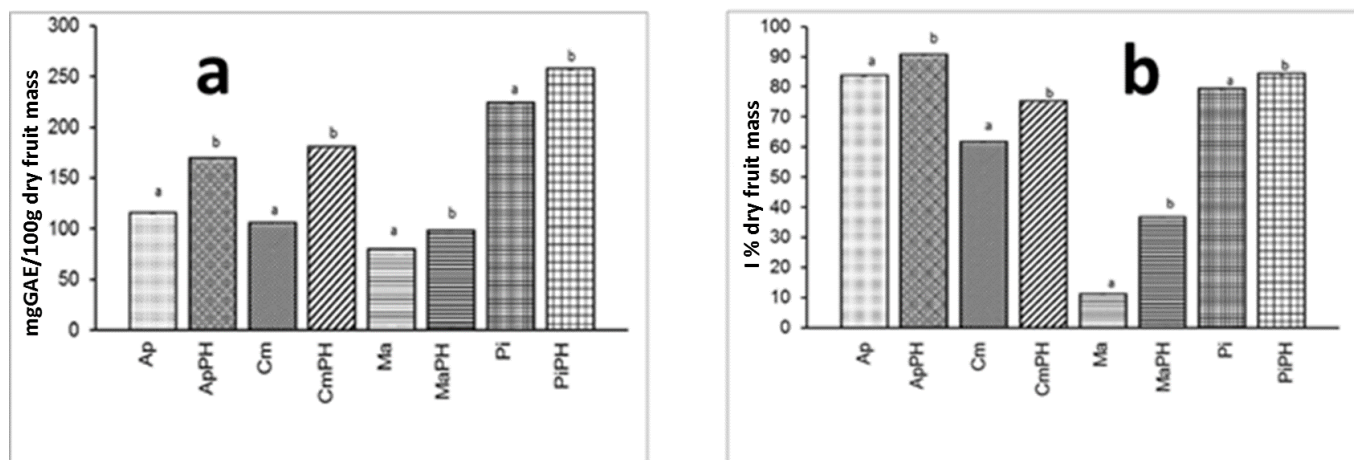
Samples/(VCRPA%)	Pi	PiPH	Ma	MaPH	Ap	ApPH	CM	CMPH
Esters and Acetates	11,63± 0,02	48,45 ±0,04	4,91±0,02	4,22±0,02	3,26±0,04	2,25±0,04	3,25±0,03	5,60±0,05
Alcohols	1,75±0,02	4,05±0,02	2,02±0,03	5,04±0,03	3,92±0,02	8,98±0,02	6,60±0,02	7,10±0,02
Ald. and ketones	7,28±0,04	8,76±0,03	7,56±0,02	8,36±0,02	15,77±0,02	49,37±0,02	9,92±0,01	12,40±0,07
Terpenoids	71,27±0,01	29,07±0,01	83,19±0,09	73,70±0,07	69,36±0,02	28,15±0,03	67,44±0,04	50,12±0,06
Others	0,89±0,02	1,91±0,02	1,28±0,08	3,91±0,06	1,88±0,04	1,86±0,03	1,92±0,05	1,23±0,05

Results are expressed as mean ± standard deviation

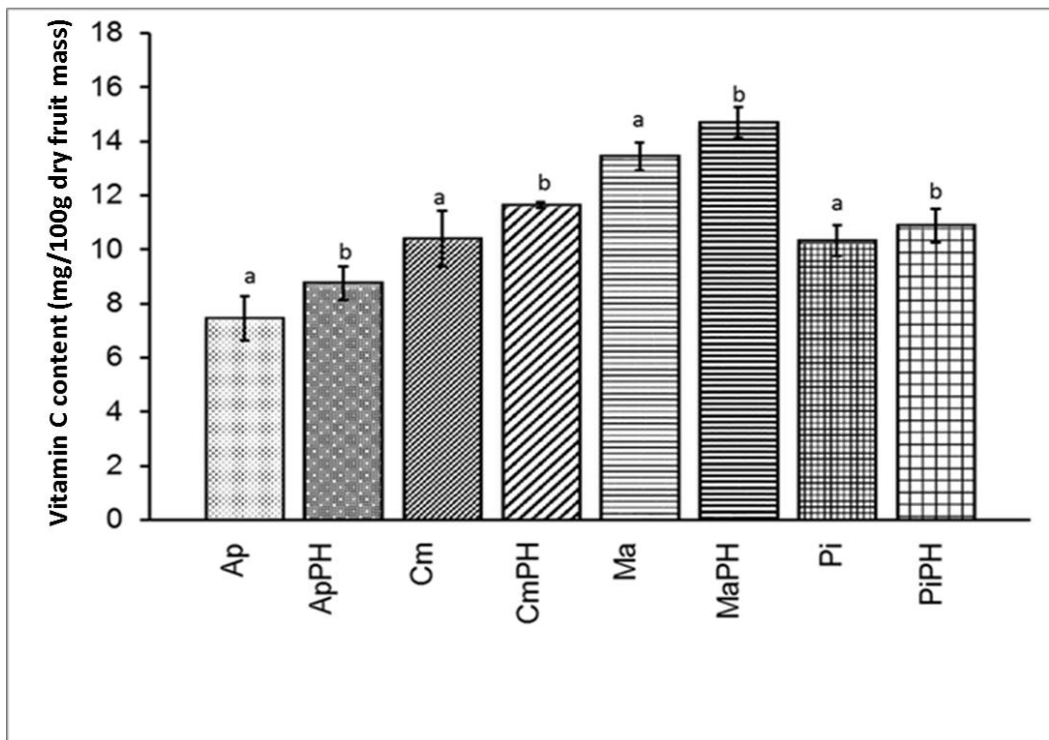


**Fig 1.** Moisture content of untreated and PH coated dehydrated fruits. For each fruit, columns with different letters are significantly different following Fisher's protected LSD test ( $P < 0.05$ ).

Figure 2

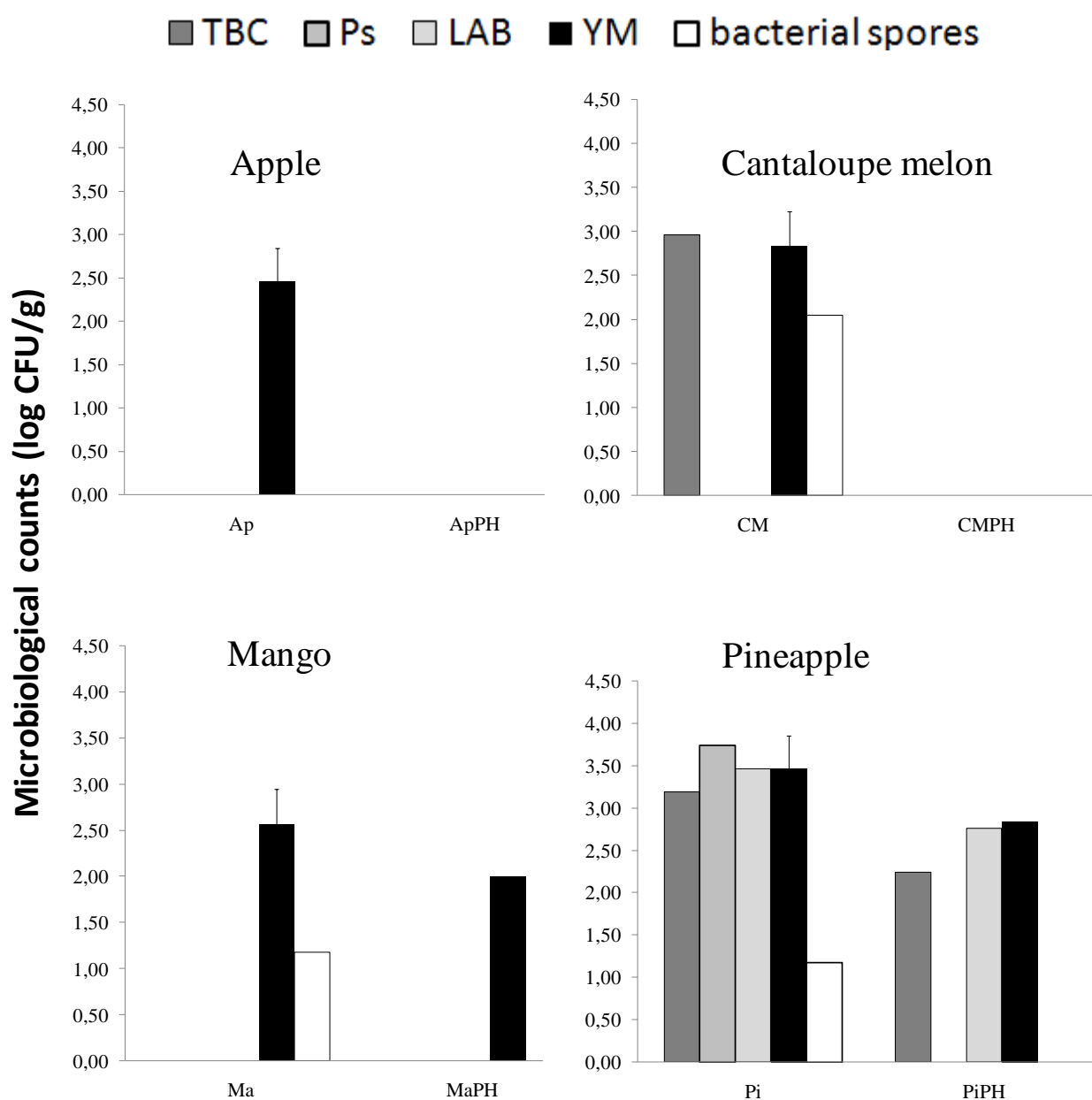


**Fig 2a,b.** Total polyphenol content (a) and antioxidant properties (b) of untreated and PH coated dehydrated fruits. For each fruit, columns with different letters are significantly different following Fisher's protected LSD test ( $P < 0.05$ ).



**Fig 3.** Vitamin C content of untreated and PH coated dehydrated fruits. Data is the average of three measurements, and the vertical bars indicate standard deviation. For each fruit, columns with different letters are significantly different following Fisher's protected LSD test ( $P < 0.05$ ).

Figure 4



**Fig. 4.** Microbiological counts (logCFU/g) of untreated and PH coated dehydrated fruits samples.

Total bacterial count (TBC, dark grey), Pseudomonas spp (Ps, grey), Yeast and Moulds (YM, black), Lactic acid bacteria (LAB, light grey), Spore formation bacteria (white).

Values indicated as 0.00 log CFU/g were lower than detection limits (TBC, Ps, LAB and YM).

Detection limits <math><2\log\text{CFU/g}</math>; Spore-forming bacteria: Detection limit <math><1\log\text{CFU/g}</math>).



Figure 5

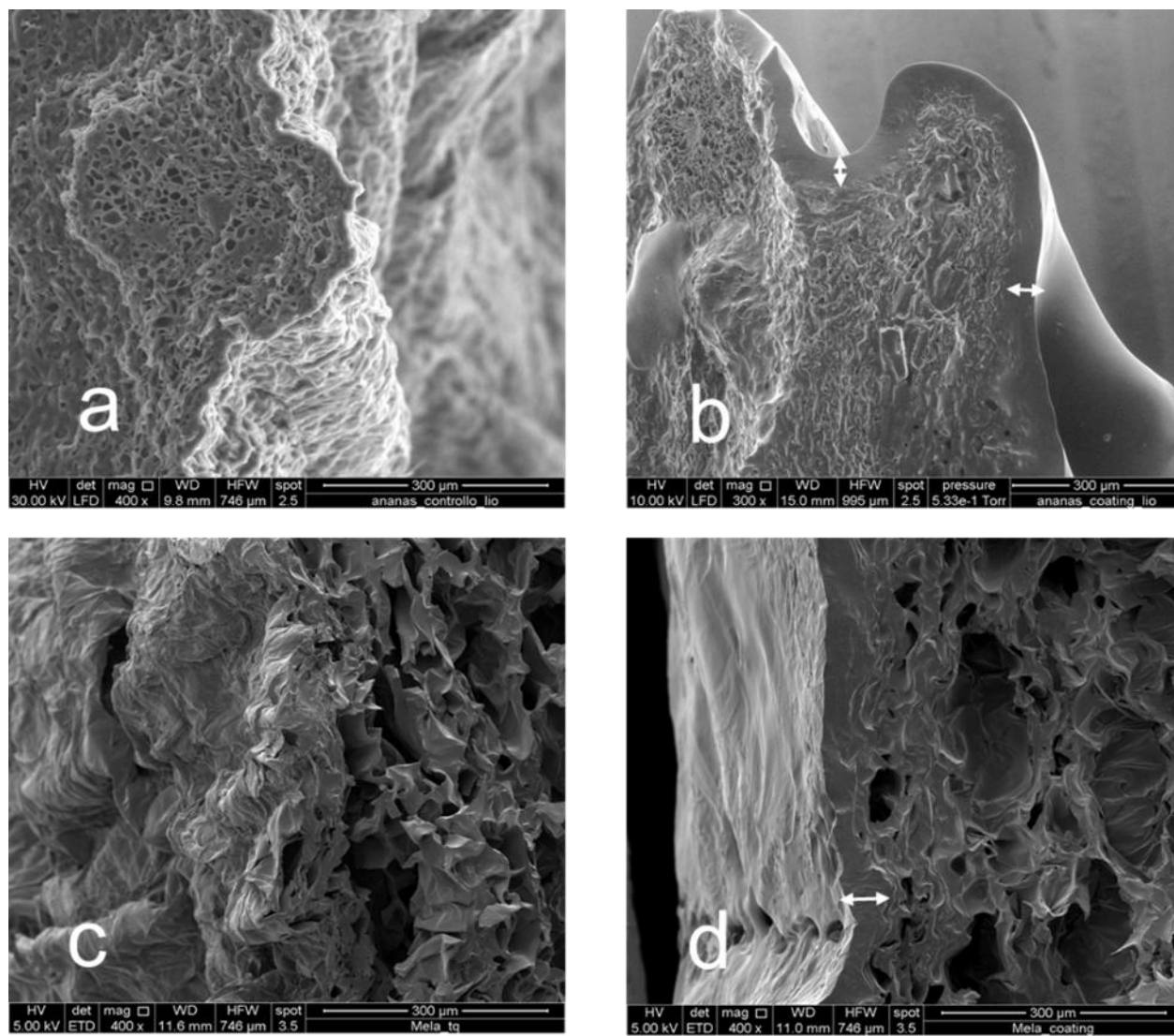


Fig 5 a,b,c,d. SEM cross-section micrographs of Pi (a), PiPH (b), Ap (c) and ApPH (d) fruit slices.

**Supplementary Material**

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