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## Highlights

- MIRS discriminated purees cooked from different apples and processing conditions.
- MIRS on purees gave robust predictions of soluble solids and acidity (RPD $\geqslant$ 3.1).
- Spectra of purees could be calculated from spectra of homogenized raw apples.
- The calculated spectra allowed acceptable predictions of puree viscosity (RPD $\geqslant$ 2.5).

## Abstract

The potential of MIRS was investigated to: i) differentiate cooked purees issued from different apples and process conditions, and ii) predict the puree quality characteristics from the spectra of homogenized raw apples. Partial least squares (PLS) regression was tested both, on the real spectra of cooked purees and their reconstructed spectra calculated from the spectra of homogenized raw apples by direct standardization. The cooked purees were well-classified according to apple thinning practices and cold storage durations, and to different heating and grinding conditions. PLS models using the spectra of homogenized raw apples can anticipate the titratable acidity (the residual predictive deviation (RPD) = 2.9), soluble solid content (RPD = 2.8), particle averaged size (RPD = 2.6) and viscosity (RPD $\geq$ 2.5) of cooked purees. MIR technique can provide sustainable evaluations of puree quality, and even forecast texture and taste of purees based on the prior information of raw materials.

**Key words**: *Malus x domestica* Borkh.; Mid infrared spectroscopy; PLS models; Direct standardization; Discriminant analysis.

## Introduction

Apple puree is one of the major industrially processed fruit products (over 0.3 million tons consumed per year in France and the world market value about 2000 million USD annually) (FranceAgriMer, 2017; Market Research Future, 2019), and can be used as the basic ingredient of jams, preserves or compotes (Defernez, Kemsley, & Wilson, 1995). The quality of apple purees is systematically influenced by both raw material characteristics (Buergy, Rolland-Sabaté, Leca, & Renard, 2020; Lan, Bureau, Chen, Leca, Renard, & Jaillais, 2021; Lan, Jaillais, Leca, Renard, & Bureau, 2020; Rembiałkowska, Hallmann, & Rusaczonek, 2007) and cooking strategies (heating, grinding speed and refining levels etc.) (Espinosa, To, Symoneaux, Renard, Biau, & Cuvelier, 2011; Oszmiański, Wolniak, Wojdyło, & Wawer, 2008; Picouet, Landl, Abadias, Castellari, & Viñas, 2009). In practical apple processing, industrial manufactures have to face the variability and heterogeneity of raw apples, optimize their processing strategies to maintain the sustainable and expected quality level of final puree products. Thus, developing rapid, efficient and integrated methods is needed to guide suitable fruit processing procedures, even to design innovative foods by using the raw material variability, and to reduce fruit wastes all along the processing chain.

Mid infrared spectroscopy (MIRS) is one of the main candidates for both the quantification and qualification of agricultural commodities and processed food (Bureau, Cozzolino, & Clark, 2019; Downey, 1998). Although MIRS presents a relatively lower ability for quantification than that of chromatography or mass spectrometry, it has the advantage of a rapid data acquisition and can provide informative fundamental vibrations of molecular bonds (Karoui, Mazerolles, & Dufour, 2003). It does require a minimal sample preparation as the measurement must be done on homogeneous samples as liquid, puree or powder due to the very low penetration of radiation into the samples. Direct MIR characterizations of raw and processed fruits have shown considerable aptitudes to evaluate soluble solids content (SSC), dry matter content (DMC), titratable acidity (TA), some individual sugars, organic acids, rheological (viscosity and viscoelasticity) and structural (particle averaged size and volume) properties (Ayvaz, Sierra-Cadavid, Aykas, Mulqueeney, Sullivan, &

Rodriguez-Saona, 2016; Lan, Renard, Jaillais, Leca, & Bureau, 2020). These studied parameters are related to the taste (SSC, DMC, TA, malic acid), the texture (viscosity, viscoelasticity, particles and cell wall contents) and the basic nutrients (fructose, sucrose and glucose) impacting in a large amount puree quality (Bureau, Ścibisz, Le Bourvellec, & Renard, 2012; Espinosa-Muñoz, Renard, Symoneaux, Biau, & Cuvelier, 2013; Fügel, Carle, & Schieber, 2005).

Currently, our interest is to determine the possibility of using this technique to anticipate the characteristics of processed materials from the data acquired on homogenized raw fruit. According to our previous studies, strong correlations of spectral, chemical and textural properties between raw apples and the corresponding purees have been pointed out (Lan, Jaillais, Leca, Renard, & Bureau, 2020; Lan, Renard, Jaillais, Leca, & Bureau, 2020). Based on that, the quality of final processed purees could be predicted by the infrared spectral information acquired on raw apples using partial least square (PLS) regression (Lan, Jaillais, Leca, Renard, & Bureau, 2020). The main drawback of this strategy is the need, for modelling, to systematically acquire the corresponding spectra on both raw and processed materials with a large number of conditions representative of the variability, giving often only rough assessments. In addition, the internal correlations of quality traits during puree processing were only confirmed under one of the most commonly used processing conditions (Lan, Jaillais, Leca, Renard, & Bureau, 2020).

Direct standardization (DS) is a simple and efficient chemometric tool for the calibration transfer between spectral measurements or between two different sets of conditions, such as the spectral calibration from the off-line to on-line spectra of olive fruits (Salguero-Chaparro, Palagos, Peña-Rodríguez, & Roger, 2013). As far as we know, this method has never been considered to bridge the spectral variations along the fruit processing chain. Our interest of this method is thus to find the spectra relationships of all processed purees and their corresponding spectral information acquired on homogenized apples, and to calculate the reconstructed processed puree spectra according to their relative spectral information acquired on apples by DS, taking into account both the variability of raw materials and of commonly used processing

conditions. If so, the predictive models of puree quality traits (biochemical and physical) using their reconstructed spectra dataset open the possibility to i) predict the properties of processed apples based on the prior information of raw materials; ii) provide sustainable and precise processing strategies to estimate the quality potential of final products, and iii) to compare *in silico* the prediction results of different processing approaches to better control the quality of fruit products.

Accordingly, this work aimed to assess the potential of MIRS to: i) detect the variability of the cooked apple purees according to the pre- and post-harvest conditions (fruit thinning and storage periods) and the main processing conditions (heating temperature and grinding speed); ii) calculate reconstructed spectra of purees taking into account the variability of raw fruits and processing conditions; and iii) characterize and anticipate the biochemical (SSC, DMC, TA, individual sugars and malic acid), rheological (viscosity and viscoelasticity) and textural (particle size and volume) properties of the processed purees.

## Materials and methods

### 2.1 Apples and purees

#### 2.1.1 Apples

The experiment was conducted on the cultivar 'Golden Delicious' in 2017 and 2019. All apples were harvested at commercial maturity from La Pugère experimental orchard (Mallemort, Bouches du Rhône, France) (**Figure 1**).

- In 2017, half of the 'Golden Delicious' apples were subjected to a commercial chemical fruit thinning (Th+) with standard fruit load (50-100 fruits/tree), the other half was not thinned (Th-), resulting in a high fruit load (150-200 fruits/tree). After harvesting, apples were processed into purees the day after harvest (T0), and after one (T1), three (T3) and six months (T6) of cold storage at 4°C.
- In 2019, the commercially thinned 'Golden Delicious' apples (Th+) were stored for up to one month (T1) at 4 °C until starch regression, then processed into purees under different processing conditions.

#### 2.1.2 Puree processing

Before processing, and for each condition, around 2 kg apples were homogenized

at 11000 rpm with an Ultraturrax T-25 (IKA, Labortechnik, GmbH, Staufen, Germany) as raw apple homogenates. Three batches of apples (3 kg each) were used to produce three puree lots for each condition. After sorting and washing, Golden Delicious apples were cored and cut in 8 portions, then processed in a multi-functional processing system (Roboqbo, Qb8-3, Bentivoglio, Italy) by different conditions (**Figure 1**):

- In 2017, all apple groups (2 thinning practices × 4 storage periods) were cooked with a standard Hot Break recipe of 95°C for 5 min at a 1500 rpm grinding speed, then cooled down to 65°C while maintaining the grinding speed. Finally, 24 different cooked purees (2 thinning practices × 4 storage periods × 3 lots) were prepared.
- In 2019, each apple group was processed with three different heating temperatures of 70°C, 83°C and 95°C for 30 min, while ground at three speed levels of 300 rpm, 1000 rpm and 3000 rpm, respectively. Totally, 27 different cooked purees (3 heating temperatures × 3 grinding speeds × 3 lots) were prepared.

Finally, all cooked purees were conditioned in two hermetically sealing plastic bags: one was cooled at room temperature (22.5 °C) before the next-day measurements of rheological, structural and some biochemical (SSC, TA, fructose, sucrose, glucose and malic acid) properties. And the other one was freeze-dried (FD) and stored at -20 °C for the determination of the content of cell wall, which are known to be a major contributor of rheological properties of apple purees (Espinosa-Muñoz, Renard, Symoneaux, Biau, & Cuvelier, 2013).

## 2.2 Determination of puree quality traits

### 2.2.1 Rheological and structural characterizations on cooked purees

The cooked puree rheological measurements were carried out using a Physica MCR-301 controlled stress rheometer (Anton Paar, Graz, Austria) and a 6-vane geometry (FL100/6W) with a gap of 3.46 mm, at 22.5 °C. The flow curves were performed after a pre-shearing period of 1 minute at a shear rate of 50 $s^{-1}$, followed by 5 minutes at rest. The viscosity was then measured at a controlled shear rate range of [10; 250] $s^{-1}$ on a logarithmic ramp. The values of viscosity at 50 $s^{-1}$ and 100 $s^{-1}$ ($\eta_{50}$ and $\eta_{100}$ respectively) were kept as final indicators of the puree viscosity linked to sensory characteristics during consumption (Engelen & de Wijk, 2012). Amplitude

172 Sweep (AS) tests were performed at an angular frequency of 10 rad./s in the
173 deformation range of [0.01; 100] %, in order to determine the linear viscoelastic range
174 of the purees and the yield stress, defined as the crossing point between the storage
175 modulus (G') and the loss modulus (G'') curves.

176 Cooked purees were diluted in distilled water to separate particles, stained with
177 calcofluor (0.1 g/L) and highlighted with a 365 nm UV lamp (Soukup, 2014). The
178 particle sizes averaged over volume d(4:3) (de Brouckere mean) and over surface area
179 d(3:2) (Sauter mean) were measured by a laser granulometer (Rawle, 2003)
180 (Mastersizer 2000, Malvern Instruments, Malvern, UK).

181 **2.2.2 Biochemical analyses on cooked purees**

182 SSC was determined with a digital refractometer (PR-101 ATAGO, Norfolk, VA,
183 USA) and expressed in °Brix at 20 °C. TA was determined by titration up to pH 8.1
184 with 0.1 mol/L NaOH and expressed in mmol $H^+$/kg of fresh weight (FW) using an
185 autotitrator (Methrom, Herisau, Switzerland). Individual sugars and malic acid were
186 quantified using colorimetric enzymatic kits, according to the manufacturer's
187 instructions (R-biopharm, Darmstadt, Germany). The content of glucose, fructose,
188 sucrose and malic acid were expressed in g/kg FW. These measurements were
189 performed with a SAFAS flx-Xenius XM spectrofluorimeter (SAFAS, Monaco) at 570
190 nm for the sugars and 450 nm for malic acid. The DMC was estimated from the weight
191 of freeze-dried samples upon reaching a constant weight (freeze-drier, 5 days). Cell
192 wall materials of purees were isolated using the alcohol insoluble solids method
193 proposed by Renard (2005) and the cell wall contents (AIS contents) were expressed in
194 both FW and dry matter weight (DW).

195 **2.3 Spectrum acquisition on raw apple homogenates and cooked purees**

196 The MIR spectra were acquired at 23 °C using a Tensor 27 FTIR spectrometer
197 (Bruker Optics®, Wissembourg, France) equipped with a horizontal attenuated total
198 reflectance (ATR) sampling accessory and a deuterated triglycine sulphate (DTGS)
199 detector. The samples were placed at the surface of a zinc selenide crystal (ATR-ZnSe)
200 with six internal reflections. Spectra with 32 scans for ATR-ZnSe were collected from
201 4000 $cm^{-1}$ to 650 $cm^{-1}$ with a 4 $cm^{-1}$ resolution and were corrected against the

background spectrum of air. Three replications of spectral measurement were done for each homogenized raw apples and each cooked apple puree.

The whole spectral dataset of MIR is described in **Figure S1**. It included i) 81 spectra of raw apple homogenates, of which 72 spectra acquired in 2017 (3 apple batches × 2 fruit thinning conditions × 4 storage times × 3 spectral replicates) and 9 spectra acquired in 2019 (3 apple batches × 3 spectral replicates); and ii) 153 spectra of cooked apple purees, containing 72 spectra acquired in 2017 (2 fruit thinning conditions × 4 storage times × 3 processing lots × 3 spectral replicates) and 81 spectra acquired in 2019 (3 heating temperatures × 3 grinding levels × 3 processing lots × 3 spectral replicates).

### 2.4 Statistical analyses of reference data

The reference data of cooked purees processed in 2017 and 2019 are presented as the mean values and the data dispersion within our experimental dataset expressed as standard deviation values (SD). After the Shapiro-Wilk tests, the references data of processed purees affected by fruit thinning and storage times were normal distributed ($\alpha$=0.05), but not for the dataset of heating temperature and grinding effects during puree processing. Thus, analysis of variance (ANOVA) was carried out to determine the significant differences of cooked purees due to fruit thinning and storage times applied on raw apples (**Table S1**) using XLSTAT (version 2018.5.52037, Addinsoft SARL, Paris, France) data analysis toolbox. Kruskal-Wallis tests were performed to evaluate the effects of heating temperature and grinding levels during puree processing (**Table S2**).

### 2.5 Spectra transferred by direct standardization (DS)

In this study, DS was used to find the relationship between the spectra matrices of all cooked purees ($\boldsymbol{P}$) and their corresponding spectra of raw apple homogenates ($\boldsymbol{F}$), taking into account the effects of raw material variability and processing conditions. The DS transfer works were performed in R software (version 4.0.2) (R Core Team, 2019) following a previous report (Ji, Viscarra Rossel, & Shi, 2015):

$$\boldsymbol{P} = \boldsymbol{F}\boldsymbol{B} + \boldsymbol{E} \quad (1)$$

where $\boldsymbol{B}$ is the transfer matrix ($\lambda \times \lambda$) presenting the variations in both $\boldsymbol{F}$ and $\boldsymbol{P}$,

$\boldsymbol{E}$ is the residual matrix used to correct the baseline difference. $\boldsymbol{F}$, $\boldsymbol{P}$ and $\boldsymbol{E}$ matrices have the same size $n \times \lambda$, where $n$ presents the numbers of transfer spectra and λ is the number of wavenumbers between 1800 and 900 $cm^{-1}$.

First, to compute the transfer $\boldsymbol{B}$ and error $\boldsymbol{E}$ matrices, the whole MIR spectral dataset ($\boldsymbol{P}$ and $\boldsymbol{F}$) was divided into: the calibration matrices, presenting the first two batches of raw apple homogenates ($\boldsymbol{Fc}$) and the first two lots of cooked purees ($\boldsymbol{Pc}$), and the validation matrices with the third batch of raw apple homogenates ($\boldsymbol{Fv}$) and the third lot of cooked purees ($\boldsymbol{Pv}$) (**Figure S1**).

In a second step, DS was performed separately on the calibration matrices of raw apple homogenates ($\boldsymbol{Fc}$) and cooked purees ($\boldsymbol{Pc}$) in 2017 ($\boldsymbol{Fc}_{2017}$ and $\boldsymbol{Pc}_{2017}$) and 2019 ($\boldsymbol{Fc}_{2019}$ and $\boldsymbol{Pc}_{2019}$) (**Figures S1 and 2**):

- the calibration matrices of apples ($\boldsymbol{Fc}_{2017}$) and purees ($\boldsymbol{Pc}_{2017}$) were processed to obtain the $\boldsymbol{B_0}$ and $\boldsymbol{E_0}$ related to the effects of raw materials on the processed purees as follows:

$$\boldsymbol{Pc}_{2017} = \boldsymbol{Fc}_{2017}\boldsymbol{B_0} + \boldsymbol{E}_0 \quad (2)$$

Both ($\boldsymbol{Fc}_{2017}$) and ($\boldsymbol{Pc}_{2017}$) have the same size $n \times \lambda$ , where $n$ = 48; (2 thinning practices × 4 storage periods × 2 apple batches/ puree lots × 3 spectral replicates.

- the calibration matrices of apples ($\boldsymbol{Fc}_{2019}$) and purees ($\boldsymbol{Pc}_{2019}$) were performed for each puree processing condition, as follows:

$$\boldsymbol{Pc}_{\boldsymbol{2019}}^{(\boldsymbol{i})} = \boldsymbol{Fc}_{2019}\boldsymbol{B}_i + \boldsymbol{E}_i \quad (3)$$

where $i$ from 1 to 9, corresponding to 9 different processing conditions (3 heating temperatures × 3 grinding speeds). To each spectral replicate of $\boldsymbol{Fc}_{2019}$ corresponds nine spectra according to each processing condition ($\boldsymbol{Pc}_{2019}$). All the spectra of $\boldsymbol{Pc}_{2019}$ matrix corresponding to the same processing conditions were gathered in a specific matrix $\boldsymbol{Pc}_{\boldsymbol{2019}}^{(\boldsymbol{i})}$. The size of this matrix (one for each processing condition) is equal to that of raw apple homogenates $\boldsymbol{Fc}_{2019}$ ($n$ = 6 spectra (2 apple batches × 3 spectral replicates) × λ).

Thirdly, once all the transfer $\boldsymbol{B}$ ($\boldsymbol{B}_0$ and $\boldsymbol{B}_i$) and error $\boldsymbol{E}$ ($\boldsymbol{E}_0$ and $\boldsymbol{E}_i$) matrices were computed, they were used to calculate the cooked puree reconstructed calibration

and validation spectra matrices, as follows (**Figure 2**):

$$\boldsymbol{Tc}_{2017} = \boldsymbol{Fc}_{2017}\boldsymbol{B}_0 + \boldsymbol{E}_0 \quad (4)$$

$$\boldsymbol{Tv}_{2017} = \boldsymbol{Fv}_{2017}\boldsymbol{B}_0 + \boldsymbol{E}_0 \quad (5)$$

$$\boldsymbol{Tc}_{2019} = \boldsymbol{Fc}_{2019}\boldsymbol{B}_i + \boldsymbol{E}_i \quad (6)$$

$$\boldsymbol{Tv}_{2019} = \boldsymbol{Fv}_{2019}\boldsymbol{B}_i + \boldsymbol{E}_i \quad (7)$$

Finally, the reconstructed calibration and validation spectral matrices of cooked puree, $\boldsymbol{Tc}$ ($\boldsymbol{Tc}_{2017} + \boldsymbol{Tc}_{2017}$) and $\boldsymbol{Tv}$ ($\boldsymbol{Tv}_{2017} + \boldsymbol{Tv}_{2019}$) of the two years (2017 and 2019) were obtained with the same sizes of the real spectral matrices of cooked puree, $\boldsymbol{Pc}$ and $\boldsymbol{Pv}$, for the further multivariate regressions.

### 2.6 Multivariate regression

Spectral pre-processing and multivariate data analysis were performed with Matlab 7.5 (Mathworks Inc. Natick, MA, USA) software using the SAISIR package (Cordella & Bertrand, 2014). After pretests of several pre-processing treatments (baseline correction, standard normal variate (SNV) and a derivative transform calculation using Savitzky–Golay method (window size = 11, 21, 31) applied on several different spectral regions, the best results of prediction and discrimination were obtained on the range 1800-900 $cm^{-1}$, which has been already highlighted (Lan, Renard, Jaillais, Leca, & Bureau, 2020). Principal Component Analysis (PCA) and Factorial Discriminant Analysis (FDA) were applied on SNV pre-treated spectra of cooked purees to detect their differences related to the variability of both, raw apples and processing conditions. The specificity and sensitivity values of FDA discriminations were calculated by the already reported method of Nargis et al. (2019).

PLS models were developed using the SNV pre-processed puree spectra (1800-900 $cm^{-1}$) of the calibration set $\boldsymbol{Pc}$ and the DS transferred spectra of purees ($\boldsymbol{Tc}$), corresponding to the same reference dataset. The two calibration matrices of cooked purees included a total of 102 spectra (48 spectra in 2017: 2 thinning practices × 4 storage periods × 2 lots × 3 spectral replicates, and 54 spectra in 2019: 3 heating temperatures × 3 grinding speeds × 2 lots × 3 spectral replicates). Then, the developed PLS models were applied on their corresponding validation spectra sets of $\boldsymbol{Pv}$ and $\boldsymbol{Tv}$, with a total of 51 spectra in 2017: 2 thinning practices × 4 storage periods × 1 lot

× 3 spectral replicates and in 2019: 3 heating temperatures × 3 grinding speeds × 1 lot × 3 spectral replicates. PLS model performance was assessed using the determination coefficient of calibration ($R_c^2$) and validation ($R_v^2$), the root-mean-square error of validation (RMSEV), the number of latent variables for calibration (LVs), the residual predictive deviation of validation set (RPD), as described by Nicolai et al. (2007). The linkable spectral regions of the acceptable PLS models presenting RPD values higher than 2.5 (Nicolai, Beullens, Bobelyn, Peirs, Saeys, Theron, et al., 2007) were displayed based on their β- coefficients (**Tables 1 and 2**).

## 3. Results and discussion

### 3.1 Variability of cooked purees based on their MIR Spectra

#### 3.1.1 Variability induced by the raw materials

According to ANOVA (F-values), fruit thinning applied on apples during their growth in orchard resulted in a significant variation ($p < 0.001$) of viscosity ($\eta_{50}$ and $\eta_{100}$), viscoelasticity (G', G'', yield stress), particle sizes (d4:3 and d3:2) and biochemical compositions (DMC, SSC, TA, pH, malic acid, sucrose, fructose and AIS) of the cooked purees. Particularly, the impact of thinning on the viscosity, DMC and SSC of purees was higher than the effect of post-harvest storage at 4°C (**Table S1**). Purees processed from thinned apples (Th+) had higher viscosity values ($\eta_{50}$ and $\eta_{100}$) and bigger particle sizes (d4:3) than those from the non-thinned apples (Th-), observed after the three months of cold storage (T3 and T6) (Buergy, Rolland-Sabaté, Leca, & Renard, 2020). Moreover, an intensive decrease of average particle sizes (d 4:3) was observed in the purees cooked with the apples stored one month at 4°C (T1) for both 'thinning' (Th+) and 'non-thinning' (Th-) treatments. PCA applied on the spectra of cooked purees in 2017 showed a good ability to detect the effects of treatments applied on raw apples (fruit thinning and storage periods) (**Figure 3a and 3b**). The effect of thinning on the first principal component (PC1 90.1%) was much higher than that of storage on the second principal component (PC2 6.9%), which was in line with our previous results (Lan, Renard, Jaillais, Leca, & Bureau, 2020). In addition, the increase of the band at 1022 $cm^{-1}$ and the decrease of the bands at 1061-1065 $cm^{-1}$, attributed to sucrose and fructose respectively (Bureau, Cozzolino, & Clark, 2019), were the major

contributors of the observed discriminations on the two PCs (**Figure 3c and 3d**).

3.1.2 Variability induced by processing conditions

The different grinding speeds affected significantly ($p < 0.05$) the viscosity ($\eta_{50}$ and $\eta_{100}$), viscoelasticity (G' and G'') and particle size (d4:3 and d3:2) of the cooked purees (**Table S2**). Particularly, the increase of grinding speed significantly ($p < 0.001$) decreased viscosity ($\eta_{50}$ and $\eta_{100}$), viscoelasticity (yield stress, G' and G'') and particle sizes (d4:3) which was observed at each tested temperature. From macroscopic images of purees (data not shown), the larger particles disappeared with increasing grinding speeds, which was enough to cause a decrease in the puree viscosity (Espinosa-Muñoz, Renard, Symoneaux, Biau, & Cuvelier, 2013). Inversely, the increasing heating temperatures induced no significant ($p > 0.05$) changes of puree viscosity ($\eta_{50}$ and $\eta_{100}$) and viscoelasticity (yield stress, G' and G''). The highest heating temperature (95°C) resulted in a significant ($p < 0.05$) increase of DMC and SSC and a decrease of TA and malic acid. Consequently, the changes of grinding speed during the puree processing significantly modified the structural properties and viscoelastic behaviors of purees, whereas heating temperature affected strongly the biochemical composition of purees.

FDA performed on the cooked puree spectra in 2019 successfully classified the processing changes induced at the different heating temperatures (**Figure 4a**) and grinding speeds (**Figure 4b**). The samples cooked at 95 °C were well-separated from the other two conditions (4 factors, 100% of sensitivity and specificity in **Table S3a**), according to the first factorial component (F1) (**Figure 4a**). The specific bands at 1745 $cm^{-1}$ and 1539 $cm^{-1}$ were attributed to the increase of soluble pectins, probably in relationship with their solubilization in puree serum from apple cell walls, enhanced with the increasing heating temperature (Liu, Renard, Rolland-Sabaté, Bureau, & Le Bourvellec, 2020). Moreover, the negative peaks at 1057 $cm^{-1}$ and 998 $cm^{-1}$ could be due to the hydrolysis of sucrose during thermal processing, thus resulting in the increase of fructose (1022 $cm^{-1}$) and glucose (1107 $cm^{-1}$) contents.

The three different grinding levels could be discriminated according to the first two factorial components (F1 and F2) (**Figure 4b**), especially for the highest grinding speed at 3000 rpm ('G3' in **Figure 4b**) (4 factors, over 85.19% of specificity and sensitivity

in **Table S3b**). The intensive negative spectral peaks at 1558, 1539-1541, 1508 and 1458 $cm^{-1}$ along both the two discriminant factors (F1 and F2 in **Figure 4d and 4e**) were all located in the region between 1450 $cm^{-1}$ and 1600 $cm^{-1}$, which has been already attributed to the changes of particle size and rheological behavior after apple puree mechanical refining in a previous experiment (Lan, Renard, Jaillais, Leca, & Bureau, 2020). These peaks indicated the decrease of particle size (d4:3 and d3:2) and viscosity of purees with increasing grinding speeds, which was in line with our reference measurements (**Table S2**) and Espinosa et al. (2011).

Briefly, MIR technique could detect several kinds of variability sources such as thinning practices during fruit cultivation, cold storage and processing conditions (temperature and grinding) in the cooked purees. In addition, the spectral region 1450-1750 $cm^{-1}$ was validated as being a reliable analytical signal of processing linked to the textural and rheological changes in purees.

### 3.2 Prediction of quality traits of cooked purees by MIRS

For all developed PLS models, as expected the decreases of determination coefficients between the calibration set ($R_c^2$) and the validation set ($R_v^2$) were observed in **Table 1**. According to RPD values over 2.5 (Nicolai, Beullens, Bobelyn, Peirs, Saeys, Theron, et al., 2007), prediction was acceptable to good (RPD from 2.6 to 3.3) for viscosity ($\eta_{50}$ and $\eta_{100}$), average particle sizes (d4:3), SSC, TA, pH values and malic acid were content in cooked purees by MIRS, taking into account a large variability of raw apple materials and processing conditions.

Apparent puree viscosity at a shear rate value of 50 $s^{-1}$ ($\eta_{50}$), which has been described to be the highly correlated with the in-mouth texture perception of fluid food (Chen & Engelen, 2012), could be predicted by MIRS with a $R_v^2$ of 0.87 and a RPD of 3.2. MIR prediction of apparent puree viscosity at a single shear rate value of 100 $s^{-1}$ ($\eta_{100}$) ($R_v^2$ = 0.85, RPD= 3.0) observed here were much better than its prediction by NIRS (RPD = 1.3) (Lan, Jaillais, Leca, Renard, & Bureau, 2020), These results evidenced the possibility of MIRS to estimate puree viscosity. For the two apparent puree viscosity values measured at $\eta_{50}$ and $\eta_{100}$, the main wavenumber regions at 1718-1730 $cm^{-1}$ and 1618-1678 $cm^{-1}$ were still observed in our previous work (Lan, Renard,

Jaillais, Leca, & Bureau, 2020). This could validate the application of MIRS to predict puree viscosity by taking into account not only the raw fruit variability but also the complex effects of processing conditions. However, the predictions ($R_v^2$ < 0.81, RPD < 2.1) of the viscoelastic parameters of purees (G', G'' and yield stress) were not precise enough to estimate the viscoelastic behaviors and the moment when puree starts to flow. Indeed, heating and grinding affected puree viscoelasticity (SD values of 2362 Pa for G', 595 Pa for G'', 27.1 Pa for yield stress) and resulted in more than twice higher variations of these parameters than those induced by thinning and cold storage on raw materials (SD values of 1001 Pa for G', 234 Pa for G'' and 12.9 Pa for yield stress). These new prediction accuracies of the viscoelastic parameters of purees (G', G'' and yield stress) were not as good as our previous ones by MIRS (Lan, Renard, Jaillais, Leca, & Bureau, 2020), but could be more robust to be considered for future applications. MIR coupled with the linear regression (PLS) showed a good performance ($R_v^2$= 0.87, RPD= 3.1) to evaluate the volume average particle size of purees (d4:3), but not the surface average particle size (d3:2). Particularly, the most informative wavenumbers to evaluate puree particle size at 1701-1713 $cm^{-1}$, 1655-1668 $cm^{-1}$ and 1537-1541 $cm^{-1}$ have been already observed previously to predict puree viscosity, to discriminate the purees prepared with different grinding speeds (mentioned in **part 3.1**) and with different refining levels (Lan, Renard, Jaillais, Leca, & Bureau, 2020). Such good prediction of puree average particle size (d4:3) could not come from internal correlations with puree composition such as SSC, DMC or AIS contents because of their poor correlation ($R^2$ < 0.48), but probably some specific signals needing to be identified and confirmed. Moreover, we confirmed here the impossibility to predict the cell wall content directly in puree by MIRS, without any preparation such as freeze-drying (Lan, Renard, Jaillais, Leca, & Bureau, 2020)

A good prediction of global puree quality traits, SSC (RPD= 3.1) and DMC (RPD= 2.9), was obtained with 5 LVs (**Table 1**). The variation of SSC and DMC in purees were highly correlated ($R^2$ = 0.76), which explained the good estimations of these two parameters. Their respective fingerprint wavenumbers of SSC and DMC prediction were similar and detected at 996-1001 $cm^{-1}$, 1048-1057 $cm^{-1}$ and 1109-1112 $cm^{-1}$,

corresponding to the variations of sucrose, fructose and glucose in purees (Bureau, Cozzolino, & Clark, 2019). Moreover, the prediction of TA was excellent (RPD = 3.2), with a limited RMSEV of 7.6 mmol $H^+$/kg FW. The typical wavenumber region at 1709-1720 $cm^{-1}$ in TA prediction has already been attributed to the C=O vibration of acid group (Bureau, Quilot-Turion, Signoret, Renaud, Maucourt, Bancel, et al., 2013; Clark, 2016). Depending on the good correlations between the different contributors of apple acidity ($R^2$ = 0.81 between TA and malic acid, $R^2$ = 0.76 between TA and pH), MIRS provided an acceptable prediction of pH ($R_v^2$ = 0.83 and RPD = 2.5) and malic acid content ($R_v^2$ = 0.85 and RPD= 2.7). Despite the similar typical fingerprints observed in the β-coefficients of PLS models of malic acid and pH, the relative lower RPD values and $R_v^2$ of pH compared to malic acid were probably due to the low pH variation. Concerning the main individual sugars, acceptable prediction was obtained only for fructose (RPD = 2.6), but neither for sucrose (RPD= 1.3) nor for glucose (RPD = 1.5), which was in line with our previous results (Lan, Renard, Jaillais, Leca, & Bureau, 2020). The lower concentration of glucose (10.4-25.4 g/kg FW) than the other individual sugars (34.9-98.7 g/kg FW of fructose, 39.1-118.5 g/kg FW of sucrose) led to its worse prediction by MIR results. A higher internal biochemical correlation between the major compounds (SSC, TA) and fructose ($R^2$ = 0.79 for SSC and fructose, $R^2$ = 0.76 for TA and fructose) in apple purees might explain the better prediction of fructose than the one of sucrose ($R^2$ = 0.58 for SSC and sucrose, $R^2$ = 0.44 for sucrose and TA).

Briefly, MIR technique can provide a simultaneous and robust estimation of biochemical compositions (dry matter, soluble solids, titratable acidity, pH, malic acid and fructose), rheological behaviors (viscosity at $\eta_{50}$ and $\eta_{100}$) and particle size (d4:3) of apple purees, taking into account the large variability along the apple puree production chain (agricultural practices, post-harvest storage and processing conditions).

### 3.3 Reconstructed spectra for prediction of puree quality traits

In this part, MIR prediction models were developed using the reconstructed spectra of the calibration set of cooked purees ($\boldsymbol{Tc}$), only done for the well-predicted parameters

mentioned in **part 3.2**, which were $\eta_{50}$ and $\eta_{100}$, d4:3, SSC, DMC, TA, pH, malic acid and fructose. Then, these models were applied on the validation reconstructed spectra of the cooked purees ($\boldsymbol{Tv}$).

Overall, based on the PLS regression applied on the puree reconstructed spectra, acceptable predictions were obtained (RPD > 2.5) for rheological ($\eta_{50}$, $\eta_{100}$), structural (d4:3) and global biochemical (SSC, DMC, TA) parameters (**Table 2**). In contrast, predictions appeared not acceptable for malic acid (RPD = 2.3), fructose (RPD = 1.7) and pH (RPD = 2.1). Compared to the previous prediction from the real puree spectra (**Table 1**), lower $R_v^2$ and higher LVs have been generally obtained for all parameters giving some lower prediction performance (**Table 2**). Particularly, the use of the reconstructed spectra showed a good ability to predict puree viscosity parameters ($\eta_{50}$ and $\eta_{100}$) with $R_v^2$ > 0.82, RPD > 2.5 and prediction errors (RMSEV) of 0.21 Pa.s and 0.10 Pa.s for $\eta_{50}$ and $\eta_{100}$, respectively. These results were close to those from the real spectra of purees (**Table 1**). The fingerprint wavenumbers used in the PLS models were similar for both reconstructed and real spectra, mainly 1718-1734 $cm^{-1}$, 1616-1336 $cm^{-1}$ and 1547-1553 $cm^{-1}$ as described in **Part 3.2**. Although a relative lower $R_v^2$ and RPD ($R_v^2$ = 0.84 and RPD = 2.6) were obtained for particle size (d4:3) compared to the results on the real puree spectra (**Table 1**), the consistent fingerprints were highly related to the puree texture such as 1701-1715 $cm^{-1}$, 1537-1541 $cm^{-1}$ and 1101-1107 $cm^{-1}$. These prediction performances revealed for the first time the possibility to evaluate the variation of averaged particle sizes in the cooked purees based on the MIR information of the corresponding raw apple homogenates. Considering the other global quality parameters, acceptable predictions were obtained for SSC ($R_v^2$ = 0.85 and RPD = 2.8) and DMC ($R_v^2$ = 0.84 and RPD = 2.6) contents. The specific wavenumbers in the ranges 997-1001 $cm^{-1}$ and 1048-1057 $cm^{-1}$ for sucrose and in the ranges 1009-1112 $cm^{-1}$ for fructose mainly contributed to the PLS models for both reconstructed and real spectra. These ranges have been already mentioned to be linked to these sugars (Bureau, Cozzolino, & Clark, 2019), which are the main ones in apples. For acidity, the reconstructed spectra gave an excellent prediction of TA ($R_v^2$ = 0.86 and RPD = 2.9), using the spectral regions between 1709-1720 $cm^{-1}$ of the typical C=O absorption

(Clark, 2016). Consequently, PLS applied on the reconstructed MIR spectra calculated from the spectra of raw apple homogenates showed the possibility to directly predict the viscosity, averaged particle sizes, SSC, DMC and TA of cooked purees.

Several initial attempts have been tested to monitor the quality of cooked food from infrared information of their raw materials with the objectives to predict the texture of cooked poultry pectoralis major muscles (Meullenet, Jonville, Grezes, & Owens, 2004), of cooked rice (Windham, Lyon, Champagne, Barton, Webb, McClung, et al., 1997) and of apple purees (Lan, Jaillais, Leca, Renard, & Bureau, 2020). In these studies, the spectra matrix of the raw materials and the reference data of the corresponding processed materials were used to calibrate models. The predictions thus obtained are mainly due to the strong internal correlations of quality traits between materials before and after processing, which could provide semi-quantitative prediction accuracy for practical uses. However, the internal correlations of quality traits during fruit processing still remain unreliable when using a large variability of raw materials and various industrial processing systems (Lan, Jaillais, Leca, Renard, & Bureau, 2020). Further, such a direct modelling method requires a necessary step of acquisition of the infrared information on raw material batches for each processing condition, in order to obtain the same matrix sizes of spectra and reference data for calibration.

Here, a potential strategy has been firstly proposed to build reconstructed MIR spectra of processed purees from the spectra of raw apple homogenates using a spectral transfer method. The high consistency of the specific fingerprints used in the PLS models obtained for both the real spectra and the reconstructed spectra, confirmed our choice for this modelling strategy. Compared to the direct modelling method, a great advantage of using spectral transfer strategy is that the calibration dataset only needs the infrared information and reference data of several processed purees and just a limited number of spectra of corresponding raw apples. For example, in our dataset of 2019, the reconstructed spectra of 27 different processed purees could be transferred from only 3 corresponding spectra of the same apple batches.

After a simple scanning of raw apple homogenates by MIRS, our models revealed the possibility to i) predict the quality of apple purees, such as viscosity, SSC and TA

501 using a standard processing recipe (95 °C for 5 mins and grinding at 1500 rpm), even
502 though a large variability of raw apples was used (different fruit thinning and cold
503 storage periods); and ii) to monitor and anticipate the organoleptic properties of cooked
504 purees under different processing strategies, which is relevant for the processors and
505 market. For example, a higher viscosity and acidity in-mouth feeling (predicted $\eta_{50}$ =
506 1.42 ± 0.09 Pa.s, predicted TA = 65.8 ± 3.5 meq/kg FW) were predicted with the recipe
507 at 83 °C for 30 min and grinding speed of 1000 rpm than with the recipe at 95 °C for
508 30 min and grinding speed of 3000 rpm (predicted η 50 = 0.98 ± 0.14 Pa.s, predicted
509 TA = 56.4 ± 4.5 meq/kg FW).

510 **Conclusion**

511 As far as we know, this is the first study that shows the ability of MIRS to estimate
512 the quality of processed fruit products taking into account a large variability coming
513 from agricultural practices, post-harvest storage and processing conditions along the
514 whole processing chain. MIR technique provided reliable assessment of viscosity,
515 averaged particle sizes and major compositions (SSC, DMC, TA and malic acid) of
516 apple purees.

517 Further, a simple spectroscopic transfer method (direct standardization) was
518 applied for the first time to develop the reconstructed spectra of purees from their
519 corresponding spectra of raw apple homogenates. MIRS coupled with PLS regression
520 obtained acceptable predictions of TA, DMC, SSC, viscosity (η 50 and η 100) and
521 averaged particle sizes of the final puree based on their reconstructed spectra. With a
522 simple scanning of raw apple homogenates, MIR technique opens the possibility to i)
523 predict the quality of final purees under a standard processing procedure, which is
524 beneficial for fruit processing sustainability; and even ii) to monitor the texture and
525 tastes of purees under different processing conditions for a better management.

526

**Acknowledgements**

The authors thank Patrice Reling, Barbara Gouble, Marielle Boge, Caroline Garcia and Gisèle Riqueau (INRAE, SQPOV unit) for their technical help. The 'Interfaces' project is an Agropolis Fondation Flashship project publicly funded through the ANR (French Research Agency) under "Investissements d'Avenir" programme (ANR-10-LABX-01-001 Labex Agro, coordinated by Agropolis Fondation). Weijie Lan was supported by a doctoral grant from Chinese Scholarship Council.

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**Figure captions:**

**Figure 1.** Experimental scheme for apple production, puree preparation and the sample characterization by infrared spectroscopy and reference measurements.

**Figure 2.** Overview of the applied methodology to exploit reconstructed MIR spectra of purees and multivariate regression.

**Figure 3.** Principal Component Analysis on the SNV pre-treated MIR spectra (900-1800 $cm^{-1}$) of purees cooked with thinned (Th+) and non-thinned (Th-) 'Golden Delicious' apples stored at 4°C during 0, 1, 3 and 6 months (T0, T1, T3 and T6): (**a**) the scores plot of the first two components (PC1 and PC2) related to fruit thinning; (**b**) the scores plot of the first two components (PC1 and PC2) related to storage periods; (**c**) the loading plot of PC1; (**d**) the loading plot of PC2.

**Figure 4.** Maps of Factorial Discriminant Analysis (FDA) performed on the SNV pre-treated MIR spectra (900-1800 $cm^{-1}$) of purees cooked with: (**a**) three different temperatures (70 °C, 83 °C and 95 °C) and (**b**) three grinding speeds (G0 at 300 rpm, G1 at 1000 rpm and G3 at 3000 rpm); (**c**) the first factorial score ('F1') of heating temperature discrimination; (**d**) the first factorial score ('F1') of grinding discrimination; (**e**) the second factorial score ('F2') of grinding discrimination.

**Figure S1**. Overview of MIR spectra pre-processing, direct standardization (SD) and multivariate regression

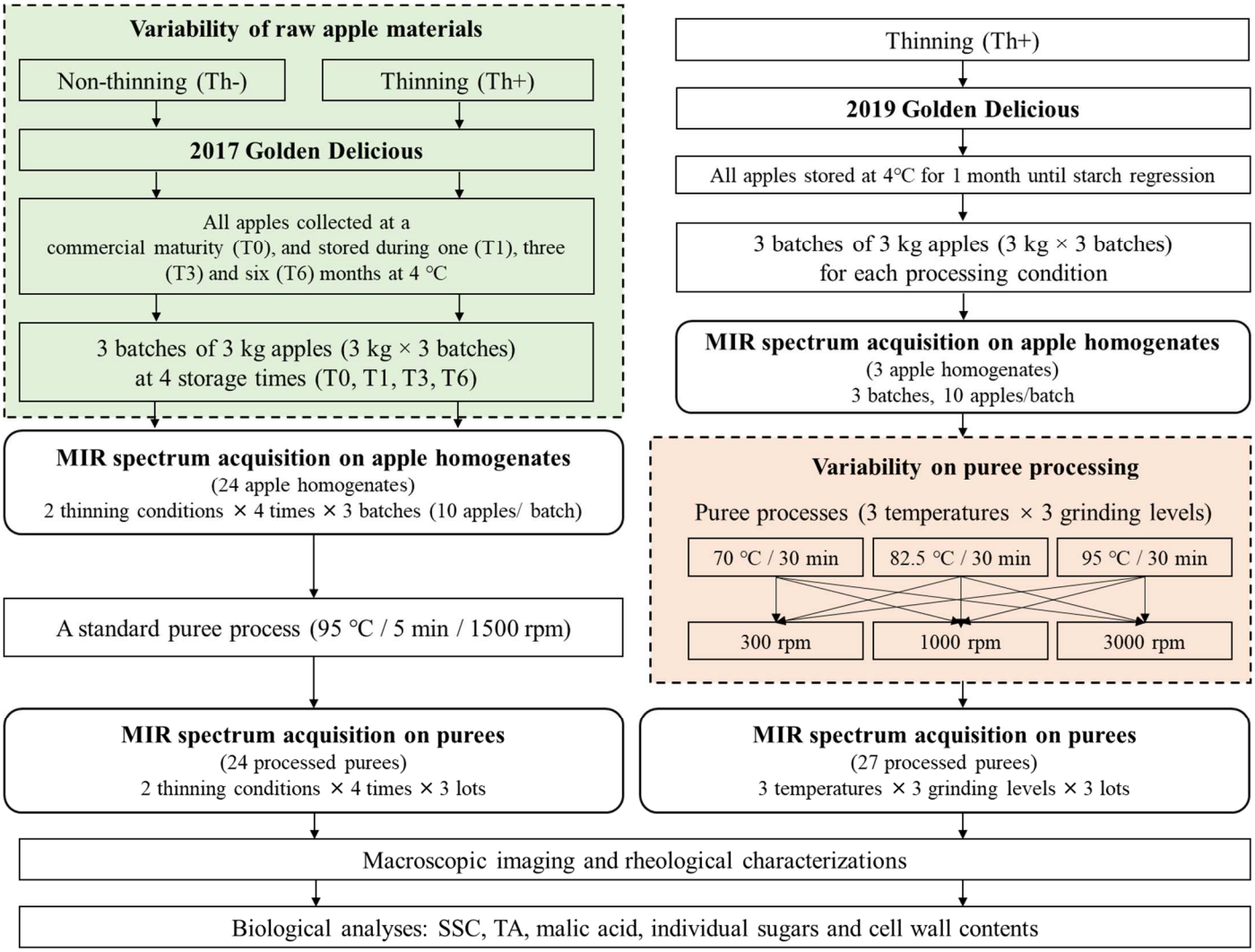
Variability of raw apple materials
Non-thinning (Th-)
Thinning (Th+)
2017 Golden Delicious
All apples collected at a commercial maturity (T0), and stored during one (T1), three (T3) and six (T6) months at 4 °C
3 batches of 3 kg apples (3 kg × 3 batches) at 4 storage times (T0, T1, T3, T6)
MIR spectrum acquisition on apple homogenates
(24 apple homogenates)
2 thinning conditions × 4 times × 3 batches (10 apples/ batch)
A standard puree process (95 °C / 5 min / 1500 rpm)
MIR spectrum acquisition on purees
(24 processed purees)
2 thinning conditions × 4 times × 3 lots
Thinning (Th+)
2019 Golden Delicious
All apples stored at 4°C for 1 month until starch regression
3 batches of 3 kg apples (3 kg × 3 batches) for each processing condition
MIR spectrum acquisition on apple homogenates
(3 apple homogenates)
3 batches, 10 apples/batch
Variability on puree processing
Puree processes (3 temperatures × 3 grinding levels)
70 °C / 30 min
82.5 °C / 30 min
95 °C / 30 min
300 rpm
1000 rpm
3000 rpm
MIR spectrum acquisition on purees
(27 processed purees)
3 temperatures × 3 grinding levels × 3 lots
Macroscopic imaging and rheological characterizations
Biological analyses: SSC, TA, malic acid, individual sugars and cell wall contents

**Figure 1**

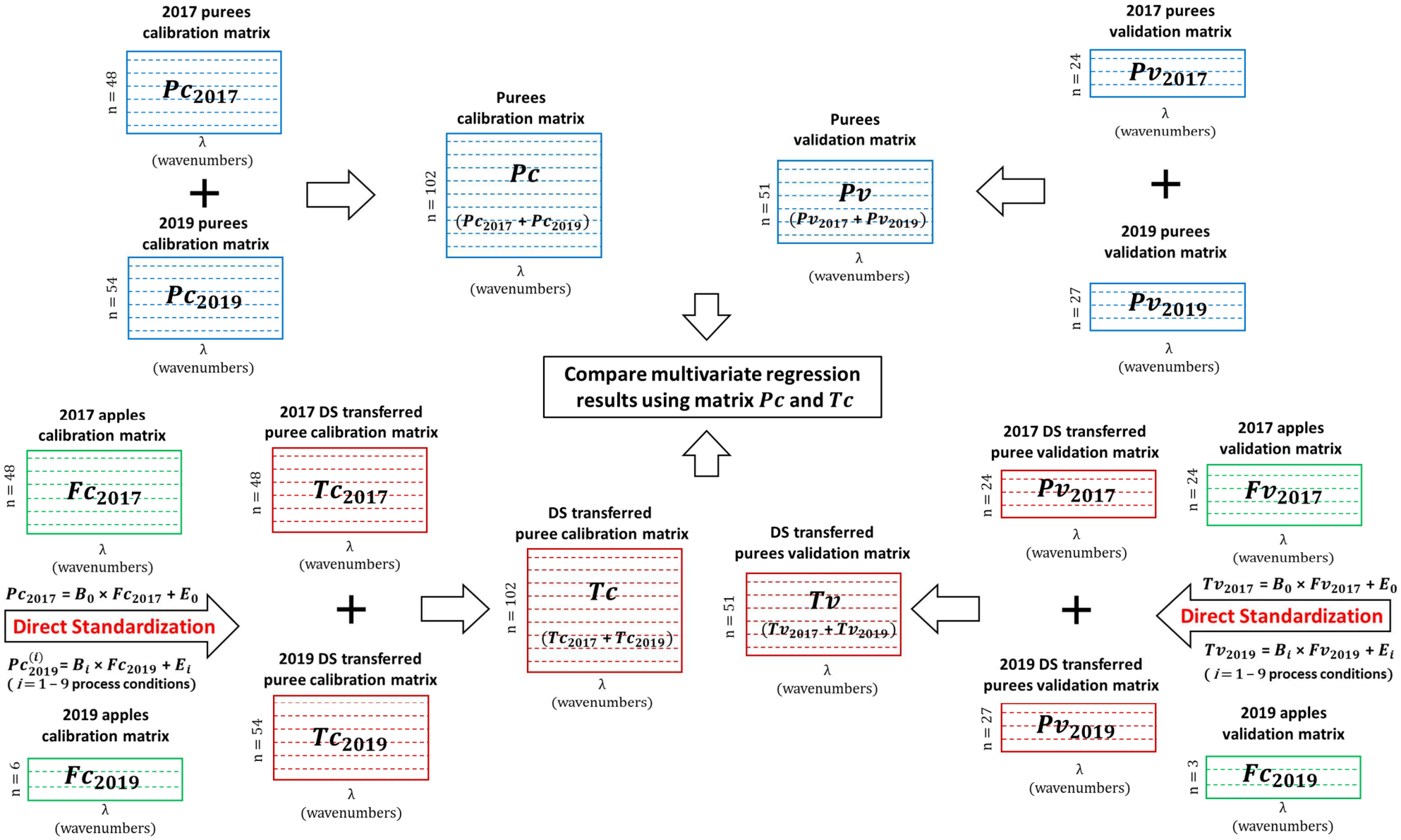
2017 purees calibration matrix
n = 48
$Pc_{2017}$
λ (wavenumbers)
+
2019 purees calibration matrix
n = 54
$Pc_{2019}$
λ (wavenumbers)
Purees calibration matrix
n = 102
$Pc$
$(Pc_{2017} + Pc_{2019})$
λ (wavenumbers)
Purees validation matrix
n = 51
$Pv$
$(Pv_{2017} + Pv_{2019})$
λ (wavenumbers)
2017 purees validation matrix
n = 24
$Pv_{2017}$
λ (wavenumbers)
+
2019 purees validation matrix
n = 27
$Pv_{2019}$
λ (wavenumbers)
Compare multivariate regression results using matrix $Pc$ and $Tc$
2017 apples calibration matrix
n = 48
$Fc_{2017}$
λ (wavenumbers)
$Pc_{2017} = B_0 \times Fc_{2017} + E_0$
Direct Standardization
$Pc^{(i)}_{2019} = B_i \times Fc_{2019} + E_i$
( $i = 1 - 9$ process conditions)
2019 apples calibration matrix
n = 6
$Fc_{2019}$
λ (wavenumbers)
2017 DS transferred puree calibration matrix
n = 48
$Tc_{2017}$
λ (wavenumbers)
+
2019 DS transferred puree calibration matrix
n = 54
$Tc_{2019}$
λ (wavenumbers)
DS transferred puree calibration matrix
n = 102
$Tc$
$(Tc_{2017} + Tc_{2019})$
λ (wavenumbers)
DS transferred purees validation matrix
n = 51
$Tv$
$(Tv_{2017} + Tv_{2019})$
λ (wavenumbers)
2017 DS transferred puree validation matrix
n = 24
$Pv_{2017}$
λ (wavenumbers)
+
2019 DS transferred purees validation matrix
n = 27
$Pv_{2019}$
λ (wavenumbers)
2017 apples validation matrix
n = 24
$Fv_{2017}$
λ (wavenumbers)
$Tv_{2017} = B_0 \times Fv_{2017} + E_0$
Direct Standardization
$Tv_{2019} = B_i \times Fv_{2019} + E_i$
( $i = 1 - 9$ process conditions)
2019 apples validation matrix
n = 3
$Fc_{2019}$
λ (wavenumbers)

**Figure 2**

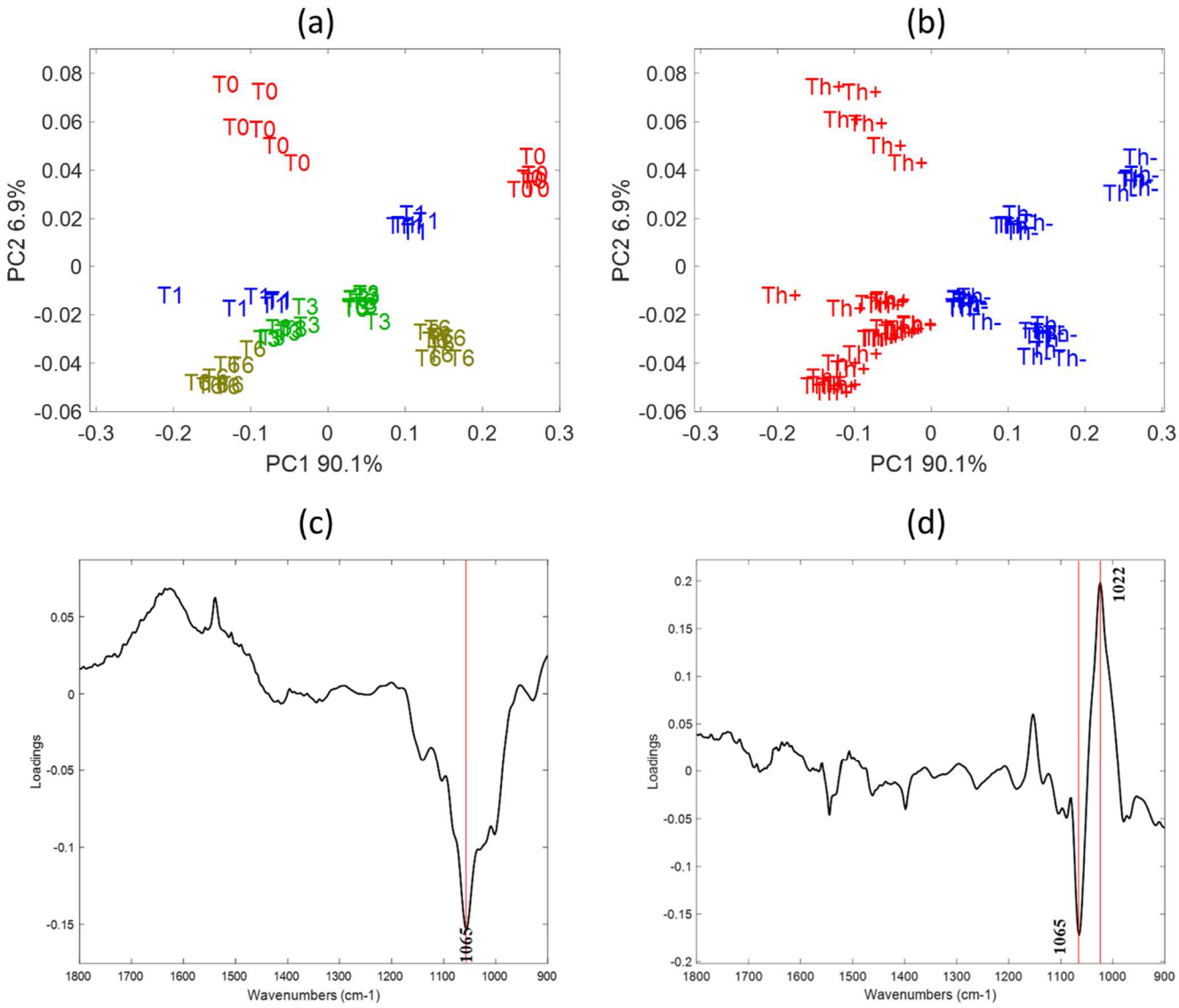


**Figure 3**

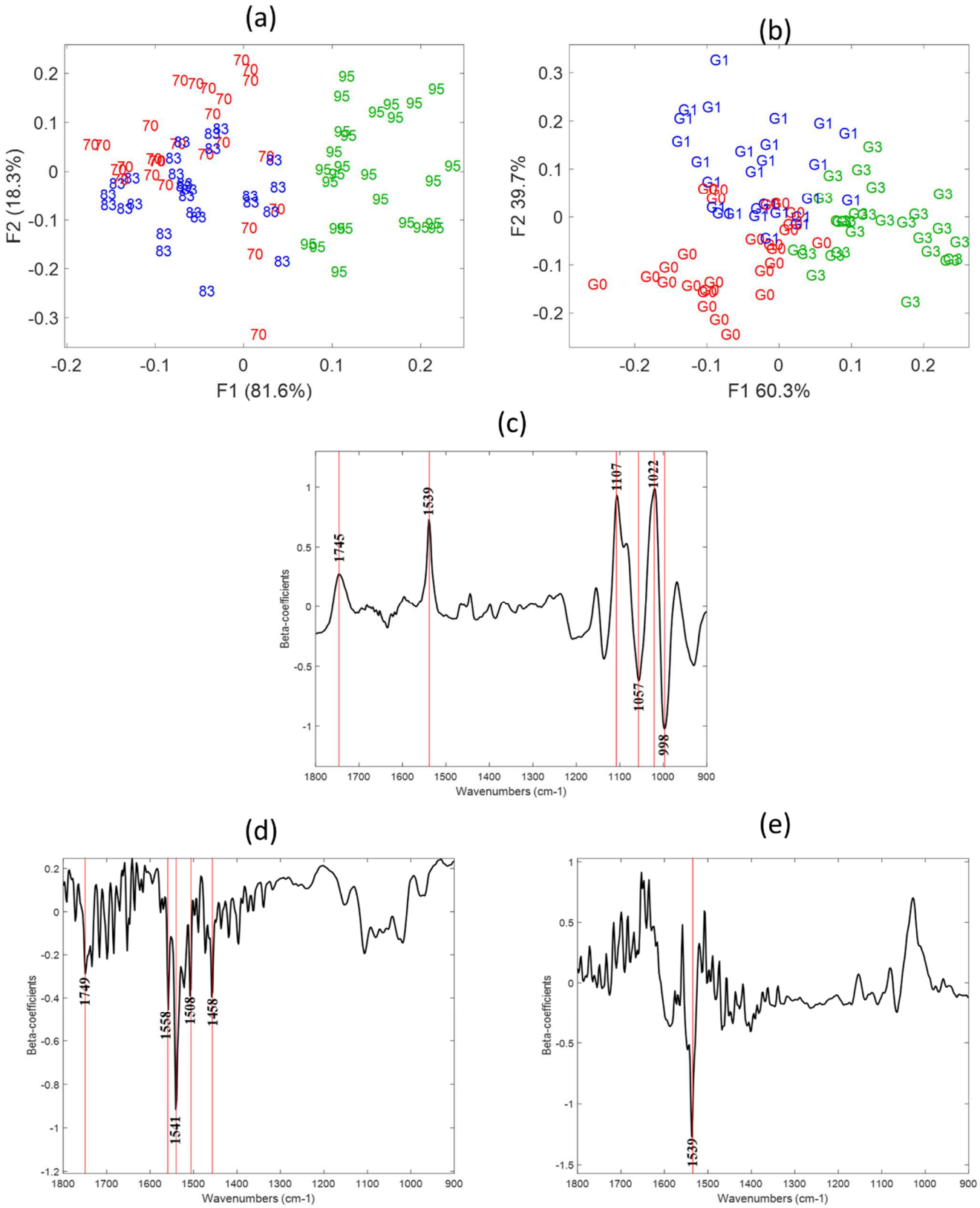
(a)
F2 (18.3%)
F1 (81.6%)
(b)
F2 39.7%
F1 60.3%
(c)
Beta-coefficients
Wavenumbers (cm-1)
1745
1539
1107
1022
1057
998
(d)
Beta-coefficients
Wavenumbers (cm-1)
1749
1558
1541
1508
1458
(e)
Beta-coefficients
Wavenumbers (cm-1)
1539

**Figure 4**

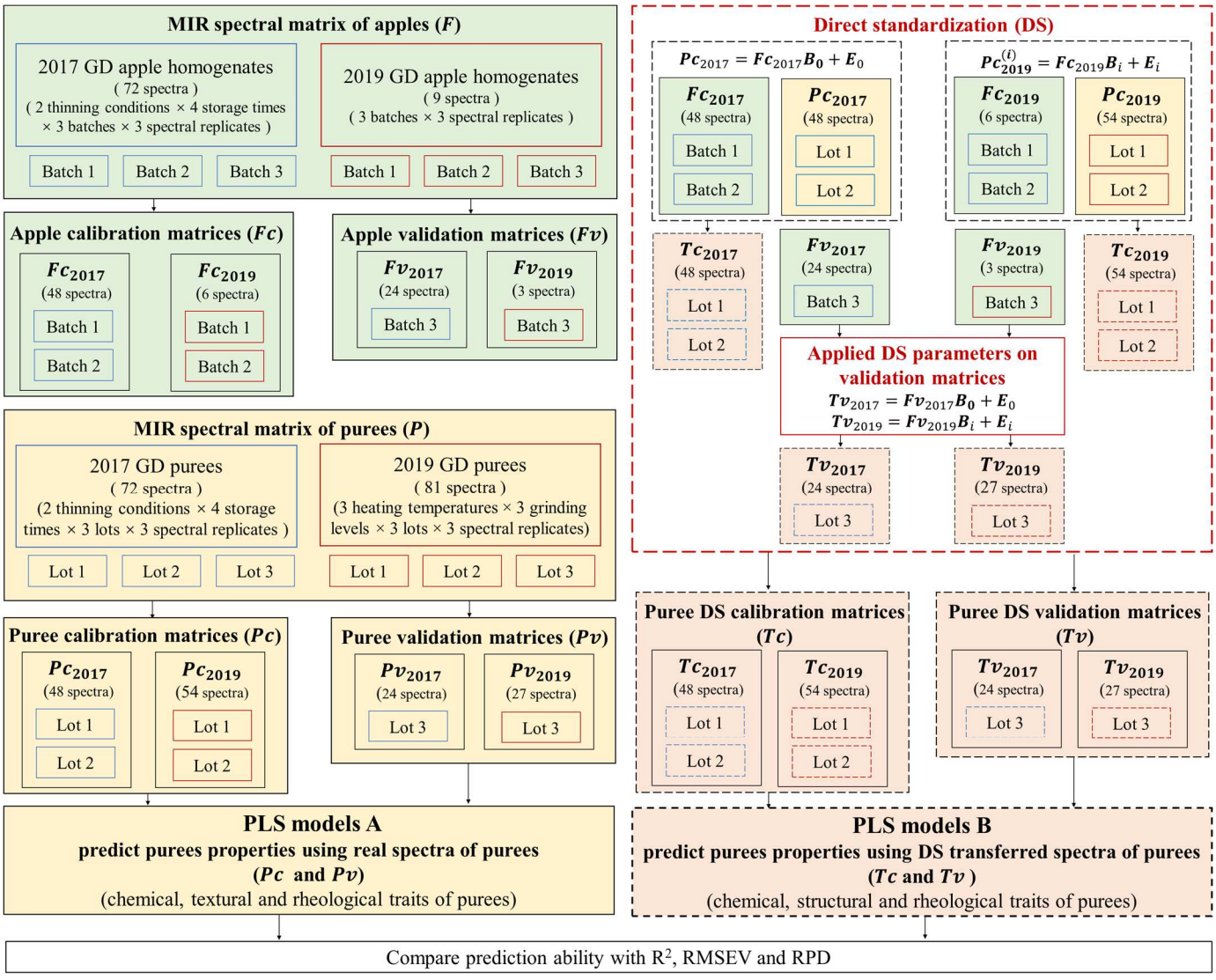
MIR spectral matrix of apples (F)
2017 GD apple homogenates
( 72 spectra )
( 2 thinning conditions × 4 storage times × 3 batches × 3 spectral replicates )
Batch 1
Batch 2
Batch 3
2019 GD apple homogenates
( 9 spectra )
( 3 batches × 3 spectral replicates )
Batch 1
Batch 2
Batch 3
Apple calibration matrices (Fc)
Fc2017 (48 spectra)
Batch 1
Batch 2
Fc2019 (6 spectra)
Batch 1
Batch 2
Apple validation matrices (Fv)
Fv2017 (24 spectra)
Batch 3
Fv2019 (3 spectra)
Batch 3
MIR spectral matrix of purees (P)
2017 GD purees
( 72 spectra )
(2 thinning conditions × 4 storage times × 3 lots × 3 spectral replicates )
Lot 1
Lot 2
Lot 3
2019 GD purees
( 81 spectra )
(3 heating temperatures × 3 grinding levels × 3 lots × 3 spectral replicates)
Lot 1
Lot 2
Lot 3
Puree calibration matrices (Pc)
Pc2017 (48 spectra)
Lot 1
Lot 2
Pc2019 (54 spectra)
Lot 1
Lot 2
Puree validation matrices (Pv)
Pv2017 (24 spectra)
Lot 3
Pv2019 (27 spectra)
Lot 3
PLS models A
predict purees properties using real spectra of purees (Pc and Pv)
(chemical, textural and rheological traits of purees)
Direct standardization (DS)
Pc2017 = Fc2017B0 + E0
Fc2017 (48 spectra)
Batch 1
Batch 2
Pc2017 (48 spectra)
Lot 1
Lot 2
Pc2019(i) = Fc2019Bi + Ei
Fc2019 (6 spectra)
Batch 1
Batch 2
Pc2019 (54 spectra)
Lot 1
Lot 2
Tc2017 (48 spectra)
Lot 1
Lot 2
Fv2017 (24 spectra)
Batch 3
Fv2019 (3 spectra)
Batch 3
Tc2019 (54 spectra)
Lot 1
Lot 2
Applied DS parameters on validation matrices
Tv2017 = Fv2017B0 + E0
Tv2019 = Fv2019Bi + Ei
Tv2017 (24 spectra)
Lot 3
Tv2019 (27 spectra)
Lot 3
Puree DS calibration matrices (Tc)
Tc2017 (48 spectra)
Lot 1
Lot 2
Tc2019 (54 spectra)
Lot 1
Lot 2
Puree DS validation matrices (Tv)
Tv2017 (24 spectra)
Lot 3
Tv2019 (27 spectra)
Lot 3
PLS models B
predict purees properties using DS transferred spectra of purees (Tc and Tv )
(chemical, structural and rheological traits of purees)
Compare prediction ability with R2, RMSEV and RPD

**Figure S1**

**Table 1** Prediction of biochemical, structural and rheological properties of apple purees using PLS regression based on their MIR spectra between 900-1800 $cm^{-1}$.

| Parameter | Range | SD | $R_c^2$ | $R_v^2$ | RMSEV | RPD | LVs | Linkable regions ($cm^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| η 50 | 0.57-2.28 | 0.39 | 0.91 | 0.87 | 0.19 | 3.2 | 7 | 1780-1788, 1718-1734, 1659-1678, 1636-1616, 1549-1558, 1157-1163 |
| η 100 | 0.26-1.22 | 0.27 | 0.89 | 0.85 | 0.07 | 3.0 | 8 | 1780-1788, 1718-1734, 1659-1678, 1636-1616, 1549-1558, 1157-1163 |
| AS-G' (Pa) | 1069-11154 | 2362 | 0.85 | 0.75 | 854 | 2.0 | 12 | / |
| AS-G" (Pa) | 210-2707 | 595 | 0.87 | 0.73 | 298 | 1.9 | 12 | / |
| yield stress | 8.7-131 | 27.1 | 0.86 | 0.81 | 12.8 | 2.1 | 11 | / |
| d 4:3 | 196-920 | 197 | 0.90 | 0.87 | 65 | 3.1 | 8 | 1780-1788, 1701-1713, 1649-1653, 1537-1541, 1101-1107, 1032-1043 1011-1026 |
| d 3:2 | 44-360 | 60.1 | 0.76 | 0.62 | 41.7 | 1.4 | 11 | / |
| AIS (DM) | 133.1-193.6 | 12.4 | 0.79 | 0.50 | 7.6 | 1.4 | 10 | / |
| AIS (FW) | 21.8-14.1 | 3.4 | 0.80 | 0.50 | 2.2 | 1.5 | 6 | / |
| DMC (g/g FW) | 0.15-0.23 | 0.02 | 0.87 | 0.84 | 0.01 | 2.9 | 5 | 997-1001,1051-1057, 1101-1109 |
| SSC (°Brix) | 12.6-18.6 | 1.9 | 0.91 | 0.86 | 0.6 | 3.1 | 5 | 997-1001,1051-1057, 1101-1109 |
| TA (mmol $H^+$/kg FW) | 5.1-73.5 | 25.5 | 0.89 | 0.86 | 7.6 | 3.2 | 6 | 1713-1709, 1105-1109, 1016-1018, 1074-1072, 1038-1042 |
| pH | 3.6-4.4 | 0.2 | 0.89 | 0.83 | 0.1 | 2.6 | 7 | 1713-1709, 1105-1109, 1016-1018, 1074-1072, 1038-1042 |
| malic (g/kg FW) | 2.4-7.0 | 1.2 | 0.88 | 0.85 | 0.4 | 2.7 | 6 | 1721-1709, 1105-1109, 1016-1018, 1074-1072 |
| fructose (g/kg FW) | 34.9-98.7 | 16.0 | 0.89 | 0.84 | 6.1 | 2.6 | 9 | 1709-1713, 1259-1265, 1105-1109, 1074-1080,1038-1042, 1016-1020, 970-974 |
| sucrose (g/kg FW) | 39.1-118.5 | 18.5 | 0.79 | 0.64 | 14.0 | 1.3 | 9 | / |
| glucose (g/kg FW) | 10.4-25.4 | 3.6 | 0.74 | 0.68 | 2.4 | 1.5 | 7 | / |

Note: Puree spectra and references data from 'Golden Delicious' apples, including variability of two different thinning conditions, cold storage (during 0, 1, 3 and 6 months), three heating temperatures (70, 83 and 95 °C) and three grinding levels (300, 1000, 3000 rpm). All results based on the SNV pre-treated MIR spectra at 900-1800 $cm^{-1}$. $R_c^2$: determination coefficient of the calibration set; $R_v^2$: determination coefficient of the validation set; RPD: the residual predictive deviation of validation set; the linkable regions based on the β-coefficients of PLS models with the RPD values higher than 2.5; "/" presented the unacceptable results with the RPD values lower than 2.5.

673 **Table 2** Prediction of biochemical, structural and rheological properties of apple purees using PLS regression based on their reconstructed MIR spectra of raw
674 apple homogenates between 900-1800 $cm^{-1}$.

| Parameter | Range | SD | $R_c^2$ | $R_v^2$ | RMSEV | RPD | LVs | Linkable regions ($cm^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| η 50 | 0.57-2.28 | 0.39 | 0.85 | 0.82 | 0.21 | 2.5 | 8 | 1720-1734, 1636-1614, 1556-1560, 1547-1533, 1506-1510, 1448-1470, 1157-1169 |
| η 100 | 0.26-1.22 | 0.27 | 0.86 | 0.83 | 0.10 | 2.6 | 9 | 1720-1734, 1661-1675, 1636-1616, 1549-1558, 1507-1512,1445-1468, 1157-1163 |
| d 4:3 | 196-920 | 197 | 0.89 | 0.84 | 76 | 2.6 | 9 | 1740-1745, 1701-1715, 1645-1659, 1583-1587, 1537--1541, 1508-1510, 1452-1470, 1100-1112 |
| DMC (g/g FW) | 0.15-0.23 | 0.02 | 0.87 | 0.84 | 0.01 | 2.6 | 6 | 1161-1165, 1101-1107, 1084-1090, 1051-1063, 989-1001 |
| SSC (°Brix) | 12.6-18.6 | 1.9 | 0.89 | 0.85 | 0.7 | 2.8 | 5 | 1101-1112, 1084-1090, 1051-1069, 997-1001 |
| TA (mmol $H^+$/kg FW) | 5.1-73.5 | 25.5 | 0.88 | 0.86 | 8.9 | 2.9 | 7 | 1715-1710, 1107-1113, 1082-1086, 1059-1063, 1038-1042, 1001-993 |
| pH | 3.6-4.4 | 0.2 | 0.84 | 0.79 | 0.1 | 2.1 | 8 | 1715-1709, 1105-1110, 1016-1018, 1074-1072, 1038-1042 |
| malic (g/kg FW) | 2.4-7.0 | 1.2 | 0.87 | 0.82 | 0.6 | 2.3 | 9 | 1713-1709, 1105-1109, 1080-1088, 1058-1064, 1016-1018, 1001-998 |
| fructose (g/kg FW) | 34.9-98.7 | 15.0 | 0.82 | 0.72 | 8.9 | 1.7 | 11 | / |

675 Note: Puree spectra and references data from 'Golden Delicious' apples, including variability of two different thinning conditions, cold storage (during 0, 1, 3
676 and 6 months), three heating temperatures (70, 83 and 95 °C) and three grinding levels (300, 1000, 3000 rpm). All results based on the SNV pre-treated MIR
677 spectra at 900-1800$cm^{-1}$. $R_c^2$: determination coefficient of the calibration set; $R_v^2$: determination coefficient of the validation set; RPD: the residual predictive
678 deviation of validation set; the linkable regions based on the β-coefficients of PLS models with the RPD values higher than 2.5; "/" presented the unacceptable
679 results with the RPD values lower than 2.5.

680

**Table S1** Biochemical, structural and rheological data of apple purees and ANOVA results.

| Fruit thinning | Storage periods | $\eta_{50}$ Pa.s | $\eta_{100}$ Pa.s | G' Pa | G'' Pa | Yield stress Pa | d 4:3 - | d 3:2 - | DMC g/g FW | SSC °Brix | TA mmol H+/kg FW | glucose g/kg FW | fructose g/kg FW | sucrose g/kg FW | malic acid g/kg FW | pH | AIS mg/g FW | AIS mg/g DW |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Th- | T0 | 1.28 | 0.77 | 3127.8 | 626.7 | 47.5 | 909.9 | 251.5 | 0.19 | 13.4 | 58.1 | 18.9 | 50.5 | 66.7 | 4.5 | 3.7 | 164.5 | 31.6 |
| | T1 | 1.13 | 0.70 | 1960.2 | 466.7 | 21.9 | 694 | 351.9 | 0.19 | 15.0 | 54.4 | 15.4 | 49.4 | 59.1 | 2.8 | 3.8 | 147.2 | 27.6 |
| | T3 | 0.87 | 0.55 | 1849 | 453 | 13.9 | 339.8 | 205.9 | 0.20 | 14.1 | 46.7 | 18.6 | 84.1 | 84.8 | 3.6 | 4.0 | 140.0 | 27.3 |
| | T6 | 0.92 | 0.50 | 1816 | 427 | 14 | 316.1 | 223.6 | 0.19 | 13.8 | 26.8 | 23.0 | 85.1 | 77.3 | 2.7 | 4.4 | 145.7 | 27.6 |
| Th+ | T0 | 1.75 | 0.97 | 3375.1 | 816.4 | 52.1 | 831.6 | 231.6 | 0.21 | 15.5 | 70.9 | 23.5 | 85.3 | 64.4 | 5.5 | 3.6 | 163.3 | 33.9 |
| | T1 | 1.54 | 0.94 | 2783.7 | 639.5 | 25.2 | 489 | 261.8 | 0.21 | 17.6 | 69.3 | 16.8 | 80.3 | 115.9 | 5.6 | 3.8 | 150.9 | 31.9 |
| | T3 | 1.25 | 0.70 | 2517.6 | 609 | 22.3 | 405.1 | 228.3 | 0.22 | 16.9 | 59.9 | 13.8 | 88.0 | 102.5 | 4.9 | 3.8 | 143.3 | 31.6 |
| | T6 | 1.60 | 0.88 | 3168.2 | 751.7 | 33.9 | 393.5 | 255.1 | 0.23 | 17.5 | 34.7 | 23.8 | 95.7 | 44.0 | 3.6 | 4.3 | 150.3 | 34.8 |
| Storage time | significance | *** | *** | *** | *** | *** | *** | *** | ** | ** | *** | *** | *** | *** | *** | *** | *ns* | * |
| | F-values | *20.4* | *13.6* | *24.7* | *15.0* | *72.8* | *216.1* | *41.5* | *6.6* | *8.3* | *279.4* | *74.7* | *76.2* | *38.5* | *13.8* | *436.3* | *1.8* | *4.3* |
| Fruit thinning | significance | *** | *** | *** | *** | *** | *** | * | *** | *** | *** | *ns* | *** | ** | *** | *** | ** | ** |
| | F-values | *138.2* | *61.5* | *67.3* | *91.5* | *29.6* | *47.2* | *5.5* | *157.7* | *115.7* | *176.9* | *1.3* | *187.8* | *13.8* | *57.9* | *48.8* | *15.9* | *10.3* |

Data expressed in fresh weight (FW) or dry weight (DW); values correspond to the mean of 3 puree replications (3 kg per replication). Raw apples were stored at 4°C: from harvest (T0) and during one (T1), three (T3) and six months (T6). Two conditions of fruit load during cultivation: non-thinning with 100% number of apples (Th-) and thinning with 50% number of apples (Th+) per tree. In grey, two way- ANOVA results obtained for Golden Delicious purees. ns, *, **, ***: Non significant or significant at $P < 0.05$, 0.01, 0.001 respectively.

686 **Table S2** Biochemical, textural and rheological data of apple purees and results of Kruskal-Wallis non-parametric test.

| Temperatures | Grinding speeds | $\eta_{50}$ | $\eta_{100}$ | G' | G'' | Yield stress | d 4:3 | d 3:2 | DMC | SSC | TA | glucose | fructose | sucrose | malic acid | pH | AIS | AIS |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| °C | rpm | Pa.s | Pa.s | Pa | Pa | Pa | - | - | g/g FW | °Brix | mmol H+/kg FW | g/kg FW | g/kg FW | g/kg FW | g/kg FW | | mg/g FW | mg/g DW |
| | 300 | 1.27 | 0.89 | 9629.8 | 2389.5 | 97.7 | 583.8 | 103.6 | 0.17 | 13.8 | 63.6 | 16.1 | 67.9 | 70.9 | 6.3 | 3.9 | 26.8 | 158.0 |
| 70 | 1000 | 1.42 | 0.91 | 3295.4 | 768.8 | 38.8 | 633.7 | 274.8 | 0.16 | 13.8 | 67.8 | 18.0 | 66.2 | 76.6 | 6.4 | 3.9 | 28.0 | 171.4 |
| | 3000 | 0.64 | 0.40 | 1111.3 | 215.6 | 10.2 | 353.5 | 207.6 | 0.17 | 14.4 | 69.1 | 17.3 | 65.5 | 86.0 | 6.6 | 3.9 | 28.1 | 165.1 |
| | 300 | 1.23 | 0.93 | 8437.5 | 2078.9 | 97.9 | 553.0 | 212.8 | 0.18 | 14.9 | 59.9 | 14.6 | 67.7 | 70.6 | 5.8 | 3.8 | 29.5 | 166.7 |
| 83 | 1000 | 1.38 | 0.87 | 3036.9 | 764.4 | 35.6 | 647.8 | 324.8 | 0.16 | 13.4 | 69.8 | 17.4 | 71.9 | 75.2 | 5.2 | 3.9 | 25.6 | 159.6 |
| | 3000 | 0.91 | 0.54 | 1312.2 | 259.9 | 11.8 | 297.4 | 192.4 | 0.17 | 14.8 | 70.3 | 16.6 | 66.8 | 76.1 | 5.7 | 3.9 | 28.2 | 162.6 |
| | 300 | 1.93 | 1.16 | 3708.2 | 1101.3 | 30.0 | 492.9 | 262.1 | 0.17 | 14.9 | 60.2 | 17.0 | 72.6 | 66.9 | 4.8 | 3.9 | 27.9 | 160.8 |
| 95 | 1000 | 1.44 | 0.84 | 1955.4 | 522.2 | 16.6 | 332.9 | 209.8 | 0.17 | 14.9 | 64.1 | 17.8 | 71.3 | 64.5 | 5.1 | 3.9 | 26.2 | 157.3 |
| | 3000 | 1.07 | 0.63 | 1399.4 | 362.2 | 14.0 | 206.4 | 153.2 | 0.17 | 14.8 | 62.0 | 18.9 | 69.2 | 65.8 | 5.4 | 3.8 | 26.2 | 154.1 |
| Temperature | significances | *ns* | *ns* | *ns* | *ns* | *ns* | * | *ns* | * | * | * | * | * | ** | ** | *ns* | *ns* | *ns* |
| | F-values | *4.1* | *1.3* | *0.9* | *0.4* | *1.5* | *7.0* | *1.2* | *6.3* | *6.8* | *8.4* | *6.1* | *8.9* | *9.1* | *10.2* | *2.1* | *2.3* | *4.2* |
| Grinding speeds | significances | ** | *** | *** | *** | *** | ** | ** | *ns* | *ns* | *ns* | *ns* | *ns* | *ns* | *ns* | *ns* | *ns* | *ns* |
| | F-values | *10.9* | *17.2* | *21.6* | *22.6* | *19.4* | *13.7* | *9.2* | *1.1* | *2.8* | *1.4* | *0.4* | *0.1* | *1.4* | *0.2* | *1.3* | *0.4* | *0.2* |

687 Data expressed in fresh weight (FW) or dry weight (DW); values correspond to the mean of 3 puree replications (3 kg per replication). Processing conditions variations were: three
688 heating temperatures at 70°C, 83°C and 95°C for 30 min, and three grinding speeds at 300, 1000 and 3000 rpm at each temperature. In grey, Kruskal-Wallis results obtained on Golden
689 Delicious purees. ns, *, **, ***: Non significant or significant at $P < 0.05$, 0.01, 0.001 respectively.

**Table S3** The results of sensitivity (in blue cells) and specificity (in yellow cells) from: (**a**) the FDA discrimination (4 factors) of three different heating temperatures; and (**b**) the FDA discrimination (4 factors) of three different grinding speeds.

(**a**)

| Temperatures (℃) | 70 | 83 | 95 |
|---|---|---|---|
| 70 | / | 76.67% | 100% |
| 83 | 83.33% | / | 100% |
| 95 | 100% | 100% | / |

(**b**)

| Grinding speeds (rpm) | 300 | 1000 | 3000 |
|---|---|---|---|
| 300 | / | 75.00% | 85.19% |
| 1000 | 76.92% | / | 87.10% |
| 3000 | 85.19% | 100% | / |