1	INFLUENCE OF APPLE PEEL POWDER ADDITION ON THE PHYSICO-CHEMICAL
2	CHARACTERISTICS AND NUTRITIONAL QUALITY OF BREAD WHEAT COOKIES
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4	Running title: Apple peel powder improves cookies quality
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6	Gjore Nakov ^{a*} , Andrea Brandolini ^b , Alyssa Hidalgo ^c , Nastia Ivanova ^a , Marko Jukić ^d ,
7	Daliborka Koceva Komlenić ^d , Jasmina Lukinac ^d
8	
9	^a University of Ruse "Angel Kanchev", branch Razgrad, blv. Aprilsko vastanie, 47 7200
10	Razgrad, Bulgaria.
11	^b Consiglio per la ricerca in agricoltura e l'analisi dell'economia agraria - Centro di Ricerca
12	Zootecnia e Acquacoltura (CREA-ZA), Via Forlani 3, 26866 S. Angelo Lodigiano (LO), Italy.
13	^c Department of Food, Environmental and Nutritional Sciences (DeFENS), Università degli
14	Studi di Milano, via Celoria 2, 20133 Milano, Italy.
15	^d Faculty of Food Technology Osijek, Josip Juraj Strossmayer University of Osijek, str. Franje
16	Kuhača 20, HR-31000 Osijek, Croatia.
17	

- 18 * Corresponding author; e-mail: gnakov@uni-ruse.bg; tel: +359882956044. ORCID iD: 0000-
- 19 0002-3780-8412

20 Abstract

21 Apple peel, a food industry by-product, is rich in fibre, polyphenols and minerals, and is a potentially attractive ingredient for bakery products. To evaluate the effect of wheat cookies 22 23 enrichment with apple peel powder (APP) six types of cookies with increasing APP percentage (0%, 4%, 8%, 16%, 24% and 32%) were produced. The traits analysed were: 24 pasting parameters; chemical properties (moisture, ash, lipid, protein, fibre and total 25 polyphenols content); antioxidant capacity (DPPH and FRAP methods); physical attributes 26 (width, thickness, volume and CIE lab colour); and sensory characteristics (external 27 appearance, internal structure, texture, odour, taste and aroma). Statistical analysis included 28 29 analysis of variance (ANOVA) followed by Fisher's Least Significant Difference test (p < 0.05). The APP-enriched cookies had significantly higher moisture, ash, lipid, fibre, total 30 polyphenols and antioxidant capacity than the control bread wheat cookies. The addition of 31 32 APP did not modify the physical characteristics and improved the sensorial quality of the products. The addition of 24% APP gave the cookies with the best overall quality. 33 34

35 Keywords

Antioxidant capacity; apple peel powder; cookies; fibre; polyphenols; sensory quality.
37

38 INTRODUCTION

39 Every year the food industry produces millions of tonnes of by-product waste that could be

40 used as a source of high-quality components (e.g. proteins, fibres, polysaccharides,

41 antioxidants, aromatic compounds, etc.) for the preparation of innovative foods,

42 pharmaceutics, cosmetics and other products (Lauková et al. 2016; Socaci et al. 2017).

Apple (Malus domestica Borkh) is widely grown in temperate climates all over the world; 4.9 43 44 million ha were planted with apple trees in 2017, producing over 83 million tonnes of fruits (FAO 2019). About 18% of world total production is processed into drinks, syrups, jams, 45 purees, etc. (Rabetafika et al., 2014), generating a vast amount of by-products whose disposal 46 may lead to environmental problems and adds extra costs to the food manufacturers. 47 Apple industrial waste, mainly pomace from the juice and cider industry or peel from the 48 49 preparation of canned apples, sauces, purees, dried rings, etc., has been proposed as a commodity, suitable to generate high added value by-products (Rabetafika et al., 2014). 50 Compared to the pomace, the peel is richer in lipids, ash and soluble dietary fibre (Rabetafika 51 52 et al., 2014), as well as in flavonoids (Rupasinghe et al. 2008; Wolfe and Liu 2003; Wolfe et al. 2003) and phenolic acids (Rabetafika et al., 2014). Phenolics and dietary fibre exert a 53 positive influence on the prevention of several diseases (Hidalgo et al. 2018; Mendoza-Wilson 54 55 et al. 2016; Rupasinghe et al. 2008) and have a positive effect on human health (Dahl et al., 2015; Henríquez et al. 2010). Furthermore, from a technological perspective, the dietary fibre 56 increases water and oil holding capacity, emulgation and gel forming (changing texture, 57 colour and aroma of foods), stabilises products with high content of fats and emulsions 58 (increasing their shelf life) (Karovičová et al. 2015), and lowers the energy of the final 59 60 products (Lauková et al. 2016). The increased sensibility and interest of the consumers in the nutritional quality of foods and 61

62 their potential benefits on the human health has led to a strong demand for functional

63 products, rich in natural antioxidants and dietary fibre (Rupasinghe et al. 2008). Baked

64 products, popular, ready-to-eat and consumed on a daily basis, are an ideal target for nutritive

65 improvement (Martins et al. 2017). While some information is available on the addition of

apple peel to muffins (Rupasinghe et al. 2008; Rupasinghe et al. 2009) or of apple pomace to

67 cookies (Lauková et al. 2016), no information on the utilisation of apple peel to improve the

68 nutritional quality of cookies is available. The exploitation of apple peel as an ingredient of

69 healthy foods can bring considerable benefits to human health (Chen et al. 1988);

additionally, the use of this by-product would provide to the apple industry an efficient and

71 environment-friendly solution for waste disposal (Rupasinghe et al. 2008).

Aim of our research was therefore to study the effect of the addition of dry apple peel powder

in different quantity (0%, 4%, 8%, 16%, 24% and 32%) on the physico-chemical, nutritional

74 and sensorial characteristics of wheat cookies.

75

76 MATERIALS AND METHODS

77 Materials

The type-550 white wheat flour used for cookies manufacturing was produced by the Sofia 78 Mel mills (Sofia, Bulgaria). The apple peel powder (APP) was prepared from apples of 79 80 Idared, a variety with red peel, tart and juicy white flesh. The apples were harvested in 2017 in Razgrad Province (43°32'00" N, 26°31'00" E), Bulgaria. After careful washing, the apples 81 82 were manually peeled; the peels were blanched in hot water for 30 s to stop enzymatic darkening, washed and dried for 48 hours at 60 °C in a UFE 500 oven (Memmert GmbH, 83 Schwabach, Germany). The dried peels were finely ground (size < 0.5 mm) with an IKA 84 85 MF10 grinder (IKA®-Werke GmbH & Co. KG, Staufen, Germany) and stored in sealed bags at 4°C until cookies manufacturing. All the other ingredients were obtained from shops in 86 Razgrad (Bulgaria). 87

88

89 **Cookies preparation**

90 Cookies were prepared according to method 10-50D (AACC International 2000), with minor

91 modifications. The control cookies were prepared from 100 g bread wheat flour, 33.0 g

92 glucose solution (5.93 g/L), 2.5 g baking soda, 2.1 g salt, 64 g butter and 36.0 mL water,

while in the enriched cookies 4%, 8%, 16%, 24% and 32% of the flour was replaced by equal 93 94 amounts of apple peel powder. The percentages used were chosen based on literature review information (e.g. Rupasinghe et al., 2008), preliminary tests and previous experience with 95 other by-products powders. The ingredients were kneaded for 300 s with an electronic mixer 96 (Stand Mixer ELITE STM-0248, Timetron Bulgaria, Sofia, Bulgaria). The dough was stored 97 in a refrigerator at 8 °C for 30 min, then flattened to 18 mm with a rolling pin and cut with a 98 99 die cutter (internal diameter: 44 mm). The cookies were baked for 10 min at 205 °C in an AEG BES351110M oven (AEG, Bulgaria), cooled at room temperature for 30 min and stored 100 at -20 °C until analysis. Six types of cookies were produced: control (100% wheat flour) and 101 enriched with increasing percentages (4%, 8%, 16%, 24% 32%) of apple peel powder. Two 102 sets of 10 cookies were prepared for each flour or mixture. 103

104

105 Pasting properties of raw materials

Pasting properties of flour and mixtures were examined using a Micro Visco-Amylo-Graph
(Brabender OHG, Duisburg, Germany) according to Nakov et al. (2018). The following
parameters were determined: peak viscosity, breakdown, setback and final viscosity. The
measurements were performed in duplicate; the results are presented in Brabender Units
(BU).

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112 Chemical characteristics

The moisture of flour, apple peel powder and baked cookies was evaluated according to
method 6540:1980 (International Standard Organisation 1980). Ash content was assessed
according to method 5984:2002 (International Standard Organisation 2002). Protein content
was determined as in Maehre et al. (2018). Lipid content was measured gravimetrically after
Soxhlet extraction of the acid hydrolysed sample (method 136, ICC 1995). Fibre content was

assessed as detailed in method 32-07.01 (AACC International 2000). Total carbohydratecontent was computed by difference.

120

121 Total polyphenol content and antioxidant activity

- 122 Total polyphenols content was determined on methanol extracts with the Folin-Ciocalteu
- method according to Slinkard and Singleton (1977) and expressed as mg gallic acid
- equivalent (GAE)/100 g dry matter (DM). The antioxidant capacity was tested using the 2,2-
- diphenyl-1-picrylhydrazyl (DPPH) radical cation following Brand-Williams et al. (1995), and
- the ferric reducing antioxidant power (FRAP) as described by Benzie and Strain (1996); the
- results are reported in μ g trolox equivalent (TE)/g DM.
- 128

129 Cookies physical characteristics

130 Cookie width was measured as the average of five cookies, and cookie thickness was

- 131 measured as the average of five stacked cookies, following method 10-50D (AACC
- 132 International 2000); both measurements are in mm. The volume was computed from the width

and thickness parameters, considering the cookies as cylinders. Two replicates of each

- 134 measurement were performed. The colour of the cookies in CIE $L^* a^* b^*$ system was
- assessed with a Chroma Meter CR-400 colorimeter (Konica Minolta, Tokyo, Japan) with the

136 use of illuminant D65 on two sets of five random cookies.

137

138 Cookies sensory analysis

139 Sensory analytical tests, aimed at objectively evaluating the characteristics of the cookies,

- 140 were performed at the University of Ruse "Angel Kanchev" Branch Razgrad, using the
- 141 methodology first described by Popov-Raljić et al. (2005). Twenty trained assessors
- 142 participated (14 females and 6 males, aged 21 to 32 years) to the sensory analysis. The

assessors were trained according to the guidelines given in the International standard ISO 143 144 8586 (2012) and selected according to three criteria: having a preference for the short dough cookies, eating cookies at least once a month, and having interest in participating in the study. 145 Informed consent from the participants was obtained, according to the guidelines on Ethics 146 and Food-Related research defined by the European Union (Alfonsi et al. 2012). External 147 appearance, internal structure, texture, odour, taste and aroma were scored from 1 to 5, where 148 1 is extreme dislike and 5 is extreme like; the final quality score was computed as the average 149 of the five traits scored. 150

151

152 Statistical analysis

153 Analysis of variance (ANOVA) and Fisher's Least Significant Difference test (LSD) at

154 p<0.05 were performed with the softwares XLSTAT 2017 (Addinsoft Inc., Long Island City,

155 NY, USA) and Office Excel 2013 (Microsoft, Redmond, WA, USA).

156

157 RESULTS AND DISCUSSION

158 Flour and apple peel powder composition

159 The characteristics of the bread wheat white flour and apple peel powder used in the

160 preparation of the cookies are reported in Table 1. The apple peel powder had lower proteins

- 161 content but higher ash, lipid, fibre, total polyphenols concentrations as well as superior
- 162 antioxidant capacity than the white flour, with values like those by Rupasinghe et al. (2008).

163 Table 1. Composition (mean ± standard deviation) and antioxidant activity (DPPH and FRAP
164 methods) of white bread wheat flour and apple peel powder. The means are computed from
165 three repetitions. GAE: gallic acid equivalent; TE: trolox equivalent.

	White flour	Apple peel		
	white nour	powder		
Moisture (g/100 g)	12.01±0.07	3.23±0.09		

Ash (g/100 g DM)	0.50 ± 0.07	4.78±0.01
Lipid (g/100 g DM)	1.38 ± 0.14	10.15±0.24
Protein (g/100 g DM)	13.21±0.60	3.20±0.98
Soluble fibre (g/100 g DM)	4.71±0.10	32.03±1.44
Insoluble fibre (g/100 g DM)	2.50±0.09	11.86±0.31
Total fibre (g/100 g DM)	7.21±0.54	43.89±1.85
Total carbohydrates	84.91±1.25	81.87±1.19
Total phenols (µg GAE/100 g DM)	101.4±18.22	1086.7±15.50
DPPH (µg TE/g DM)	96.6±0.50	228.1±1.42
FRAP (µg TE/g DM)	378.0±11.3	7142.3±218.6

168 **Pasting properties**

169 Table 2 shows several visco-amylographic parameters (peak viscosity, breakdown, setback, 170 final viscosity and pasting temperature) of the bread wheat flour and the five mixtures with increasing apple peel powder concentrations. The ANOVA (not presented) highlighted the 171 existence of significant differences (p<0.05) among the six samples for all the parameters. 172 Increasing the quantity of apple peel powder in the mixture led to a significant decrease of 173 peak viscosity, breakdown, setback and final viscosity values, whereas pasting temperature 174 increased. Similar conclusions were reached by Mir et al. (2015) when studying the pasting 175 176 properties of a mixture of wheat flour and apple pomace, leading them to point out that the increase in apple pomace lowered the starch content of the mixture, influencing the pasting 177 properties. An additional reason may be the high fibre (mostly pectin) content of apple peel 178 powder, which absorbs water and reduces the water available to starch granules, thus limiting 179 their swelling. Such a hypothesis was proposed by Sudha et al. (2007) after noticing that peak 180 viscosity of a suspension declined after increasing apple pomace content. Interestingly, a 181 decrease in peak and final viscosity was observed also by Rojas et al. (1999) after adding 182 pectin to wheat flour. 183

Table 2. Mean values (±standard deviation) of the pasting parameters of the six mixtures.
APP: apple peel powder; BU: Brabender units. The means are computed from three
repetitions. Values in the same column with different exponents are significantly different
(p<0.05) following Fisher`s LSD test.

APP	Peak viscosity BU	Breakdown BU	Setback BU	Final viscosity BU	Pasting temperature °C
0%	320.0 ^a ±4.04	208.0 ^a ±2.63	167.0 ^a ±5.66	279.0 ^a ±7.07	$61.1^{d} \pm 0.71$
4%	$291.5^{b} \pm 7.78$	182.0 ^{ab} ±2.79	162.0 ^{ab} ±0.00	271.5 ^{ab} ±4.95	$61.2^{d}\pm0.35$
8%	275.5 ^{bc} ±0.71	161.0 ^{bc} ±4.24	$154.0^{ab} \pm 1.31$	$268.5^{abc}{\pm}6.36$	62.1°±0.00
16%	$261.0^{\circ}\pm1.41$	151.5 ^c ±4.95	$158.5^{b}\pm0.71$	$268.0^{abc} \pm 4.24$	$62.4^{bc} \pm 0.07$
24%	$260.0^{c} \pm 1.52$	$142.5^{c}\pm2.12$	140.0°±2.83	$260.0^{bc} \pm 0.00$	$63.1^{ab} \pm 0.07$
32%	$259.0^{\circ}\pm1.40$	138.0 ^c ±1.41	137.5°±0.71	258.5°±0.71	63.8 ^a ±0.42

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190 **Physical characteristics**

In cookies, size, shape and colour are important choice elements for the consumer and are 191 192 strongly influenced by ingredients and production condition. The ANOVA (not presented) demonstrated the existence of significant differences (p<0.05) among samples for thickness, 193 194 but not for width; the volume, computed from these parameters, varied among samples; 195 significant differences (p <0.05) for all the colour parameters were also observed. Table 3 reports the mean values of the physical parameters. As hinted from the ANOVA, cookies 196 width was similar among samples (minimum value: 4.10 cm, maximum value 4.19 cm). On 197 the contrary, the thickness declined with the increase of apple peel powder in the samples, 198 passing from 2.51 cm (control) to 1.69 cm (cookie enriched with 32% apple peel powder). 199 Hence, the volume gradually declined from the control to the 32% APP cookies. A possible 200 reason for this trend is the high fibre content of APP that, when used to substitute bread wheat 201 flour, leads to a dilution of the storage proteins content and furthermore interferes with the 202 formation of the gluten matrix, thus giving smaller biscuits. In fact, Rupasinghe et al. (2009) 203 experienced a height reduction in muffins with 16%-32% APP compared to the all-bread-204

wheat control, while Lauková et al. (2016) observed a decrease of cookies volume after the

addition of 5, 10 and 15% apple powder. Similarly, Sudha et al. (2007) prepared cakes

207 containing increasing apple pomace percentages (up to 30%) and noticed that their volume

decreased from 850 cm^3 (control) to 620 cm^3 (30% apple pomace).

Baking affected the colour of the cookies, as shown by the results reported in Table 3 and by

the images in Supplementary Figure 1.

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Table 3. Mean values (±standard deviation) of the physical characteristics of the six cookie
 types. APP: apple peel powder. The means are computed from three repetitions. Values in the
 same column with different exponents are significantly different (p<0.05) following Fisher's
 LSD test.

216	APP	Width	Thickness	Volume	e Colour		
217		(cm)	(cm)	(cm ³)	L^*	<i>a</i> *	b^*
210	0%	4.10 ± 0.07	2.51 ^a ±0.01	34.5 ^a ±0.55	$75.6^{a}\pm0.85$	$2.6^{d}\pm 0.13$	32.8 ^a ±0.41
218 219	4%	4.15±0.02	$2.30^{b}\pm0.02$	31.4 ^{ab} ±0.38	$71.9^{b} \pm 1.11$	$3.0^d \pm 0.52$	$29.5^{b} \pm 1.86$
220	8%	4.16±0.02	$2.29^{b} \pm 0.02$	31.2 ^{ab} ±0.77	69.7 ^c ±2.21	$4.4^{c}\pm1.60$	28.5 ^{bc} ±0.15
221	16%	4.17±0.02	2.16 ^c ±0.22	29.3 ^b ±3.00	69.7°±2.03	4.3°±0.42	$27.9^{bcd} \pm 0.42$
223	24%	4.17±0.02	$1.84^d \pm 0.02$	24.9°±0.12	$62.1^d \pm 0.35$	$6.7^{b}\pm 0.68$	27.2 ^{cd} ±1.28
224	32%	4.19±0.02	1.69 ^e ±0.01	22.3°±0.93	$59.2^{e}\pm0.89$	$8.5^{a}\pm0.82$	$26.2^{d}\pm 0.99$

225 The control cookies, prepared with 100% bread wheat white flour, were the most luminous $(L^*=75.6)$ and the samples became darker with increasing apple peel powder additions; in 226 fact, the cookie with 32% APP had the lowest L^* (59.2). The b^* parameter followed a similar 227 pattern: the control had the highest score (32.8), while the increase of apple peel powder in 228 the cookies led to b^* as low as 26.2 in the 32% APP-enriched sample, indicating a decline of 229 the yellowish tinge of the cookies. Conversely, the a^* values increase with the addition of 230 apple peel powder, denoting a change from greenish to magenta hues. The addition of food 231 industry by-products such as APP, apple pomace or sour cherry cake has a darkening effect 232 on bakery products (Mir et al. 2015; Tumbas Šaponjac et al. 2016; Rupasinghe et al. 2009), 233 possibly because these ingredients are darker than the wheat flour and/or, being rich in 234

polyphenols, favour enzymatic darkening reactions (Mir et al. 2015). Recently, Pasqualone et

al. (2019) reported a similar darkening in cookies after the addition of phenolic-rich acorn

flour and attributed it to effect of polyphenoloxidase on the phenolic compounds.

238

239 Chemical characteristics

240 The ANOVA for moisture, ash, lipid, protein and fibre content showed significant differences 241 between cookie types. Fig. 1 depicts some chemical parameters of the different samples. The 242 moisture content (Fig. 1A) was low in the control (17.0%), and progressively grew with the increase in APP content, reaching 22.9% in cookies with 32% apple peel powder. This trend 243 is probably linked to the strong water binding properties of apple fibre (Chen et al. 1988) and 244 has been observed by other authors (Sudha et al. 2007; Rupasinghe et al. 2008). The ash 245 content is shown in Fig. 1B; the cookies without APP had the lowest content (1.81%) and the 246 addition of apple peel powder significantly increased (+21%) ash concentration, as reported 247 for muffins by Rupasinghe et al. (2008). The lipid content of the cookies is shown in Fig. 1C. 248 249 The control had the lowest concentration (18.1 g/100 g DM); adding APP increased it, up to 250 23.61 g/100 g DM in products with 32% APP, corresponding to a +30% variation. Different lipids are added in cookies manufacturing because they improve dough plasticity, air 251 252 retention, appearance and taste, but apple peel is particularly rich in unsaturated galactolipids 253 and phospholipids (Wang and Faust 1992). The protein content (Fig. 1D) showed a drastic reduction (-58%), from 19.37 g/100 g DM in the 100% white bread wheat flour cookies to 254 8.09 g/100 g DM in the 32% APP cookies. Proteins concentration and quality are other 255 relevant quality determinants in cookies, as during baking they coagulate and strengthen the 256 gluten structure. A protein content decrease in muffins after the addition of apple peel powder 257 is reported by Rupasinghe et al. (2008) and was attributed to the substitution of wheat flour 258 with the fibre-rich apple peel. 259







265 Dietary fibre content is depicted in Fig. 2A. In the control cookies, the soluble component

represented the majority (57.9%) of total fibre. A steady increase in total fibre was recorded

with the APP enrichment of cookies, passing from 2.2 g/100 g DM (control) to 10.4 g/100 g

268 DM (32% APP); interestingly, the soluble fraction showed a stronger raise (from 2.3 to 7.8

g/100 g DM) than the insoluble fraction (from 0.9 to 2.5 g/100 g DM), and in the cookies with

the highest APS addition represented 73% of the total fibre. An analogous trend was reported

by Rupasinghe et al. (2008) in muffins: their total fibre rose from 1.3 to 7.6% DM after the

incorporation of up to 24% APP and the soluble fibre fraction increased more than the

- insoluble fraction. The samples with 4 and 8% APP can be characterized as "fibre source",
- while the samples with 16, 24 and 32% APP can be labelled as "high fibre source" products
- according to Regulation (EC) No 1924/2006 of the European Parliament on Nutrition and
- 276 Health Claims Made on Foods.

Overall, the total carbohydrate concentration passed from 60.7 g/100 g DM in the control to 66.1g/100 g DM in the 32% APP-enriched cookies; however, the increase was in digestible fibre, which, compared to starch, has lower energetic value (FAO, 2003), helps to control the glycemic index (Alongi et al., 2019) and increases satiety (Burton-Freeman et al., 2017; Ye et al., 2015).

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283 Total polyphenols content and antioxidant capacity

The ANOVA for total polyphenols content and antioxidant capacity (not presented) showed 284 significant differences between cookie types. The total polyphenols content in the control 285 286 (Fig. 2B) was 146.15 µg GAE/g. Even a small addition (4%) of APP increased TPC content (250.6 μ g GAE/g), and the cookies with 32% APP reached a TPC content of 622.12 μ g 287 GAE/g. Fresh apples (110-357 mg/100 g), and fresh apple peels (309-589 mg/100 g) are a 288 289 good source of polyphenols (Wolfe et al. 2003); variety, ripeness, growing area, weather and management conditions modify their content and composition, but the polyphenols contained 290 291 in apples are among the best natural antioxidants (Mendoza-Wilson et al. 2016). Thus, the 292 utilisation of apple peel as an ingredient in sweet food products like cookies and muffins is an efficient and easy way to improve their nutritional characteristics and salutistic effects. The 293 294 antioxidant capacity (Fig. 2 C and 2D), determined with the DPPH and FRAP radicals, was 295 lowest (84.3 µg TE/g DM and 264 µg TE/g DM, respectively) in the APP-less control cookies. The increase of APP content led to a strong increase in the antioxidant capacity, up 296 to 126.7 µg TE/g DM (DPPH) and 2280.9 µg TE/g DM (FRAP) in cookies with 32% APP. 297 Rupasinghe et al. (2008) found that APP significantly increased both polyphenols content and 298 antioxidant capacity in muffins. Similarly, Hidalgo et al. (2018) reported a similar sharp 299 300 increase in TPC content (from 79.8 to 153.7 mg GAE/kg DM) and in FRAP antioxidant capacity (from 2.0 to 8.5 mmol TE/kg DM, i.e. from 508 to 2133 mg TE/g DM) after the 301

addition of microencapsulate beetroot pomace extracts to bread wheat water biscuits. Thermal
treatment during manufacturing can influence polyphenols composition and antioxidant
capacity of the products; for example, low moisture and high temperature (around 200 °C)
favour the transformation of quercetin into other compounds (Zhang et al. 2014), while
thermal treatments release bound phenolic acids from cell walls (Zielinski et al. 2001),
improving their bioavailability.



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Figure 2. Fibre (A), carbohydrates (B), total phenolic content (TPC; C) and antioxidant
capacity (DPPH and FRAP methods; D and E, respectively) of the six cookie types. I: control
(100% white wheat flour); II: 4% APP; III: 8% APP; IV: 16% APP; V: 24% APP; and VI:
32% APP. APP: apple peel powder. The bars indicate the standard deviations. Different
letters indicate significant differences (p≤0.05) among samples.

315 Sensory characteristics

316 Sensory characteristics are often the main reason to choose a product (Nakov et al. 2017). The results of the sensory analysis carried out on the six cookie types are shown in Fig. 3A. In 317 318 general, the addition of APP improved the aroma and the taste of the cookies without penalising internal structure, odour and appearance; in fact, only for texture the control 319 formulation outscored the APS-enhanced cookies. The addition of 24% APP gave the best 320 results for appearance, internal structure, texture and (together with 16% and 32% APP 321 cookies) taste, arguably the most important factor to consider when creating a new food 322 product (Rupasinghe et al. 2008). Overall, the 24% addition of APP gave the best cookies 323 (Fig. 3B), with an average score of 4.4 out of 5. Sudha et al. (2007), performing a sensory 324 analysis on muffins prepared with the addition of 8%, 16% and 24% apple pomace, concluded 325 that replacing wheat flour with up to 20% apple pomace did not worsen the products and gave 326 327 a pleasant fruity flavour.



Figure 3. Sensory analysis traits results (A) and overall quality (B) of the six cookie types. I: control (100% white wheat flour); II: 4% APP; III: 8% APP; IV: 16% APP; V: 24% APP; and VI: 32% APP. APP: apple peel powder. The sensory traits (y axis) vary from 1 (very poor) to 5 (excellent). The bars indicate the standard deviations. Different letters indicate significant differences ($p \le 0.05$) among samples.

337 CONCLUSIONS

Apple peel is an underutilised food industry by-product, and its disposal can be difficult and 338 expensive. Nevertheless, apple peel is rich in fibre, polyphenols and minerals, making it a 339 potentially attractive ingredient for bakery products. Our research demonstrated that APP-340 enriched cookies had higher moisture, ash, lipid, fibre, total polyphenols and antioxidant 341 342 capacity than the control cookies. Given the high fibre content, APP cookies can be characterized as "fibre source" and "high fibre source" products according to European 343 Community Regulation No 1924/2006. Furthermore, the addition of APP did not worsen the 344 345 physical characteristics of the products and helped to improve their sensorial quality. Overall, the addition of 24% APP gave the cookies with the best combination of physical, chemical 346 and sensorial qualities. APP can be considered a valuable source of polyphenols and dietary 347 348 fibre for the manufacturing of new bakery products with functional and nutraceutical properties; additionally, its utilisation will contribute to alleviate environmental concerns 349 350 associated with its disposal.

351

352 Conflict of interest

- 353 The Authors declare no conflict of interest.
- 354

355 **REFERENCES**

- AACC International (2000). Approved Methods of Analysis, 10th Ed. Methods 10-50.05 and
 32-07.01. AACC International, St. Paul, MN, USA.
- 358 Alfonsi A, Coles D, Halse C, Koppel J, Ladikas M, Schmucker von Koch J, Schroeder D,
- Sprumont D, Verbeke W and Zaruk D. (2012). Guidande note: ethics and food-related research.

- 360 European Group on Ethics in Science and New Technologies.
 361 http://ec.europa.eu/research/participants/data/ref/fp7/89847/research-food_en.pdf
- Alongi M., Melchior S and Anese M. 2019. Reducing the glycemic index of short dough
 biscuits by using apple pomace as a functional ingredient. *LWT-Food Science and Technology*,
 100, 300-305.
- Benzie I and Strain J. (1996). The ferric reducing ability of plasma (FRAP) as a measure of
 "antioxidant power": the FRAP assay. *Analytical Biochemistry* 239: 70-76.

367 Brand-Williams W, Cuvelier ME and Berset C. (1995). Use of a free radical method to evaluate

antioxidant activity. *LWT – Food Science and Technology*, 28: 25-30.

- Burton-Freeman B., Liyanage D, Rahman S and Edirisinghe I. (2017). Ratios of soluble and
- 370 insoluble dietary fibers on satiety and energy intake in overweight pre-and postmenopausal
- women 1. *Nutrition and healthy aging* 4: 157-168.
- 372 Chen H, Rubenthaler GL, Leung H and Baranowski JD. (1988). Chemical, physical and baking
- properties of apple fibre compared with wheat and oat bran. *Cereal Chemistry* 65: 244–247.
- 374 Dahl WJ and Stewart ML. (2015). Position of the Academy of Nutrition and Dietetics: health
- 375 implications of dietary fiber. Journal of the Academy of Nutrition and Dietetics, 115(11): 1861-

376 1870.

- FAO (2003). Food and Nutrition Paper 77. Food energy methods of analysis and conversion
- factors. Report of a Technical Workshop, Rome, 3-6 December 2002. Rome, 2003. Food and
- 379 *Agriculture Organization of the United Nations.*
- **380** FAO (2019). FAOstat-Food and Agriculture Organization of the United Nations statistics.
- 381 http://www.fao.org/faostat Accessed 20 June 2019.
- Henríquez C, Speisky H, Chiffelle I, Valenzuela T, Araya M, Simpson R and Almonacid S.
- 383 (2010). Development of an ingredient containing apple peel, as a source of polyphenols and
- dietary fiber. *Journal of Food Science* 75: 172-181.

- Hidalgo A, Brandolini A, Čanadanovic-Brunet J, Ćetkovic G and Tumbas-Šaponjac V. (2018).
- Microencapsulates and extracts from red beetroot pomace modify antioxidant capacity, heat
 damage and colour of pseudocereals-enriched einkorn water biscuits. *Food Chemistry* 268: 40-
- **388** 48.
- 389 ICC (1995). Standard method 136. In: ICC Standard Methods, 2nd supplement., *International*390 *Association for Cereal Science and Technology. Vienna, Austria.*
- International Standard Organisation (1980). ISO 6540:1980. Maize Determination of moisture
 content (on milled grains and on whole grains). *ISO, Vernier, Geneva, Switzerland*.
- International Standard Organisation (2002). ISO 5984:2002. Animal feeding stuffs Determination of crude ash. *ISO, Vernier, Geneva, Switzerland*.
- 395 International Standard Organisation (2012). ISO 8586:2012. Sensory analysis General
- 396 guidelines for the selection, training and monitoring of selected assessors and expert sensory
- 397 assessors. ISO, Vernier, Geneva, Switzerland.
- Karovičová J, Kohajdová Z, Minarovičová L and Kuchtová V. (2015). The chemical
 composition of grape fibre. *Potravinarstvo* 9: 56-57.
- 400 Lauková M, Kohajdová Z and Karovičová J. (2016). Effect of hydrated apple powder on dough
- 401 rheology and cookies quality. *Potravinarstvo* 10: 506-511.
- 402 Maehre KH., Dalheim L., Edvinsen KG., Elvevol OE and Jensen JI. (2018). Protein
 403 Determination Method Matters. *Foods* 7:5.
- 404 Martins ZE, Pinho O and Ferreira IMPLVO. (2017). Food industry by-products used as
- 405 functional ingredients of bakery products. *Trends in Food Science & Technology* 67:106-128.
- 406 Mendoza-Wilson AM, Castro-Arredondo IS, Espinosa-Plascencia A, Robles-Burgueño MR,
- 407 Balandrán-Quintana RR and Bermúdez-Almada MC. (2016). Chemical composition and
- 408 antioxidant-prooxidant potential of a polyphenolic extract and a proanthocyanidin-rich fraction
- 409 of apple skin. *Heliyon* 2: 00073.

- Mir SA, Bosco SJD, Shah MA, Santhalakshmy S and Mir MM. (2017). Effect of apple pomace
 on quality characteristics of brown rice-based cracker. *Journal of the Saudi Society of*
- 412 Agricultural Sciences 16: 25-32.
- 413 Nakov G, Brandolini A, Ivanova N, Stamatovska V and Dimov I. (2018). The effect of einkorn
- 414 (*Triticum monococcum* L.) whole meal flour addition on physico-chemical characteristics,
- biological active compounds and in vitro starch digestion of cookies. *Journal of Cereal Science*83: 116-122.
- 417 Nakov G, Koceva Komlenić D, Ivanova N, Damyanova S, Godjevargova T and Šušak A.
- 418 (2017). Sensory analysis of biscuits from einkorn flour, barley flour, einkorn flakes and wheat
- 419 flour in different proportions and different sugars. Proc. 9th International Congress FLOUR-
- 420 BREAD 2017-11th Croatian Congress of Cereal Technologists: 105-114.
- 421 Pasqualone A, Makhlouf FZ, Barkat M, Difonzo G, Summo C, Squeo G and Caponio F. (2019).
- 422 Effect of acorn flour on the physico-chemical and sensory properties of biscuits. *Heliyon* 5(8):
- **423** 02242.
- 424 Popov-Raljić J, Laličić J, Gorjanović R and Sikimić, V. (2005). Predlog Mogućnosti Senzornog
- 425 vrednovanja brašneno'konditorskih proizvoda I tupu tvrdog keksa, krekera i slanog peciva
- 426 (Suggestions for sensory evaluation of flour, confectionery products and dough of hard biscuits,
- 427 crackers and salty pastries). *Žito-hleb*, 32: 179-184.
- 428 Rabetafika HN, Bchir B, Blecker C and Richel A. (2014). Fractionation of apple by-products
- 429 as source of new ingredients: Current situation and perspectives. *Trends in Food Science and*
- 430 *Technology* 40: 99-114.
- 431 Regulation (EC) No 1924/2006 of the European Parliament and of the Council of 20 December
- 432 2006 on nutrition and health claims made on foods.
- 433 Rojas AJ, Rosell MC and Benedito de Barber C. (1999). Pasting properties of different wheat
- 434 flour-hydrocolloid systems. *Food Hydrocolloids* 13: 27-33.

- Rupasinghe VHP, Wang L, Huber MG and Pitts LN. (2008). Effect of baking on dietary fibre
 and phenolics of muffins incorporated with apple skin powder. *Food Chemistry* 107: 12171224.
- 438 Rupasinghe VHP, Wang L, Pitts LN and Astatkie T. (2009). Baking and sensory characteristics
- 439 of muffins incorporated with apple skin powder. *Journal of Food Quality* 32: 685–694.
- 440 Slinkard K and Singleton VL. (1977). Total phenol analyses: automation and comparison with
- 441 manual methods. *American Journal of Enology and Viticulture* 28: 49-55.
- 442 Socaci AS, Fărcaș CA, Vodnar CD and Tofană M. (2017). Food wastes as valuable sources of
- bioactive molecules. In: Superfood and functional food The development of superfoods andtheir roles as medicine. *IntechOpen*: 77-93.
- 445 Sudha LM, Baskaran V and Leelavathi K. (2007). Apple pomace as a source of dietary fiber
- and polyphenols and its effect on the rheological characteristics and cake making. *Food Chemistry* 104: 686–692.
- 448 Tumbas Šaponjac V, Ćetkovic G, Čanadanović-Brunet J, Pajin B, Djilas S, Petrović J,
- 449 Lončarević I, Stajčić S and Vulić J. (2016). Sour cherry pomace extract encapsulated in whey
- and soy proteins: incorporation in cookies. *Food Chemistry* 207: 27–33.
- 451 Wang SY and Faust M. (1992). Variation in lipid composition of apples in relation to watercore.
- 452 *Journal of the American Society of Horticultural Science* 117: 829-833.
- 453 Wolfe K and Liu RH. (2003). Apple peels as a value-added food ingredient. Journal of
- 454 *Agricultural and Food Chemistry* 51: 76–83.
- Wolfe K, Wu X and Liu RH. (2003). Antioxidant activity of apple peels. *Journal of Agricultural and Food Chemistry* 51: 609–614.
- 457 Ye, Z., Arumugam, V., Haugabrooks, E., Williamson, P. and Hendrich, S. (2015). Soluble
- 458 dietary fiber (Fibersol-2) decreased hunger and increased satiety hormones in humans when
- 459 ingested with a meal. *Nutrition research* 35: 393-400.

- Zhang X, Chen F and Wang M. (2014). Antioxidant and antiglycation activity of selected
 dietary polyphenols in a cookie model. *Journal of Agricultural and Food Chemistry* 62: 1643–
 1648.
- 463 Zielinski H, Kozlowska H and Lewczuk B. (2001). Bioactive compounds in the cereal grains
- 464 before and after hydrothermal processing. *Innovative Food Science & Emerging Technologies*
- 465 2: 159-169.