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1 **Visible, Near- and Mid- infrared spectroscopy coupled with an innovative**
2 **chemometric strategy to control apple puree quality**

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28 **Highlights**

29 Vis-NIRS and MIRS coupled with PLS can detect the cultivar composition of mixed
30 purees.

31 MIRS evaluated with a high confidence the quality characteristics of formulated
32 purees by PLS.

33 Spectra of individual puree cultivars can be used to control the quality of formulated
34 apple purees.

35 MIRS coupled with MCR-ALS can reconstruct the puree mixture using the
36 concentration profiles.

37

38

39 **Abstract**

40 Vis-NIRS, MIRS, and a combination of both coupled with PLS and machine learning
41 were applied to i) trace the composed proportions of different apple varieties in
42 formulated purees and ii) predict the quality characteristics of formulated purees from
43 spectral information of initial puree cultivars. The PLS models could estimate
44 proportions of each apple cultivar in puree mixtures using MIR spectra
45 (RMSEP<8.1%, RPD> 3.6), especially for Granny Smith (RMSEP=2.7%, RPD=11.4).
46 The concentration profiles from multivariate curve resolution-alternative least squares
47 (MCR-ALS) made possible to reconstruct spectra of formulated purees. MIRS
48 technique was evidenced to predict the final puree quality, such as viscosity
49 (RPD>4.0), contents of soluble solids (RPD=4.1), malic acid (RPD=4.7) and glucose
50 (RPD=4.3), based only on the spectral data of composed puree cultivars. Infrared
51 technique should be a powerful tool for puree traceability, even for multicriteria
52 optimization of final products from the characteristics of composed puree cultivars
53 before formulation.

54

55 **Key word:** *Malus domestica* Borkh., Vis-NIR, MIR, machine learning, MCR-ALS

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59 **1. Introduction**

60 Apple puree is an ideal source of healthy constituents such as polyphenols and fibers
61 (Le Bourvellec et al., 2011) and ~~antioxidants such as polyphenols with their major~~
62 ~~polymeric form, procyanidins (Loncaric, Dugalic, Mihaljevic, Jakobek, & Pilizota,~~
63 ~~2014; Oszmiański, Wolniak, Wojdyło, & Wawer, 2008; Rembiałkowska, Hallmann, &~~
64 ~~Rusaczonek, 2007).~~ It can be used as an intermediate for smoothies, fruit sauce, pie
65 fillings and fruit-based baby food (Opatová, Voldřich, Dobiáš, & Čurda, 1992). The
66 ~~industrial production of apple purees consists typically in cooking at 93–98°C for~~
67 ~~about 4–5 min, refining to remove seeds and skin pieces and then pasteurization at~~
68 ~~90°C around 20 min to obtain a shelf life of 6 months at room temperature~~
69 ~~(Oszmiański, Wolniak, Wojdyło, & Wawer, 2008).~~ Puree quality characteristics vary
70 with fruit genetics (Rembiałkowska, Hallmann, & Rusaczonek, 2007), storage
71 (Loncaric, Dugalic, Mihaljevic, Jakobek, & Pilizota, 2014), cooking parameters
72 (Picouet, Landl, Abadias, Castellari, & Viñas, 2009), grinding intensity (Espinosa et
73 al., 2011) and refining (Lan, Jaillais, Leca, Renard, & Bureau, 2020). In order to reach
74 an apple puree with anticipated and constant taste and texture, a mixture of
75 proportions of different apple varieties is generally done, presenting also the most
76 economic and efficient strategies for manufacturers (O'sullivan, 2016). ~~Most papers~~
77 ~~dealing with the apple processing have not considered this practice insofar as they~~
78 ~~have been focused only on one apple cultivar (Espinosa et al., 2011; Picouet, Landl,~~
79 ~~Abadias, Castellari, & Viñas, 2009; Keenan, Brunton, Butler, Wouters, & Gormley,~~

2011).—Thus, developing rapid and reliable approaches to determine the puree
formulation, including fruit cultivars and the proportions of each one, could be highly
beneficial for fruit processed products and traceability control.

Infrared spectroscopy (visible-near and mid infrared) known as a rapid, relatively
cheap, easy-to-use, non-destructive and automatable technique, has been applied for
the quality analysis of apple based products, such as juices (Kelly & Downey, 2005;
León, Kelly, & Downey, 2005; Reid, Woodecock, O'Donnell, Kelly, & Downey, 2005)
and wine (Peng, Ge, Cui, & Zhao, 2016). For fruit purees, the studies have mainly
aimed at detecting adulterations in mixed purees of different fruit species (Contal,
León, & Downey, 2002; Defernez, Kemsley, & Wilson, 1995; Kemsley, Holland,
Defernez, & Wilson, 1996). Particularly, the MIR technique combined with partial
least squares discrimination analysis (PLS-DA) detects the presence of apple starting
at 20% in apple-raspberry mixed purees (Kemsley, Holland, Defernez, & Wilson,
1996). Similar detectable limits are obtained using Vis-NIRS coupled with a principal
component analysis (PCA) and a linear discriminant analysis (LDA) in
apple-strawberry mixed purees (Contal, León, & Downey, 2002). The infrared
spectroscopy (Vis-NIR and MIR) appears as a potential tool to access the composition
of purees prepared with several fruit species. However, so far, there has been no
attempt to use such approaches for more advanced works on purees of apples only,
but resulting from various proportions of different cultivars.

Further, for fruit processors, the ever-increasing variability of raw fruits may mean
that their empirical knowhow may not be sufficient to produce expected and constant

102 final purees. The challenge is therefore to provide specific guidance for formulation of
103 final purees based on information of individual batches of single cultivar puree.
104 Multivariate curve resolution-alternative least square (MCR-ALS) has been widely
105 used to simultaneously elucidate the pure spectra of different species present in
106 processed products and their concentration profiles (de Juan & Tauler, 2006), such as
107 edible oils from different vegetable sources (Le Dréau, Dupuy, Artaud, Ollivier, &
108 Kister, 2009) and fruit juices with various organic acids (Silva, Lourenço, & de
109 Araujo, 2018). The interest of this approach is to reconstruct the spectra of final
110 processed products (in our case, formulated purees) according to the relative spectra
111 of individual components (single cultivar purees) by MCR-ALS. If so, the predictive
112 models of processed puree quality traits (physical and chemical) using the
113 reconstructed spectra dataset could open the possibility to provide a multicriteria
114 optimization of puree formulation based on the prior information of single cultivar
115 purees.

116 Partial least squares (PLS), a typical linear algorithm, has been used to successfully
117 determine the global quality parameters of apple purees using NIRS information, such
118 as titratable acidity, dry matter and soluble solids (Lan, Jaillais, Leca, Renard, &
119 Bureau, 2020). However, the overlapping of absorption bands linked to non-linear
120 rheological variations gave poor prediction of puree's texture by PLS regression.
121 Machine learning approaches, such as random forest (RF) and Cubist, have been
122 specially constructed to address large and complex nonlinear systems. Indeed, RF
123 algorithm allows a better detection of adulteration in formulated oils than PLS (de

124 Santana, Borges Neto, & Poppi, 2019). Cubist regression working as decision tree
125 models, gives a higher prediction accuracy than RF and PLS regression in palm-based
126 cooking oil (Goh et al., 2019).

127 Accordingly, Vis-NIRS, MIRS and the combination of both (CB) infrared spectra
128 coupled with machine learning (RF and Cubist) and PLS regressions were applied in
129 our work on apples to: i) assess the possibility to detect the proportions of specific
130 cultivar purees in the formulated purees and evaluate the limits of the detection; ii)
131 build models to evaluate the quality parameters of formulated purees obtained from
132 different proportions of single cultivar purees; and then iii) use information of single
133 cultivar purees to reconstruct spectra of formulated purees by MCR-ALS and
134 investigate the possibility to develop regression models to guidance the quality of
135 final purees.

136 **2. Material and methods**

137 **2.1 Apple purees**

138 **2.1.1 Purees processing**

139 Apples of four varieties: ‘Golden Delicious’(GD), ‘Granny Smith’(GS),
140 ‘Braeburn’(BR) and ‘Royal Gala’(GA) (all abbreviations are shown in **Table 1**) were
141 harvested at a commercial maturity from La Pugère experimental orchard (Mallemort,
142 Bouches du Rhône, France) in 2019, and stored for up to 2 months at 4 °C and around
143 90% relative humidity to ensure starch regression. After sorting and washing, on three
144 consecutive weeks, a batch of each apple cultivar (2 kg) was processed into purees in
145 a multi-functional processing system (Roboqbo, Qb8-3, Bentivoglio, Italy) following

146 a Hot Break recipe: cooked at 95°C for 5 min at a 1500 rpm grinding speed, then
147 cooled down to 65°C while maintaining the grinding speed. Finally, processed purees
148 were conditioned in two hermetically sealed cans: one was cooled in a cold room
149 (4°C) before formulation, while the other was stored at -20°C for biochemical
150 measurement of individual sugars (fructose, sucrose and glucose) and malic acid.

151 **2.1.2 Puree formulations**

152 After processing the single-cultivar purees, a total of 6 experimental groups (named A
153 to E) were prepared, each, with two apple cultivars (**Figure 1**). Each group (A-F)
154 included 9 samples with different formulated proportions of weight, was divided into
155 two subsets: the first including 6 proportions (10%-90%, 25%-75%, 50%-50%,
156 75%-25%, 90%-10%, 95%-5%) for the modeling set, while the second with 3
157 proportions (80%-20%, 33%-67%, 14%-86%) for the external prediction set. Finally,
158 spectral measurements (Vis-NIR and MIR), chemical (soluble solids, titratable acidity,
159 pH, dry matter) and physical (color and rheological tests) characterizations were
160 performed on each sample (single and formulated purees).

161 **2.2 Determination of quality traits**

162 **2.2.1 Physical characterizations**

163 The puree color was determined three times through a dedicated glass cuvette using a
164 CR-400 chromameter (Minolta, Osaka, Japan) and expressed in the CIE 1976 L*a*b*
165 color space (illuminant D65, 0° view angle, illumination area diameter 8 mm). The
166 puree rheological measurements, as flow curves, were carried out using a Physica
167 MCR-301 controlled stress rheometer (Anton Paar, Graz, Austria) and a 6-vane

168 geometry (FL100/6W) with a gap of 3.46 mm, at 22.5°C. The flow curves were
169 performed after a pre-shearing period of 1 minute at a shear rate of 50 s⁻¹, followed by
170 5 minutes at rest. The viscosity was then measured at a controlled shear rate range of
171 [10; 250] s⁻¹ on a logarithmic ramp. The values of viscosity at 50 s⁻¹ and 100 s⁻¹ (η_{50}
172 and η_{100} respectively) were kept as final indicators of the puree texture linked to
173 sensory characteristics during consumption (Chen & Engelen, 2012).

174 **2.2.2 Biochemical analyses**

175 Soluble solids content (SSC) was determined with a digital refractometer (PR-101
176 ATAGO, Norfolk, VA, USA) and expressed in °Brix at 20°C. Titratable acidity (TA)
177 was determined by titration up to pH 8.1 with 0.1 mol/L NaOH and expressed in
178 mmol H⁺ kg⁻¹ of fresh weight (FW) using an autotitrator (Methrom, Herisau,
179 Switzerland). Individual sugars and malic acid were quantified using colorimetric
180 enzymatic kits (glucose: No. 10716251035; fructose: No. 10139106035, sucrose: No.
181 10716260035, malic acid: No. 10139068035) according to the manufacturer's
182 instructions (R-biopharm, Darmstadt, Germany), respectively. The content of glucose,
183 fructose, sucrose and malic acid were expressed in g kg⁻¹ FW. These measurements
184 were performed with a SAFAS flx-Xenius XM spectrofluorimeter (SAFAS, Monaco)
185 at 570 nm for the sugars and 450 nm for malic acid. The dry matter content (DMC)
186 was estimated from the weight of freeze-dried samples upon reaching a constant
187 weight (freeze-drier, 5 days). The individual sugars (fructose, glucose, sucrose) and
188 malic acid contents of formulated puree samples were calculated based on the
189 measured values of processed single cultivar purees.

190 **2.3 Spectrum acquisition**

191 The Vis-NIR spectral data of purees was acquired with a multi-purpose analyzer
192 spectrometer (Bruker Optics®, Wissembourg, France) at 23°C, which provides
193 diffuse reflectance measurements with a spectral resolution of 8 cm⁻¹ from 12500 to
194 4000 cm⁻¹ (wavelength from 400 to 2500 nm). For each spectrum, 32 scans were
195 recorded and averaged. The spectral acquisition and instrument adjustments were
196 controlled by OPUS software Version 5.0 (Bruker Optics®). Puree were transferred
197 into 10 mL glass vials (5 cm height x 18 mm diameter) which were placed on the
198 automated sample wheel of the spectrophotometer. Each puree sample was measured
199 three times on different aliquots. A reference background measurement was
200 automatically activated before each data set acquisition using an internal Spectralon
201 reference.

202 The MIR spectra of purees was collected at 23°C using a Tensor 27 FTIR
203 spectrometer (Bruker Optics®, Wissembourg, France) equipped with a horizontal
204 attenuated total reflectance (ATR) sampling accessory and a deuterated triglycine
205 sulphate (DTGS) detector. Three replications of spectral measurement were
206 performed on different aliquots. The purees were placed at the surface of a zinc
207 selenide (ATR-ZnSe) crystal with six internal reflections. Spectra with 32 scans for
208 ATR-ZnSe were collected from 4000 cm⁻¹ to 650 cm⁻¹ with a 4 cm⁻¹ resolution and
209 were corrected against the background spectrum of air.

210 The whole spectral dataset of Vis-NIR or MIR included 36 spectra (3 replicates × 3
211 processing weeks × 4 varieties) of single-cultivar purees, 324 spectra of formulated

212 purees spectra for the modelling set (3 replicates \times 3 processing weeks \times 6 formulated
213 puree groups \times 6 proportions) and 162 spectra for the external prediction set (3
214 replicates \times 3 processing weeks \times 6 formulated puree groups \times 3 proportions)
215 described in 2.2.1 and **Figure 1**.

216 **2.4 Statistical analyses of reference data**

217 After ensuring normal distribution with a Shapiro-Wilk test ($\alpha=0.05$), the reference
218 data of processed purees were presented as mean values and the data dispersion
219 within our experimental dataset expressed as standard deviation values (SD). Analysis
220 of variance (ANOVA) was carried out to determine the significant differences due to
221 the different single apple varieties (**Table S-1**) or formulated puree groups (**Table S-2**)
222 using XLSTAT (version 2018.5.52037, Addinsoft SARL, Paris, France) data analysis
223 toolbox. And the pairwise comparison between means was performed using Tukey's
224 test. Principal component analysis (PCA) was carried out on all reference data of
225 single-cultivar purees or of formulated purees to evaluate their discriminant
226 contributions using Matlab 7.5 (Mathworks Inc. Natick, MA, USA) software.

227 **2.5 MCR-ALS and spectra reconstruction**

228 MCR-ALS (multivariate curve resolution-alternative least square) is an effective
229 multivariate self-modelling curve resolution method developed by Tauler (de Juan &
230 Tauler, 2006). The relative contributions given by MCR-ALS were obtained for both,
231 the Vis-NIR (400-2500 nm) and MIR (900-1800 cm^{-1}) spectral information, using the
232 formulated purees and their corresponding single-cultivar purees (**Figure 2**). For the
233 formulated samples, one matrix D ($n \times \lambda$) was made up with the number of samples (n)

234 and the intensity at each wavenumbers or wavelengths (λ). The S^T matrix ($s \times \lambda$) is the
 235 spectroscopic matrix describing the ‘pure’ infrared spectra (λ) of all single-cultivar
 236 purees (s). The D matrix can be mathematically decomposed into the individual
 237 contributions related to the spectral information of ‘pure’ purees in matrix S^T
 238 according to Eq. (1) and is interactively transformed using an alternative least square
 239 (ALS) procedure as Eq (2).

$$240 \quad \mathbf{D} = \mathbf{C}\mathbf{S}^T + \mathbf{E} \quad (1)$$

$$241 \quad \mathbf{C} = \mathbf{R}(\mathbf{S}^T)^+ \quad (2)$$

242 Matrix C ($n \times q$) is the concentration matrix describing the contribution of every
 243 single-cultivar purees (q) in reconstructed purees (n). E is the error matrix that
 244 provides the data variation not explained by their contributions. The matrix $(S^T)^+$ is the
 245 pseudo-inverse matrix of S^T . A general constraint used in curve resolution method is
 246 the non-negativity on the concentration profiles.

247 Once the concentration profiles (matrix C) for each single-cultivar spectrum,
 248 including Golden Delicious (C_{GD}), Granny Smith (C_{GS}), Braeburn (C_{BR}) and Royal
 249 Gala (C_{GA}), were obtained, they were used to reconstruct a new spectroscopic matrix
 250 R ($n \times k$) for monitoring all formulated purees. Each row R_i . ($i=1, \dots, n$) was made up
 251 of a reconstructed spectrum. And each column R_j ($j=1, \dots, k$) gave the reconstructed
 252 spectral intensity at a wavenumber of MIRS or a wavelength of Vis-NIRS based on
 253 the corresponding pure puree spectra of Golden Delicious (λ_{GD}), Granny Smith (λ_{GS}),
 254 Braeburn (λ_{BR}) and Royal Gala (λ_{GA}), following Eq (3).

$$255 \quad \mathbf{R} = \mathbf{C}_{GD}\lambda_{GD} + \mathbf{C}_{GS}\lambda_{GS} + \mathbf{C}_{BR}\lambda_{BR} + \mathbf{C}_{GA}\lambda_{GA} \quad (3)$$

256 **2.6 Spectral multivariate regression**

257 Spectral pre-processing and multivariate regression were performed with several
258 packages ('prospectr' (Stevens & Ramirez-Lopez, 2013), 'pls' (Mevik, Wehrens, &
259 Liland, 2019), 'Cubist' (Kuhn, Weston, Keefer, Coulter, & Quinlan, 2014) and 'caret'
260 (Kuhn, 2015)) of the R software (version 2.6.2) (R Core Team, 2019). As
261 demonstrated in previous works (Bureau et al., 2013; Ncama, Opara, Tesfay, Fawole,
262 & Magwaza, 2017), the wavelengths from 400 to 2500 nm of Vis-NIR and the
263 wavenumbers from 900 to 1800 cm^{-1} in MIR were selected (**Figure 3**). For all
264 spectral datasets, standard normal variate (SNV), resampling (intervals= 5, 10, 15),
265 and derivative transform calculation (Savitzky–Golay method, gap size = 11, 21, 31,
266 41) of first or second order were compared before multivariate regression. SNV
267 pre-processing applied on the Vis-NIR and MIR data had the best performances to
268 predict puree quality and was then systematically used.

269 The partial least square (PLS), Cubist and RF regression models were developed to i)
270 detect the proportions of each apple varieties in puree samples (**Table 2**) and predict
271 the quality characteristics of formulated purees based on ii) the acquired Vis-NIR,
272 MIR and their combined infrared spectra (CB) (**Table 3**) or iii) the reconstructed
273 Vis-NIR, MIR and CB spectra (**Table 4**). All aforementioned spectral matrices
274 (Vis-NIRS, MIRS and CB) corresponded to the same reference dataset. The set of all
275 modelling spectra (324 spectra) was randomly split, with two-thirds of the dataset
276 (216 spectra) used for calibration and a third (108 spectra) for internal validation.
277 Then, calibrated models were further validated with the external prediction set (162

278 spectra). The procedure was repeated 10 times in order to obtain the dispersion of
279 values giving an idea of the model stability and robustness. The developed models
280 performance was then described by the 10-times averaged values of the determination
281 coefficients of internal validation (R_v^2) and external prediction (R_p^2), root mean
282 square error of prediction (RMSEP), RPD (Residual Predictive Deviation) value as
283 described by Nicolai (Nicolai et al., 2007). During model training, the variable
284 importance (VIP) for each puree characteristics were computed using the ‘varImp’
285 function by ‘caret’ package in R software (Kuhn, 2015), which could be applied both
286 on PLS and machine learning regressions (Parmley, Higgins, Ganapathysubramanian,
287 Sarkar, & Singh, 2019).

288 **3. Results and discussion**

289 **3.1 Characteristics of single-cultivar purees and formulated purees**

290 After puree processing, the four different cultivars provided a large variability of
291 appearance, in particular color and texture (**Figure S-1**). According to PCA results
292 taking into account their rheological and biochemical characteristics (**Figure S-2**),
293 ‘Royal Gala’ (GA) purees were clearly discriminated from the other purees along the
294 first principal component (PC1), with significantly ($p < 0.001$) lower TA, pH, glucose,
295 malic acid and viscosity (η_{50} and η_{100}) (**Table S-1**). Particularly, the values of
296 viscosity at a shear rate of 50 s^{-1} (η_{50}), which is commonly used to describe the
297 in-mouth texture perception of fluid foods (Chen & Engelen, 2012), were much more
298 lower in GA purees ($547 \pm 13 \text{ Pa}\cdot\text{s}^{-1}$) than in ‘Golden Delicious’ (GD) ($839 \pm 53 \text{ Pa}\cdot\text{s}^{-1}$)
299 and ‘Granny Smith’ (GS) ($904 \pm 31 \text{ Pa}\cdot\text{s}^{-1}$) purees (**Table S-1**). As expected, the

300 viscosity and global quality (SSC and TA) of the formulated purees were affected
301 when prepared with GA purees (**Figure S-3**). For example, the formulated GA-GD
302 (group C) or GA-GS purees (group E) provided a high range of viscosity (**Figure**
303 **S-3c and d**) and composition (**Figure S-3e and f**), but with a limited variation of
304 color (a^* and b^* values) (**Figure S-3a and b**).

305 Remarkable changes ($p < 0.001$) of color parameters (L^* , a^* and b^*) allowed the
306 separation of 'Braeburn' (BR) purees and the others along the second principal
307 component (**Figure S-2 and Table S-2**). Particularly the redness (a^* values) of
308 formulated puree groups (**Figure S-3a**), the admixture of BR (groups B, D and F)
309 introduced more intensive variations (from -4.33 to 2.35) than the others (groups A, C
310 and E, from -4.77 to -1.52). The limited variations of yellowness (b^* values) in
311 formulated GD-GA purees resulted in differences below the visual detection threshold
312 (**Figure S-3b**). Consequently, different strategies of puree formulation, especially the
313 mixtures with 'Royal Gala' or with 'Braeburn' purees, could provide variability in
314 taste, texture and color.

315 **3.2 Characteristics of formulated purees: determination of composed** 316 **single-cultivar puree proportions**

317 In this part, the ability of SNV pre-processed Vis-NIR, MIR and CB coupled with
318 PLS, Cubist and RF regressions was compared to estimate the proportions of
319 single-cultivar in all formulated purees (**Table 2**).

320 Both, Vis-NIR and MIR techniques were potentially able to estimate the proportions
321 of single-variety puree in the formulated purees with good models presenting robust

322 determination coefficients for both internal validation (R_v^2) and external validation
323 (R_v^2), acceptable RMSEP (<10%) and RPD values at least higher than 2.5 (Nicolai et
324 al., 2007). For Vis-NIR technique, two regression methods, PLS and RF, showed an
325 acceptable ability to estimate proportions of GS ($R_P^2 > 0.92$, RPD > 3.4, RMSEP <
326 9.2%) and of BR ($R_P^2 > 0.95$, RPD > 4.2, RMSEP < 7.9%) varieties in all formulated
327 purees, based on the VIP wavelengths at 412 nm, 524 nm and 672 nm (**Figure S-4b**
328 **and c**). The predictive errors obtained here for the mixture of two cultivars of the
329 same species, apple, were lower than those obtained earlier for the mixture of two
330 species, namely apple/raspberry (11.3%) (Contal, León, & Downey, 2002). The
331 poor Vis-NIRS prediction results for GD (RMSEP > 17.4%, RPD < 1.7) and GA
332 (RMSEP > 16.2%, RPD < 2.1) were probably due to their similar color (**Figure S-1**).
333 As the VIP wavelengths of Vis-NIR models were mainly dominated in the visible
334 spectral region (412-672 nm), the color variations were not enough to be used for
335 prediction of proportions in formulated purees, especially in the group C (GD-GA)
336 (**Figure S-3a & b**).

337 MIR provided a better prediction of the proportions of single-cultivar purees in the
338 formulated purees than Vis-NIR. Moreover, the regression method affected the
339 prediction results of MIR. PLS gave better prediction results (RMSEP < 8.1%, RPD >
340 3.6) than Cubist (RMSEP < 15.1%, RPD > 2.3) and RF (RMSEP < 10.6%, RPD > 2.7).
341 Particularly, MIRS combined with PLS reached the lowest determination error
342 (RMSEP = 2.7%, RPD = 11.4) for GS compared with other cultivars (GD, GA, BR).
343 The highest VIP values (**Figure S-5c**) at 1723 cm^{-1} , 1065 cm^{-1} and 1034 cm^{-1}

344 attributed respectively to malic acid, fructose and glucose (Bureau, Cozzolino, &
345 Clark, 2019; Clark, 2016), were consistent with the existence of marked differences in
346 chemical composition (SD and significance) between purees containing GS (**Table**
347 **S-2**). The excellent PLS predictions obtained for BR (RMSEP=4.3%, RPD=7.7) were
348 based on the VIP wavenumbers at 998 cm^{-1} and 1084 cm^{-1} related to sucrose and
349 fructose (Bureau, Cozzolino, & Clark, 2019) (**Figure S-5b**). Besides the
350 aforementioned spectral signal, the satisfactory assessments of GD and GA
351 proportions (RMSEP<8.1%, RPD>3.6) were linked to the MIRS region between 1750
352 and 1650 cm^{-1} related to organic acids, pectins, proteins, phenolics and absorbed water
353 (**Figure S-5a & d**) (Abidi, Cabrales, & Haigler, 2014; Canteri, Renard, Le Bourvellec,
354 & Bureau, 2019; Kačuráková et al., 1999).

355 The CB spectra, including Vis-NIR and MIR regions, coupled with PLS (RPD>2.8,
356 RMSEP<11.5%) and RF (RPD>3.0, RMSEP<9.5%) provided a satisfactory
357 assessment of the proportions of single-cultivar purees (**Table 2**). However, the results
358 on CB were not as good as for MIR only.

359 Consequently, to predict proportions of single-cultivar purees, Vis-NIR was suitable
360 for the formulated samples presenting large diversity in the color range, with the use
361 of Braeburn and Granny Smith apples for example, and under vacuum processing
362 conditions providing a good puree color preservation. MIRS coupled with PLS was
363 evidenced as a powerful tool to provide excellent estimations of puree proportions,
364 mainly based on differing concentrations of individual sugars and acid. Combining
365 Vis-NIR and MIR did not improve prediction.

366 **3.3 Characteristics of formulated purees: prediction of quality traits**

367 As previously, the different spectral areas, Vis-NIR, MIR or CB, of all formulated
368 purees coupled with the different regression methods, PLS, Cubist and RF, were
369 compared for their ability to predict color, rheological and biochemical characteristics
370 of formulated purees (**Table 3**). MIR spectra coupled with PLS obtained the best
371 predictions in comparison with Vis-NIR and CB, except for color. Indeed concerning
372 the color parameters, a good prediction of a^* values was obtained for all spectral areas
373 with a RPD decreasing order Vis-NIR (RPD>4.0), CB (RPD>3.6) and MIR (RPD>3.3)
374 for both PLS and machine learning regressions (Cubist and Random forest).
375 Particularly, the best prediction of a^* values was obtained on CB with PLS models
376 ($R_p^2=0.96$, RPD=5.0), slightly better than in Vis-NIR ($R_p^2=0.95$, RPD=4.7).
377 MIR spectra coupled with PLS gave the best prediction ($R_p^2>0.90$, RPD>4.1) of the
378 rheological parameters (η_{50} and η_{100}) (**Table 3**). The identified VIP wavenumbers
379 were 1026, 1065, 1113 and 1720 cm^{-1} (**Figure S-6**). These dominant carbohydrate
380 bands centered at 1000-1200 cm^{-1} , associated with C-OH and C-O-C vibration of
381 glucose and fructose (Bureau, Cozzolino, & Clark, 2019), have also been identified to
382 predict viscosity of tomato purees (Ayvaz et al., 2016). And an acceptable estimation
383 of DMC was observed for all developed MIR models (RMSEP< 0.003, RPD>2.7).
384 For biochemical parameters, MIR coupled with PLS allowed a very good prediction
385 of SSC (RMSEP=0.1, RPD=5.1) in accordance with previous results of apple and
386 tomato purees (Ayvaz et al., 2016; Lan, Renard, Jaillais, Leca, & Bureau, 2020). In
387 apples, SSC is strongly correlated to the presence of sugars, namely fructose, sucrose

388 and glucose. The two main sugars, fructose and sucrose, were satisfactorily predicted
389 with PLS (RPD>3.0) and the non-linear regressions, Cubist and RF (RPD>2.9).
390 However, MIR could not predict the glucose content (RPD<2.4) (**Table 3**).

391 Considering the different expressions of acidity such as pH, TA and malic acid content,
392 MIR coupled with PLS provided their excellent prediction with $R_p^2>0.92$ and
393 RPD>4.0. It can be noticed that Vis-NIRS gave also acceptable prediction of TA and
394 malic acid ($R_p^2>0.87$, RPD>2.9), better than our previous results in NIRS on apple
395 purees (Lan, Jaillais, Leca, Renard, & Bureau, 2020).

396 In comparison with Vis-NIRS and MIRS, the slight improvements of using the
397 combined spectra (CB) concerned only the prediction of a^* values (**Table 3**).

398 Combining Vis-NIRS and MIRS spectra offered little improvement or even degraded
399 the results in comparison with MIRS alone for analyzing puree viscosity and chemical
400 variations, (**Table 3**). These conclusions were in accordance with previous works on
401 forage (Reeves, 1997) and beers (Iñón, Garrigues, & Guardia, 2006). They can be
402 explained by i) the limited ability to balance the important variables after combination
403 of two spectral domains with different resolutions (**Figure S-6**); and ii) the
404 involvement of non-relevant or unimportant spectral regions which disturbed the
405 calibration modelling by producing more noise.

406 In summary, MIRS coupled with PLS had promising ability to well estimate viscosity,
407 a^* color parameter, DMC, SSC, pH, TA, malic acid, sucrose and glucose of
408 formulated purees, but not for fructose. Acceptable assessments of a^* , TA, malic acid

409 and glucose were obtained with the Vis-NIR region, in which sensors could be easily
410 adapted for fruit processing.

411 **3.4 Characteristics of formulated purees: prediction of quality traits based on the** 412 **reconstructed spectra**

413 In order to compute the concentration profiles of relevant single-cultivar puree
414 compositions, MCR-ALS was applied on the Vis-NIR and MIR spectra of all
415 formulated purees and of the four single-cultivar purees, using two approaches: the 54
416 averaged formulated puree spectra and the 4 averaged single-cultivar puree spectra of
417 a) each week or b) over the three weeks. These two methods (a and b) obtained
418 similar concentrations, indicating their robustness over different processing weeks.
419 Results are only shown for method b taking into account different processing periods
420 (**Table S-3**). Based on that, in total 486 spectra of formulated purees were
421 reconstructed based on their corresponding 36 single-cultivar spectra (4 varieties x 3
422 replications x 3 weeks).

423 Accurate predictions of the concentrations were obtained with MIRS. These
424 predictions were highly related to the proportions of the single-cultivar purees (**Table**
425 **S-3**). However, the results were not acceptable with Vis-NIRS (**Table S-3**). The
426 limited ability of Vis-NIRS was due to the high similarity in color between GA and
427 GD and so a poor prediction of the proportions GA/GD in formulated purees (**Table**
428 **2**). The concentration profiles of MIRS in each group (A-E) appeared to follow a
429 non-linear relationship along the variation of puree proportions.

430 Prediction models were then developed using these reconstructed MIR spectra and the

431 reference data characterized on the formulated purees (**Table 4**). Overall,
432 reconstructed MIR spectra with PLS regression better predicted the puree
433 characteristics than Cubist and RF regressions. What stands out in these results was
434 the highly accurate PLS predictions ($R_p^2 > 0.85$, $RPD > 4.0$) of rheological parameters
435 (η_{50} and η_{100}) from reconstructed spectra (**Table 4**), which were close to those
436 obtained from the spectra of formulated purees ($R_p^2 > 0.90$, $RPD > 4.1$) (**Table 3**).
437 Particularly, similar MIRS fingerprint wavenumbers were obtained in reconstructed
438 spectra and directly on formulated purees described above, mainly 1720, 1113, 1065
439 and 1026 cm^{-1} related to acid and sugars (Bureau, Cozzolino, & Clark, 2019). The
440 prediction of DMC was acceptable ($RPD > 2.5$) as mentioned above with real spectra
441 in **Table 3**. For color, a good prediction of a^* value was obtained with both, PLS
442 ($R_p^2 = 0.92$, $PRD = 3.5$) and machine learning methods ($R_p^2 > 0.89$, $PRD > 3.2$) but not for
443 L^* and b^* . For SSC, although the slight lower R_p^2 and RPD values than the best
444 results obtained directly on MIR spectra ($RMSEP = 0.13$, $RPD = 5.1$) (**Table 3**), the PLS
445 and Cubist models had an acceptable ability to estimate it for all formulated purees
446 ($RMESP < 0.20$, $RPD > 4.1$) (**Table 4**). Considering the global acidity parameters,
447 acceptable PLS predictions ($R_p^2 > 0.88$, $PRD > 3.2$) were obtained for pH and TA, with
448 a lower performance than directly on real spectra ($R_p^2 > 0.92$, $PRD > 4.0$ in **Table 3**).
449 For individual sugars and acids, PLS models showed an excellent prediction of
450 glucose and malic acids ($R_p^2 > 0.94$, $RPD > 4.3$), and an acceptable prediction of sucrose
451 ($R_p^2 = 0.86$, $RPD = 2.8$) but not for fructose ($RPD < 2.5$). The specific wavenumbers at
452 1034 cm^{-1} for glucose, 1723 cm^{-1} for malic acid and 998 cm^{-1} for sucrose, mainly

453 contributed to the PLS models both from reconstructed spectra and directly on puree
454 spectra. The decrease of prediction accuracy was possibly owing to the non-negativity
455 of the concentration profiles which could constrain the spectral reconstruction (Le
456 Dréau, Dupuy, Artaud, Ollivier, & Kister, 2009). Briefly, MIR spectra coupled with
457 the concentration profiles of MCR-ALS showed a potential way to directly estimate
458 the viscosity, a^* color parameter, SSC, TA, malic acid, pH, fructose and glucose for
459 formulated purees depending only on the spectral information of the single-cultivar
460 purees.

461 Compared to the previous prediction models obtained on the real spectra of
462 formulated purees (**Table 3**), highly consistent specific fingerprints and acceptable
463 prediction results (**Table 4**) provided a justifiable explanation to use the spectra
464 reconstruction of formulated purees from spectra of single-cultivar purees. MCR-ALS
465 has been used in other ways to identify precisely the chemical species or track their
466 evolutions (Garrido, Rius, & Larrechi, 2008; de Juan & Tauler, 2006). Here, it was
467 firstly used with the concentration profiles to reconstruct spectra of processed
468 products based on the spectra of raw materials.

469 **4. Conclusion**

470 This was the first detailed work to show the ability of infrared spectroscopy coupled
471 to suitable chemometric methods as a powerful tool to trace different composed
472 cultivars and estimate their corresponding compositions in apple purees. Moreover, an
473 innovative chemometric method based on MCR-ALS was developed to reach
474 simultaneous targets in terms of composition (in % of different cultivars) and

475 physico-chemical properties (rheology, SSC, TA, DMC) of final puree products. As
476 far as we know, this was the first report concerning the control of the final fruit
477 product quality variations depending on the spectral information of the initial purees
478 using a spectral reconstruction approach.

479 Vis-NIR on formulated purees could detect the composed single cultivars purees with
480 large color differences, such as ‘Granny Smith’ ($R_p^2 > 0.92$, $RPD > 3.4$, $RMSEP < 9.2\%$)
481 and ‘Braeburn’ ($R_p^2 > 0.95$, $RPD > 4.2$, $RMSEP < 7.9\%$), but not for ‘Golden
482 Delicious’ and ‘Royal Gala’. MIR had the potential to trace the composed apple
483 varieties with the excellent evaluations of ‘Granny Smith’ and ‘Braeburn proportions’
484 ($RMSEP < 4.3\%$, $RPD > 7.7$) and the satisfactory assessments of ‘Golden Delicious’
485 and ‘Royal Gala’ proportions ($RMSEP < 8.1\%$, $RPD > 3.6$). And MIR could also predict
486 the internal quality (SSC, TA, DMC, viscosity, pH, fructose, malic acid) of formulated
487 purees coupled with PLS and machine learning regressions.

488 Innovatively, MIR technique opens the possibility to control and guidance the final
489 puree characteristics by simply scanning the single-cultivar apple purees, in order to
490 maintain the product quality or to drive the development of new products in apple
491 industry. For instance, after acquiring MIR spectra of the four single-cultivar purees,
492 our developed PLS models might be used in industry: i) to formulate purees with
493 defined SSC and viscosity (e.g. 15.0 ± 0.3 °Brix and 1500 ± 100 Pa.s⁻¹, which might
494 be reached with the formulate solutions as 75% GD-25% GS, 80% GD-20% BR and
495 90% GD-10% GA purees); or ii) to compare *in silico* the results of different puree
496 formulation strategies, such as 33.3% GD and 66.6% GS purees (low redness, high

497 acidity and viscosity) versus another strategy of 80% BR and 20% GA purees (more
498 redness, low acidity, low viscosity), depending on the market.

499 Further challenging works will be to investigate the possibility to reconstruct spectra
500 of final processed purees based on spectra acquired directly on raw apples to provide
501 non-destructive information guidance.

502

503

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512

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629

1 **Figure captions:**

2 **Figure 1.** Experimental scheme of purees reformation, quality characterizations and
3 spectral acquisition.

4 **Figure 2.** Process of VIS-NIRS and MIRS data by multivariate curve resolution-
5 alternative least square (MCR-ALS) and spectral reconstruction of reformulates puree
6 samples.

7 **Figure 3.** Overview of the applied methodology of VIS-NIR and MIR spectra pre-
8 processing and multivariate regression.

9

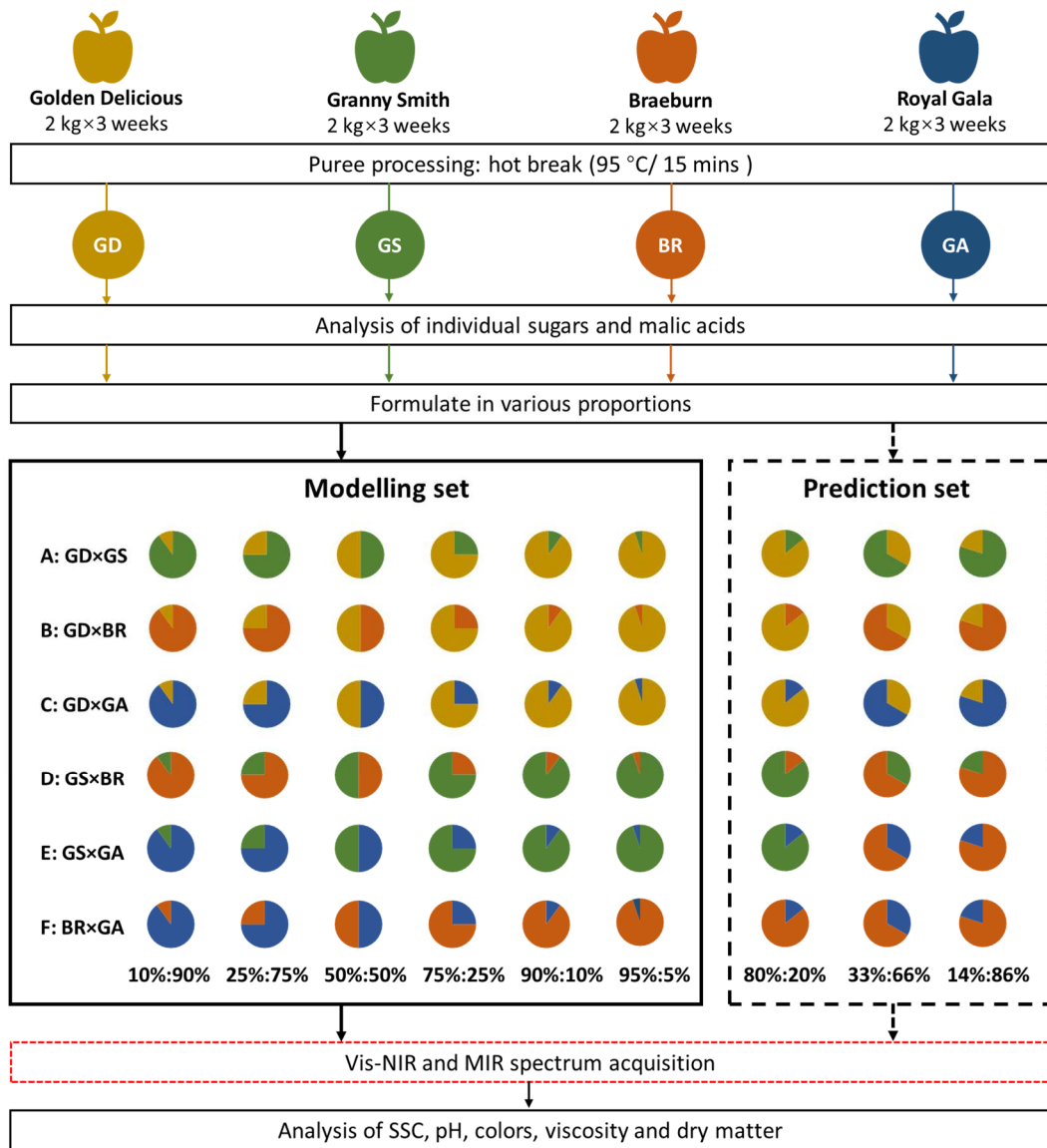
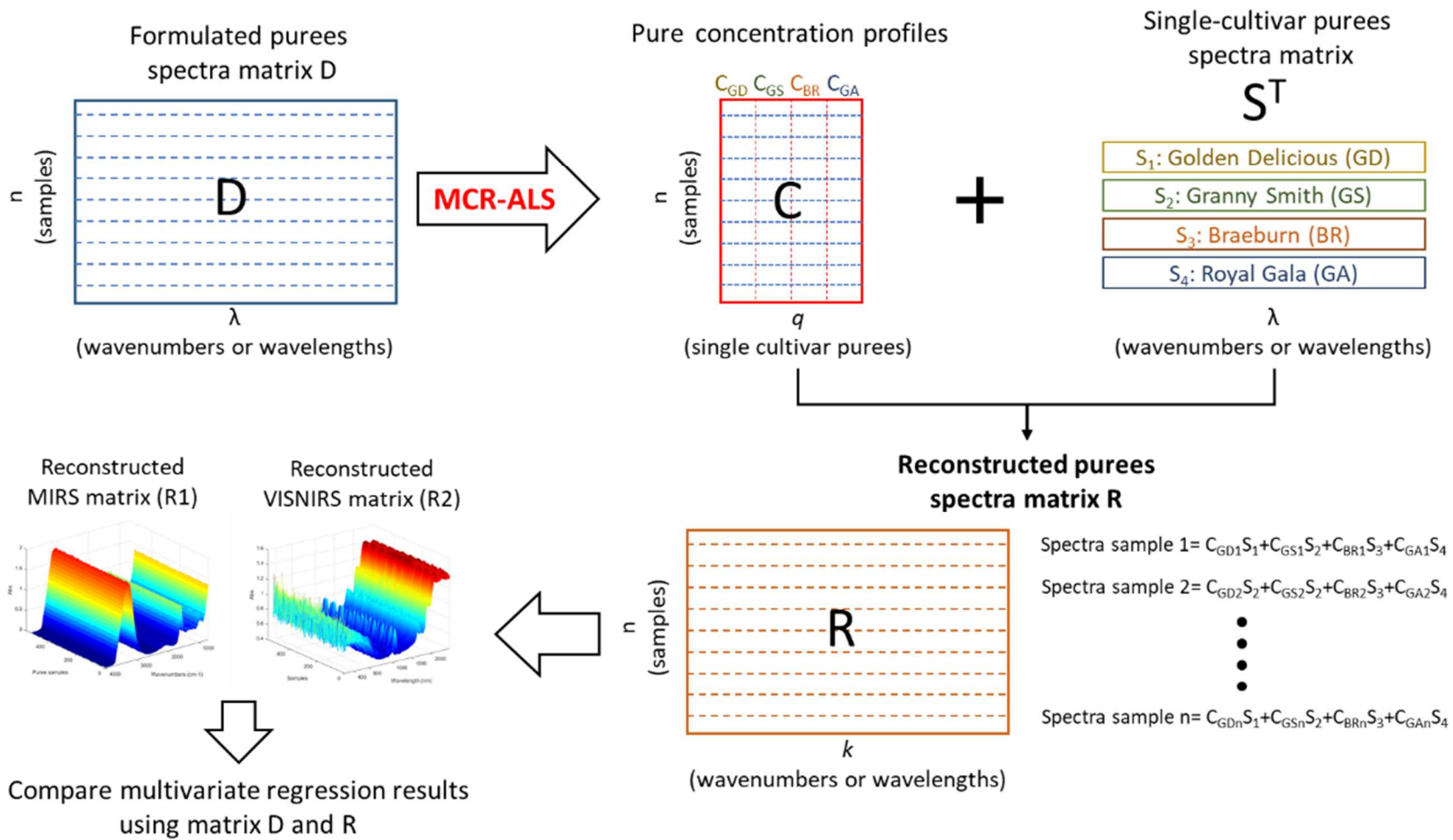


Figure 1



13

14 **Figure 2**

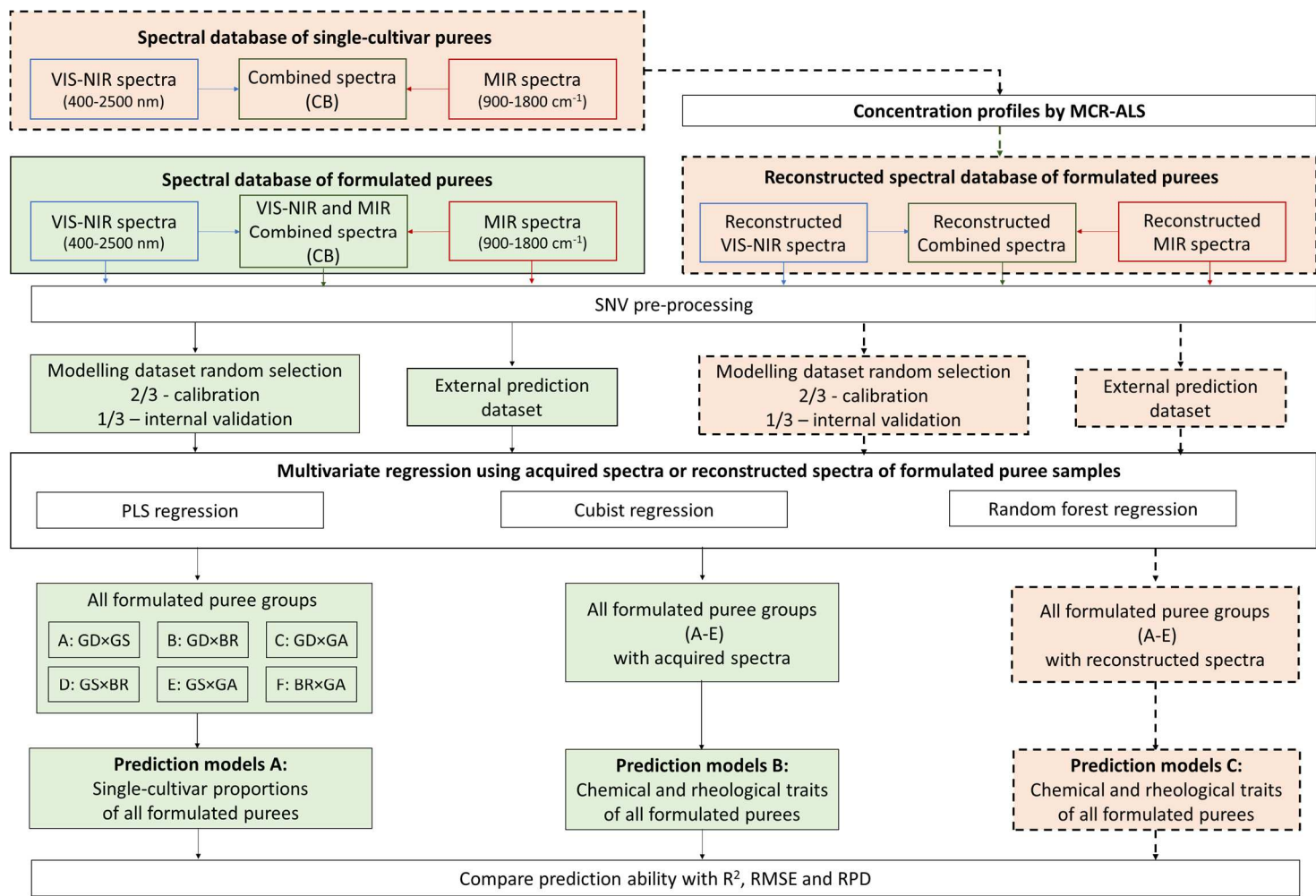


Figure 3

1 **Table 1.** The common names and their abbreviations used in this study

Common names	Abbreviations
'Golden Delicious' purees	GD
'Granny Smith' purees	GS
'Braeburn' purees	BR
'Royal Gala' purees	GA
partial least square	PLS
random forest regression	RF
combination of Vis-NIR and MIR	CB
multivariate curve resolution-alternative least square	MCR-ALS
dry matter contents	DMC
soluble solid contents	SSC
titratable acidity	TA
purees viscosity at a control share rate of 50 s ⁻¹	η_{50}
purees viscosity at a control share rate of 100 s ⁻¹	η_{100}
standard deviation values	SD
Principal component analysis	PCA
fresh weight	FW
standard normal variate	SNV
determination coefficients of internal validation	R_v^2
determination coefficients of external prediction	R_p^2
root mean square error of prediction	RMSEP
variable importance	VIP
Residual Predictive Deviation	RPD

2

3 **Table 2.** Prediction of the proportions (%) of single-cultivar purees in all formulated purees based on VIS-NIR (400- 2500 nm), MIR (900- 1800
 4 cm^{-1}) and their combined spectra (CB; VIS-NIR-MIR). Comparison of three regression models (PLS, Cubist and Random forest)

Single-cultivar	Spectra	PLSR				Cubist				Random forest			
		R_v^2	R_p^2	RMSEP	RPD	R_v^2	R_p^2	RMSEP	RPD	R_v^2	R_p^2	RMSEP	RPD
GD	Vis-NIR	0.66	0.60	19.0	1.5	0.82	0.58	19.4	1.5	0.88	0.64	17.4	1.7
	MIR	0.94	0.92	8.1	3.6	0.95	0.86	11.3	2.6	0.94	0.87	10.6	2.7
	CB	0.91	0.88	10.3	2.8	0.93	0.82	12.7	2.3	0.96	0.90	9.5	3.0
BR	Vis-NIR	0.97	0.95	7.5	4.4	0.98	0.93	8.7	3.8	0.97	0.95	7.9	4.2
	MIR	0.99	0.98	4.3	7.7	0.98	0.97	5.2	6.3	0.98	0.95	7.6	4.3
	CB	0.99	0.98	5.0	6.6	1.00	0.97	5.6	5.9	0.98	0.97	6.1	5.4
GS	Vis-NIR	0.93	0.92	9.2	3.4	0.97	0.89	10.5	3.0	0.97	0.94	8.2	3.8
	MIR	0.99	0.99	2.7	11.4	0.99	0.93	8.1	3.8	0.98	0.97	5.3	5.8
	CB	0.99	0.98	4.3	7.3	0.99	0.98	4.9	6.4	0.98	0.97	5.8	5.4
GA	Vis-NIR	0.79	0.65	16.2	2.1	0.67	0.68	20.2	1.7	0.75	0.73	18.5	1.9
	MIR	0.96	0.94	7.4	4.7	0.90	0.82	15.1	2.3	0.91	0.90	10.3	3.4
	CB	0.89	0.83	11.5	3.0	0.88	0.79	16.2	2.2	0.94	0.92	9.4	3.7

5 Notes: single-cultivar purees of Golden Delicious named 'GD', Braeburn named 'BR', Granny Smith named 'GS', Royal Gala named 'GA'. All results corresponded
 6 to the averaged values of 10 replicates. R_v^2 : determination coefficient of the validation test (internal); R_p^2 : determination coefficient of the prediction test (external);
 7 RMSEP: root mean square error of prediction test (external) expressed as the puree proportions (%); RPD: the residual predictive deviation of prediction test. (external).

8

9 **Table 3.** Prediction of chemical and rheological parameters of all formulated purees using Vis-NIR (400-2500 nm), MIR (900-1800 cm⁻¹) or their
 10 combined spectra (CB) and regression methods, PLS, Cubist or Random forest.

Parameter	Spectra	Range	SD	PLSR				Cubist				Random forest			
				R _v ²	R _p ²	RMSEP	RPD	R _v ²	R _p ²	RMSEP	RPD	R _v ²	R _p ²	RMSEP	RPD
L*	Vis-NIR			0.81	0.70	0.8	1.6	0.87	0.63	0.9	1.4	0.88	0.75	0.6	1.9
	MIR	41.6-48.6	1.5	0.88	0.80	0.6	2.0	0.96	0.83	0.6	2.2	0.94	0.80	0.6	2.2
	CB			0.89	0.79	0.6	2.1	0.95	0.79	0.6	1.9	0.94	0.83	0.5	2.4
a*	Vis-NIR			0.97	0.96	0.4	4.7	0.98	0.94	0.5	4.0	0.96	0.94	0.5	4.1
	MIR	(-4.8)-2.4	2	0.96	0.94	0.5	4.0	0.98	0.92	0.5	3.6	0.97	0.91	0.6	3.3
	CB			0.98	0.96	0.4	5.0	0.99	0.93	0.5	3.6	0.98	0.94	0.5	4.1
b*	Vis-NIR			0.62	0.55	1.2	1.5	0.76	0.46	1.5	1.3	0.72	0.53	1.3	1.4
	MIR	9.6-18.4	1.7	0.67	0.56	1.2	1.5	0.86	0.48	1.4	1.3	0.84	0.62	1.1	1.6
	CB			0.67	0.53	1.3	1.5	0.88	0.46	1.4	1.3	0.81	0.57	1.2	1.5
Viscosity η ₅₀	Vis-NIR			0.79	0.81	54.6	2.2	0.85	0.85	49.8	2.4	0.82	0.78	57.8	2.1
	MIR	526-1029	119	0.94	0.90	29.8	4.1	0.95	0.89	39.4	3.1	0.9	0.87	43.6	2.8
	CB			0.91	0.87	43.5	2.8	0.93	0.88	43.2	2.8	0.91	0.89	42.8	2.8
Viscosity η ₁₀₀	Vis-NIR			0.73	0.74	108.0	2.0	0.87	0.79	98.9	2.2	0.82	0.75	109.3	1.9
	MIR	834-1721	210	0.94	0.91	52.0	4.1	0.96	0.86	81.2	2.6	0.90	0.88	74.4	2.9
	CB			0.88	0.87	79.6	2.7	0.91	0.87	76.5	2.8	0.91	0.88	77.3	2.8
DMC (g/g FW)	Vis-NIR			0.85	0.79	0.004	2.1	0.81	0.75	0.004	1.9	0.79	0.77	0.004	2.0
	MIR	0.14-0.17	0.009	0.93	0.89	0.003	3.1	0.91	0.88	0.003	2.7	0.93	0.90	0.003	3.0
	CB			0.85	0.83	0.003	2.5	0.96	0.83	0.003	2.5	0.93	0.87	0.003	2.8
SSC (°Brix)	Vis-NIR			0.61	0.53	0.5	1.5	0.79	0.56	0.5	1.3	0.78	0.62	0.5	1.5
	MIR	12.1-15.3	0.7	0.96	0.95	0.1	5.1	0.96	0.93	0.2	3.9	0.94	0.94	0.2	4.1
	CB			0.89	0.94	0.2	4.0	0.95	0.92	0.2	3.4	0.95	0.96	0.1	4.4

fructose (g/kg FW)	Vis-NIR			0.37	0.38	7.3	1.2	0.52	0.25	8.4	1.1	0.70	0.50	6.3	1.4
	MIR	40.2-80.3	9.1	0.82	0.78	3.7	2.4	0.93	0.81	4.0	2.2	0.92	0.70	4.8	1.8
	CB			0.67	0.56	5.8	1.5	0.83	0.74	4.4	2.0	0.91	0.76	4.4	2.0
sucrose (g/kg FW)	Vis-NIR			0.54	0.49	3.9	1.4	0.69	0.52	4.3	1.3	0.76	0.46	4.0	1.4
	MIR	33.2-57.3	5.5	0.89	0.89	1.8	3.0	0.88	0.89	1.8	2.9	0.92	0.92	1.6	3.3
	CB			0.60	0.67	3.1	1.7	0.92	0.87	2.0	2.6	0.87	0.78	2.5	2.1
glucose (g/kg FW)	Vis-NIR			0.92	0.93	1.0	3.6	0.96	0.87	1.3	2.6	0.91	0.89	1.2	2.9
	MIR	13.2-28.3	3.7	0.98	0.98	0.5	6.7	0.99	0.97	0.6	5.6	0.97	0.94	0.9	4.1
	CB			0.95	0.93	1.0	3.7	0.98	0.96	0.7	4.9	0.98	0.95	0.8	4.4
pH	Vis-NIR			0.84	0.83	0.1	2.4	0.94	0.76	0.1	2.0	0.82	0.76	0.1	2.0
	MIR	3.39-4.47	0.23	0.94	0.92	0.1	4.0	0.89	0.85	0.1	2.5	0.94	0.92	0.1	3.4
	CB			0.83	0.86	0.1	2.7	0.96	0.67	0.1	1.7	0.9	0.85	0.1	2.4
TA (meq/kg FW)	Vis-NIR			0.93	0.87	5.0	2.9	0.95	0.90	5.1	3.1	0.96	0.89	5.2	3.0
	MIR	28.0-94.8	16.2	0.99	0.96	3.5	4.3	0.99	0.94	3.9	3.9	0.96	0.91	4.7	3.3
	CB			0.95	0.91	4.9	3.1	0.98	0.95	3.8	4.0	0.96	0.9	4.8	3.2
malic acid (g/kg FW)	Vis-NIR			0.90	0.88	0.5	2.9	0.91	0.85	0.5	2.6	0.94	0.87	0.5	2.8
	MIR	3.0-8.8	1.3	0.97	0.97	0.2	5.9	0.95	0.92	0.4	3.7	0.94	0.94	0.3	4.2
	CB			0.92	0.92	0.4	3.4	0.91	0.84	0.5	2.5	0.96	0.93	0.4	3.7

11 Notes: all results corresponded to the averaged values of 10 replicates. R_v^2 : determination coefficient of the validation test (internal); R_p^2 : determination coefficient
12 of the prediction test (external); RMSEP: root mean square error of prediction test (external); RPD: the residual predictive deviation of prediction test. (external).

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14 **Table 4.** Prediction results of chemical and rheological parameters of all formulated purees from the reconstructed MIR spectra computed by the
 15 concentrations of MCR-ALS and the spectra of single-cultivar purees.

Parameter	Range	SD	PLSR				Cubist				Random forest			
			R_v^2	R_p^2	RMSEP	RPD	R_v^2	R_p^2	RMSEP	RPD	R_v^2	R_p^2	RMSEP	RPD
L*	41.6-48.6	1.5	0.91	0.86	0.5	2.4	0.9	0.83	0.6	1.9	0.86	0.78	0.6	2.1
a*	(-4.8)-2.4	2	0.92	0.92	0.5	3.5	0.94	0.89	0.6	3.2	0.93	0.91	0.6	3.4
b*	9.6-18.4	1.7	0.62	0.59	1.2	1.6	0.56	0.48	1.2	1.5	0.58	0.54	1.2	1.5
Viscosity η_{50}	526-1029	119	0.93	0.86	32.3	4.0	0.86	0.82	45.6	3.1	0.86	0.79	47.4	2.8
Viscosity η_{100}	834-1721	210	0.94	0.85	55.5	4.0	0.86	0.83	81	2.8	0.85	0.78	85.3	2.7
DMC (g/g FW)	0.14-0.17	0.009	0.87	0.85	0.003	2.7	0.85	0.84	0.003	2.6	0.89	0.82	0.004	2.5
SSC ($^{\circ}$ Brix)	12.1-15.3	0.7	0.95	0.9	0.2	4.1	0.9	0.85	0.2	4.1	0.79	0.73	0.3	2.3
fructose (g/kg FW)	40.2-80.3	9.1	0.84	0.79	4.0	2.1	0.88	0.82	3.7	2.5	0.83	0.8	3.7	2.3
sucrose (g/kg FW)	33.2-57.3	5.5	0.88	0.86	2.0	2.8	0.87	0.85	2.1	2.7	0.88	0.83	2.1	2.7
glucose (g/kg FW)	13.2-28.3	3.7	0.94	0.94	0.9	4.3	0.97	0.9	1.1	3.2	0.93	0.94	0.9	3.7
pH	3.39-4.47	0.23	0.89	0.88	0.1	3.2	0.89	0.83	0.1	2.8	0.86	0.79	0.1	2.7
TA (meq/kg FW)	28.0-94.8	16.2	0.92	0.91	4.4	3.4	0.91	0.88	5.9	2.7	0.92	0.92	4.4	3.4
malic (g/kg FW)	3.0-8.8	1.3	0.95	0.93	0.3	4.7	0.94	0.87	0.4	3.9	0.95	0.95	0.3	4.3

16 Notes: all results corresponded to the averaged values of 10 replicates. R_v^2 : determination coefficient of the validation test (internal); R_p^2 : determination coefficient
 17 of the prediction test (external); RMSEP: root mean square error of prediction test (external); RPD: the residual predictive deviation of prediction test (external).

18

19 **Supplementary Tables:**20 **Table S-1.** Mean values with the characteristics of single-cultivar purees differed significantly using Tukey's test.

Cultivar	Viscosity η_{50}	Viscosity η_{100}	L*	a*	b*	SSC (°Brix)	DMC (g/g FW)	pH	TA (meq/kg FW)	malic acid (g/kg FW)	fructose (g/kg FW)	sucrose (g/kg FW)	glucose (g/kg FW)
GD	838.6± 69.6 a	1388.5± 138.2 ab	47.0± 0.3 a	-4.1± 0.3 c	15.8± 0.5 a	14.5± 0.4 a	0.167± 0.003 a	3.9± 0.1 b	53.9± 0.8 c	6.0± 0.1 b	72.8± 6.2 a	46.7± 6.6 a	18.7± 0.8 b
GS	904.2± 18.8 a	1501.2± 18.9 a	45.1± 0.8 b	-4.3± 0.9 c	14.4± 1.4 a	13.2± 0.3 b	0.152± 0.003 b	3.6± 0.2 c	89.1± 1.3 a	8.1± 0.5 a	58.6± 14.9 ab	37.4± 4.3 b	26.8± 1.3 a
BR	736.8± 61.1 b	1229.1± 106.2 b	42.2± 0.4 c	1.8± 0.3 a	10.2± 1.2 b	13.1± 0.5 b	0.151± 0.005 b	3.7± 0.2 bc	62.7± 1.5 b	5.9± 0.1 b	59.4± 6.6 ab	50.0± 3.7 a	17.2± 1.5 b
GA	547.1± 38.0 c	860.9± 59.5 c	45.6± 0.5 b	-2.5± 0.2 b	15.0± 0.8 a	12.4± 0.6 b	0.143± 0.008 b	4.3± 0.2 a	29.3± 1.3 d	3.5± 0.7 c	49.3± 4.0 b	36.0± 1.3 b	14.6± 1.3 c

21 Note: Data are expressed as puree fresh weight (FW) ± standard deviation. Puree cultivars: Golden Delicious ('GD'); Granny Smith ('GS'), Braeburn ('BR') and Royal Gala ('GA').

22

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Table S-2. Mean values with the characteristics of formulated puree groups differed significantly using Tukey's test.

Groups	Viscosity η_{50}	Viscosity η_{100}	L*	a*	b*	SSC (°Brix)	DMC (g/g FW)	pH	TA (meq/kg FW)	malic acid (g/kg FW)	fructose (g/kg FW)	sucrose (g/kg FW)	glucose (g/kg FW)
A: GD×GS	854.1± 70.3 a	1412.8± 124.5 a	46.3± 1.0 a	-4.5± 0.2 d	15.0± 1.2 a	13.9± 0.7 a	0.160± 0.007 a	3.7± 0.1 e	70.0± 10.5 b	7.0± 0.8 a	66.1± 6.4 a	42.3± 3.5 b	22.6± 2.8 a
B: GD×BR	767.2 ± 67.4 b	1278.8± 128.7 b	45.2± 1.6 b	-1.3± 1.9 b	13.9± 1.9 b	13.9± 0.5 a	0.160± 0.006 a	3.7± 0.1 d	58.7± 4.3 c	5.9± 0.1 b	66.4± 5.5 a	48.2± 3.1 a	18.0± 0.9 c
C: GD×GA	684.7± 90.5 c	1127.0± 172.3 cd	46.1± 0.9 a	-3.2± 0.7 c	14.7± 0.7 a	13.4± 0.7 b	0.156± 0.008 b	4.1± 0.2 a	42.7± 7.9 e	4.8± 0.8 c	61.6± 11.7 b	41.6± 4.7 b	16.8± 1.6 cd
D: GS×BR	853.8± 99.2 a	1408.5± 173.0 a	43.6± 1.0 c	-1.4± 2.0 b	12.4± 1.4 c	13.2± 0.5 b	0.154± 0.007 d	3.6± 0.1 e	76.6± 7.5 a	7.2± 0.8 a	59.0± 2.9 b	43.4± 5.1 b	22.3± 3.1 ab
E: GS×GA	743.2± 115.2 b	1209.1± 197.0 bc	45.1± 0.5 b	-3.5± 0.8 c	14.4± 1.1 ab	12.9± 0.5 c	0.150± 0.007 c	3.8± 0.2 c	60.5± 17.8 c	5.9± 1.5 b	54.2± 8.7 c	36.7± 2.0 c	21.0± 4.0 b
F: BR×GA	651.7± 86.2 c	1061.4± 161.1 d	44.1± 1.2 c	-0.3± 1.5 a	12.8± 1.7 c	12.8± 0.3 c	0.148± 0.003 c	4.0± 0.2 b	48.7± 10.2 d	4.7± 0.8 c	54.6± 7.2 c	43.3± 5.6 b	16.0± 1.1 d

24

Note: Data are expressed as puree fresh weight (FW) ± standard deviation. Puree cultivars: Golden Delicious ('GD'); Granny Smith ('GS'), Braeburn ('BR') and Royal Gala ('GA').

25 **Table S-3.** The VIS-NIR (400-2500 nm) and MIR (900-1800 cm⁻¹) spectral concentration
 26 profiles of each apple cultivar in formulated puree obtained from MCR-ALS.

Groups	Proportions	MIRS (900-1800 cm ⁻¹)				VIS-NIRS (400-2500 nm)			
		BR	GA	GD	GS	BR	GA	GD	GS
A: GD×GS	95%:5%	0.012	0.000	0.925	0.057	0.000	0.162	0.635	0.193
	90%:10%	0.010	0.000	0.911	0.073	0.000	0.275	0.381	0.333
	75%:25%	0.015	0.049	0.790	0.143	0.000	0.000	0.833	0.169
	50%:50%	0.015	0.000	0.566	0.415	0.000	0.000	0.595	0.406
	25%:75%	0.018	0.000	0.328	0.649	0.000	0.000	0.151	0.857
	10%:90%	0.075	0.000	0.100	0.809	0.000	0.000	0.254	0.731
	80%:20%	0.008	0.000	0.772	0.211	0.012	0.231	0.358	0.398
	33%:66%	0.079	0.003	0.337	0.571	0.033	0.061	0.043	0.868
	14%:86%	0.090	0.120	0.179	0.603	0.005	0.010	0.000	0.979
B: GD:BR	95%:5%	0.050	0.004	0.942	0.004	0.034	0.243	0.564	0.148
	90%:10%	0.081	0.010	0.897	0.000	0.000	0.250	0.621	0.133
	75%:25%	0.235	0.017	0.738	0.000	0.146	0.017	0.812	0.034
	50%:50%	0.495	0.004	0.492	0.000	0.252	0.000	0.764	0.000
	25%:75%	0.711	0.003	0.279	0.000	0.657	0.159	0.073	0.117
	10%:90%	0.847	0.013	0.135	0.000	0.866	0.079	0.000	0.058
	80%:20%	0.141	0.024	0.826	0.000	0.144	0.000	0.865	0.000
	33%:66%	0.515	0.000	0.425	0.058	0.599	0.164	0.103	0.133
	14%:86%	0.827	0.000	0.171	0.002	0.673	0.000	0.328	0.000
C: GD:GA	95%:5%	0.000	0.062	0.933	0.000	0.000	0.078	0.815	0.101
	90%:10%	0.048	0.047	0.897	0.005	0.000	0.000	0.995	0.000
	75%:25%	0.028	0.239	0.728	0.000	0.000	0.407	0.533	0.061
	50%:50%	0.006	0.474	0.500	0.016	0.019	0.584	0.272	0.113
	25%:75%	0.000	0.732	0.263	0.000	0.000	0.601	0.416	0.000
	10%:90%	0.000	0.907	0.087	0.000	0.000	0.521	0.483	0.000
	80%:20%	0.019	0.165	0.807	0.005	0.000	0.528	0.281	0.183
	33%:66%	0.003	0.642	0.352	0.001	0.000	0.367	0.632	0.000
	14%:86%	0.000	0.836	0.142	0.021	0.000	0.549	0.461	0.000
D: GS:BR	95%:5%	0.220	0.090	0.002	0.677	0.000	0.000	0.068	0.927
	90%:10%	0.283	0.259	0.000	0.444	0.001	0.000	0.000	0.994
	75%:25%	0.403	0.163	0.013	0.410	0.205	0.000	0.011	0.768
	50%:50%	0.636	0.138	0.000	0.217	0.417	0.000	0.092	0.492
	25%:75%	0.782	0.067	0.037	0.106	0.631	0.000	0.064	0.312
	10%:90%	0.951	0.005	0.000	0.040	0.853	0.000	0.000	0.144
	80%:20%	0.341	0.162	0.000	0.488	0.143	0.000	0.031	0.822
	33%:66%	0.738	0.082	0.000	0.171	0.543	0.000	0.201	0.249
	14%:86%	0.846	0.000	0.000	0.144	0.668	0.000	0.352	0.000
E: GS:GA	95%:5%	0.000	0.265	0.000	0.725	0.000	0.000	0.142	0.849
	90%:10%	0.006	0.219	0.000	0.787	0.000	0.000	0.178	0.822
	75%:25%	0.030	0.419	0.000	0.537	0.091	0.071	0.058	0.773

	50%:50%	0.009	0.655	0.000	0.334	0.093	0.000	0.560	0.340
	25%:75%	0.003	0.820	0.000	0.169	0.024	0.436	0.348	0.195
	10%:90%	0.047	0.702	0.238	0.000	0.000	0.068	0.943	0.000
	80%:20%	0.050	0.365	0.000	0.574	0.000	0.000	0.286	0.721
	33%:66%	0.000	0.799	0.000	0.193	0.000	0.453	0.114	0.431
	14%:86%	0.000	0.991	0.000	0.000	0.000	0.428	0.530	0.046
	95%:5%	0.785	0.209	0.000	0.000	0.706	0.000	0.314	0.000
	90%:10%	0.849	0.149	0.000	0.000	0.737	0.131	0.076	0.058
	75%:25%	0.599	0.398	0.000	0.000	0.513	0.000	0.496	0.000
	50%:50%	0.406	0.593	0.000	0.000	0.292	0.620	0.000	0.097
F: BR:GA	25%:75%	0.138	0.857	0.000	0.000	0.000	0.541	0.471	0.000
	10%:90%	0.051	0.946	0.000	0.000	0.000	0.533	0.475	0.000
	80%:20%	0.732	0.263	0.000	0.000	0.649	0.194	0.000	0.162
	33%:66%	0.254	0.745	0.000	0.000	0.176	0.644	0.033	0.136
	14%:86%	0.067	0.932	0.000	0.000	0.804	0.000	0.183	0.014

27 Puree cultivars: Golden Delicious ('GD'); Granny Smith ('GS'), Braeburn ('BR') and Royal Gala
28 ('GA').
29