Heliyon 7 (2021) e05963

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Impact of sugar replacement by non-centrifugal sugar on physicochemical, antioxidant and sensory properties of strawberry and kiwifruit functional jams

L. Cervera-Chiner, C. Barrera, N. Betoret, L. Seguí *

Instituto Universitario de Ingeniería de Alimentos para el Desarrollo, Universitat Politècnica de València, Camino de Vera, s/n, Valencia 46022, Spain

ARTICLE INFO

Keywords:
Jaggery
Non-centrifugal sugar
Functional ingredients
Antioxidant properties
Food reformulation
Sugar replacement
Sustainable ingredients

ABSTRACT

Raw cane sugars have been claimed to be rich in natural phenolic compounds which, in contrast to refined sugar, may increase the nutritional value of foods and contribute to the development of healthier foods and diets. The use of non-refined cane sugars in food formulation seems an interesting option since they provide natural antioxidants with sucrose still being the major sugar present, minimizing the loss of technological properties. However, substitution of refined sugar could imply an undesired impact on physicochemical and sensory properties, conditioning consumer's acceptance. Functional jams (strawberry and kiwifruit) with a larger fruit to sugar ratio than conventional ones, in which white sugar was replaced by granulated jaggery (0, 15, 30, 45, 60 y 75 % w/w) were obtained. Impact of sugar replacement was assessed by evaluating physicochemical properties (moisture, water activity, pH, total soluble sugars, sugar profile (glucose, fructose, sucrose), and optical, rheological, mechanical and antioxidant properties). Sensory properties and microbiological stability were also determined. Jaggery improved the antioxidant properties of jams (total phenolic content, total flavonoid content, antiradical activity by the DPPH and ABTS methods), proportionally to the amount of cane sugar incorporated and more significantly in the case of kiwifruit. Other physicochemical properties were not significantly affected by jaggery, except for color. However, these differences were not crucial in the acceptability tests, since acceptance of jams containing jaggery was generally good, very good when intermediate replacement percentages were used. Conclusions of the present work suggest that granulated jaggery can be used to formulate sugar-rich food products such as jams in order to increase their nutritional value, with little impact on physicochemical properties and good consumer acceptance.

1. Introduction

Unrefined derivatives of sugarcane have gained interest in recent years due to their nutraceutical properties attributed to sugarcane phenolic content. Phenolic constituents naturally occurring in sugarcane have been proved to be present in several non-refined derivatives such as brown sugar, jaggeries, muscovado sugar or cane honey, in a concentration related to processing and degree of refining (Lee et al., 2018; Seguí et al., 2015). Particularly, the flavones tricin, apigenin, luteolin and their derivatives, as well the hydroxycinnamic acids caffeic, chlorogenic, and ferulic have been identified in significant amounts (Barrera et al., 2020; Duarte-Almeida et al., 2011; Duarte-Almeida et al., 2006). Tricin and apigenin are the most abundant phenolics in raw sugars, both considered important bioactive constituents of foods which postulate as nutraceuticals, antiproliferative and chemopreventive agents (Barrera et al., 2020). Despite the search for non-caloric and non-cariogenic alternatives to sugar and excess sugar intake concern in high consuming countries, white sugar continues to be an ingredient extensively used in the food industry due to its sweetening capacity but also to its technological properties (Harish-Nayaka et al., 2009; Payet et al., 2006). In fact, removal of sucrose from food formulation modifies not only sweetness and flavor, but also physicochemical properties and shelf life, for which replacement usually requires other actions such as the incorporation of additives, preservatives and/or changes in processing conditions. In some cases, only partial replacement is feasible due to the loss of technological properties.

The use of natural additives and ingredients is a current trend for the food industry, consumers showing an increasing interest for natural food and ingredients (Battacchi et al., 2020). In addition, reformulation of processed foods implies a unique opportunity to positively impact on people's health by improving the nutritional characteristics of foods which are regularly consumed. This addresses the FAO sustainable

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* Corresponding author.
E-mail address: lusegil@upvnet.upv.es (L. Seguí).

https://doi.org/10.1016/j.heliyon.2021.e05963
Received 21 August 2020; Received in revised form 20 November 2020; Accepted 8 January 2021

development goal “Ensuring healthy lives and promoting well-being for all at all ages” (FAO Sustainable Development Goals).

From a nutritional point of view, sugars contribute mostly to the energetic value of foods. The use of unrefined cane sugars in food formulation is an interesting option since they may provide natural antioxidants removed during the refining process, with sucrose still being the major sugar, for which loss of technological properties might be minimized. Among the different commercial alternatives, granulated jaggery has been proposed as the best substitutive to refined sugar due to dosing facility, richer phenolic profile, and good antioxidant properties (Seguí et al., 2015; Barrera et al., 2020). According to this, the use of non-centrifugal sugar in food formulation would imply a more nutritious product, but the partial or total substitution of refined sugar could involve an undesired impact on physical and sensory properties, and therefore condition consumer's acceptance.

Jams are fruit preserves with a high sugar content. These are prepared by boiling fruit pulp with sugar, pectin, acid and other ingredients (preservative, coloring and flavoring agents) to a reasonably thick gel consistency (Belovic et al., 2017; Peinado et al., 2012). Total soluble solids content of the finished jam should be between 60% and 65% or greater and the product should contain at least 45% fruit (CODEX, 2009). Nevertheless, the term “jam” is generally used in the literature to refer also to fruit preserves with lower sugar content when a healthier product is being formulated (low-caloric, dietetic, functional) (Basu et al., 2011; Belovic et al., 2017). Apart from fruit itself, sugar is the main ingredient of jams. In recent years, however, an increased health concern, together with a higher incidence of non-communicable diseases related to obesity and metabolic syndrome, have increased the demand for reduced or no-sugar added products, leading manufacturers to develop healthier alternatives to sugar-based foods. Accordingly, total or partial replacement of sucrose by non-caloric sweeteners or other sugars (e.g. fructose, isomaltulose, sucralose, sorbitol, steviosides) to obtain jams or spreads has been proposed in the literature (Basu et al., 2011; Basu et al., 2013; Peinado et al., 2012; Belovic et al., 2017).

The aim of the present work was to evaluate the impact of partial replacement of sucrose by granulated jaggery on strawberry and kiwifruit jams attributes. In addition to the use of jaggery sugar, jams were formulated with a larger amount of fruit in order to contribute to a reduced sugar content and increased dry matter, together with the consequent increase in the bioactive compounds naturally present in the fruit. Impact of replacement is evaluated in terms of physicochemical, including antioxidant, microbiological, and sensory properties.

2. Materials and methods

2.1. Raw materials

Strawberries (*Fragaria x ananassa*) and kiwifruits (*Actinidia deliciosa*) with an appropriate maturity level were purchased (7.8 ± 1.4 Brix degrees and a pH of 3.6 ± 0.2 in the case of strawberries, and 13.3 ± 0.4 Brix degrees and a pH of 3.40 ± 0.11 in the case of kiwifruits). Whole fruits were kept under refrigeration for no more than 4 days until jams manufacturing. Among the different non-refined alternatives commercially available, granulated jaggery (Ecuador OXFAM Intermón) was chosen due to its optimum physicochemical and antioxidant properties, as well as its dosing ease (Seguí et al., 2015; Barrera et al., 2020). To ensure proper consistency, apple pectin (Sigma-Aldrich, Spain) was added to the raw materials as required.

2.2. Jams manufacturing

Jams were prepared in a food processor (Thermomix®, Vorwerk). Fruits were washed and inedible parts (peduncle in strawberry and skin in kiwifruit) were removed. Next, fruits were ground at 2,000 rpm for 20 s, and sugar and pectin were added to the mixture. A 2:1 w/w instead of a 1:1 w/w fruit:sugar ratio reported in traditional artisan recipes was used in the formulation of the jams, since the focus was on obtaining healthier fruit jams with reduced sugar content, increased fruit content and improved antioxidant properties. Sugar added to the formula consisted of combinations of white sugar and granulated jaggery, replacement ranging from 0% to 75%. Pectin was added in different proportions based on preliminary tests (1% and 2% (w/w) for kiwifruit and strawberry, respectively. The mixture was then cooked at 100 °C with constant stirring (200 rpm), during 20 min. After cooking, the product was hot poured into previously sterilized glass jars that, once closed, were turned upside down to ensure the lid's sterilization and vacuum generation. After 5 min, the jars were turned over again until cooling down, and stored in darkness and at room temperature until further analysis.

2.3. Analytical determinations

All analytical determinations described in this section were performed at least in triplicate within 24 h after jams manufacturing. In addition, microbial counting was also performed after 1 and 3 months of storage.

2.3.1. Moisture (x_w), water activity (a_w), total soluble solids (Brix) and pH determination

Moisture content was calculated gravimetrically after vacuum drying for 7 days (60 °C and 133 mbar) a known amount of sample (AOAC (2000) 934.06 method). Water activity was measured with an accuracy of ±0.003 in a dew point hygrometer (AquaLab 4TE). Total soluble solids content given in Brix degrees were measured in an Abbe Atago NAR-T3 table refractometer, at 20 °C. pH was measured with a calibrated Mettler Toledo S20 SevenEasy™ pH-meter.

2.3.2. Sugar profile (sucrose, fructose, glucose)

Fructose, glucose and sucrose content were determined by liquid ion exchange chromatography in a 716 Compact IC Metrohm system with a Metrosep Carb column (250 mm long and 4.6 mm internal diameter) using NaOH 0.1 M as the mobile phase (1 mL min^{-1}). Chromatograms were read with the ICnet 2.0 software (Mehtrom Ltd., Herisau, Switzerland). Measurements were performed on filtered solutions (0.45 μm) prepared by diluting 0.1 g of sample in 500 mL of deionised water. Sugars were quantified using the corresponding calibration curves of glucose, fructose and sucrose standards (purity >99.5%, Sigma Aldrich), obtained in the concentration range between 2.5 and 50 ppm.

2.3.3. Antioxidant (AO) properties

AO properties were assessed by determining total phenols, flavonoids as well as DPPH and ABTS AO activities as described in Seguí et al. (2015). Total phenolic content of 1:10 (w/v) aqueous extracts of the fruit jams was measured using the spectrophotometric method of the Folin-Ciocalteu reagent. Absorbance measurements of the mixtures obtained at 760 nm were compared to a standard curve of gallic acid (purity ≥98%, Sigma-Aldrich) ranging from 0 to 500 ppm and results were expressed as mg of gallic acid equivalents per gram of dried product (mg GAE/g dw). Flavonoid content of 1:25 (w/v) aqueous extracts of the fruit jams was measured using the colorimetric method of aluminum chloride. Absorbance at 368 nm of the mixtures were compared to a standard curve of apigenin (purity ≥95%, Sigma-Aldrich) ranging from 0 to 300 ppm, and results were given in mg of apigenin equivalents per gram of dried product (mg AE/g dw). Phenols and flavonoids spectrophotometric measurements were performed on supernatant after centrifuging at 13, 000 rpm during 5 min (MiniSpin, Eppendorf AG, Germany).

AO activity assessed by the DPPH method consisted of measuring the ability of the jams extracts (70:30% methanol:water) to inhibit DPPH in a methanol solution. For this purpose, 1:20 (w/v) jams extracts were obtained and further centrifuged (13,000 rpm, 5 min). 100 μL of supernatant were mixed with 2.9 mL of a 0.06 mM solution of DPPH in methanol and kept reacting in darkness for 2 h. Then, absorbance was measured at 515 nm and results compared to a standard curve of DPPH (purity ≥95%, Sigma-Aldrich) ranging from 0 to 20 ppm and, according to the amount of DPPH present at the beginning of the reaction, expressed as mg of reduced DPPH per gram of dried product (mg DPPHred/g dw).

Measuring the AO by ABTS method involved diluting 90 μL of extract in 2.91 mL of a 7 mM ABTS and 2.45 mM potassium persulfate solution to which phosphate buffer had been added until 0.7 absorbance at 734 nm was achieved. Extracts were obtained by diluting 1 g of each jam in 20 mL (for replacement percentages between 0 and 15%) or 40 mL (for the rest of samples) of bidistilled water and centrifuging at 13,000 rpm for 5 min. Absorbance measurements obtained after 30 min reacting in darkness were compared to a standard curve of Trolox (purity $\geq$98%, Sigma-Aldrich) ranging from 0 to 800 ppm and results were expressed as mg of Trolox equivalents per gram of dried product (mg TE/g dw).

2.3.4. Optical properties

Color of fruit jams was measured using a Minolta CM 3600D spectrocolorimeter (Konica Minolta Sensing, Inc., Japan). Color coordinates of the CIEL*a*b* color space were obtained by reflectance from the absorption spectrum provided by the equipment in the 380–770 nm range with D65 illuminant and 10° observer. The readings were made on a black background and placing each sample in standardized size plastic cuvettes (37 × 50 × 22 mm).

2.3.5. Mechanical properties

Texture of jams was analyzed by means of a back-extrusion test carried out with the ANAME Texture Analyzer TA-XT2. The test consisted of compressing along 15 mm a fixed amount of sample placed in a plastic container with a 35 mm diameter flat-base plunger at 1 mm/s deformation rate. Characteristic parameters were obtained from the force vs. time curves (Peinado et al., 2016): the maximum force required to compress the sample or firmness (F_{max} in N) and the adhesiveness or area under the curve in the negative forces period (A in N·s).

2.3.6. Rheological properties

Rheological properties were obtained with a controlled stress rheometer (RheoStress 1, Haake). All measurements were carried out at 25 °C with a plate–plate geometry system and a 2 mm gap. Steady state tests were performed with a shear rate linearly ranging from 0 to 100 s^{-1} in 3 sweeps in order to eliminate thixotropy (Peinado et al., 2015). The Herschel-Bulkey model was applied to experimental data (Eq.1), so that the flow index (n, dimensionless), the consistency (k, in Pa·s^n) and the yield shear stress (τ_0, in Pa) were obtained.

$$\tau = \tau_0 + k \cdot \gamma^n \tag{1}$$

where τ is the shear stress (in Pa) and γ is the shear rate (in s^{-1}).

2.3.7. Sensory analysis

Prior to performing the sensory evaluation, recruited volunteers were fully informed in order to make an informed decision on their participation an give their informed consent.

Sensorial attributes of fruit jams were evaluated 24 h after their preparation. Each sample was evaluated by a semi-trained panel of 15 members aged between 25 and 50, which were identified as regular consumers of fruit jams and familiarized with sensory analysis. To prevent fatigue, each panelist tested 3 of the 6 formulations prepared for each kind of fruit. Samples were provided on white plastic glasses with a 3-digit random number code. Panelists were asked to evaluate acceptance of color, aroma, texture (spreadability), flavor, and global acceptance using a 9-point hedonic scale (1 = dislike extremely; 9 = like extremely). Likewise, specific basic flavors such as sweetness and acidity were also evaluated. To assess spreadability, salt free biscottes and a small plastic spoon were also provided. In addition, a section of comments was included in the sensory analysis sheet. Samples were provided at room temperature. Panelists were given room-temperature water to cleanse the palate between samples. Sensorial evaluation was carried out in individual booths with an ambient temperature of 25 ± 2 °C, placed in a tasting room at the facilities of the IUIAD (UPV).

2.3.8. Microbial counts

Yeast and molds and total aerobic mesophilic bacteria were evaluated in freshly prepared fruit jams (24 h) and in jams stored for 1 and 3 months at room temperature, by serial dilution in peptone water and further surface plating on Sabouraud Agar and Plate Count Agar (Scharlab), respectively. 2 g of jam were mixed with 18 mL of sterilized peptone water in a Stomacher, and serially diluted to 10^{-3}. 0.1 mL of the last two dilutions were plated and incubated at 30 °C (PSelecta, model Incudigt), 72 h for total aerobic mesophylls, and 3–5 days for yeasts and molds.

2.3.9. Statistical analysis

Differences among means was evaluated with Statgraphics Centurion XVI by performing analysis of variance (ANOVA) with a 95% confidence level.

3. Results and discussion

3.1. Moisture content (x_w), water activity (a_w), total soluble solids (Brix), pH and sugar profile (sucrose, fructose, glucose)

Moisture, water activity, Brix degrees and pH values of kiwifruit and strawberry jams as affected by the replacement of white sugar by granulated jaggery are given in Table 1. As expected, soluble solids content

Table 1. Moisture content (x_w), water activity (a_w), total soluble solids (Brix) and pH of kiwifruit (K) and strawberry (S) jams formulated with different percentage of replacement of granulated jaggery (0–75%). Mean value of three replicates ±standard deviation.

sample	x_w (g w/g total)	a_w	Brix	pH
K0	0.454 ± 0.003^c	0.92 ± 0.012^c	49.5 ± 0.15^b	3.58 ± 0.04^a
K15	0.4542 ± 0.0008^c	0.9254 ± 0.0007^c	49.6 ± 0.15^b	3.6 ± 0.10^{bc}
K30	0.425 ± 0.002^b	0.9127 ± 0.0004^b	52.3 ± 0.3^d	3.60 ± 0.02^a
K45	0.399 ± 0.003^a	0.9096 ± 0.0008^a	54.6 ± 0.10^e	3.63 ± 0.012^{ab}
K60	0.45 ± 0.02^c	0.9257 ± 0.0004^c	50.0 ± 0.3^c	3.69 ± 0.02^c
K75	0.489 ± 0.006^d	0.932 ± 0.002^d	46.67 ± 0.06^a	3.75 ± 0.010^d
S0	0.510 ± 0.005^c	0.933 ± 0.004^a	46.5 ± 0.6^{cd}	3.45 ± 0.012^a
S15	0.498 ± 0.003^b	0.935 ± 0.0010^a	46.73 ± 0.06^d	3.56 ± 0.02^b
S30	0.502 ± 0.003^b	0.9380 ± 0.0002^b	46.4 ± 0.3^{cd}	3.62 ± 0.03^b
S45	0.522 ± 0.003^d	0.9430 ± 0.0008^c	44.27 ± 0.06^a	3.70 ± 0.04^c
S60	0.510 ± 0.008^c	0.9420 ± 0.0007^c	45.6 ± 0.5^b	3.76 ± 0.06^{cd}
S75	0.490 ± 0.005^a	0.9410 ± 0.0006^c	46.0 ± 0.3^{bc}	3.82 ± 0.05^d

[abc] For each fruit, different letters in the same column indicate statistical significant differences with a 95% confidence level (p-value < 0.05).

(Brix degrees) were lower than in conventional jams due to the reduced proportion of sugar in the recipe. The addition of jaggery did not have a proportional impact on Brix degrees. Although a difference in the glucose and fructose contents coming from jaggery (Seguí et al., 2015) could be partially responsible for differences in the refractometric measurements, results variability could also be explained by differences in the raw materials and manufacturing process. Moisture content and water activity values were in the range of the published for similar products with reduced sucrose content (García-Martínez et al., 2002; Peinado et al., 2013). Although adding jaggery to the recipe slightly increased a_w in the case of strawberry, this was not appreciated for kiwifruit.

Sugar profile of jams is presented in Figure 1, where the amount of glucose, fructose and sucrose in each product is reported. Adding jaggery to the recipe was expected to increase the fructose and glucose content in the final product due to the higher content of inverted sugar in jaggeries (Seguí et al., 2015); however, it was observed that the amount of inverted sugar was higher in jams with a lower percentage of replacement, this being more evident in the case of kiwifruit. This result may be explained in terms of sucrose inversion occurring at lower pH. In fact, jams exhibited a slightly pH increase with the addition of granulated jaggery, which is in line with the remaining content of lime used in the manufacture of non-centrifugal sugar. As indicated, this was more significant for kiwifruit than for strawberry, which is consistent with the lower pH of the raw material. Nevertheless, differences between both fruit jams could also be attributed to the higher proportion of inverted sugar naturally present in kiwifruit as compared to strawberry (Wang et al., 2020; Akšić et al., 2019). In addition, sucrose inversion is also affected by initial inverted sugar content together with low pH (Andrews et al., 2002). On the other hand, it is also hypothesized that sucrose present in the refined sugar added to the recipe could be more susceptible to hydrolysis during jam manufacturing than sucrose present in jaggery. This could be explained by the fact that sucrose contained in jaggery is part of a mixture of sugars which results from previous hydrolysis reactions (ripening processes in the plant and sugar manufacturing process), it being less prone to hydrolysis during jam manufacturing. This effect would be more remarkable when the fruit used for jam manufacturing is more acid and has higher inverted sugar content, as in the case of kiwifruit.

3.2. Antioxidant (AO) properties

In the present research, granulated jaggery is used as a white sugar replacer with the aim of increasing nutritional properties of jams, due to the presence of specific phenolic constituents such as flavones (tricin, apigenin, luteolin and their derivatives) and hydroxycinnamic acids (caffeic, chlorogenic, ferulic) in non-centrifugal derivatives of sugarcane (Duarte-Almeida et al., 2011; Barrera et al., 2020). As expected, increasing the percentage of granulated jaggery in the jams increased both total phenol and flavonoid content as well as ABTS and DPPH AO activities (Figure 2). This improvement in the overall AO properties was particularly evident in the case of kiwifruit jams, difference that could be attributed to the naturally lower content in AO of kiwifruit (total phenols ranging from 0.492 to 1.31 mg GAE/g, according to Liu et al. (2019) as compared to strawberry (total phenols ranging from 1.6 to 2.5 mg GAE/g, as reported by Chaves et al. (2017). This is confirmed when comparing AO properties of control samples. Main impact was observed in the flavonoids content (particularly in kiwifruit jams), which is in accordance with the flavonoid content of sugarcane and jaggeries.

Improvement of the antioxidant properties of jams was also noticed when examining AO activities by the DPPH and ABTS methods. This was more significant in the case of kiwifruit, in which AO activity increased up to three times when replacing 75% of the white sugar by granulated jaggery (DPPH assay). In contrast, strawberry jams did not exhibit such an enhancement, it being even not noticeable by the DPPH assay in which AO activity almost remained the same. Together with phenols and flavonoid results, AO activity results suggest that improvement of AO properties of fruit jams formulated with granulated jaggery strongly depend on the type of fruit used as raw material. Strawberry is a fruit characterized by a high phenolic content exhibiting significant AO properties, for which including jaggery in the formulation of these jams did not have a significant impact, at least when an increased fruit to sugar ratio is used in jam formulation, as in the present work. It remains to be determined whether using a common 1:1 ratio for traditional jam manufacturing results in significantly increased AO activities not only for kiwifruit, but also for strawberry. Nevertheless, and despite not having such an impact on AO activities, strawberry jam experimented a significant increase in the phenols and flavonoids content when jaggery was used in the formulation (Figure 2). This result suggests that granulated jaggery phenolics are not as effective in scavenging free radicals (especially those of DPPH) as strawberry phenolics ones. This is in accordance with the literature (Rice-Evans et al., 1997), where it is reported that total AO activity of flavones and hydroxycinnamic acids (main phenolic compounds present in granulated jaggery) range from 1.4 to 2.1 μM and from 1.3 to 2.2 μM, respectively; whereas that of anthocyanidins (of which strawberries are an important source) range from 1.8 to 4.4 μM.

3.3. Optical, mechanical and rheological properties

Physical properties have a risk to be affected by sugar being replaced by non-centrifugal sugar, which needs to be evaluated. This is of relevance in the case of color, due to the intense brown coloration of jaggery, which can lead to consumer rejection by failing to identify the fruit used as raw material. L*, a* and b* values measured in the jams obtained confirmed the relevance of these color changes (Figure 3). Increasing jaggery content implied darker samples (decrease in the lightness value L*), as well as an evolution towards brownish hues. In kiwifruit jams, it being a green fruit (L* = 40.17 ± 0.02, a* = -1.557 ± 0.006 and b* = 30.70 ± 0.02 according to Benlloch-Tinoco et al. (2014), the browning implied a significant increase in a* coordinate. In strawberry jams, it being a red fruit (L* = 33 ± 3, a* = 20 ± 3 and b* = 8 ± 2 according to Contreras et al. (2008), the browning implied a significant decrease in

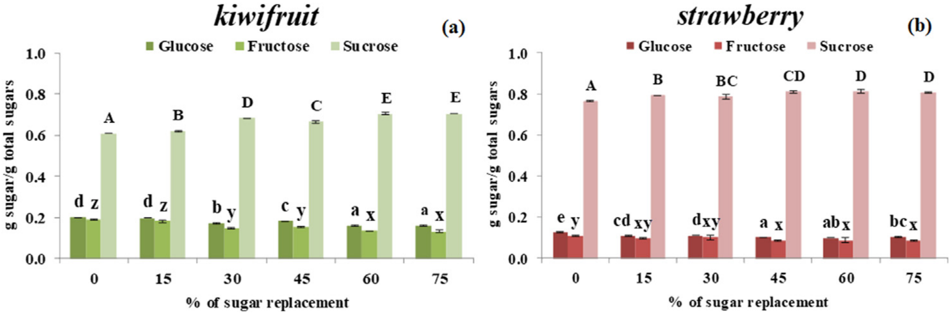

Figure 1. Sugar profile (g sugar/g total sugars) of kiwifruit (a) and strawberry (b) jams as affected by the replacement of white sugar by granulated jaggery (0–75%). Different letters for each series indicate statistically significant differences (p-value < 0.05): $^{ABC...}$ sucrose series; $^{abc...}$ glucose series; $^{xyz...}$ fructose series. Error bars represent the standard deviation of three replicates.

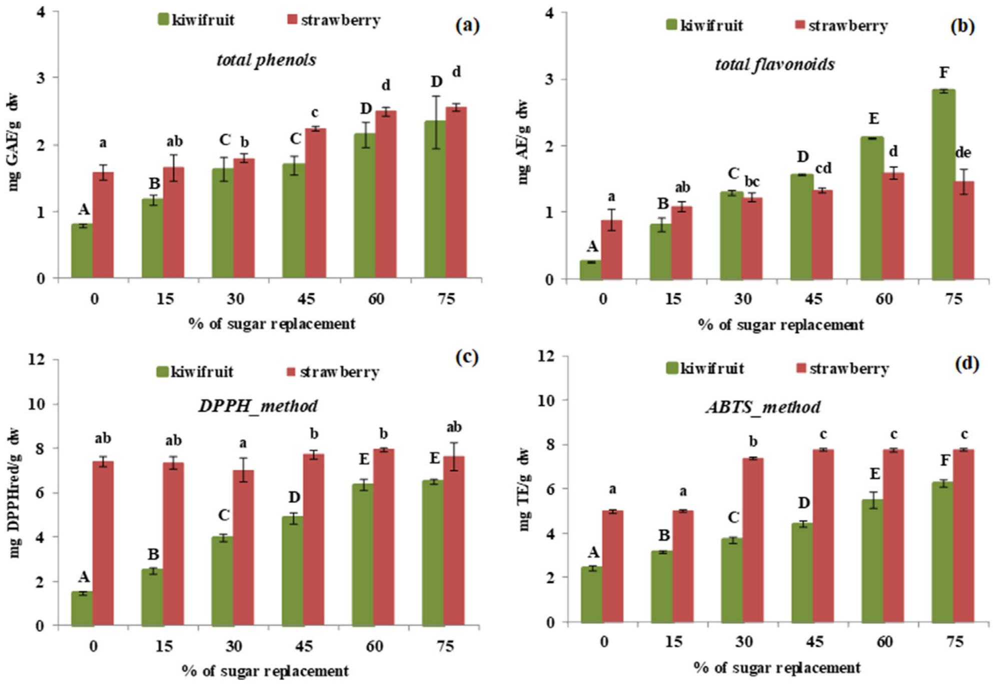

Figure 2. Total phenols (mg GAE/g dw) (a), total flavonoids (mg AE/g dw) (b) and antiradical activity measured by the DPPH (mg DPPHred/g dw) (c) and ABTS (mg TE/g dw) (d) methods of kiwifruit and strawberry jams as affected by the % of replacement of white sugar by granulated jaggery (from 0 to 75%). Different letters for each series indicate statistically significant differences (p-value < 0.05): $^{ABC...}$ kiwifruit series; $^{abc...}$ strawberry series. Error bars represent the standard deviation of three replicates.

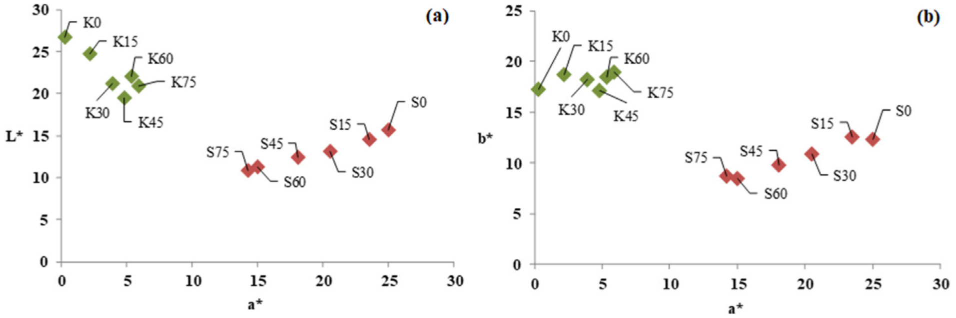

Figure 3. Colorimetric maps (a) L* vs. a* and (b) b* vs. a*, of kiwifruit (K) and strawberry (S) jams as affected by percentage (%) of replacement of white sugar by granulated jaggery (from 0 to 75%). The number after the K/S indicates percentage of replacement.

Table 2. Rheological (n, K and τ_0) and mechanical properties (F_{max} and A) of kiwifruit (K) and strawberry (S) jams formulated with different percentages of granulated jaggery (from 0 to 75%). Mean value of three replicates ±standard deviation.

sample	*rheological properties*				*mechanical properties*	
	n	K (Pa·s^n)	τ_0 (Pa)	R^2	F_{max} (N)	A (N·s)
K0	0.66 ± 0.18^{b}	5 ± 4^{a}	20 ± 5^{b}	0.876	0.44 ± 0.04^{b}	1.08 ± 0.06^{b}
K15	0.45 ± 0.06^{a}	8 ± 3^{a}	$8.6 \pm 1,5^{a}$	0.988	0.47 ± 0.02^{b}	1.21 ± 0.05^{c}
K30	0.54 ± 0.03^{ab}	8 ± 2^{a}	8.9 ± 0.6^{a}	0.997	0.48 ± 0.04^{b}	1.26 ± 0.05^{c}
K45	0.568 ± 0.006^{ab}	10.2 ± 0.3^{a}	10.5 ± 1.2^{a}	0.998	0.544 ± 0.006^{c}	1.46 ± 0.02^{d}
K60	0.38 ± 0.04^{a}	7.79 ± 1.09^{a}	15 ± 6^{ab}	0.923	0.48 ± 0.02^{b}	1.11 ± 0.03^{b}
K75	0.39 ± 0.02^{a}	5.9 ± 0.5^{a}	6.8 ± 1.3^{a}	0.912	0.396 ± 0.012^{a}	0.897 ± 0.014^{a}
S0	0.5011 ± 0.0004^{a}	16.4 ± 0.3^{a}	5.48 ± 0.09^{a}	0.996	0.47 ± 0.02^{a}	1.407 ± 0.005^{ab}
S15	0.490 ± 0.003^{a}	17.2 ± 1.4^{ab}	6.3 ± 0.5^{ab}	0.990	0.509 ± 0.013^{abc}	1.52 ± 0.04^{bc}
S30	0.523 ± 0.004^{b}	21 ± 2^{c}	9.1 ± 1.2^{c}	0.977	0.48 ± 0.03^{bc}	1.52 ± 0.08^{bc}
S45	0.52 ± 0.010^{b}	16.59 ± 1.07^{a}	6.0 ± 0.4^{a}	0.987	0.47 ± 0.02^{a}	1.36 ± 0.05^{a}
S60	0.5259 ± 0.00014^{bc}	19.93 ± 1.07^{bc}	7.98 ± 1.02^{bc}	0.984	0.53 ± 0.03^{c}	1.51 ± 0.03^{bc}
S75	0.534 ± 0.006^{c}	21.8 ± 0.9^{c}	9.3 ± 0.3^{c}	0.980	0.52 ± 0.03^{bc}	1.55 ± 0.10^{c}

^{abc}For each type of fruit, different letters in the same column indicate significant differences with a 95% confidence level (p-value < 0.05).

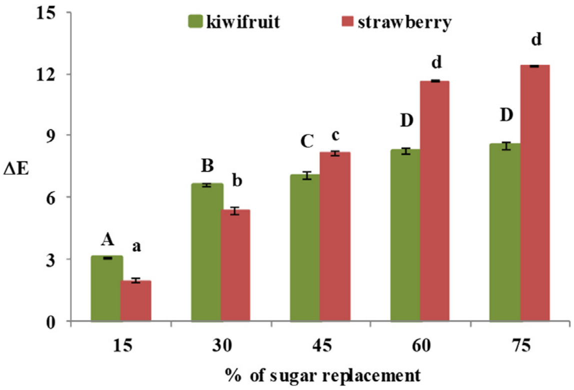

Figure 4. Color difference of kiwifruit and strawberry jams as affected by percentage (%) of replacement of white sugar by granulated jaggery (from 0 to 75%). Different letters for each series indicate statistically significant differences ($p\text{-value} < 0.05$): $^{ABC...}$ kiwifruit series; $^{abc...}$ strawberry series. Error bars represent the standard deviation of four replicates.

both a* and b* coordinates. Color differences (ΔE) are plotted in Figure 4. It was at low percentages of replacement that the color of kiwifruit jams was more significantly affected (15 and 30%), whereas strawberry jams experimented a more gradual color change, differences not being significant between 60% and 75% of replacement.

Mechanical and rheological properties (Table 2) were more determined by the fruit and the amount of pectin added to the formula than by jaggery addition. Although slightly higher for strawberry, mechanical properties (firmness and adhesiveness) of jams were of the same order for both fruits. Strawberry jams were formulated with twice the pectin than kiwifruit ones, but the amount of naturally occurring pectin or fibre in the fruits, as well as pH are factors responsible for mechanical properties. Compared to commercial products (dietetic and non-dietetic jams) analysed by García-Martínez et al. (2002), the jams obtained in the present work required less force to reach a given deformation. If consumer acceptance is low due to textural characteristics, it could be solved by adding more pectin and/or reducing the pH, which contributes to strengthen the network of crosslinked pectin molecules (Peinado et al., 2015).

Rheological results indicate that jams behaved as rigid bodies at low stresses ($\tau < \tau_0$) but as shear thinning or pseudoplastic non-Newtonian

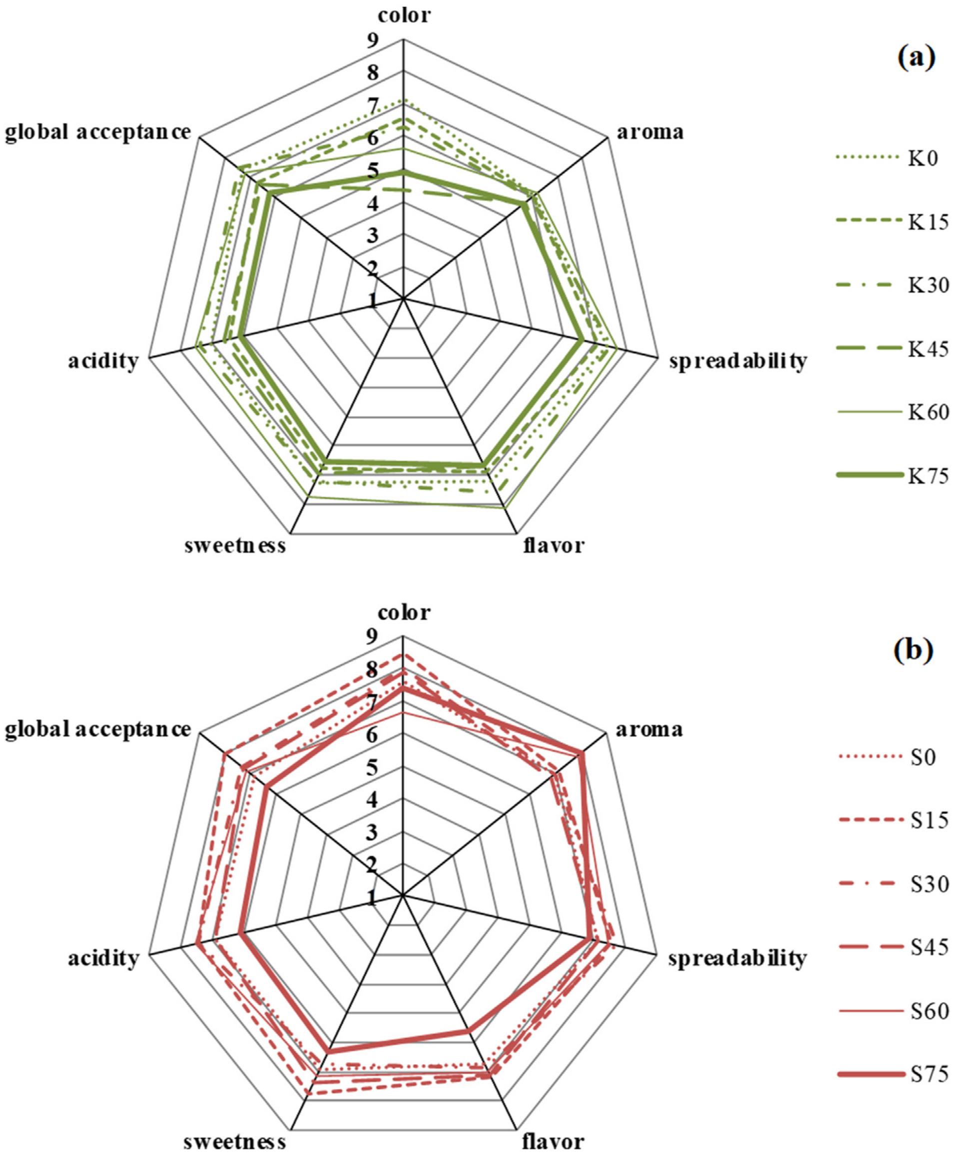

Figure 5. Sensory properties of kiwifruit (a) and strawberry (b) jams as affected by percentage of replacement of white sugar by granulated jaggery (from 0 to 75%). The number after the K (kiwifruit) or S (strawberry) indicates percentage of replacement.

fluids ($n < 1$) at high ones ($\tau \geq \tau_0$) and so their rheological behavior was well described by Herschel–Bulkley model ($R^2 \geq 0.9$). When evaluating rheological properties of fruit spreads, other authors have confirmed better fitting to Herschel–Bulkley model than others such as power law or Bingham (Sorour et al., 2016). Rheological parameters obtained showed significant differences among some samples, but no specific effect could be attributed to refined sugar replacement by granulated jaggery. Some measurements variability could be due to the presence of seeds, especially in the case of kiwifruit. When comparing percentages of replacement, the yield shear stress values generally indicate no differences in the samples spreadability or ability to be uniformly deformed and spread at the end use temperature (Basu et al., 2017). Consistency coefficients (k) were significantly higher for strawberry jams, which could be attributed to the higher pectin content of those samples.

3.4. Sensory evaluation

Results of the sensory evaluation of jams are summarized in Figure 5. With regard to the control samples, strawberry jam (S0) was preferred to kiwifruit one (K0). As deduced from the comments section, this could be due to tasters being more familiar with strawberry jams, along with the fact that they negatively scored the presence of seeds in the kiwifruit jam. According to the scores given, including jaggery in the formulation of jams results in products potentially acceptable by consumers. When compared to control samples, no specific relationship was found between percentage of replacement and acceptance of samples, but it was appreciated that medium percentages of replacement scored better than control for some attributes, while high percentages (75%) scored the worst for most attributes and both jams, although still acceptable.

The addition of jaggery had a better impact for strawberry than kiwifruit, mainly because of the color attribute, the sample with the highest replacement (K75) scoring 2 points below the control one (K0). Other attributes such as spreadability, sweetness, and acidity, along with global acceptance were only negatively affected at the highest replacement (75%), 60 and 45% scoring better than control ones. In contrast, tasters had a different perception regarding the impact of jaggery on strawberry jam, the addition of small amounts of jaggery improved color perception (S15 and S30), whereas the highest replacement (S75) was perceived quite similar to the control one. However, these results are in contrast with analytical determinations since color differences (ΔE) were more significant for strawberry jams (Figure 4).

In general, sensory attributes of strawberry jams with jaggery were positively appreciated by tasters, except for the highest percentage of replacement (75%). Some tasters indicated that jaggery adds toasted and liquorice notes to strawberry jams, improving their aroma. Contrarily, this was negatively evaluated in the case of kiwifruit. As for global acceptance, jams with intermediate percentages of replacement were the ones preferred.

3.5. Microbial stability

Total aerobic mesophylls, molds and yeasts were evaluated in freshly prepared fruit jams (24 h) and in jams stored for 1 and 3 months at room temperature. As expected, the thermal treatment proved effective and no microbial growth was detected in the jams ($<10^2$ cfu/g for molds and yeast and $< 10^3$ cfu/g). Microbial counts maintained at the previous levels along storage, except for mesophyll counts in strawberry jams in which counting increased to 10^4 cfu/g, probably due to their higher pH and a_w. Although this fact does not pose a risk to consumer, preservatives such as ascorbic acid or potassium sorbate could be added to the jam formulation to guarantee microbiological stability.

4. Conclusions

Replacing refined sugar by granulated jaggery in jams formulation has been proved to increase their antioxidant properties. It is therefore confirmed that reformulation of processed foods may have an impact on health by improving the nutritional characteristics of frequently consumed processed foods. An increase in the antioxidant properties has been achieved for both fruits, however, potential improvement strongly depends on the fruit used as the raw material. Intermediate percentages of replacement implied a significant increase in the antioxidant parameters evaluated, with little impact on the other physicochemical properties evaluated as well as on sensory attributes. Medium percentages of replacement were more appreciated than control ones, and some tasters valued the particular toasted and liquorice aroma notes provided by jaggery. On the contrary, the potential negative impact on color needs to be considered, especially in those fruits which are naturally less intense. It is concluded from the present investigation that it has been possible to reformulate jams to obtain products with improved functional properties by partially replacing white sugar by granulated jaggery, with little impact on technological and physicochemical properties and with good consumer's acceptance. The use of natural sustainable ingredients rich in natural bioactive compounds in food reformulation is confirmed as an opportunity to contribute to the concept of sustainable diets.

Declarations

Author contribution statement

L. Cervera-Chiner: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

C. Barrera, L. Seguí: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

N. Betoret: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Funding statement

This work was supported by Universitat Politècnica de València (PAID-06-10-2420) and Generalitat Valenciana (GV/2013/047), Spain.

Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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