

# Accepted Manuscript

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Aislinn M. Richardson, Andrey A. Tyuftin, Kieran N. Kilcawley, Eimear Gallagher, Maurice G. O' Sullivan, Joseph P. Kerry



PII: S0023-6438(18)30344-X

DOI: [10.1016/j.lwt.2018.04.038](https://doi.org/10.1016/j.lwt.2018.04.038)

Reference: YFSTL 7049

To appear in: *LWT - Food Science and Technology*

Received Date: 8 December 2017

Revised Date: 12 April 2018

Accepted Date: 13 April 2018

Please cite this article as: Richardson, A.M., Tyuftin, A.A., Kilcawley, K.N., Gallagher, E., O' Sullivan, M.G., Kerry, J.P., The impact of sugar particle size manipulation on the physical and sensory properties of chocolate brownies, *LWT - Food Science and Technology* (2018), doi: 10.1016/j.lwt.2018.04.038.

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## **The impact of sugar particle size manipulation on the physical and sensory properties of chocolate brownies**

Aislinn M. Richardson<sup>1</sup>, Andrey A. Tyuftin<sup>1</sup>, Kieran, N. Kilcawley<sup>3</sup>, Eimear Gallagher<sup>4</sup>  
Maurice G.O' Sullivan<sup>2\*</sup>, Joseph P. Kerry<sup>1\*</sup>

<sup>1</sup> Food packaging group, School of Food & Nutritional Sciences, University College Cork, College Road, Cork, Ireland

<sup>2</sup> Sensory group, School of Food & Nutritional Sciences, University College Cork, College Road, Cork, Ireland

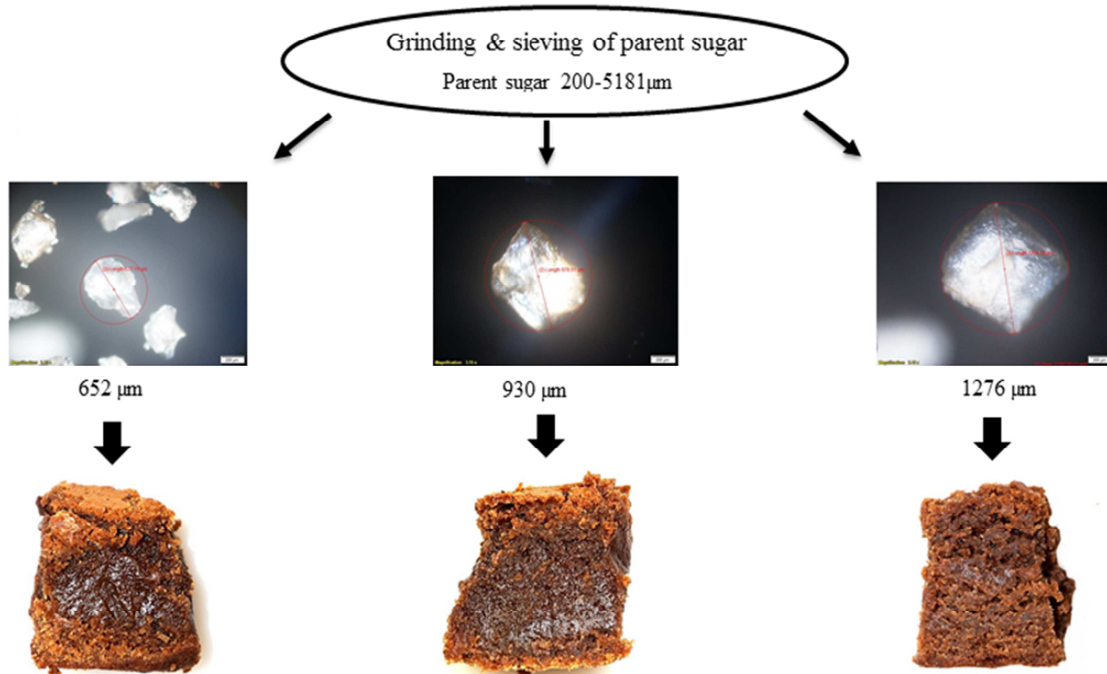
<sup>3</sup>Teagasc, Moorepark, Fermoy, Cork, Ireland

<sup>4</sup>Teagasc, Ashtown, Dublin Ireland

\*Corresponding authors, Tel: +353 (0)21 4903798, Fax: +353 (0)21 4276318, E-mail:

<sup>1</sup>[Joe.Kerry@ucc.ie](mailto:Joe.Kerry@ucc.ie) (J.P. Kerry) Tel: +353-(0)21-490-3544, E-mail:

<sup>2</sup>[maurice.osullivan@ucc.ie](mailto:maurice.osullivan@ucc.ie) (M.G. O'Sullivan).



Impact of sugar particle size on the physical properties and sensory attributes

**Graphical abstract**

ACCEPTED

1 **The impact of sugar particle size manipulation on the physical and sensory**  
2 **properties of chocolate brownies**

3 Aislinn M. Richardson<sup>1</sup>, Andrey A. Tyuftin<sup>1</sup>, Kieran, N. Kilcawley<sup>3</sup>, Eimear Gallagher<sup>4</sup>  
4 Maurice G.O' Sullivan<sup>2\*</sup>, Joseph P. Kerry<sup>1\*</sup>

5 <sup>1</sup> Food packaging group, School of Food & Nutritional Sciences, University College Cork,  
6 College Road, Cork, Ireland

7 <sup>2</sup> Sensory group, School of Food & Nutritional Sciences, University College Cork, College  
8 Road, Cork, Ireland

9 <sup>3</sup>Teagasc, Moorepark, Fermoy, Cork, Ireland

10 <sup>4</sup>Teagasc, Ashtown, Dublin Ireland

11 \*Corresponding authors, Tel: +353 (0)21 4903798, Fax: +353 (0)21 4276318, E-mail:

12 <sup>1</sup>[Joe.Kerry@ucc.ie](mailto:Joe.Kerry@ucc.ie) (J.P. Kerry) Tel: +353-(0)21-490-3544, E-mail:

13 <sup>2</sup>[maurice.osullivan@ucc.ie](mailto:maurice.osullivan@ucc.ie) (M.G. O'Sullivan).

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## 18 **Highlights**

- 19 • Small sugar particles increase sweetness intensity perception in chocolate brownies.
- 20 • Small sugar particles enhance the soft and moist texture of chocolate brownies.
- 21 • Sugar particle size manipulation significantly affects appearance, texture and colour
- 22 liking of chocolate brownies.

23

## 24 **Abstract**

25 The overall objective of this research was to assess the effect of sugar particle size  
26 manipulation on the physical and sensory properties of chocolate brownies. A control sugar  
27 (commercially available, 200-5181  $\mu\text{m}$ ) and four of its sieved sugar separates (mesh size of  
28 710, 500, 355 and 212  $\mu\text{m}$ ) were determined by grinding and sieving. The particle diameter  
29 and diameter distributions of the control sugar and each sugar fraction were measured. As a  
30 result, five sugar treatments were determined for chocolate brownie formulations; Control  
31 ( $C_{200-5181 \mu\text{m}}$ ), Large-particle replacement ( $LPR_{924-1877 \mu\text{m}}$ ), Medium-particle replacement  
32 ( $MPR_{627-1214 \mu\text{m}}$ ), Small-particle replacement ( $SPR_{459-972 \mu\text{m}}$ ) and a known MIX sample.  
33 Samples were tested using sensory (hedonic & intensity), instrumental (texture and colour)  
34 and compositional analyses (moisture and fat). Brownie samples containing the smallest  
35 sugar fraction ( $SPR_{459-972 \mu\text{m}}$ ) were perceived as significantly sweeter than any other sample  
36 ( $p < 0.05$ ). Brownies containing this fraction were also the softest and moistest samples  
37 ( $p < 0.05$ ). Texture liking was significantly associated with the  $LPR_{924-1877 \mu\text{m}}$  brownie  
38 ( $p < 0.05$ ). Darkness of brownie samples increased ( $p < 0.05$ ) as sugar particle size decreased.  
39 Therefore, sugar particle size alteration affects the physical and sensory properties of

40 chocolate brownies and could be used as a viable approach to reduce sugar in confectionery-  
41 type products.

42

### 43 **Keywords**

44 Sugar fraction, sensory analysis, texture, colour, sieved sugar

45

### 46 **1. Introduction**

47 The consumption of free and refined sugar in the diet is one of the main causes for obesity in  
48 society today (Hu & Malik 2010; MacGregor & Hashem, 2014). Free and refined sugar  
49 include; monosaccharides and disaccharides added to foods and beverages by the  
50 manufacturer, cook or consumer, and sugars naturally present in honey, syrups, fruit juices  
51 and fruit juice concentrates, as defined by the World Health organisation (WHO, 2015). A  
52 recent review involving a meta-analysis of randomised controlled trials and prospective  
53 cohort studies has established that intake of sugars is a determinant of body weight, with a  
54 clear positive association between higher intakes of sugars, body fat and long-term weight  
55 gain in adults (Te Morenga, Mallard, Mann & Morenga, 2013). In Ireland the Irish  
56 Universities Nutrition Alliance (IUNA, 2011) reported that free sugars account for 14.6% of  
57 the total energy intake of Irish adults who participated in the study. According to the Healthy  
58 Ireland survey (2015), 37% of adults that participated were overweight and a further 23%  
59 were obese. Obesity is a strong risk factor for type 2 diabetes (Chan, Rimm, Colditz,  
60 Stampfer & Willett, 1994; Rosner, Speizer & Manson, 1997) and the Slán (2007) study  
61 reported that the estimated prevalence of pre-diabetes among over 45 year olds in Ireland was  
62 19.8% (Morgan et al. 2008). A new guideline was published by the WHO in March 2015

63 which strongly recommends that adults and children reduce their daily intake of free sugars to  
64 less than 10% of their total energy intake. A conditional recommendation was also made to  
65 further reduce free sugar intake to below 5% of total energy intake (WHO 2015).

66 Confectionery and snacks account for 9% of carbohydrate intake in Irish adults, therefore  
67 reducing sugar in these products would be a significant development in reducing the dietary  
68 intake of sugar (Irish Universities Nutrition Alliance (IUNA) 2011). However, sugar has a  
69 huge part to play in the sensory properties of confectionery products, such as cake and cake-  
70 like products. Consequently, reducing sugar in these products presents a huge challenge for  
71 the food industry. Sugar is responsible for the sweetness in cakes and muffins and sucrose is  
72 the most commonly used sugar in cake making (Bennion and Bamford, 2013). According to  
73 Martínez-Cervera, Sanz, Salvador & Fiszman, 2012), sugar inhibits or reduces gluten  
74 development during cake batter mixing by competing with gluten proteins for water and thus,  
75 acts as a tenderiser of baked goods. The incorporation of air during batter creaming is  
76 facilitated by the addition of sucrose (Shepard & Yoell, 1976). This lightens the batter and air  
77 pockets formed during creaming expand and lift the batter, causing it to rise during baking.  
78 Sugar binds moisture and moisture content varies between the different types of sugar, for  
79 example liquid sugars contain more moisture than brown sugar and brown sugar contains  
80 more moisture than crystalline white sugar (Manley, 2011). Therefore sugar is not only  
81 responsible for the sweetness of cakes, but contributes significantly and positively to the  
82 sensory and physical properties of cakes.

83 Recent reports indicate that the global sugar substitutes market is valued at around \$11.5  
84 billion and it is expected to grow up to \$14 billion by 2019 (Markets and markets. 2015). The  
85 inclination for combination of non-nutritive sugar substitutes with sugar alcohols to produce a  
86 low-calorie bakery product has increased, with artificial sweeteners such as aspartame and  
87 sucralose providing sweetness and sugar alcohols providing the bulking properties. However,

88 controversy exists over the use of artificial sweeteners in foods and beverages (Suez et al.  
89 2014; Azad et al. 2016). In a recent US Mintel survey it was found that 64% of respondents  
90 indicated they were concerned about the safety of “artificial” sweeteners. (Gardner et al.  
91 2012). This is an important finding for the food industry if companies are to implement  
92 replacement strategies using artificial sweeteners. Therefore, it is necessary to pursue other  
93 strategies for sugar reduction/replacement in such products.

94 In this study, a new strategy of sugar reduction based on the manipulation of sugar particle  
95 size is proposed. From extensive review of the scientific literature, we have not been able to  
96 detect research investigating the effect of sugar particle size on sweetness perception and  
97 overall acceptability of cakes. Sugar particle size has been shown to affect flour cookie  
98 quality (Kissel, Marshall, Yamazaki, 1973). Manley, (2011) reported that sucrose crystal size  
99 and their rate of dissolution affects the appearance and crunchiness of baked biscuits. Rama  
100 et al. (2013) conducted a study on salt particle size manipulation and found that smaller salt  
101 crystals increased salt perception in fried sliced potato crisps in a controlled chewing  
102 environment. This proves that salt size manipulation can be used to reduce salt in crisp  
103 products. Based on these findings, we hypothesise that smaller sugar particles increase  
104 sweetness perception in chocolate brownies.

105 The primary objective of this study was to determine the effect of sugar particle size on the  
106 sensory (hedonic, descriptive) and physical properties of chocolate brownies and to determine  
107 if this approach might constitute an effective strategy for reducing sugar levels in  
108 confectionery-type products.

109

## 110 **2. Materials and methods**



111 Food ingredients used in this trial, included; Light golden soft brown sugar (1.1% moisture,  
112 98% sucrose, cane molasses and invert sugars, Siucra brand, UK); Irish Creamery butter  
113 (81% total fat, 65.4% of which were saturated, Dunnes stores, Ireland); Cream plain flour  
114 (1.4% fat, 82.7% carbohydrate, 2% of which sugars, 3.4% fibre, 11.7% protein and 0.81%  
115 salt, Odlums, Ireland); Free range eggs (Upton, Ireland); Dark chocolate (34.7% total fat,  
116 55.8% carbohydrate, 97.2% of which sugars, 3.6% protein and 0.1% salt, Homecook wonder  
117 bar, Ireland). Food products were all purchased from a local supermarket and stored under  
118 refrigerated or cool, dry conditions where appropriate prior to sample preparation.

119

## 120 2.1 Sieving

121 Sugar was stored at ambient temperatures of 20°C prior to grinding and sieving. Sugar was  
122 dried at 70°C for one hour (h) in an oven (Binder, ED 115, Germany) to reduce moisture  
123 content for more effective sieving. Moisture content was obtained for the sugar, both before  
124 and after drying, using methodologies described below. Moisture content (%) was kept  
125 constant at 0.5% for all sugar fractions. Dried Sugar was ground by hand using rolling pins  
126 and mechanically sieved through a sequence of sieves (90, 180, 212, 355, 500, 710, 1,180 and  
127 2,360  $\mu\text{m}$ ) set in a sieve shaker (Endeotts Octagon 200 London, England). Sieving was  
128 carried out in batches of 200g of sugar for 10 minutes (min) at 5-mm amplitude and particle  
129 size distributions of the sugar, both before and after grinding, were obtained using this  
130 method. For the purpose of the baking trials, four sugar-sieve separates were established; 212,  
131 355, 500, and 710 $\mu\text{m}$ . The un-ground, un-sieved, commercially-available parent sugar was  
132 used as the control. Several separations were carried out until 1kg sugar quantities were  
133 available for all size ranges.

134

## 135 2.2 Measurement of particle size

136 Sugar-sieve separates obtained by milling and sieving were analysed by microscopy using a  
137 light microscope (Olympus BX-61 Tokyo, Japan) and cellSens™ standard software (version  
138 510\_UMA\_Database\_cellsens19-Krishna-en\_00). The average particle size and distributions  
139 of the parent sugar and each separate (212, 355, 500 and 710  $\mu\text{m}$ ) were determined by  
140 obtaining and recording the 2D longest diameter of 100 particles per fraction in transparent  
141 light mode. Particle images within each fraction were captured using a microscope digital  
142 camera lens (Olympus DP73 Tokyo, Japan).

143

## 144 2.3 Chocolate brownie preparation

145 Three independent batches of brownies for all experimental treatments were manufactured in  
146 the preparation area of the sensory science laboratory, University College Cork. For the  
147 purpose of the experiment, samples were identified as follows; C<sub>200-5181  $\mu\text{m}$</sub>  (Control), LPR<sub>924-  
148 1877  $\mu\text{m}$</sub>  (large-particle replacement), MPR<sub>627-1214  $\mu\text{m}$</sub>  (medium-particle replacement), SPR<sub>459-972  
149  $\mu\text{m}$</sub>  (small-particle replacement) and MIX (mix of 50% SPR, 40% MPR and 10% of the finer  
150 particle size captured by the 212  $\mu\text{m}$  sieve mesh size). Dark chocolate (175g) and butter  
151 (175g) were melted in a heat stable bowl for one min in a microwave oven. The melted  
152 mixture was stirred for 30 seconds before sugar (250g) was added and stirred by hand for  
153 another one min. Eggs (180g) were beaten in a separate bowl and added to the mixture. All of  
154 the ingredients were stirred by hand for one min until the flour (115g) was sieved into the  
155 mixture. Mixture was stirred by hand until smooth (two mins). The batter was poured into  
156 tinfoil trays (16.5x24cm) and batches were baked for 30 min at 180° C in a Zanussi  
157 convection oven (C. Batassi, Conegliano, Italy). Batches of brownies were left to set for 30  
158 min in the tray before being removed and cut into Individual brownie pieces (45x45mm).

159 Chocolate brownies were placed on a rack for cooling for one hour before being removed and  
160 placed into plastic containers for storage prior to testing.

161

## 162 2.4 Sensory analysis

### 163 *Sensory affective evaluation*

164 Sensory acceptance testing (SAT) was carried out in the panel booths of the sensory science  
165 laboratory, food science building, University College Cork according to international  
166 standards (ISO 11136:2014). SAT took place over three separate days as three independent  
167 trials were carried out for all five treatments. SAT was conducted according to the methods of  
168 Stone, Bleibaum & Thomas, (2012a) using a total of 70 untrained assessors (n=70) all of  
169 which were regular consumers of chocolate brownies and had experience with SAT. Samples  
170 (2x2x2cm) were assigned a randomised three-digit code and presented in duplicate (Stone,  
171 Bleibaum & Thomas, 2012b). Thus, each samples was evaluated 140 times (70 x 2). Sessions  
172 were carried out at room temperature under white light and sensory evaluators were  
173 instructed to use the water provided to cleanse their palates between tastings. Participants  
174 used the Hedonic descriptors summarised in Table 1 to rate chocolate brownie samples.  
175 Assessors were asked to indicate their degree of liking for samples on a 10cm continuous  
176 line-scale ranging from 0 (extremely dislike) at the left to 10 (extremely like) at the right  
177 (Rodrigue, Guillet, Fortin & Martine 2000; Fellendorf, O' Sullivan, Kerry, 2016). Overall  
178 acceptability was also evaluated using the scale from 0 cm, extremely unacceptable to 10 cm,  
179 extremely acceptable.

180

### 181 *Ranking descriptive analysis*

182 Following sensory acceptance testing assessors were trained and participated in a separate  
183 ranking descriptive analysis (RDA) according to the method of Richter, Almeida, Prudencio  
184 & Benassi, 2010. Training and RDA took place three separate times as three independent  
185 trials were carried out for all five treatments. Sensory descriptors were selected from panel  
186 discussion as the most appropriate and reflected the main variation in the samples profiled.  
187 The consensus list of sensory descriptors (Table 1, intensity), were measured on a 10 cm  
188 continuous line scale with the term “none” used as the anchor point for the 0 cm end of the  
189 scale to “extreme” for the 10 cm end of the scale ( Fellendorf, O’ Sullivan, Kerry, 2016).  
190 Each trained panellist was asked to rank each sample for each attribute. RDA was carried out  
191 in the panel booths of the sensory science laboratory, food science building, University  
192 College Cork using 70 trained assessors in total over three separate sessions (25+25+20=70).  
193 The samples (2x2x2cm) were served coded in randomised order and presented  
194 simultaneously to assessors (Stone, Bleibaum & Thomas, 2012b) in duplicate.

195

## 196 2.5 Chocolate brownie images

197 Photographs were taken of the chocolate brownie samples portioned out for sensory analysis  
198 using a digital camera (Nikon D3200, Japan).

199

## 200 2.6 Instrumental analysis

### 201 *Texture*

202 As outlined already three independent trials were carried out for all five treatments. Two  
203 chocolate brownies (45x45mm) from the centre of each batch tray were used for texture  
204 analysis (3 x 2 = 6). Texture profile analysis (TPA) was carried out on samples using a

205 Texture Analyser 16 TA-XT2I (Stable Micro Systems, Surrey, UK). A 50% double  
206 compression test was carried out on each sample to a height of 2cm with a 75mm diameter  
207 flat-ended cylindrical probe (P/75), at a speed of 1mm/s with a 5 sec waiting time between  
208 the two cycles. This was carried out in accordance with the method of Martínez-Cervera,  
209 Sanz, Salvador & Fiszman, (2012).

210

### 211 *Colour*

212 Two chocolate brownies (45x45mm) from the top right of each batch tray were used for  
213 colour analysis. Crust and crumb colour characteristics were assessed by the CIE  $L^*a^* b^*$   
214 method. Lightness  $L^*$  was defined by means of a Minolta CR-200B Chroma Meter (Minolta  
215 Camera Co. Ltd., Osaka, Japan). The  $L^*$  parameter ( $L^*=0$  [Black],  $L^*=100$  [White]) for  
216 crust was measured at two separate points directly from the top of each individual brownie  
217 sample. The brownie samples were cut horizontally to remove the crust and crumb colour  
218 was measured directly at two separate points. As a result four measurements for crust colour  
219 and four measurements for crumb colour were taken for each treatment and as three trials  
220 were carried out for each treatment results for crust and crumb colour represent a mean of  
221 eight measurements.

222

### 223 2.7 Moisture and fat

224 Two chocolate brownies (45x45mm) from each independent trial (three) were used for  
225 moisture and fat determination ( $3 \times 2 = 6$ ). Samples were homogenised for compositional  
226 analysis using Büchi Mixer B-400 (Büchi Labortechnik AG, Switzerland). Moisture content  
227 was determined using the CEM SMART system and fat was determined using the SMART  
228 Trac system (CEM GmbH, kamp-Lintfort, Germany). Two fibreglass pads were placed in the

229 drying chamber of the CEM SMART system and the weight of the pads were tared.  
230 Homogenised samples (2-4g) were weighed accurately on the fibreglass pads and afterwards  
231 one pad was placed over the sample and pressed together. Percent of fats was determined by  
232 wrapping the fibreglass pads with the sample in a sheet of Smart Trac film. Wrapped samples  
233 were placed in Smart Trac tube and positioned in the Smart Trac NMR unit. Percentage fat is  
234 displayed after roughly 5 min. Methods were carried out in accordance with that of Bostian,  
235 Fish, Webb & Arey, (1985) with slight modifications for confectionery samples.

236

## 237 2.8 Statistical analysis

238 As stated previously all chocolate brownie formulations were prepared in three independent  
239 batch trials and two independent samples for each experimental treatment from each of these  
240 batches were assessed for each parameter, providing a total of 6 independent samples, unless  
241 stated otherwise above. Raw data obtained from sensory evaluation was coded into Microsoft  
242 excel and analysed using ANOVA- Partial Least Squares Regression (APLSR) using  
243 Unscrambler software version 10.3 (CAMO ASA, Trondheim, Norway). The X-matrix was  
244 defined as the different sample treatments. The Y – matrix contained the sensory variables of  
245 the design. To achieve significant results for the relationships determined in the quantitative  
246 APLSR, regression coefficients were analyzed by jack-knifing which is based on cross-  
247 validation and stability plots (Martens & Martens, 2000). Statistical significance for sensory  
248 data was defined as  $P < 0.05$ - $0.01$  (significant),  $P < 0.01$ - $0.001$  (highly significant) and  $P < 0.001$   
249 (extremely significant). Texture and compositional data were presented as a mean of six  
250 values  $\pm$  standard deviation. Colour (crust and crumb) data was presented as a mean of eight  
251 values  $\pm$  standard deviation. One-way ANOVA was used to compare the means of the data  
252 obtained from instrumental analysis and compositional analysis. Tukey's post-hoc test was

253 used to adjust for multiple comparisons between treatment means using SPSS statistics 20  
254 software (IBM, Armonk, NY, USA).

255

### 256 **3. Results and discussion**

#### 257 3.1 Particle size distribution

258 Particle size distribution (PSD) of brown sugar, both before and after grinding, can be seen in  
259 Fig 1. It is clear from this chart that significant particle size differences exist within the parent  
260 sugar employed for this trial. Sugar particles captured by 710, 500 and 355  $\mu\text{m}$  sieves  
261 increased by 34.1%, 17.4% and 7.1%, respectively after grinding. No particles  $>2,360 \mu\text{m}$   
262 were present after grinding and particles captured by the sieve with the second largest  
263 aperture (1,180  $\mu\text{m}$ ) decreased by 21.31%. Finer sugar particles  $<212 \mu\text{m}$  were present after  
264 grinding. Visual representation of the particle diameter distribution of control sugar and  
265 individual sugar separates can be seen in Fig 2. Particle size diameter differences between  
266 control sugar and sugar separates were evident. Control sugar had the widest particle size  
267 distribution as expected with particles ranging from 200-5181  $\mu\text{m}$ . After grinding and  
268 separation, particle size distribution within each fraction became smaller, in the range of 924-  
269 1877  $\mu\text{m}$  for LPR, 627-1214  $\mu\text{m}$  for MPR and 459-972  $\mu\text{m}$  for SPR. Particle size ranges and  
270 mean sizes for control sugar and each sieved sugar-separate are shown in Table 2.  
271 Microscopic images for brown sugar particles with 2D diameters for different mesh sizes are  
272 represented in Fig 3.

273

#### 274 3.2 Sensory analysis

275 A total of 54.8% of the sensory evaluators who participated in this study were female and  
276 45.2% were male. Ages of assessors ranged from 18-45. Significance of estimated regression  
277 coefficients for the relationship of sensory terms and chocolate brownies are presented in  
278 Table 3.

279 The control sample ( $C_{200-5181 \mu m}$ ) was negatively associated with crust darkness, ( $p < 0.001$ ).  
280 Brownie samples containing  $LPR_{924-1877 \mu m}$  were significantly positively associated with  
281 colour liking ( $p < 0.05$ ) whereas brownie samples containing  $SPR_{459-972 \mu m}$  were negatively  
282 associated ( $p < 0.05$ ).

283 Chocolate brownie samples with  $LPR_{924-1877 \mu m}$  were positively associated with texture liking  
284 ( $p < 0.01$ ). Samples containing  $SPR_{459-972 \mu m}$  were extremely associated with having a moist  
285 texture ( $p < 0.001$ ). These samples were also significantly negatively associated with texture  
286 hardness ( $p < 0.01$ ). Therefore brownie samples containing this fraction ( $SPR_{459-972 \mu m}$ ) were  
287 perceived as the softest and moistest samples. In contrast control samples ( $C_{200-5181 \mu m}$ ) were  
288 perceived as the hardest samples ( $p < 0.05$ ). It has been observed that replacement of sucrose  
289 with different fibres increases crumb firmness in muffins (Struck, Gundel, Zahn & Rohm,  
290 2016). The authors cite air cell incorporation as a contributing factor to mechanical  
291 resistance. The presence of larger sugar particles in the  $C_{200-5181 \mu m}$  sample in this study could  
292 have impacted upon air cell incorporation and could therefore be contributing to the increased  
293 hardness observed in samples.

294 The chocolate brownie samples containing the smallest sugar fraction ( $SPR_{459-972 \mu m}$ ) were  
295 perceived as significantly sweeter than any other sample ( $p < 0.05$ ). This finding is in  
296 agreement with results obtained for salt crystal size manipulation, with smaller salt particles  
297 being shown to increase saltiness perception in crisps (Rama et al., 2013).



298 Images of chocolate brownie samples divided out for sensory analysis can be seen in Fig 4.  
299 Visual variation in brownie texture was evident. In agreement with the sensory data presented  
300 in this study, brownie samples containing SPR<sub>459-972 μm</sub> had the greatest moist appearances.

301

### 302 3.3 Texture and colour analysis

303 Texture profile analysis (TPA) results are shown in Table 4. In agreement with sensory data,  
304 chocolate brownie samples containing SPR<sub>459-972 μm</sub> were the softest samples with the force  
305 ( $45.1 \pm 2.42$  N) required to compress brownie samples being lower ( $p < 0.05$ ) than determined  
306 for any other sample. Contradictory to the sensory data, brownie samples containing LPR<sub>924-</sub>  
307 <sub>1877 μm</sub> were the hardest samples ( $69.2 \pm 2.12$  N) ( $p < 0.05$ ). As sugar particle size decreased,  
308 hardness values decreased significantly ( $p < 0.05$ ) with the exception of the Control and Mix  
309 samples. These results are similar to mean cake strength results obtained by Dozan, Benković  
310 & Bauman (2014), who found that cake strength increased with increasing sugar particle size  
311 due to the force required for crystal breakage, as well as cake breakage. Similarly, Dozan,  
312 Benković & Bauman (2014) demonstrated that the force required to compress cakes with  
313 larger crystals was greater than the force required to compress cakes with smaller crystals. In  
314 our study, chewiness values (N-mm) varied significantly between samples. Chocolate  
315 brownies containing SPR<sub>459-972 μm</sub> were found to have the lowest value ( $4.2 \pm 0.23$  N-mm) for  
316 chewiness (chewiness hardness x cohesiveness x springiness) and different ( $p < 0.05$ ) from all  
317 other samples. Brownie samples containing MPR<sub>627-1214 μm</sub> presented the second lowest value  
318 ( $p < 0.05$ ) for chewiness ( $5.0 \pm 0.50$  N-mm) and samples containing a mix of sugar particle  
319 sizes (MIX) obtained the third lowest value ( $p < 0.05$ ) for chewiness  $6.2 \pm 0.13$  N-mm).  
320 Control<sub>200-5181 μm</sub> and LPR<sub>924-1877 μm</sub> brownie samples were not significantly different from  
321 each other with regards chewiness, but both samples presented the highest values ( $p < 0.05$ ).

322 The slightly higher chewiness values ( $9.8 \pm 0.12$  N-mm) associated with chocolate brownie  
323 samples containing  $LPR_{924-1877 \mu m}$  could be a reason why these samples were liked so much  
324 in terms of texture and also may be a reason why these samples were not perceived correctly  
325 as the hardest samples as determined during sensory evaluation (see Table 3). No significant  
326 differences were observed between brownie samples with respect to other physical product  
327 parameters such as adhesiveness, springiness, cohesiveness, or resilience.

328 In accordance with sensory data, control brownie samples had the lightest crust, which was  
329 different ( $p < 0.05$ ) from any other sample (Table 5). Trends showed that as sugar particle size  
330 decreased, darkness of crust colour increased. The control sample also had the lightest crumb  
331 colour ( $p < 0.05$ ) compared to all other brownie samples, with the exception of those samples  
332 containing  $LPR_{924-1877 \mu m}$ . Trends showed that as sugar particle size decreased, darkness of  
333 crumb colour increased, with samples containing  $SPR_{459-972 \mu m}$  having the darkest crumb  
334 colour ( $24.4 \pm 1.81$ ). The darker crumb and crust colour can be associated with the lower  
335 melting point of smaller sugar crystals which can caramelize quicker than larger crystals. The  
336 darker crumb colour of  $SPR_{459-972 \mu m}$  could be a reason why this sample was negatively  
337 associated with colour liking as determined by sensory evaluation.

338

### 339 3.4 Moisture and fat content

340 As anticipated, fat (%) did not vary between samples as fat content remained constant in  
341 samples during baking. The average fat content determined in baked brownies ranged from  
342 26.24 to 27.64% as shown in Table 6. However, moisture content varied significantly  
343 between samples. As sugar particle size decreased, moisture content increased in brownie  
344 samples, with the exception being that of the MIX sample.  $Control_{200-5181 \mu m}$  and  $LPR_{924-1877}$   
345  $\mu m$  brownie samples had the lowest moisture content and were different ( $p < 0.05$ ) from

346 samples containing MPR<sub>627-1214 μm</sub> and SPR<sub>459-972 μm</sub>, but not significantly different from the  
347 MIX sample. Chocolate brownie samples containing SPR<sub>459-972 μm</sub> had the highest ( $p<0.05$ )  
348 moisture content ( $13.0 \pm 0.84$ ) compared to all other brownie samples, with the exception of  
349 samples containing MPR<sub>627-1214 μm</sub> which had the second highest moisture content ( $p<0.05$ ).

350

## 351 **Conclusion**

352 This work demonstrates that sugar particle size manipulation has a significant impact on the  
353 physical and sensory properties of chocolate brownies. Chocolate brownies formulated with  
354 LPR<sub>924-1877 μm</sub> received the highest scores for liking of texture, appearance and colour. Thus,  
355 replacement of the parent sugar with this experimental fraction improved acceptance of the  
356 final product. Therefore, sugar within this size range could be used to improve the texture and  
357 appearance of low-sugar or partially-replaced sugar in confectionery-type products.

358 Chocolate brownies prepared with the smallest sugar particle size (SPR<sub>459-972 μm</sub>) were the  
359 softest and moistest of all samples as supported by sensory, instrumental and compositional  
360 analysis. This is an important finding as sugar within this size range could be employed to  
361 retain moisture and softness in low sugar/low fat confectionery type products. Chocolate  
362 brownies formulated with the smallest sugar particles were perceived as the sweetest  
363 samples. Based on these findings sugar particle size reduction would permit sugar reduction  
364 as sweetness perception is increased in samples with smaller sugar particles. Further research  
365 needs to be carried out to demonstrate this finding further. In conclusion, sugar particle size  
366 reduction increases the sensory perception of sweetness in chocolate brownies and could be  
367 used as a viable technological approach to effectively reduce the sugar content of  
368 confectionery-type products and be of benefit to the baking industry in the formulation of  
369 low-calorie, clean-label baked goods.

**370 Funding**

371 The funding for this research was provided by the Food Institutional Research  
372 Measure (FIRM), (Grant number 14F 812), administered by the Department of Agriculture,  
373 Food and the Marine.

374

**375 Conflicts of interest**

376 The authors declare that no conflicts of interests exist.

377

**378 References**

- 379 Azad, M.B., Sharma, AK., de Souza, RJ., Dolinsky, VW., Becker, AB.,  
380 Mandhane, PJ., Turvey, SE., Subbarao, P., Lefebvre, DL., Sears, MR.,  
381 2016. Association between artificially sweetened beverage consumption  
382 during pregnancy and Infant body mass index. *JAMA Pediatrics*, 170(7),  
383 662–670.
- 384 Bennion, E.B. and Bamford, G.S., 2013. Sugars. In A.J Bent (Eds), *The*  
385 *technology of cake making*. Aylesbury: L. Hill, 84–99.
- 386 Bostian, M. L., Fish, D. L., Webb, N. B., & Arey, J.J., 1985. Automated  
387 methods for determination of fat and moisture in meat and poultry products:  
388 Collaborative study. *Journal of the Association of Official Analytical*  
389 *Chemists*, 68(5), 876–880.

- 390 Chan, J.M., Rimm, E.B., Colditz, G.A., Stampfer, M.J., Willett, W.C., 1994.  
391 Obesity, fat distribution, and weight gain as risk factors for clinical diabetes  
392 in men. *Diabetes Care*, 17(9), 961–969.
- 393 Dozan, T., Benković, M. & Bauman, I., 2014 Sucrose particle size reduction -  
394 Determination of critical particle diameters causing flowability difficulties.  
395 *Journal of Hygienic Engineering and Design*, 3–10.
- 396 Fellendorf, S., O' Sullivan, M.G., Kerry, J.P., 2016. Effect of using replacers on  
397 the physiochemical properties and sensory quality of low salt and low fat  
398 white puddings. *European Food Research and Technology*, 242, 2105-  
399 2118.
- 400 Gardner, C., Wylie-Rosett, J., Gidding, S.S, Steffen, L.M., Johnson, R.k.,  
401 Reader, D., Lichtenstein, A.H. (2012). Nonnutritive sweeteners: Current use  
402 and health perspectives: A scientific statement from the American heart  
403 association and the American diabetes association. *Circulation*, 126(4),  
404 509–519.
- 405 Healthy Ireland, (2015). *Healthy Ireland Survey 2015: Summary of Findings*,
- 406 Hu, F.B. & Malik, V.S., 2010. Sugar-sweetened beverages and risk of obesity  
407 and type 2 diabetes: Epidemiologic evidence. *Physiology and Behavior*,  
408 100(1), 47–54.
- 409 Irish Universities Nutrition Alliance (IUNA), 2011. National Adult Nutrition

- 410 Survey (NANS) Summary Report. , (March 2011), 37.  
411 [http://www.iuna.net/wp-content/uploads/2010/12/National-Adult-Nutrition-](http://www.iuna.net/wp-content/uploads/2010/12/National-Adult-Nutrition-Survey-Summary-Report-March-2011.pdf)  
412 [Survey-Summary-Report-March-2011.pdf](http://www.iuna.net/wp-content/uploads/2010/12/National-Adult-Nutrition-Survey-Summary-Report-March-2011.pdf) /Accessed April 5, 2017.
- 413 ISO, 2014. ISO 11136: 2014 <https://www.iso.org/ics/67.240/x/> /Accessed July  
414 18, 2017.
- 415 Kissel, L., Marshall, BD & Yamazaki, W., 1973. Effect of Variability in Sugar  
416 Granulation on the Evaluation of Flour Cookie Quality. *American*  
417 *association of cereals chemists*, 50, 10.
- 418 MacGregor, G.A. & Hashem, K.M., 2014. Action on sugar-lessons from UK  
419 salt reduction programme. *The Lancet*, 383(9921), 929–931.
- 420 Manley, D., 2011. Sugars and syrups as biscuit ingredients. In D. Manley,  
421 (Eds). *Manleys's Technology of Biscuits, crackers and cookies* 143–159.
- 422 Markets and markets., 2015. Sugar substitutes market  
423 [http://www.marketsandmarkets.com/Market-Reports/sugar-substitute-](http://www.marketsandmarkets.com/Market-Reports/sugar-substitute-market-1134.html)  
424 [market-1134.html](http://www.marketsandmarkets.com/Market-Reports/sugar-substitute-market-1134.html) /Accessed July 20, 2017.
- 425 Martens, H., Martens. M., 2000. Modified Jack-knife estimation of parameter  
426 uncertainty in bilinear modelling by partial least squares regression (PLSR),  
427 *Food Quality and Preference* 11 (1) 5–16.
- 428 Martínez-Cervera, S., Sanz, T., Salvador, A., Fiszman, S.M., 2012. Rheological,  
429 textural and sensorial properties of low-sucrose muffins reformulated with

- 430 sucralose/polydextrose. *LWT - Food Science and Technology*, 45(2), 213–  
431 220.
- 432 Te Morenga, L., Mallard, S., Mann, J., Morenga, L.T., 2013. Dietary sugars and  
433 body weight: systematic review and meta-analyses of randomised  
434 controlled trials and cohort studies. *BMJ (Clinical research ed.)*, 346.
- 435 Morgan, K., McGee H., Watson D., Perry I., Barry M., Shelley E., Harrington  
436 J., Molcho M., Layte R., Tully N., van Lente E., Ward M., Lutomski J.,  
437 Conroy R., Brugha R., 2008. SLAN 2007: Survey of lifestyle, attitudes &  
438 nutrition in Ireland: main report,  
439 <http://epubs.rcsi.ie/cgi/viewcontent.cgi?article=1002&context=psycholrep/>  
440 Accessed 14 February 2016.
- 441 Rama, R., Chiu, N., Carvalho Da Silva, M., Hewson, L., Hort, J., Fisk, I.D.,  
442 2013. Impact of salt crystal size on in-mouth delivery of sodium and  
443 saltiness perception from snack foods. *Journal of Texture Studies*, 44(5),  
444 338–345.
- 445 Richter, V., Almeida, T., Prudencio, S., Benassi, M., 2010. Proposing a ranking  
446 descriptive method. *Food quality and preference*, 21 (6), 611-620.
- 447 Rosner, B.A., Speizer, F.E. & Manson, J.E., 1997. Body Fat Distribution and  
448 Risk of Non-Insulin-dependent Diabetes Mellitus in Women. *American*  
449 *Journal of Epidemiology*, 145(7), 614–619.

- 450 Shepard, IS & Yoell, R., 1976. Cake emulsions. In *Food emulsions*. Marcel  
451 Dekker, New York, 216–275.
- 452 Stone, H., Bleibaum, R.N., Thomas, H.A., 2012a. Affective testing. In: Stone,  
453 H., Bleibaum, R.N., Thomas, H.A (Eds.), *Sensory Evaluation Practices*,  
454 fourth ed. Elsevier Academic Press, USA, 291-325.
- 455 Stone, H., Bleibaum, R., Thomas, H.A., 2012b. Test strategy and design of  
456 experiments. In: Stone, H., Bleibaum, R.N., Thomas, H.A. (Eds.), *Sensory  
457 Evaluation Practices*, fourth ed. Elsevier Academic Press, USA, 117-157.
- 458 Struck, S., Gundel, L., Zahn, S., Rohm, H., 2016. Fiber enriched reduced sugar  
459 muffins made from iso-viscous batters. *LWT - Food Science and  
460 Technology*, 65,32–38 <http://dx.doi.org/10.1016/j.lwt.2015.07.053>.
- 461 Suez, J, Korem, T., Zeevi, D., Ziberman-Schapira, G., Thaïss, C.A., Mazza, O.,  
462 Israeli, D., Zmora, N., Gilad, S., Weinberger, A., Kuperman, Y., Harmelin,  
463 A., Kolodkin-Gal, I., Shapiro, H., Halpern, Z., Segal, E., Elinav, E., 2014.  
464 Artificial sweeteners induce glucose intolerance by altering the gut  
465 microbiota. *Nature*, 514(7521), 181–186.
- 466 WHO, 2015. WHO Guideline: “Sugars intake for adults and children.” *Agro  
467 Food Industry Hi-Tech*, 26(4), 34–36.



**Table 1**

Hedonic and sensory descriptors used in sensory evaluation questionnaires

Descriptors	Scale	Definition
<b>Hedonic</b>		
Appearance	0 = extremely dislike 10 = extremely like	
Flavour	0 = extremely dislike 10 = extremely like	
Texture	0 = extremely dislike 10 = extremely like	
Colour	0 = extremely dislike 10 = extremely like	
Overall acceptability	0 = extremely unacceptable 10 = extremely acceptable	
<b>Intensity</b>		
<i>Appearance</i>		
Crust darkness	0 = none 10 = extreme	Degree of darkness of crust
<i>Texture</i>		
Hardness	0 = none 10 = extreme	Force needed to compress the sample
Moisture	0 = none 10 = extreme	Moist/wet texture in mouth
<i>Flavour</i>		
Sweet taste	0 = none 10 = extreme	Flavour sensation associated with sucrose
Brownie flavour	0 = none 10 = extreme	Characteristic chocolate brownie flavour
Off flavour	0 = none 10 = extreme	Off-flavour (rancid)
Aftertaste	0 = none 10 = extreme	A taste remaining in the mouth after eating

**Table 2**Particle size ranges of parent sugar and sugar sieve separates ( $\mu\text{m}$ )

Sugar-sieve separates ( $\mu\text{m}$ )	Sample	Particle size ranges ( $\mu\text{m}$ )	Average particle size ( $\mu\text{m}$ )
Parent sugar	Control (C)	200-5181	1533
710	LPR	924-1877	1276
500	MPR	627-1214	930
355	SPR	459-972	652
212	10% of MIX	330-700	479

MIX sample was made up of 50% SPR, 40% MPR and 10% of the finer particle size captured by the 212 $\mu\text{m}$  sieve mesh size

**Table 3**

Significance of estimated regression coefficients (ANOVA values) for the relationship of sensory terms (hedonic & intensity) and Chocolate Brownies prepared with varying sugar particle sizes

Sample	Appearance Liking	Flavour Liking	Texture Liking	Colour Liking	Overall acceptability	Crust darkness	Hardness	Moisture	Sweet taste	Brownie Flavour	Off flavour	Aftertaste
Control <sub>200-5181 μm</sub>	0.289	0.478	0.366	0.669	0.463	-0.003**	0.021*	0.061	0.519	0.695	0.574	0.886
LPR <sub>924-1877 μm</sub>	0.041*	0.186	0.012**	0.050*	0.359	0.809	0.134	0.507	0.433	0.431	0.897	0.530
MPR <sub>627-1214 μm</sub>	0.607	0.102	0.439	0.939	0.258	0.104	0.662	0.602	0.432	0.192	0.422	0.449
SPR <sub>459-972 μm</sub>	0.187	0.657	0.134	-0.012**	0.877	0.413	-0.002**	0.001***	0.045*	0.850	0.370	0.502
MIX	0.521	0.996	0.720	0.439	0.655	0.595	0.980	0.544	0.105	0.272	0.805	0.413

Significance of regression coefficients\*= $P \leq 0.05$ , \*\*= $P \leq 0.01$ , \*\*\*= $P \leq 0.001$ . – indicates whether the correlation is negatively correlated. LPR; large particle replacement MPR; medium particle replacement and SPR; small particle replacement

**Table 4**

Texture profile analysis (TPA) values for Chocolate brownies made with decreasing sugar particle size.

Sample	TPA					
	Hardness (N)	Adhesiveness	Springiness (mm)	Cohesiveness (n/a)	Chewiness (N-mm)	Resilience (n/a)
Control <sub>200-5181μm</sub>	54.5 ± 1.45 <sup>b</sup>	-0.0 ± 0.73 <sup>a</sup>	0.5 ± 0.74 <sup>a</sup>	0.3 ± 0.05 <sup>a</sup>	9.1 ± 0.80 <sup>d</sup>	0.1 ± 0.01 <sup>a</sup>
LPR <sub>924-1877μm</sub>	69.2 ± 2.12 <sup>c</sup>	-0.0 ± 0.59 <sup>a</sup>	0.5 ± 0.06 <sup>a</sup>	0.3 ± 0.04 <sup>a</sup>	9.8 ± 0.12 <sup>d</sup>	0.1 ± 0.01 <sup>a</sup>
MPR <sub>627-1214μm</sub>	52.0 ± 2.75 <sup>b</sup>	-0.0 ± 0.01 <sup>a</sup>	0.3 ± 0.04 <sup>a</sup>	0.3 ± 0.03 <sup>a</sup>	5.0 ± 0.50 <sup>b</sup>	0.1 ± 0.01 <sup>a</sup>
SPR <sub>459-972μm</sub>	45.1 ± 2.42 <sup>a</sup>	-1.0 ± 1.17 <sup>a</sup>	0.3 ± 0.07 <sup>a</sup>	0.3 ± 0.05 <sup>a</sup>	4.2 ± 0.23 <sup>a</sup>	0.1 ± 0.02 <sup>a</sup>
MIX	53.4 ± 1.72 <sup>b</sup>	-0.0 ± 0.81 <sup>a</sup>	0.3 ± 0.04 <sup>a</sup>	0.3 ± 0.05 <sup>a</sup>	6.2 ± 0.13 <sup>c</sup>	0.1 ± 0.02 <sup>a</sup>

<sup>abc</sup> mean values (± standard deviation) in the same row bearing different superscripts are significantly different,  $P < 0.05$ .

**Table 5**

Colour lightness values (L\*) for Chocolate brownies made with decreasing sugar particle size

Sample	Colour (L*)	
	Crust	Crumb
Control <sub>200-5181μm</sub>	39.8 ± 1.85 <sup>a</sup>	29.0 ± 1.51 <sup>a</sup>
LPR <sub>924-1877μm</sub>	34.2 ± 0.93 <sup>b</sup>	27.1 ± 1.82 <sup>ab</sup>
MPR <sub>627-1214μm</sub>	33.2 ± 1.22 <sup>b</sup>	25.1 ± 0.80 <sup>b</sup>
SPR <sub>459-972μm</sub>	33.1 ± 1.95 <sup>b</sup>	24.4 ± 1.81 <sup>b</sup>
MIX	32.6 ± 0.80 <sup>b</sup>	25.4 ± 1.73 <sup>b</sup>

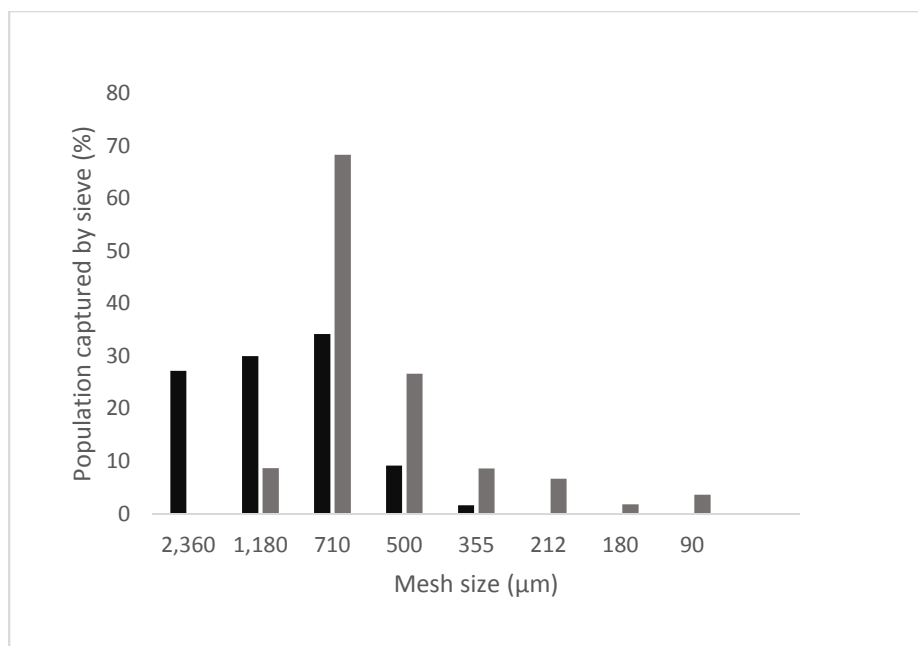
<sup>abc</sup> mean values (± standard deviation) in the same row bearing different superscripts are significantly different,  $P < 0.05$ .

**Table 6**

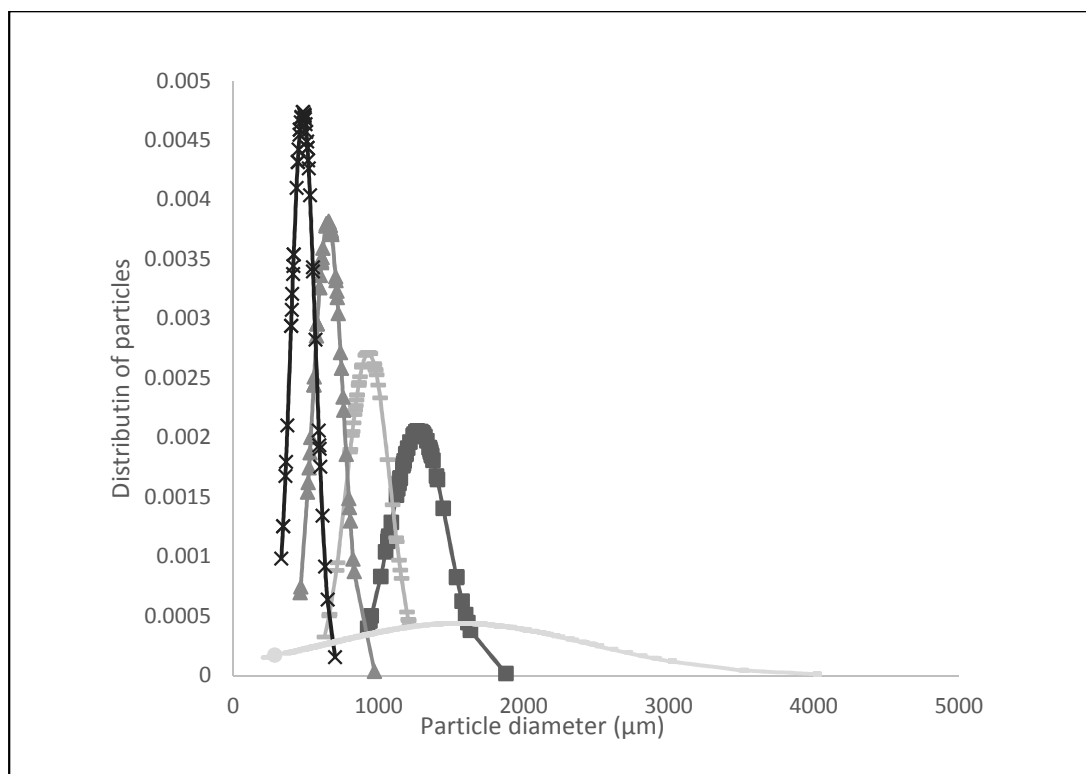
Moisture and fat of chocolate brownie samples prepared with decreasing particle size

Sample	% Moisture	%Fat
Control <sub>200-5181<math>\mu</math>m</sub>	9.2 $\pm$ 0.80 <sup>a</sup>	26.3 $\pm$ 1.87 <sup>a</sup>
LPR <sub>924-1877<math>\mu</math>m</sub>	9.9 $\pm$ 1.08 <sup>a</sup>	27.6 $\pm$ 1.68 <sup>a</sup>
MPR <sub>627-1214<math>\mu</math>m</sub>	11.8 $\pm$ 1.03 <sup>bc</sup>	26.6 $\pm$ 1.95 <sup>a</sup>
SPR <sub>459-972<math>\mu</math>m</sub>	13.0 $\pm$ 0.84 <sup>c</sup>	26.9 $\pm$ 1.28 <sup>a</sup>
MIX	10.3 $\pm$ 0.70 <sup>ab</sup>	26.2 $\pm$ 0.77 <sup>a</sup>

<sup>abc</sup> mean values ( $\pm$  standard deviation) in the same row bearing different superscripts are significantly different,  $P < 0.05$

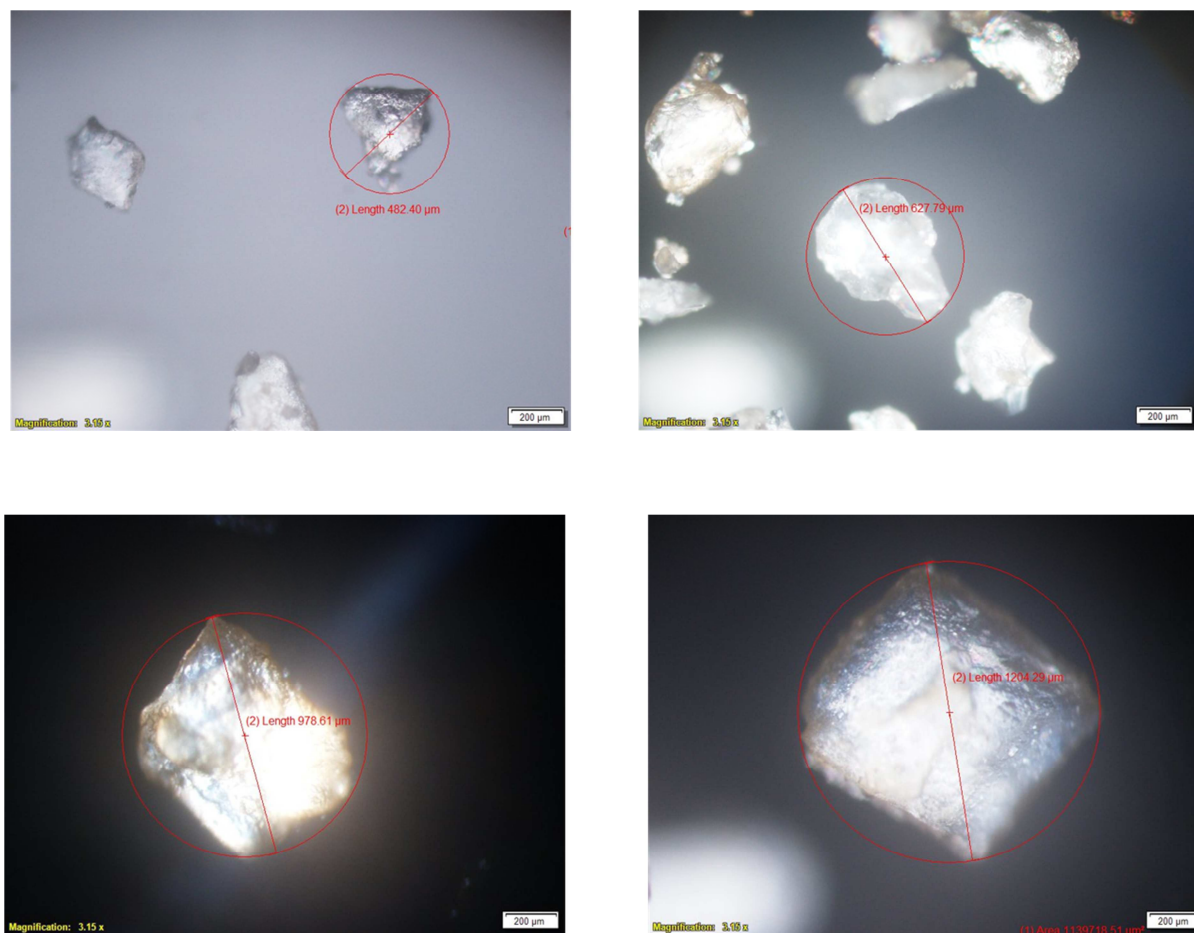


**Fig 1.** Particle size distribution of dried brown sugar (200g, 0.5% moisture) before (■) and after (■) grinding.



**Fig 2.** Particle diameter distribution of parent brown sugar before grinding: control (—○—) and sugar-sieve separates; 710 (—■—), 500 (—□—), 355 (—▲—) and 212 μm (—×—) after grinding.





**Fig 3.** Microscopic images of brown sugar particles captured by different sieve apertures after grinding. From top left, 212μm and 355μm apertures and from bottom left 500μm and 710μm apertures. Red line across particle indicates diameter of the particle.



**Fig 4.** Cross section images of chocolate brownies (2 x 2 x 2 cm). Samples were taken from the upper right midsection of each batch tray. From left: Control, LPR, MPR, SPR and MIX sample.

**Highlights**

- Small sugar particles increase sweetness intensity perception in chocolate brownies.
- Small sugar particles enhance the soft and moist texture of chocolate brownies.
- Sugar particle size manipulation significantly affects appearance, texture and colour liking of chocolate brownies.