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An eco-friendly approach for analysing sugars, minerals, and colour in brown sugar using digital image processing and machine learning

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ABSTRACT

Brown sugar is a natural sweetener obtained by thermal processing, with interesting nutritional characteristics. However, it has significant sensory variability, which directly affects product quality and consumer choice. Therefore, developing rapid methods for its quality control is desirable. This work proposes a fast, environmentally friendly, and accurate method for the simultaneous analysis of sucrose, reducing sugars, minerals and ICUMSA colour in brown sugar, using an innovative strategy that combines digital image processing acquired by smartphone cell with machine learning. Data extracted from the digital images, as well as experimentally determined contents of the physicochemical characteristics and elemental profile were the variables adopted for building predictive regression models by applying the kNN algorithm. The models achieved the highest predictive capacity for the Ca, ICUMSA colour, Fe and Zn, with coefficients of determination (R^2) ≥ 92.33 %. Lower R^2 values were observed for sucrose (81.16 %), reducing sugars (85.67 %), Mn (83.36 %) and Mg (86.97 %). Low data dispersion was found for all the predictive models generated (RMSE < 0.235). The AGREE Metric assessed the green profile and determined that the proposed approach is superior in relation to conventional methods because it avoids the use of solvents and toxic reagents, consumes minimal energy, produces no toxic waste, and is safer for analysts. The combination of digital image processing (DIP) and the kNN algorithm provides a fast, non-invasive and sustainable analytical approach. It streamlines and improves quality control of brown sugar, enabling the production of sweeteners that meet consumer demands and industry standards.

1. Introduction

Brown sugar is a minimally processed sweetener that contains a range of nutrients and phytochemicals naturally found in sugar cane (Weerawatanakorn et al., 2016). It is obtained through a thermal process that involves the evaporation and concentration of sugarcane juice (Jader et al., 2018). However, these processes can cause undesirable chemical reactions, such as non-enzymatic browning through the Maillard reaction and caramelization. These reactions are responsible for the dark colour of brown sugar and any changes in its organoleptic properties, such as colour, texture, and flavour, as well as its nutraceutical properties (Asikin et al., 2014; Takahashi et al., 2016). These changes can affect the perception of product quality and the consumer's purchasing decision (Alarcón et al., 2021). Consequently, alterations in the composition and physicochemical properties of brown sugar can provide valuable insights for quality control and the development of food and cosmetic products that utilise this sweetener as an ingredient.

Quality control is an essential step in ensuring the integrity and safety of the product. It provides information on the industrial processes used in certain food products, as well as technological procedures and raw materials. In addition, it helps prevent possible fraud (de Araújo

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Gomes et al., 2023). In order to ensure that food products comply with the standards established by legislation and meet consumer needs and requirements, various physicochemical characteristics are analysed by both the industry and inspection bodies. This analysis is conducted in accordance with the standards set out in legislation (BRASIL, 1978; Codex Alimentarius Commission, 2019; MAPA, 2018).

Conventional methodologies used in the sugar industry for quality control of brown sugar rely on off-line analytical techniques. These techniques require specific equipment and preparation steps for each physicochemical parameter evaluated (Codex Alimentarius Commission, 2019; Mehrotra & Siesler, 2003). However, these methodologies have several drawbacks, including the use of toxic reagents, generation of large volumes of waste, and being time-consuming and expensive (Meenu et al., 2021). As a result, they have a significant environmental impact. To overcome the limitations of traditional methods, non-destructive techniques can be an effective strategy for evaluating and selecting a more sustainable analytical approach. Currently, there is a growing trend towards assessing the environmental impact of analytical methods and adopting green metrics to verify the sustainability of each stage of the process or to guide the choice of method to be employed (Imam & Abdelrahman, 2023; López-Lorente et al., 2022; Shi et al., 2023). The application of Green Chemistry metrics to assess the environmental impact of a process is an interesting strategy when proposing new analytical methodologies. It facilitates the implementation of more sustainable practices, enables the comparison of different available methods and aids in selecting of the most eco-friendly option (López-Lorente et al., 2022).

In this context, it is essential to develop accessible analytical strategies that can reduce the costs associated with conventional analytical methods while being more environmentally friendly. Green Analytical Chemistry-based analytical methods, such as the use of digital images for pattern recognition and variable selection techniques for multivariate calibration, have been widely used for this purpose in various matrices. These methods comprise colour histograms extracted from digital images and hyperspectral images (Ballesteros et al., 2021; Dong et al., 2022; He et al., 2023; Macedo dos Santos-Tonial et al., 2023; Meenu et al., 2021; Tsakiridis et al., 2023).

Recent literature reports several studies that have employed the combination of digital image processing (DIP) or spectral data and machine learning (ML) algorithms. These studies produce mathematical models that correlate results from conventional analyses with samples or spectral images to create classification and prediction models that meet various needs (Alves et al., 2024; Cruz-Fernández et al., 2017; Perin et al., 2020; Schlesner et al., 2022; Schuastz Breda et al., 2024).

In a previous study conducted by our research group, models were developed to classify brown sugar into colour classes based on smartphone images and chemical features (Alves et al., 2024). This study demonstrated that the integration of digital image processing techniques, machine learning, and chemical composition analysis is a promising approach for the classification of brown sugar samples based on their colour (Alves et al., 2024). Nevertheless, studies focusing on the prediction of various physicochemical properties of brown sugar using smartphone images combined with machine learning algorithms have not yet been published in the literature. In the field of sugar quality control, predictive modelling has been limited to chemometric/machine learning tools in combination with spectroscopic measurements and/or hyperspectral images (de Almeida et al., 2018; Ramírez-Morales, Rivero, Fernández-Blanco, & Pazos, 2016; da Silva et al., 2022). The use of digital image processing methods is less common.

Pattern recognition algorithms are increasingly popular in the food sector due to their ability to solve complex food authenticity problems. The most widely adopted algorithms include support vector machine (SVM), k–nearest neighbours (kNN), classification and regression tree (CART), and random forest (RF), which are used in both classification and prediction models (Jiménez-Carvelo et al., 2019; Wu et al., 2008). Among these, the use of kNN is emphasised due to its advantages over other methods. It is less sensitive to outliers, can adapt to new data and has easy interpretation. Furthermore, it is effective in analysing high-dimensional data sets with small sample sizes and very attributes. It has a high capacity to deal with non-linear data, reduces overfitting and shows high adaptability and ability to learn complex patterns in the data, ensuring good prediction and generalization capabilities even with small sample sizes (Deng et al., 2016; Mahesh, 2020; Tan, Pang-Ning, Michael Steinbach, 2016; Zhang, 2020).

The kNN algorithm has been widely adopted for developing predictive models in various food matrices for different purposes. Davies et al. (2022) predicted the added sugar content of packaged products from information available on food labels, while Keramat-Jahromi et al. (2021) generated a prediction model between the moisture index obtained in a hybrid convective hot air dryer (EHD) and the colour attribute obtained from digital images of dehydrated date fruit chips.

This work presents the development of predictive models using the kNN algorithm to establish a correlation between the physicochemical and elemental composition of brown sugar and its digital images. The aim is to provide a more environmentally friendly quality control strategy for sugars, minerals, and colour for this type of sweetener. The greenness indices of the developed approach and of the conventional methods were evaluated using the greenness metric AGREE.

2. Material and methods

2.1. Sampling

During the 2017–2018 period, 34 samples of Brazilian brown sugar from various producers were obtained from commercial establishments. The samples were coded according to their state of origin and Arabic numerals in the order in which they were received in the laboratory. The samples were collected from various regions of Brazil, including the Midwest (n = 4), Southeast (n = 9), South (n = 19), North (n = 1), and Northeast (n = 1). Given that the South region represents the predominant centre of brown sugar production in Brazil, duplicate or triplicate batches were selected for sampling from four producers located in this region of the country. In other regions, the production of brown sugar tends to be more limited, conducted on a smaller scale with a seasonal focus, which presents a challenge in obtaining replicate batches. In view of this, for the other regions, a sample was taken from each producer.

2.2. Physicochemical and elemental composition

The brown sugar colour was determined using the international colour unit for sugar products, in accordance with the standard protocol of ICUMSA (International Commission for Uniform Methods of Sugar Analysis), as described by Alves et al. (2024). For this purpose, 0.5 g of the brown sugar sample was dissolved in 0.1 mol L^{-1} triethanolamine buffer. The mixture was vacuum filtered using 0.45 μm hydrophilic glass fibre membranes and then sonicated in an ultrasonic bath (Elmasonic P-30H; Analitica, Brazil). The absorbance was then measured at 420 nm using a UV–VIS spectrophotometer (SP2000 UV, Spectrum). Colour quantification was performed using the ICUMSA colour unit equation ($IU = absorbance \times \frac{1000}{b \times C}$). All tests were performed in duplicate.

Fructose, glucose, and sucrose were quantified using a High–Performance Liquid Chromatography system (HPLC–RID), adapted from Santos et al. (2016). First, 100 mg of the brown sugar samples were dissolved in 10 ml of acetonitrile (75 %, v/v). The resulting solution was then centrifuged for 10 min and filtered through a 0.22 μm PTFE membrane filter. An ultrapure Tracer Excel 120 APS silica column (5.0 μm, 250 mm × 4.6 mm) was used for sugar separation. The chromatographic system consisted of a quaternary pump and an RID refractive index detector (Gilson 132). The pump operated in isocratic mode with a mobile phase of acetonitrile: water 75:25 (v/v), and the flow rate was maintained at 1.0 mL min^{-1}. The composition and concentration of the sugars were determined by plotting the peak area against the concentration of the respective sugar standards (fructose, glucose and sucrose), and the results were expressed in g per 100 g of brown sugar. All tests were performed in triplicate.

The concentrations of zinc (Zn), iron (Fe), manganese (Mn), magnesium (Mg), and calcium (Ca) in the brown sugar samples were determined following the methodology described by dos Santos et al. (2019) without any modifications. The brown sugar samples (0.100 ± 0.01 g) were pretreated with an extraction solution of HNO_3 and H_2O_2 in a ratio of 60:40 % (v/v) under previously optimized and validated extraction conditions with a sonication time of 60 min. The samples were then analysed using a flame atomic absorption spectrophotometer (FAAS) (AA 220, Varian). All tests were performed in triplicate.

2.3. Digital images acquisition and processing

Approximately 100 g of brown sugar was placed evenly in a transparent plastic container. A total of 102 digital images were taken, with three photos taken for each brown sugar sample, which were compared with the values of each experimental measurement of the physicochemical characteristic being evaluated. As the instrumental measurements of the minerals and sugars were taken in triplicate, the photos of each brown sugar sample were also taken in triplicate. The photos were taken using an 8.0-megapixel cell phone camera (resolution: 1280 × 720 pixels) positioned 0.3 m away from the sample area to ensure measurement reproducibility. The photos were taken under ambient light and saved in jpeg format. The region of interest (ROI) was cropped digitally to a size of 200 x 200 pixels using the free license software Gimp® v. 2.10.32 (GIMP, 2022). No further image manipulation was required. The regions of interest (ROIs) were imported into the free software Chemostat® V. 2 (Helfer et al., 2015). The software converted the ROIs into intensity data for the red, green, and blue components of the RGB colour scale. These data were then used to generate a colour index histogram (R, G, B, H, S, V, L, and I). The colour data matrix generated, with the frequency of pixels for each colour tone per image, was used for creating the predictive models using the WEKA v. 3.8.6 code packages (Frank, Hall & Witten, 2016). The kNN algorithm was used to build regression models by selecting the relevant colour channels through a genetic filter. Fig. 1 illustrates the process of combining digital image processing with machine learning.

2.4. Modelling using the kNN algorithm

Prior to the modelling stage, the presence of outliers in the brown sugar physicochemical composition data was assessed by a *Grubbs* test at a 95 % confidence level using Minitab v. 16.2.1 software (MINITAB, 2010). Values identified as outliers were removed from the modelling. Next, variance inflation factor (VIF) analysis was applied to all the data using the R v. 4.2.1 software (Team R Core, 2022) to assess which parameters of colour index histogram (R, G, B, H, S, V, L, and I) did not have a significant multicollinearity effect on the regression predictors. The colour parameters with VIF values of less than 10.0 were used to build the calibration/training and validation/testing models.

Spearman's correlation analysis was utilized to determine the significance of the correlation between the R, H and S variables extracted from the images and selected by VIF analysis and the assessed physicochemical parameters (elements, sugars and ICUMSA colour). The data were divided into two exploratory multivariate analyses (Principal Component Analysis) based on significant linear correlations ($p < 0.05$). The scaling data used an index based on the sum of the colour matrix parameters extracted from images, as well as the physicochemical and elemental compositions for each of the brown sugar samples. The classes were divided into C1 – C6, based on the increasing intervals of this index.

The kNN algorithm used to predict the contents of the components (ICUMSA colour, reducing sugars, sucrose, and elements such as Ca, Mg, Fe, Mn, and Zn) in brown sugar was run using the WEKA code packages v. 3.8.6. For the modelling, the data were separated into training/calibration (70 % of the data, $n = 71$) and verification/test (30 % of the data, $n = 31$) sets. Cross-validation was applied to the data set using the k-fold method where $k = 10$. The performance of the predictive models was measured by the following metrics: coefficient of determination (R^2), root mean square error (RMSE) and mean absolute error (MAE) (Equations (1) – (3). The evaluation metrics were automatically generated by the software WEKA v. 3.8.6.

$$R^2 = \left\{ \frac{\sum_{i=1}^{n}\left(0_i - \overline{0}\right)\left(P_i - \overline{P}\right)}{\sqrt{\sum_{i=1}^{n}\left(0_i - \overline{0}\right)^2}\sqrt{\sum_{i=1}^{n}\left(P_i - \overline{P}\right)^2}} \right\}^2 \quad (1)$$

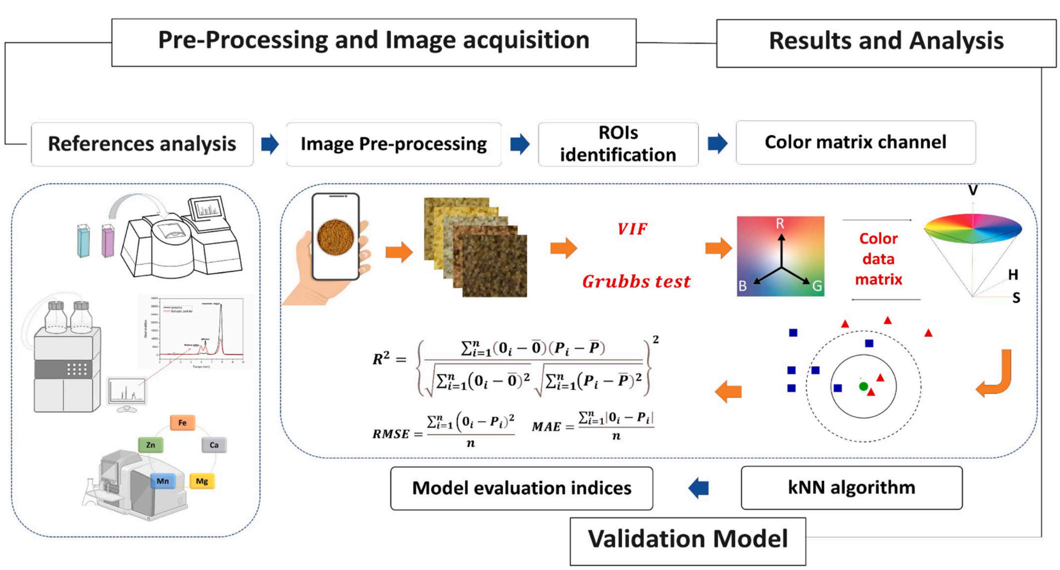


Fig. 1. Diagram illustrative of the adopted approach for acquiring and processing of the digital images of brown sugar samples, as well as predictive models generated using the kNN algorithm.

$$RMSE = \frac{\sum_{i=1}^{n}(0_i - P_i)^2}{n} \tag{2}$$

$$MAE = \frac{\sum_{i=1}^{n}|0_i - P_i|}{n} \tag{3}$$

These metrics are commonly used to evaluate the performance of regression models, where 0_i are the actual observed values, P_i are the values estimated by the model, O¯ is the mean of the actual observations, P¯ is the mean of the observations predicted by the model, and n is the number of data points.

3. Results and discussion

3.1. Relation of information's extracted from images and the composition of brown sugar

Brown sugar has a wide range of shades, ranging from light yellow/amber to dark brown. This variability in its colour can be attributed to several factors, such as the chemical composition of the sugar cane, production, and processing practices, among others (Alves et al., 2024). Therefore, it is important to consider the association of these parameters, starting with the appropriate selection of colour channels, which are obtained by converting images into colour spaces. In this system, each parameter contains information that contributes to describing the predictive response. Therefore, the VIF analysis selected the colour parameters R, H, and S from the colour index histogram (R, G, B, H, S, V, L, and I) as the best descriptors for predicting the concentration of physicochemical parameters evaluated in brown sugar samples from the digital images acquired by smartphone.

A Principal Component Analysis (PCA) was conducted to identify relationships between the colour of brown sugar samples, specifically the R, H, and S colour channels, and their composition (physicochemical and elemental). Fig. 2a displays the principal components PC1 and PC2, which account for 64 % of the variability in the data. These variables were selected based on the results of Spearman linear correlation (data not shown).

Fig. 2a shows a direct correlation between the ICUMSA colour and all investigated elements. Additionally, an inverse correlation is observed between the physicochemical composition and the R and H colour channels, as demonstrated by PC1. As the values of the Hue parameter, which describes the dominant spectral colour component, and the red colour (R parameter) increase or decrease, the ICUMSA colour tends to exhibit an antagonistic behaviour. The impact of Ca, Fe, and Zn contents on the colour of brown sugar is widely acknowledged in the literature. These elements can form complexes or pigments that give the sugar its characteristic colour, or act as catalysts in the hydrolysis of sucrose, leading to an increase in the content of reducing sugars. These reducing sugars are precursors to the non-enzymatic browning reactions of sugar (Alloway, 2008; Alves et al., 2024; Swietlik, 2010). Furthermore, the presence of Ca, Mg, and Fe elements may indicate contamination during brown sugar processing from the sugar 'liming' stage or sugar cane milling. This can result in an excessive incorporation of these elements into the final product, which gives the brown sugar a darker colour (Alves et al., 2023, 2024).

It is important to note that current national and international quality control legislations do not mandate the determination of element levels in brown sugar. Nevertheless, the role that elements play in the development of colour in this sweetener indicates that this should be considered a recommended practice in the quality control of brown sugar (Alves et al., 2024; Bento, 2009; Clarke et al., 1997; Feron & Groten, 2002; Riffer, 1988). Moreover, an understanding of the elemental composition of brown sugar can provide a competitive advantage in the sweetener market. This underscores the product as a nutrient-rich alternative, thereby indicating superior quality and nutritional value in comparison to other sweeteners, such as refined white sugar.

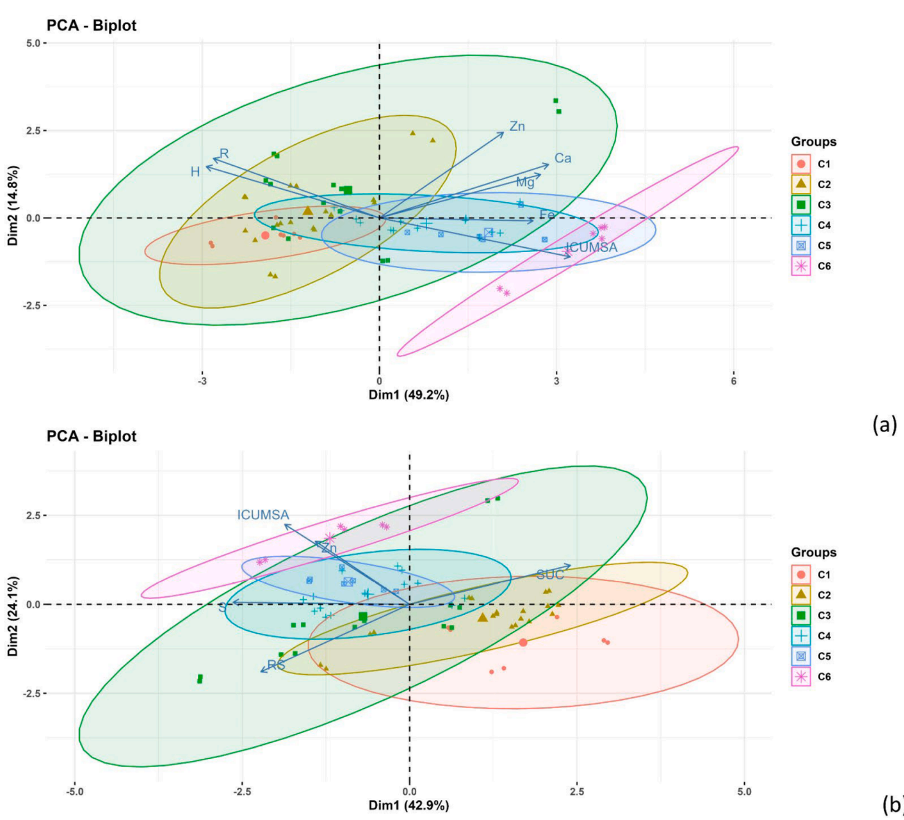


Fig. 2. Projection of physicochemical variables and brown sugar samples onto principal components PC1 and PC2: (a) R and H colour channels with ICUMSA colour, Ca, Fe, Zn, Mg, and (b) S colour channel with ICUMSA colour, sucrose, reducing sugars and Zn, respectively.

PC1 and PC2 together account for 67 % of the variability in the data for the S parameter in relation to ICUMSA colour, sucrose, reducing sugars, and Zn (Fig. 2b). A direct correlation was found between the S parameter and ICUMSA colour, reducing sugars, and Zn levels, while an inverse correlation was found between this parameter and sucrose. Based on these results, it seems that the S parameter, which indicates the saturation of colour the brown sugar samples, is more relevant in correlating with the physicochemical composition of this sweetener. This parameter may be supporting the colour variation from yellow/amber to dark brown, which are the most common colours in commercial brown sugars (Shrivastav et al., 2016).

It is widely accepted that reducing sugars are one of the main precursors of brown sugar colouration resulting from thermal processing (Bento, 2009; Riffer, 1988). When heated, the solution of reducing sugars can form high molecular weight pigments called melanoidins through the Maillard reaction between amino acids and carbonyl compounds (Bento, 2009). Variations in the concentration of these compounds, as well as the time and temperature used during the processing of brown sugar, can affect its final colour. The same pattern can be observed with Zn, which plays a crucial role in various complex reactions, including carbohydrate metabolism, photosynthesis, protein synthesis, and enzyme system structure (Alloway, 2008). This metal may also participate in Maillard reactions, leading to the browning of the product through catalysis, such as the sucrose hydrolysis reaction (Swietlik, 2010).

3.2. Predictive models for the quality control of brown sugar using DIP–kNN algorithm

In this study, the association between brown sugar colour and its physicochemical characteristics was investigated as a strategy to develop predictive models for sucrose, reducing sugars, ICUMSA colour, and certain elements (Ca, Mg, Fe, Mn, and Zn). This was achieved by combining the information extracted from the images with the supervised machine learning algorithm kNN.

The performance metrics of the predictive models generated by the kNN algorithm for the physicochemical parameters in relation to the colour matrix can be seen in Table 1, while the scatter plots of the values observed by conventional methods versus the values predicted by the predictive models, are illustrated in Fig. 3. Among these metrics, R^2 is the correlation between the predicted value and the true value of the model, where a value > 80 % indicates an adequate model. Meanwhile, RSME indicates the predictive ability of the model for unknown samples, where obtaining a low value is preferable (Tian et al., 2022).

The generated predictive models exhibited high coefficients of determination (R^2) values, with 100 % for the calibration/training model and ≥ 81.16 % for the testing/validation model, and low data scatter (RMSE < 0.235). This demonstrates a good linear agreement between the values predicted by DIP–kNN strategy and those determined by conventional methods. The models generated by the kNN algorithm achieved higher prediction values for the variables Ca (R^2 = 97.67 % and RSME = 0.139), ICUMSA colour (R^2 = 94.01 % and RSME = 0.154), Fe (R^2 = 92.33 % and RSME = 0. 191) and Zn (R^2 = 95.32 % and RSME = 0.045) (Fig. 3). This indicates the robustness of the predictive models and the good accuracy of the kNN algorithm in predicting the concentrations of the elements and of physicochemical properties of the brown sugar samples. These performance metrics were similar to those reported in the literature by other researchers for the determination of Brix, polarization (POL), and sucrose contents in sugar by combining hyperspectral imaging or spectroscopy with multivariate/machine learning techniques (de Almeida et al., 2018; Henrique da Silva Melo et al., 2022). However, in this work, greater scatter was observed in the testing/external validation phase for sucrose and reducing sugar contents (Fig. 3), indicating a lower predictive capacity of the model for these physicochemical parameters. This variability can be attributed to the lower correlation of sucrose and reducing sugars with brown sugar colour in comparison to the other compounds assessed (elements and ICUMSA colour) in this work. It is concluded that this behaviour may be due to the interaction, to a greater or lesser extent, of sucrose and reducing sugars with other components that influence the final colour of brown sugar. Examples include the sucrose degradation reaction, the formation of melanoidins, the complexation of metal ions, among others (Asikin et al., 2014; Eggleston et al., 2021; Lindeman & O'Shea, 2004; Raes et al., 2014).

The Codex Alimentarius Commission (2019) does not have specific analytical methods for the determination of the physicochemical and elemental composition of brown sugar. Instead, the document recommends general analysis methods, such as colour analysis using the spectrophotometric method. Although there is no specific method for determining sucrose and reducing sugars in brown sugar, a widely accepted analytical method is high-performance liquid chromatography (HPLC) (Jalaludin & Kim, 2021). Fehling's titrimetric method is a commonly used technique for determining reducing sugars (IAL. Instituto Adolfo Lutz, 2008). However, the methods used to determine the degree of polarization and Brix are typically associated with the analysis of white or refined sugar, rather than brown sugar (MAPA, 2018). Our work highlights the need to explore the use of digital image processing combined with chemometric tools/machine learning in the analysis of sweeteners such as brown sugar. This approach offers new perspectives for research and provides a single analytical strategy for quality control of a wide range of brown sugar samples, making it suitable for application in industrial environments.

3.3. Environmental impact assessment of the DIP–kNN strategy

The greenness profile of conventional brown sugar quality control methods with the predictive models generated by the DIP–kNN strategy using the AGREE metric were compared. The literature commonly uses greenness metrics such as NEMI, modified NEMI, GAPI, Eco-scale, AMVI, AMGS, AGREE and AGREEprep (Imam & Abdelrahman, 2023; Kannaiah et al., 2021; Kowtharapu et al., 2023; Sajid & Płotka-Wasylka, 2022; Shi et al., 2023). The AGREE greenness metric was selected due to its simplicity, informative and easy-to-understand results, and superior performance compared to other metrics described above (Pena-Pereira et al., 2020).

Table 1
Performance metrics for training and testing models for physicochemical and elemental composition analysis of the brown sugar.

Parameters	Training/Calibration model	Testing/Validation model		
	R^2 (%)	*R^2 (%)*	*RMSE (%)*	*MAE (%)*
Mn (mg 100 g^{-1}; n = 3)	100.0	83.36	0.210	0.106
Zn (mg 100 g^{-1}; n = 3)	100.0	95.32	0.045	0.022
Fe (mg 100 g^{-1}; n = 3)	100.0	92.33	0.191	0.110
Mg (mg 100 g^{-1}; n = 3)	100.0	86.97	0.225	0.085
Ca (mg 100 g^{-1}; n = 3)	100.0	97.67	0.139	0.064
ICUMSA colour (I.U; n = 2)	100.0	94.01	0.155	0.091
Sucrose (g 100 g^{-1}; n = 3)	100.0	81.16	0.021	0.014
Reducing sugars (g 100 g^{-1}; n = 3)	100.0	85.67	0.235	0.125

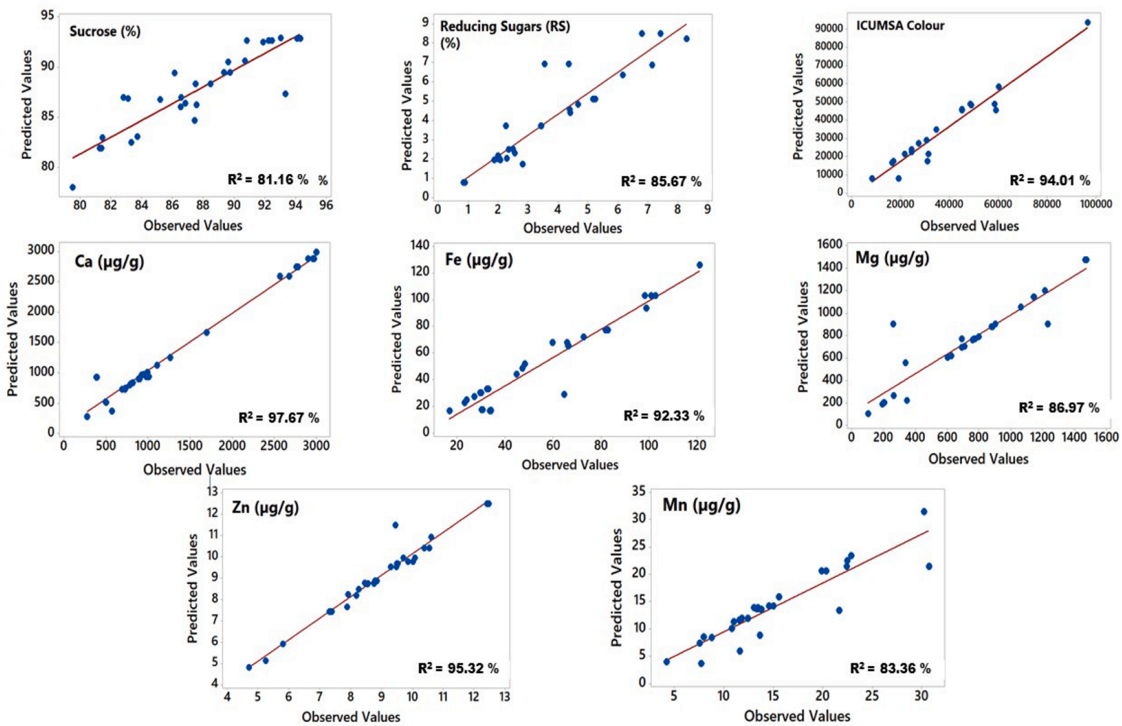


Fig. 3. Plots of the reference and predicted values of the validation set for the predictive models created with the kNN algorithm.

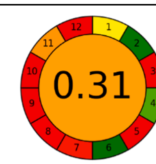
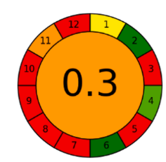
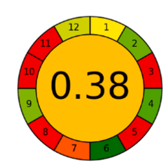
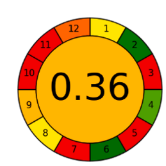
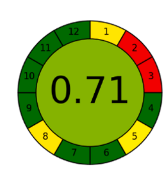
The AGREE calculator is based on the 12 criteria of Green Analytical Chemistry (GAC), each assigned a weight that directly influences the greenness of the evaluated analytical methodology (Pena-Pereira et al., 2020). The analytical method's greenness is represented by a circle coloured from green to yellow to red, indicating adherence level to the 12 GAC criteria, with a score from 0 to 1. A score of 0 represents the least eco-friendly method, while a score of 1 represents the most eco-friendly method (Imam & Abdelrahman, 2023). The main advantage of this greenness metric is that it includes a pictogram indicating the influence of each criterion, allowing the researcher to easily determine the environmental impact of the analytical method being evaluated.

The overall AGREE score for the DIP–kNN strategy (0.71) was higher than those observed for conventional methods (0.36 for HPLC–RID, 0.38 for UV–VIS spectrophotometry, and 0.30 and 0.31 for FAAS), demonstrating its superior sustainability and environmental friendliness (Table 2).

The DIP–kNN adheres best to criteria 4, 6, 7, 9, 10, 11 and 12. These aspects make this analytical strategy more environmentally friendly by avoiding the use of toxic solvents and reagents, minimizing energy expenditure (criteria 4, 9 – 11), avoiding derivatization reactions (criterion 6), not generating toxic wastes (criterion 7), and ensuring greater analyst safety (criterion 12).

For all the conventional methods evaluated (determination of ICUMSA colour by UV–VIS spectrophotometry, sugars by HPLC–RID and elements by FAAS), their compliance was strongest in criteria 2, 4 and 6. These criteria correspond to the use of a minimum amount of sample, fewer steps involved in the methodology, reduced use of reagents and lower energy expenditure, and the non-use of derivatization reactions, respectively. Also noteworthy among these analyses is the determination of ICUMSA colour, which adheres to criterion 9 by requiring minimal energy expenditure and criterion 12 by ensuring greater analyst safety. The AGREE metric's greenness score for conventional methodologies is reduced due to factors such as sample granulometry, the volume of toxic and non-renewable reagents used, the amount of waste generated, and the time required for sample extraction and analysis (refer to Table 2). In this sense, we highlight the analysis of elements by

Table 2
Comparison between the greenness profiles of the conventional methods and of the DIP–kNN strategy.

Analytical methods	Physicochemical parameter	AGREE pictogram
FAAS (Section 2.2)	*Mn (mg 100 g^{-1}; n = 3)* *Zn (mg 100 g^{-1}; n = 3)* *Fe (mg 100 g^{-1}; n = 3)*	
FAAS (Section 2.2)	*Mg (mg 100 g^{-1}; n = 3)* *Ca (mg 100 g^{-1}; n = 3)*	
UV–VIS Spectrophotometry (Section 2.2.)	*ICUMSA Colour (I.U; n = 2)*	
HPLC-RID (Section 2.2)	*Sucrose (g 100 g^{-1}; n = 3)* *Fructose and Glucose (g 100 g^{-1}; n = 3)*	
DIP-kNN (Sections 2.3 and 2.4)	*Sugars (sucrose e reducing sugars)* *Elements (Ca, Mg, Fe, Mn and Zn)* *ICUMSA Colour*	

FAAS and the sugars analysis using HPLC–RID. It is important to note that although chromatography is an analytical technique that consumes less energy, it uses potentially toxic organic solvents, which results in the generation of wastes (Kannaiah et al., 2021), while FAAS uses flammable gases, consumes more energy, and generates waste from the acid pre-treatment of the samples (Hill & Fisher, 2017; Miller-Ihli & Baker, 2017). These methodological requirements reduce the eco-friendliness of conventional methods.

3.4. Limitations and future tendencies

There is currently a global trend in the development of analytical strategies for the classification of food and beverages and the prediction of their physicochemical properties that incorporate machine learning algorithms (Alves et al., 2024; Jiménez-Carvelo et al., 2019; Meenu et al., 2021). However, the application of these methods in quality control and food inspection is still limited, emphasizing the need for further research to disseminate them. Food safety and contamination from inadequate hygiene, pesticides, bacteria, chemical additives and heavy metals are extremely important due to their direct impact on human and animal health (Khalaf et al., 2023).

Although the analytical strategy proposed in this paper (DIP-kNN) is very promising compared to conventional methods, like any analytical methodology it has its limitations. Currently, one of the biggest challenges in implementing it in a laboratory is ensuring the reproducibility of image acquisition in different environments, as images can be captured under different lighting conditions and with different smartphones. This means that the image processing steps vary significantly depending on the type of images captured, the resources available and the researcher's goal (Capitán-Vallvey et al., 2015; Rateni et al., 2017).

Another aspect that requires further research is the development of software's or apps for smartphone that enables the automatic capture and processing of images and the subsequent modelling of data. To this end, images captured with smartphones can be converted into digital values such as GSV (grayscale values), HSL (hue-saturation-brightness), RGB (red–green–blue) and HSV (hue-saturation-value) (Fan et al., 2021). In the literature, some authors have already conducted research in this direction for disease diagnosis, pathogen detection and food monitoring. Theses uses images captured by smartphones and/or optical devices, processed by existing applications or sent remotely via Bluetooth, micro-USB and Wi-Fi (Khalaf et al., 2023; Mousavizadegan et al., 2024; Sood & Singh, 2021). This enables the construction of models or the verification of concentration (predictive model) and classification on computers with greater data processing capacity. For example, it is possible to obtain an image of a food sample in real time and at any location and send it for processing and determination of concentration or classification as needed, using applications focused on food quality such as Clarifruit, FruitSize, Tuber Ruler, Pollenyzer or even a QR code sticker (Schwartzer & Boyarski, 2019; Borlinghaus et al., 2023; Pounds et al., 2022; Veloo et al., 2024).

Considering all the above aspects, the implementation of this strategy can facilitate the release of batches in the food industry, identify non-compliant batches or problems in the production process and help with food quality controls. In addition, it can serve as a preliminary method to verify the chemical composition of a particular product (Pandey et al., 2023). These measures can lead to lower production costs and better compliance with the Sustainable Development Goals, which makes them very attractive. These results can also be quickly utilized by new startup companies.

4. Conclusions

The main aspects to be highlighted about the strategy adopted in this work for the quality control of brown sugar are:

a) Variations in the physicochemical composition of brown sugar, as well as its processing, have a direct influence on the colouration and, consequently, on the quality of the product, which affects the consumer's purchase decision or its industrial application.
b) The combination of digital image processing and machine learning is an approach that could automate and optimize the quality control of brown sugar. This combination not only makes it possible to automate procedures, but also paves the way for the integration of different fields of study in the creation of new evaluation and classification methodologies for this sweetener, which stand out from conventional analysis methods.
c) The models generated by the kNN algorithm, showed high accuracy in predicting Ca, ICUMSA colour, Fe and Zn contents, while greater dispersion in the data was observed for sugars, which may be correlated to the presence of other components that impact on the final colour of brown sugar, such as sucrose degradation, melanoidin formation and metal ion complexation.
d) When using the AGREE metric, the DIP–kNN strategy obtained an overall score of 0.71, while conventional brown sugar analysis methods ranged between 0.30 and 0.38. Using Green Chemistry metrics to assess the environmental impact of a process is a relevant approach when proposing new analytical methodologies. It facilitates the implementation of more sustainable practices, allows for the comparison of different methods, and enables the choice of the most ecological option.

CRediT authorship contribution statement

Vandressa Alves: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis. **Jeferson M. dos Santos:** Methodology, Investigation, Formal analysis. **Olga Viegas:** Methodology, Investigation. **Edgar Pinto:** Methodology, Investigation. **Isabel M.P.L.V.O. Ferreira:** Supervision, Resources, Funding acquisition. **Vanderlei Aparecido Lima:** Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Conceptualization. **Maria L. Felsner:** Writing – review & editing, Writing – original draft, Supervision, Resources, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

Alarcón, A. L., Palacios, L. M., Osorio, C., César Narváez, P., Heredia, F. J., Orjuela, A., & Hernanz, D. (2021). Chemical characteristics and colorimetric properties of non-centrifugal cane sugar ("panela") obtained via different processing technologies. *Food Chemistry, 340*. https://doi.org/10.1016/j.foodchem.2020.128183

Alloway, B. J. (2008). *Zinc in Soils and Crop Nutrition* (Second edi). IZA and IFA.

Alves, V., de Andrade, J. K., & Felsner, M. L. (2023). Green and fast ultrasound-assisted extraction procedures for Fe, Mn, Mg and Ca analysis in cane syrups by FAAS. *Journal of Food Composition and Analysis, 123*, Article 105495. https://doi.org/10.1016/j.jfca.2023.105495

Alves, V., dos Santos, J. M., Pinto, E., Ferreira, I. M. P. L. V. O., Lima, V. A., & Felsner, M. L. (2024). Digital image processing combined with machine learning: A new strategy for brown sugar classification. *Microchemical Journal, 196*, Article 109604. https://doi.org/10.1016/j.microc.2023.109604

Asikin, Y., Kamiya, A., Mizu, M., Takara, K., Tamaki, H., & Wada, K. (2014). Changes in the physicochemical characteristics, including flavour components and Maillard reaction products, of non-centrifugal cane brown sugar during storage. *Food Chemistry, 149*, 170–177. https://doi.org/10.1016/j.foodchem.2013.10.089

Ballesteros, J. I., Caleja-Ballesteros, H. J. R., & Villena, M. C. (2021). Digital image-based method for iron detection using green tea (Camellia sinensis) extract as natural colorimetric reagent. *Microchemical Journal, 160*, Article 105652. https://doi.org/10.1016/j.microc.2020.105652

Bento, L. S. M. (2009). Colorants through cane sugar production and refining. *Sugar Industry, 134*, 168–176.

Borlinghaus, P., Jung, J., & Odemer, R. (2023). Introducing Pollenyzer: An app for automatic determination of colour diversity for corbicular pollen loads. *Smart Agricultural Technology, 5*, Article 100263. https://doi.org/10.1016/j.atech.2023.100263

Capitán-Vallvey, L. F., López-Ruiz, N., Martínez-Olmos, A., Erenas, M. M., & Palma, A. J. (2015). Recent developments in computer vision-based analytical chemistry: A tutorial review. *Analytica Chimica Acta, 899*, 23–56. https://doi.org/10.1016/j.aca.2015.10.009

Clarke, M. A., Edye, L. A., & Eggleston, G. (1997). Sucrose decomposition in aqueous solution, and losses in sugar manufacture and refining. *Advances in Carbohydrate Chemistry and Biochemistry, 52*, 441–470. https://doi.org/10.1016/s0065-2318(08)60095-5

Codex Alimentarius Commission. (2019). Codex Committee on Sugars (CCS). *Analysis of Responses to CL 2018/80-CS: Draft Standard for Panela and/or Common or Vernacular Name as Known in Each Country (Non – Centrifuged Sugar)*.

Cruz-Fernández, M., Luque-Cobija, M. J., Cervera, M. L., Morales-Rubio, A., & de la Guardia, M. (2017). Smartphone determination of fat in cured meat products. *Microchemical Journal, 132*, 8–14. https://doi.org/10.1016/j.microc.2016.12.020

Davies, T., Louie, J. C. Y., Ndanuko, R., Barbieri, S., Perez-Concha, O., & Wu, J. H. Y. (2022). A Machine Learning Approach to Predict the Added-Sugar Content of Packaged Foods. *The Journal of Nutrition, 152*, 343–349. https://doi.org/10.1093/jn/nxab341

de Almeida, V. E., de Araújo Gomes, A., de Sousa Fernandes, D. D., Goicoechea, H. C., Galvão, R. K. H., & Araújo, M. C. U. (2018). Vis-NIR spectrometric determination of Brix and sucrose in sugar production samples using kernel partial least squares with interval selection based on the successive projections algorithm. *Talanta, 181*, 38–43. https://doi.org/10.1016/j.talanta.2017.12.064

de Araújo Gomes, A., Azcarate, S. M., Špánik, I., Khvalbota, L., & Goicoechea, H. C. (2023). Pattern recognition techniques in food quality and authenticity: A guide on how to process multivariate data in food analysis. *TrAC Trends in Analytical Chemistry, 164*, Article 117105. https://doi.org/10.1016/j.trac.2023.117105

Deng, Z., Zhu, X., Cheng, D., Zong, M., & Zhang, S. (2016). Efficient kNN classification algorithm for big data. *Neurocomputing, 195*, 143–148. https://doi.org/10.1016/j.neucom.2015.08.112

Dong, C., An, T., Yang, M., Yang, C., Liu, Z., Li, Y., Duan, D., & Fan, S. (2022). Quantitative prediction and visual detection of the moisture content of withering leaves in black tea (Camellia sinensis) with hyperspectral image. *Infrared Physics & Technology, 123*, Article 104118. https://doi.org/10.1016/j.infrared.2022.104118

dos Santos, J. M., de Andrade, J. K., Galvão, F., & Felsner, M. L. (2019). Optimization and validation of ultrasound-assisted extraction for the determination of micro and macro minerals in non-centrifugal sugar by F AAS. *Food Chemistry, 292*, 66–74. https://doi.org/10.1016/j.foodchem.2019.04.037

Eggleston, G., Aita, G., & Triplett, A. (2021). Circular Sustainability of Sugarcane: Natural, Nutritious, and Functional Unrefined Sweeteners That Meet New Consumer Demands. *Sugar Tech, 23*, 964–973. https://doi.org/10.1007/s12355-021-00994-4

Fan, Y., Li, J., Guo, Y., Xie, L., & Zhang, G. (2021). Digital image colorimetry on smartphone for chemical analysis: A review. *Measurement, 171*, Article 108829. https://doi.org/10.1016/j.measurement.2020.108829

Feron, V. J., & Groten, J. P. (2002). Toxicological evaluation of chemical mixtures. *Food and Chemical Toxicology, 40*, 825–839. https://doi.org/10.1016/S0278-6915(02)00021-2

Frank, E., Hall, M. A., & Witten, I. H. (2016). Online Appendix on the Weka workbench. In *Data Mining: Practical Machine Learning Tools and Techniques* (4th ed., p. 654). Morgan Kaufmann Publishers.

GIMP. (2022). *The Free & Open-Source Image Editor* (v. 2.10.32). http://gimp.org.

He, H.-J., Chen, Y., Li, G., Wang, Y., Ou, X., & Guo, J. (2023). Hyperspectral imaging combined with chemometrics for rapid detection of talcum powder adulterated in wheat flour. *Food Control, 144*, Article 109378. https://doi.org/10.1016/j.foodcont.2022.109378

Helfer, G. A., Bock, F., Marder, L., Furtado, J. C., da Costa, A. B., & Ferrão, M. F. (2015). CHEMOSTAT: EXPLORATORY MULTIVARIATE DATA ANALYSIS SOFTWARE. *Química Nova*. https://doi.org/10.5935/0100-4042.20150063

Henrique da Silva Melo, B., Figueiredo Sales, R., da Silva Bastos Filho, L., Souza Povoas da Silva, J., Gabrielle Carolino de Almeida Sousa, A., Maria Camará Peixoto, D., & Pimentel, M. F. (2022). Handheld near infrared spectrometer and machine learning methods applied to the monitoring of multiple process stages in industrial sugar production. *Food Chemistry, 369*, 130919. https://doi.org/10.1016/j.foodchem.2021.130919.

Hill, S. J., & Fisher, A. S. (2017). Atomic Absorption, Methods and Instrumentation. In *Encyclopedia of Spectroscopy and Spectrometry* (pp. 37–43). Elsevier. https://doi.org/10.1016/B978-0-12-803224-4.00099-6.

IAL. Instituto Adolfo Lutz. (2008). *Métodos Físico-Químicos para Análise de Alimentos* (4. ed).

Imam, M. S., & Abdelrahman, M. M. (2023). How environmentally friendly is the analytical process? A paradigm overview of ten greenness assessment metric approaches for analytical methods. *Trends in Environmental Analytical Chemistry, 38*, e00202.

Jader, R., Fabián, V., John, E., Sebastián, E., & Oscar, M. (2018). Thermal performance evaluation of production technologies for non-centrifuged sugar for improvement in energy utilization. *Energy, 152*, 858–865. https://doi.org/10.1016/j.energy.2018.03.127

Jalaludin, I., & Kim, J. (2021). Comparison of ultraviolet and refractive index detections in the HPLC analysis of sugars. *Food Chemistry, 365*, Article 130514. https://doi.org/10.1016/j.foodchem.2021.130514

Jiménez-Carvelo, A. M., González-Casado, A., Bagur-González, M. G., & Cuadros-Rodríguez, L. (2019). Alternative data mining/machine learning methods for the analytical evaluation of food quality and authenticity – A review. *Food Research International, 122*, 25–39. https://doi.org/10.1016/j.foodres.2019.03.063

Kannaiah, K. P., Sugumaran, A., Chanduluru, H. K., & Rathinam, S. (2021). Environmental impact of greenness assessment tools in liquid chromatography – A review. *Microchemical Journal, 170*, Article 106685. https://doi.org/10.1016/j.microc.2021.106685

Keramat-Jahromi, M., Mohtasebi, S. S., Mousazadeh, H., Ghasemi-Varnamkhasti, M., & Rahimi-Movassagh, M. (2021). Real-time moisture ratio study of drying date fruit chips based on on-line image attributes using kNN and random forest regression methods. *Measurement, 172*, Article 108899. https://doi.org/10.1016/j.measurement.2020.108899

Khalaf, E. M., Sanaan Jabbar, H., Mireya Romero-Parra, R., Raheem Lateef Al-Awsi, G., Setia Budi, H., Altamimi, A. S., Abdulfadhil Gatea, M., Falih, K. T., Singh, K., & Alkhuzai, K. A. (2023). Smartphone-assisted microfluidic sensor as an intelligent device for on-site determination of food contaminants: Developments and applications. *Microchemical Journal, 190*, 108692. https://doi.org/10.1016/j.microc.2023.108692.

Kowtharapu, L. P., Katari, N. K., Muchakayala, S. K., & Marisetti, V. M. (2023). Green metric tools for analytical methods assessment critical review, case studies and crucify. *TrAC Trends in Analytical Chemistry, 166*, Article 117196. https://doi.org/10.1016/j.trac.2023.117196

Lindeman, P. F., & O'Shea, M. G. (2004). Colorant removal during clarification and decolourisation processes. *Proceedings of the Australian Society of Sugar Cane Technologists, 26*, 51–64.

López-Lorente, Á. I., Pena-Pereira, F., Pedersen-Bjergaard, S., Zuin, V. G., Ozkan, S. A., & Psillakis, E. (2022). The ten principles of green sample preparation. *TrAC Trends in Analytical Chemistry, 148*, Article 116530. https://doi.org/10.1016/j.trac.2022.116530

Macedo dos Santos-Tonial, L., Colla, M. S., Carra, J. B., Fabris, M., & de Lima, V. A. (2023). Classification and Total Carbon Determination of the Soils Using RGB Digital Images Combined with Machine Learning. *Communications in Soil Science and Plant Analysis, 54*, 141–153. https://doi.org/10.1080/00103624.2022.2110891

Mahesh, B. (2020). Machine learning algorithms-a review. *International Journal of Science and Research (IJSR), 9*, 381–386.

MAPA. Ministério da Agricultura, P. e A. (2018). Regulamento Técnico do Açúcar. INSTRUÇÃO NORMATIVA No 47, 12–15. https://www.in.gov.br/materia/-/asset_publisher/Kujrw0TZC2Mb/content/id/39939558/do1-2018-09-06-instrucao-normativa-n-47-de-30-de-agosto-de-2018-39939440.

Meenu, M., Kurade, C., Neelapu, B. C., Kalra, S., Ramaswamy, H. S., & Yu, Y. (2021). A concise review on food quality assessment using digital image processing. *Trends in Food Science & Technology, 118*, 106–124. https://doi.org/10.1016/j.tifs.2021.09.014

Mehrotra, R., & Siesler, H. W. (2003). Application of Mid Infrared/Near Infrared Spectroscopy in Sugar Industry. *Applied Spectroscopy Reviews, 38*, 307–354. https://doi.org/10.1081/ASR-120024392

Miller-Ihli, N. J., & Baker, S. A. (2017). Atomic Spectroscopy, Food and Dairy Products Applications of. In *Encyclopedia of Spectroscopy and Spectrometry* (pp. 82–88). Elsevier. https://doi.org/10.1016/B978-0-12-803224-4.00149-7.

MINITAB. (2010). *Minitab Incorporation* (Version 16.2.2). https://www.minitab.com/pt-br/.

Mousavizadegan, M., Shalileh, F., Mostajabodavati, S., Mohammadi, J., & Hosseini, M. (2024). Machine learning-assisted image-based optical devices for health monitoring and food safety. *TrAC Trends in Analytical Chemistry, 117794*. https://doi.org/10.1016/j.trac.2024.117794

Pandey, V. K., Srivastava, S., Dash, K. K., Singh, R., Mukarram, S. A., Kovács, B., & Harsányi, E. (2023). Machine Learning Algorithms and Fundamentals as Emerging Safety Tools in Preservation of Fruits and Vegetables: A Review. *Processes, 11*, 1720. https://doi.org/10.3390/pr11061720

Pena-Pereira, F., Wojnowski, W., & Tobiszewski, M. (2020). AGREE—Analytical GREEnness Metric Approach and Software. *Analytical Chemistry, 92*, 10076–10082. https://doi.org/10.1021/acs.analchem.0c01887

Perin, E. C., Fontoura, B. H., Lima, V. A., & Carpes, S. T. (2020). RGB pattern of images allows rapid and efficient prediction of antioxidant potential in Calycophyllum spruceanum barks. *Arabian Journal of Chemistry, 13*, 7104–7114. https://doi.org/10.1016/j.arabjc.2020.07.015

Pounds, K., Bao, H., Luo, Y., De, J., Schneider, K., Correll, M., & Tong, Z. (2022). Real-Time and Rapid Food Quality Monitoring Using Smart Sensory Films with Image

Analysis and Machine Learning. *ACS Food Science & Technology, 2*, 1123–1134. https://doi.org/10.1021/acsfoodscitech.2c00124

Raes, K., Knockaert, D., Struijs, K., & Van Camp, J. (2014). Role of processing on bioaccessibility of minerals: Influence of localization of minerals and anti-nutritional factors in the plant. *Trends in Food Science & Technology, 37*, 32–41. https://doi.org/10.1016/j.tifs.2014.02.002

Rateni, G., Dario, P., & Cavallo, F. (2017). Smartphone-Based Food Diagnostic Technologies: A Review. Sensors 2017, Vol. 17, Page 1453, 17, 1453. https://doi.org/10.3390/S17061453.

Ramírez-Morales, I., Rivero, D., Fernández-Blanco, E., & Pazos, A. (2016). Optimization of NIR calibration models for multiple processes in the sugar industry. *Chemometrics and Intelligent Laboratory Systems, 159*, 45–57. https://doi.org/10.1016/j.chemolab.2016.10.003

Riffer, R. (1988). The Nature of Colorants in Sugarcane and Cane Sugar Manufacture. *Sugar Series, 9*(Issue C). https://doi.org/10.1016/B978-0-444-43020-5.50019-9. Elsevier B.V.

Sajid, M., & Płotka-Wasylka, J. (2022). Green analytical chemistry metrics: A review. *Talanta, 238*, Article 123046. https://doi.org/10.1016/j.talanta.2021.123046

Santos, J. R., Viegas, O., Páscoa, R. N. M. J., Ferreira, I. M. P. L. V. O., Rangel, A. O. S. S., & Lopes, J. A. (2016). In-line monitoring of the coffee roasting process with near infrared spectroscopy: Measurement of sucrose and colour. *Food Chemistry, 208*, 103–110. https://doi.org/10.1016/j.foodchem.2016.03.114

Schlesner, S. K., Voss, M., Helfer, G. A., Costa, A. B., Cichoski, A. J., Wagner, R., & Barin, J. S. (2022). Smartphone-based miniaturized, green and rapid methods for the colorimetric determination of sugar in soft drinks. *Green Analytical Chemistry, 1*, Article 100003. https://doi.org/10.1016/j.greeac.2022.100003

Schuastz Breda, L., Elton de Melo Nascimento, J., Alves, V., de Alencar Arnaut de Toledo, V., Aparecido de Lima, V., & Lurdes Felsner, M. (2024). Green and Fast Prediction of Crude Protein Contents in Bee Pollen based on Digital Images combined with Random Forest Algorithm. *Food Research International*, 113958. https://doi.org/10.1016/j.foodres.2024.113958.

Schwartzer A., Boyarski. R. (2019). *System and method for evaluating fruits and vegetables* (Patent No. 16 / 514,553). https://patents.justia.com/patent/10839503.

Shi, M., Zheng, X., Zhang, N., Guo, Y., Liu, M., & Yin, L. (2023). Overview of sixteen green analytical chemistry metrics for evaluation of the greenness of analytical methods. *TrAC Trends in Analytical Chemistry, 166*, Article 117211. https://doi.org/10.1016/j.trac.2023.117211

Shrivastav, P., Verma, A. K., Walia, R., Parveen, R., & Singh, A. (2016). Jaggery: A revolution in the field of natural sweeteners. *European Journal of Pharmaceutical and Medical Research, 3*, 198–202.

Sood, S., & Singh, H. (2021). Computer Vision and Machine Learning based approaches for Food Security: A Review. *Multimedia Tools and Applications, 80*, 27973–27999. https://doi.org/10.1007/s11042-021-11036-2

Swietlik, D. (2010). Zinc Nutrition in Horticultural Crops. In *Horticultural Reviews* (pp. 109–178). John Wiley & Sons, Inc.. https://doi.org/10.1002/9780470650752.ch3

Takahashi, M., Ishmael, M., Asikin, Y., Hirose, N., Mizu, M., Shikanai, T., Tamaki, H., & Wada, K. (2016). Composition, Taste, Aroma, and Antioxidant Activity of Solidified Noncentrifugal Brown Sugars Prepared from Whole Stalk and Separated Pith of Sugarcane (Saccharum officinarum L.). *Journal of Food Science, 81*, C2647–C2655. https://doi.org/10.1111/1750-3841.13531

Tan, Pang-Ning, Michael Steinbach, and V. K. (2016). *Introduction to data mining.* Pearson Education.

Team R Core, R. C. (2022). *R: A language and environment for statistical computing.* R Foundation for Statistical Computing. https://www.r-project.org/.

Tian, H., Chen, B., Yu, H., Lou, X., Li, Y., Yu, H., Chen, L., & Chen, C. (2022). Rapid detection of neutralising acid adulterants in raw milk using a milk component analyser and chemometrics. *Food Additives & Contaminants: Part A, 39*, 1501–1511. https://doi.org/10.1080/19440049.2022.2093985

Tsakiridis, N. L., Samarinas, N., Kokkas, S., Kalopesa, E., Tziolas, N. V., & Zalidis, G. C. (2023). In situ grape ripeness estimation via hyperspectral imaging and deep autoencoders. *Computers and Electronics in Agriculture, 212*, Article 108098. https://doi.org/10.1016/j.compag.2023.108098

Veloo, K., Glenn, A. E., King, A. B., Smith, B. J., Marleau, M. M., & Sankaran, S. (2024). Tuber Ruler: A mobile application for evaluating image-based potato tuber size. *Journal of Food Measurement and Characterization.* https://doi.org/10.1007/s11694-024-02542-6

Weerawatanakorn, M., Asikin, Y., Takahashi, M., Tamaki, H., Wada, K., Ho, C.-T., & Chuekittisak, R. (2016). Physico-chemical properties, wax composition, aroma profiles, and antioxidant activity of granulated non-centrifugal sugars from sugarcane cultivars of Thailand. *Journal of Food Science and Technology, 53*, 4084–4092. https://doi.org/10.1007/s13197-016-2415-5

Wu, X., Kumar, V., Ross Quinlan, J., Ghosh, J., Yang, Q., Motoda, H., McLachlan, G. J., Ng, A., Liu, B., Yu, P. S., Zhou, Z.-H., Steinbach, M., Hand, D. J., & Steinberg, D. (2008). Top 10 algorithms in data mining. *Knowledge and Information Systems, 14*, 1–37. https://doi.org/10.1007/s10115-007-0114-2

Zhang, S. (2020). Cost-sensitive KNN classification. *Neurocomputing, 391*, 234–242. https://doi.org/10.1016/j.neucom.2018.11.101