



Spectroscopic Non-targeted Techniques in Combination with Linear Discriminant Analysis for Wine Vinegar Authentication

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Abstract

This work aims at comparing different non-targeted spectroscopic techniques (i.e., UV–Vis, FT-IR, FT-NIR, and NIR spectroscopy) for the authentication of white wine vinegar. Five white wine vinegars were adulterated with two different spirit vinegars. Further twenty-five wine vinegars were analyzed to enlarge the authentic product dataset. All samples (i.e., 160) were analyzed in duplicate by UV–Vis, FT-NIR, and FT-IR spectroscopy; moreover, a handheld NIR device was tested on a subset of samples (i.e., 89). Principal component analysis revealed sample patterns related to vinegar acidity (6 or 7.1%) rather than adulteration levels. After variable selection (SELECT algorithm), linear discriminant analysis (LDA) models were developed and tested by independent external sets. The LDA models gave very high weighted correct classification rates in calibration (95.5–100.0%), cross-validation (92.4–100.0%), and prediction (90.0–100.0%) for all the spectroscopic techniques. With the portable NIR instrument, 100% correct classifications in prediction were obtained, demonstrating its suitability in vinegar authentication.

Keywords Adulteration · Discrimination · Portable device · UV–vis spectroscopy · Near-infrared spectroscopy · Infrared spectroscopy

Introduction

Historically, vinegar was considered a by-product of wine making, but nowadays, it is produced to be used mainly for food pickling and preserving, but also as disinfectant in healthcare and cleaning sectors. Besides acetic acid, a wide variety of phenolic compounds, vitamins, and other bioactive compounds are contained in vinegar, depending on the raw materials used and the fermentation conditions. Vinegars available on the market are very different in terms of quality, types, and prices, including cheap and synthetic products as well as premium traditional balsamic vinegars (Lim et al., 2019). Vinegar versatility and diverse applications have brought its global market at 1.36 billion USD in 2022, with a growth expectation to reach 1.50 billion USD by 2028. Balsamic vinegar currently dominates the global market with a market share higher than 25%. Balsamic is

followed by red wine, cider, rice, and white vinegars (Expert Market Research, 2023). In 2018, Germany, France, and Italy were the countries with the highest consumption volumes, accounting together for 49% of total consumption (Global Trade, 2023).

Due to the high variability of vinegars available on the market and the large quantities consumed in daily diet, vinegar authentication is fundamental to protect honest manufacturers, to guarantee fair trades, and to assure consumers' rights and safety. In general, the term “vinegar” refers to a liquid containing a minimum of 3.75–5.0% (w/v) acetic acid, depending on local laws and regulations. The definition includes synthetic vinegars, produced by diluting glacial acetic acid, whose market is regulated by laws. However, due to the large volumes of marketed products, there are serious concerns about fraudulent practices involving the mixing of premium products with cheap synthetic vinegars, without proper labeling (Ko et al., 2013; Lim et al., 2019). Thus, the development of rapid and easy methods of authentication is extremely important to both food industries and control bodies.

Authentication methods can be divided into targeted and non-targeted approaches. However, since vinegar is a complex

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liquid and many adulterants can be used, the application of targeted methods is often ineffective. Moreover, targeted approaches are often expensive, destructive, time- and reagent-consuming, requiring also trained personnel. As an example, Ko et al. (2013) applied site-specific natural isotope fractionation by nuclear magnetic resonance (SNIF-NMR) to examine vinegar adulteration with synthetic acetic acid, but they need to extract high-purity acetic acid by a distillation method; thus, the sample preparation is very time- and reagent-consuming. On the contrary, non-targeted techniques are non-destructive, easy, rapid, and low-cost; they have a holistic approach and aim to provide a fingerprint of the analyzed food products. Combined with the appropriate chemometric models, the fingerprint can be used to distinguish products that differ by small changes. The most important untargeted methods are based on spectroscopic techniques, such as Fourier-transform infrared (FT-IR), Fourier-transform near-infrared (FT-NIR), near-infrared (NIR), hyperspectral imaging, Raman, and nuclear magnetic resonance (NMR) (Cavdaroglu & Ozen, 2021).

Most of the studies about vinegar authentication considers high-quality product adulteration, such as Protected Designation of Origin (PDO) and Protected Geographical Indication (PGI) categories (Calle et al., 2021; Cavdaroglu & Ozen, 2021; Ríos-Reina et al., 2021). However, in food canning and preparations (e.g., dressings and sauces), industrial vinegars are usually used in large volumes; thus, an authentication method suitable also for this kind of products is necessary. For instance, white wine vinegar used for vegetable pickling can be adulterated by the cheaper spirit vinegar, which is defined as the product obtained by acetous fermentation from distilled alcohol (Codex Alimentarius Commission, 1987). Moreover, as reported in the review by Ríos-Reina et al. (2021), few studies compare the suitability of different spectroscopic techniques; the instrument selection is commonly based on its availability. At last, deeper studies on the suitability of spectroscopic portable devices are needed. Portable devices are actually important tools for fraud control, especially in an industrial perspective, being low-cost, easy to handle, and robust. The vinegar authentication methods developed so far are, on the contrary, restricted to laboratories, requiring highly sensitive and complex equipment (Ríos-Reina et al., 2021).

In this context, the aim of this work was to compare the potential of different spectroscopic techniques (i.e., UV–Vis, FT-NIR, and FT-IR spectroscopy), combined with chemometrics, in discriminating authentic white wine vinegars from products adulterated with spirit vinegars (5–25% v/v adulteration levels). Moreover, a handheld NIR device was tested on a representative number of samples, hypothesizing that it can have similar performances as the benchtop instruments. Variable selection by means of the SELECT algorithm implemented in the V-PARVUS package (Forina

et al., 1990) was tested, to develop robust and more parsimonious models, which require less computational time and can favor the development of cheaper and dedicated spectroscopic devices.

Materials and Methods

Authentic and Adulterated Vinegars

Thirty white wine vinegars were collected covering producer, origin, and acidity variability present in the Italian market. For each sample, two different aliquots were analyzed. Details about the vinegars are reported in Table S1. Twenty-five vinegars were characterized by 6% acidity, whereas the other five had 7.1% acidity. In some cases, the batch variability was considered, analyzing bottles belonging to different lot numbers (i.e., AC5, AC6, and AC26; AC1 and AC12). Five different vinegars (four at 6% and one at 7.1% acidity) were adulterated with two commercial spirit vinegars at 5, 10, 15, 20, and 25% v/v concentration. Two replicates of each adulteration were independently prepared. Thus, the total amount of samples was 160: 60 samples of authentic white wine vinegars (30 white wine vinegars \times 2 aliquots) and 100 samples of adulterated vinegars (5 white wine vinegars \times 2 adulterants \times 5 adulteration levels \times 2 adulteration replicates). The experimental plan is shown in Fig. 1.

Spectroscopic Measurements

All samples were analyzed in duplicate by applying different spectroscopic techniques: UV–Vis, FT-NIR, and FT-IR spectroscopy.

UV–Vis spectra were acquired by the double-ray V-650 spectrophotometer (Jasco Europe, Lecco, Italy) managed by the software Spectra Manager™ II (Jasco Europe, Lecco, Italy). Samples were loaded in a quartz cuvette with a pathlength of 10 mm and a volume of 3.5 mL. Spectra were collected in the range 190–900 nm, with a resolution of 2 nm and a detector speed of 400 nm/min.

The MPA FT-NIR spectrophotometer (Bruker Optics, Milano, Italy), managed by the software Opus™ (v. 6.5, Bruker Optics, Milano, Italy), was used to collect spectra in the range 12,500–4000 cm^{-1} , with 8 cm^{-1} resolution and 32 scans for both samples and background. A quartz cuvette with a pathlength of 2 mm and a volume of 700 μL was used.

FT-IR spectra were collected by a Vertex spectrophotometer (Bruker Optics, Milano, Italy) managed by the software Opus™ (v. 6.5, Bruker Optics, Milano, Italy). Measurements were performed with a multiple reflection Germanium attenuated total reflection (ATR) accessory in the 4000–800 cm^{-1} range, with a resolution of 4 cm^{-1} and 32 scans for both samples and background.

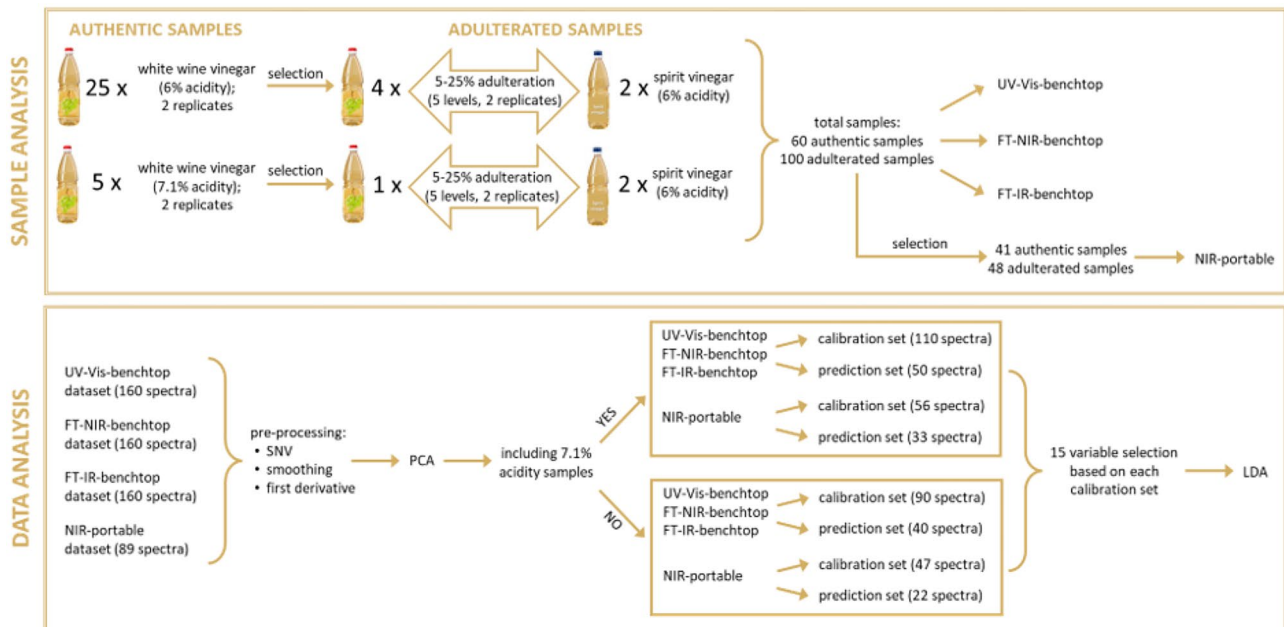


Fig. 1 Samples and data analysis workflow of the study about white wine vinegar authentication

A selected set of samples (41 authentic and 48 adulterated) was used to assess the feasibility of developing a discrimination model from data collected by using the portable instrument Polispec NIRTM (ITPhotonics, Vicenza, Italy) managed by the software poliDATA (ITPhotonics, Vicenza, Italy) implemented in a dedicated tablet. Samples were analyzed in duplicate using a prototypal liquid sample holder. The versatile sample holder permits to customize the pathlength by moving a gold plate in different predefined positions; in our experiment a 2 mm path-length was kept. Spectra were collected in the 900–1700 nm range with a resolution of 3.2 nm.

Data Analysis

The data analysis workflow is sketched in Fig. 1. The replicated spectra, collected for each sample by each instrument, were averaged and a dataset for each spectroscopic technique was obtained. Each dataset was subjected to different spectral pretreatments, namely, Standard Normal Variate (SNV), smoothing and first derivative by Savitzky-Golay filter, alone or in combination. Furthermore, a dataset excluding the vinegars at 7.1% acidity and their adulterations was created for each spectroscopic technique.

Principal Component Analysis (PCA) was performed to evaluate the variable weights and possible sample patterns based on adulteration and acidity level.

Subsequently, each dataset was divided into a calibration set (about 70% of the samples) for calibration and cross-validation (with 5 cross-validation segments) and an

external set (30% of the samples) for validation. Fifteen variables were selected by applying to the calibration sets the SELECT algorithm, implemented in the V-PARVUS package (Forina et al., 1990). At first, the algorithm selects the variable with the largest Fisher weight and decorrelates the other predictors; then, the procedure is repeated iteratively until the desired number of variables is selected (in our case 15). The obtained datasets were subjected to Linear Discriminant Analysis (LDA) using the V-PARVUS package (Forina et al., 1990) to discriminate authentic from adulterated samples, thus considering two classes. Model performances were evaluated in terms of correct classification ability in calibration, cross-validation, and prediction. The correct classification ability was calculated for each class or as weighted classification ability according to the equation reported by Grassi et al. (2019). Performances of the models obtained with the data collected by the different instruments were compared for predictive abilities by McNemar test applying the “testcholdout” function implemented in MATLAB environment (v. 2020a, MathWorks, Inc., Natick, MA, USA) as reported by Grassi et al. (2018).

Results and Discussion

Spectral Inspection

As for UV–Vis analysis, the most informative region was found in the UV range (Fig. 2A). Two strong absorption bands characterized the collected spectra at 210–215 and

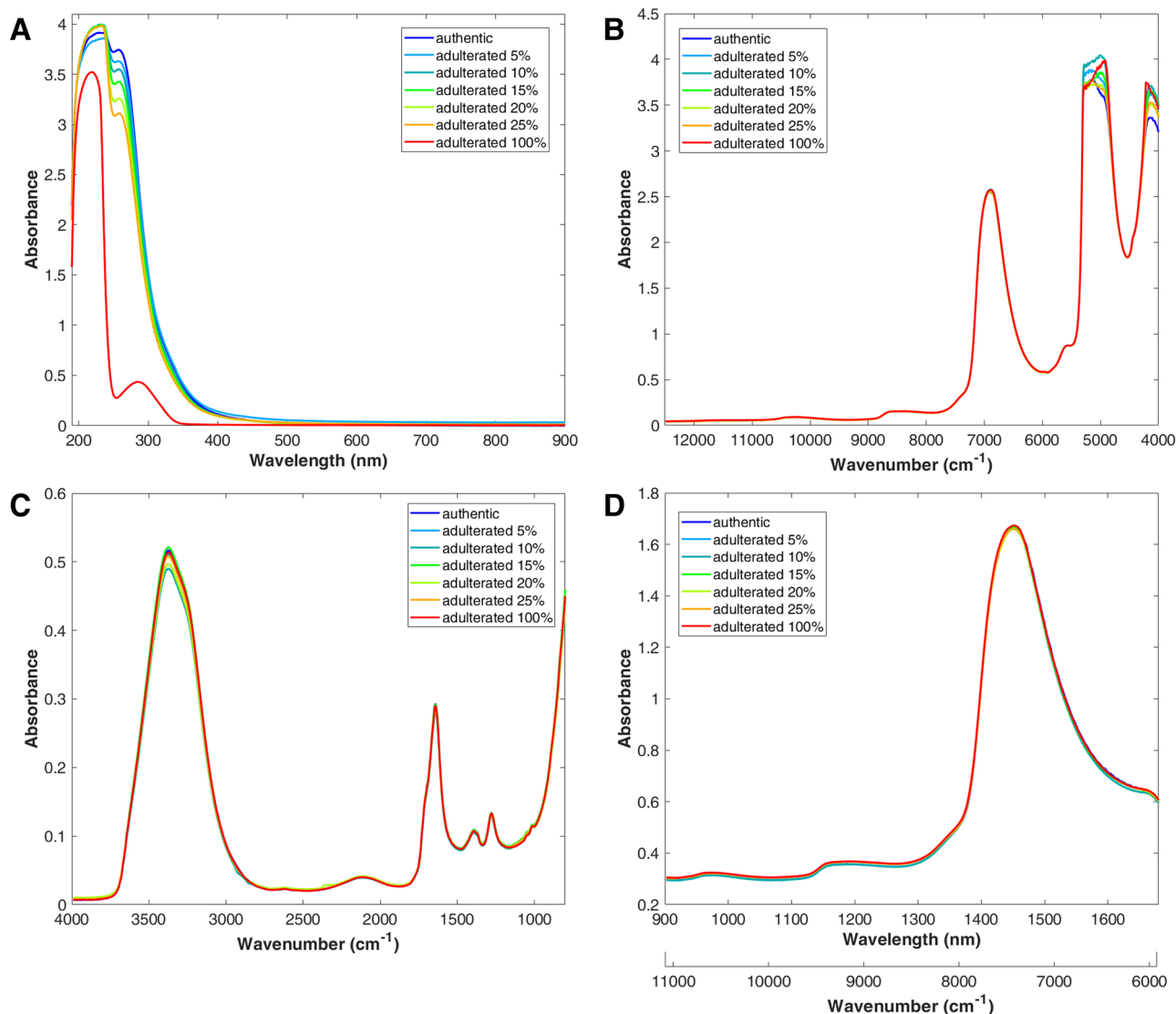


Fig. 2 Spectra collected for the white wine vinegar sample AC1 and its adulteration with the spirit vinegar AE1 at different levels: **A** UV–Vis spectra, **B** FT-NIR spectra, **C** FT-IR spectra, **D** portable NIR spectra

285–290 nm. The first band is possibly related to free amino acids and the latter linked to the presence of organic acids, such as acetic, malic, citric, gallic, tannic, and lactic acid (Yalçın et al., 2021). Amino acids and organic acids are both more abundant in wine vinegars rather than in spirit vinegars (Ousaaid et al., 2021), thus justifying the higher absorptions in the spectra of authentic and adulterated samples rather than in those of spirit vinegars (adulterated 100%).

The FT-NIR spectra (Fig. 2B) were characterized by a band at 6900 cm^{-1} and two saturated broad bands at $5300\text{--}4600\text{ cm}^{-1}$ and around 4100 cm^{-1} , related to overtone and combination vibrations of the O–H bonds. Moreover, a shoulder was present at 5600 cm^{-1} , due to O–H bonds possibly associated with sucrose, fructose, and glucose. A weak peak at 6000 cm^{-1} could be attributed to the first overtone of

--CH_3 stretching or C–H groups of chemical aromatic compounds (Ríos-Reina et al., 2021). In any case, no relevant differences between authentic and adulterated vinegars could be appreciated. Similarly, the portable NIR spectra (Fig. 2D), covering a reduced range (900–1700 nm, a.k.a. $11,100\text{--}5900\text{ cm}^{-1}$), showed a characteristic broad band at 1450 nm (6900 cm^{-1}) linked to the first overtone of O–H bonds (Ríos-Reina et al., 2021). A visible change in slope among the different adulteration levels was detected between 1520 and 1660 nm.

No relevant or systematic differences were visible in the FT-IR spectra collected for authentic and adulterated vinegars (Fig. 2C). A change in absorbance was highlighted for the band at $3750\text{--}2750\text{ cm}^{-1}$, attributed to –OH group of water and C–H stretching of acetic acid, and some

differences were observed for the band at $1700\text{--}1600\text{ cm}^{-1}$, associated with C–O stretching of aldehydes (Ríos-Reina et al., 2017). In the fingerprint region ($1500\text{--}800\text{ cm}^{-1}$), the two small shoulders at 1065 and 1030 cm^{-1} are associated with O–H and $-\text{CH}_2$ groups of sugars (Ríos-Reina et al., 2017).

Spectral Exploration and Variable Selection

Data exploration was performed by PCA on the averaged spectra collected by each instrument, after the application of different preprocessing strategies. The PC1 vs. PC2 score plots of raw and pretreated UV–Vis spectra did not show a clear separation between adulterated and authentic samples. However, after first derivative transformation (Savitzky-Golay filter, 5 window points, third polynomial order) in

the PC2 vs. PC3 score plot (Fig. 3A), it was possible to find a sample separation based on acidity along PC2. In fact, the box in Fig. 3A indicates samples with 7.1% acidity, which were characterized by positive PC2 scores and distributed along PC3 from low to high adulteration levels following the arrow direction.

The FT-NIR spectra were reduced in the most informative spectral ranges ($9000\text{--}5300\text{ cm}^{-1}$; $4600\text{--}4300\text{ cm}^{-1}$). No sample distribution based on authenticity was appreciable in score plots for any of the investigated preprocessing. The score plot obtained by exploring raw data in the PC1 vs. PC2 space (Fig. 3B) showed a sample grouping as a function of acidity. Indeed, the samples characterized by high acidity (AC04, AC08, AC13, AC14, and AC15), as well as their adulterated mixtures (AC04 adulterated at 5, 10, 15, 20, and 25% v/v), formed a separated group highlighted in the box,

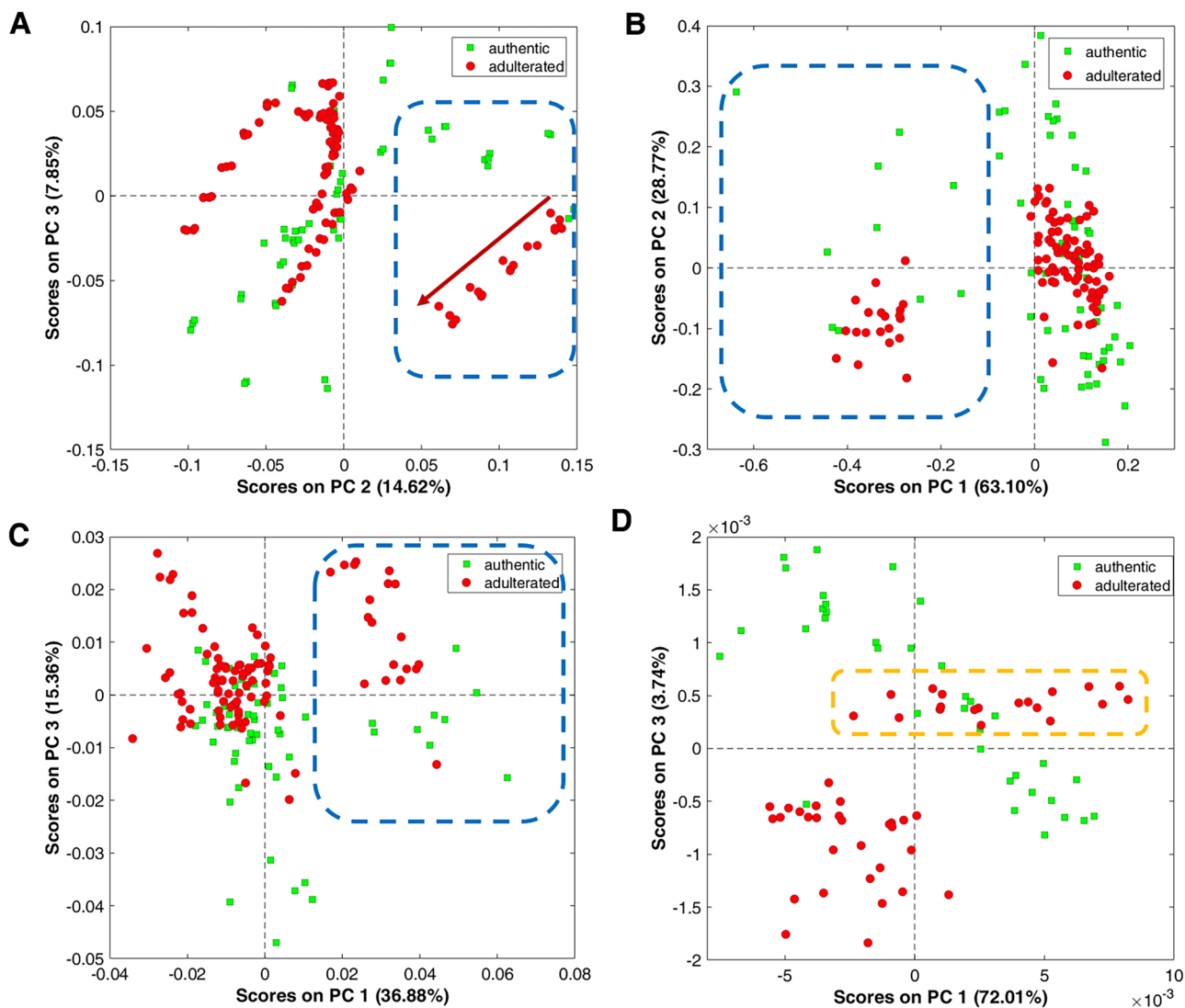


Fig. 3 Score plot of the PCA performed on the data collected for wine vinegar authentication by **A** UV–Vis, **B** FT-NIR, **C** FT-IR spectroscopy, and **D** portable NIR device

characterized by negative PC1 scores. Actually, the ability of NIR and Vis–NIR spectroscopy to analyze pH and acidity of vinegars was already demonstrated (Bao et al., 2014; Liu et al., 2011).

When considering the selected region of the FT-IR spectra (2200–800 cm^{-1}), the most relevant information was retrieved by the PC1 vs. PC3 score plot obtained from the raw spectra (Fig. 3C). The group of samples with positive PC1 scores marked with a dashed box in Fig. 3C is the authentic and adulterated vinegars characterized by 7.1% acidity.

The PC1 vs. PC3 score plot of first derivative spectra (Savitzky-Golay filter, 11 window points, third polynomial

order) collected by the portable NIR device for the selected set of samples (41 authentic and 48 adulterated) showed a good separation of authentic and adulterated vinegars (Fig. 3D). Most adulterated samples were positioned in the third quadrant of the plane (negative PC1 and PC3 scores) except for a group of samples (marked in the dashed box) at low adulteration levels (5 and 10% v/v).

As expected, the relevant signals for sample distribution were mainly related to acidity, as highlighted especially in the loading plots related to UV–Vis, FT-NIR, and FT-IR data. The loading plot of the PCA applied to the UV–Vis data transformed in first derivative (Fig. 4A) showed that the most relevant region was located in the range from 240

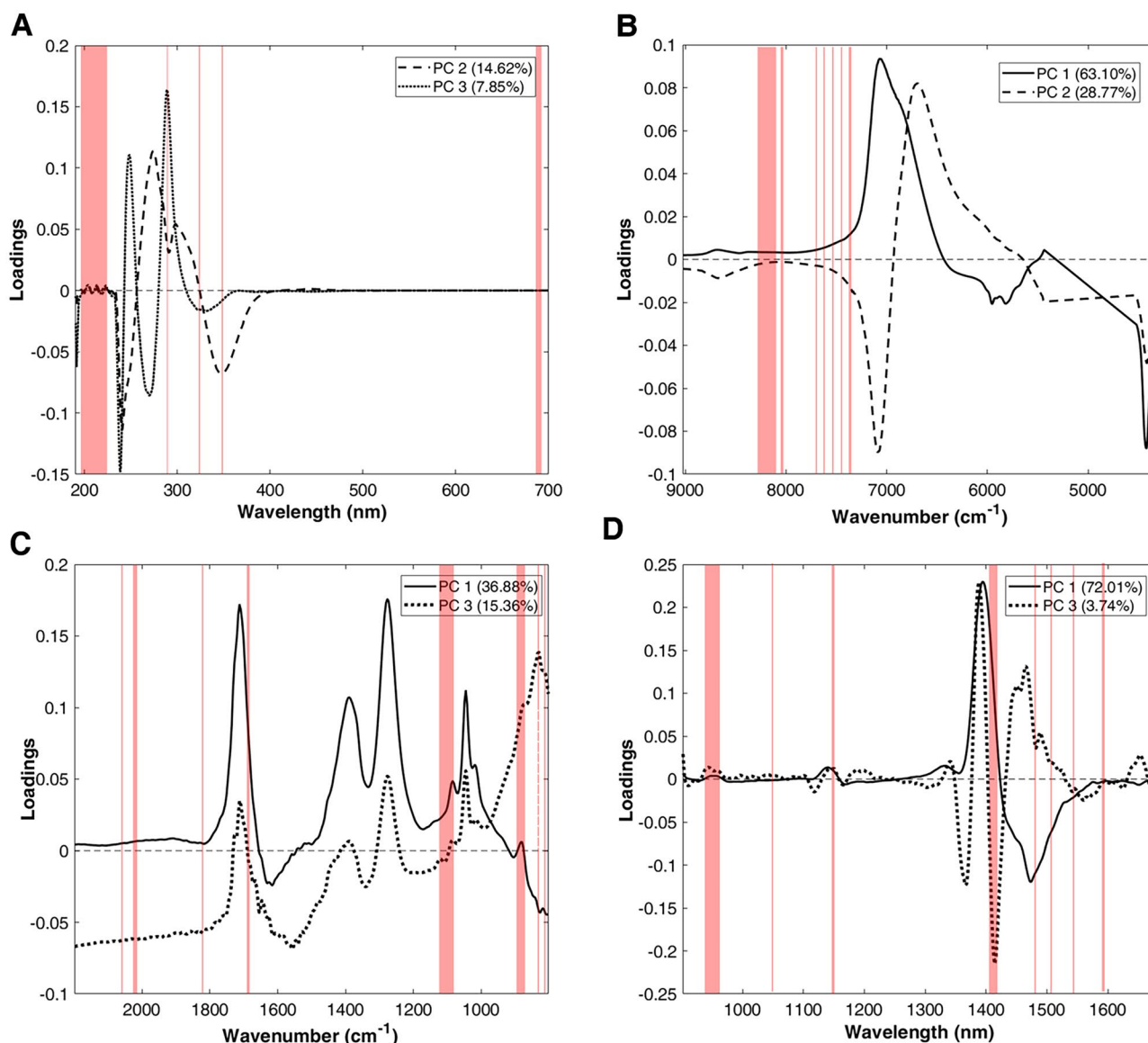


Fig. 4 Loading plot of the PCA performed on the data collected for wine vinegar authentication by **A** UV–Vis, **B** FT-NIR, **C** FT-IR spectroscopy, and **D** portable NIR device. Highlighted bands correspond to variables selected for the development of classification models

to 350 nm, which includes the absorptions related to organic acids, such as acetic, malic, citric, gallic, tannic, and lactic acid formed during vinegar fermentation (Yalçın et al., 2021). In the visible range, from 400 to 700 nm, no signal was significant for sample discrimination, thus confirming that the vinegars were comparable by visual appearance.

The highest effect on sample distribution observed with FT-NIR raw spectra was related to the signal at 6900 cm^{-1} corresponding to the first overtone of the O–H bonds (Ríos-Reina et al., 2018), which had high PC2 weight in the loading plot (Fig. 4B). This is mainly related to water and organic acids, thus explaining the sample grouping based on acidity levels observed in the score plot (Fig. 3B). Furthermore, PC2 loadings were characterized by negative signals around $5800\text{--}6200\text{ cm}^{-1}$ and 4300 cm^{-1} , related to phenolic aromatic compounds (Ríos-Reina et al., 2018) and C–H stretching of acetic acid (Chung & Ku, 2003), respectively.

The PC1 and PC3 loadings obtained from the PCA applied to the FT-IR fingerprint region (Fig. 4C) were both characterized by a high weight of the signals at 1713, 1393, 1277, 1045, and 833 cm^{-1} . A study by Ríos-Reina et al. (2017), comparing the FT-IR vinegar absorption with a standard of acetic acid diluted in water, assigned the signal around 1711 cm^{-1} to the C=O group of acetic acid and the two bands near 1400 and 1290 cm^{-1} to the C–O stretching and the C–O–H in-plane bending, respectively. The signals around 1045 and 1015 cm^{-1} were found only in vinegar samples and assigned to alcohol compounds, aldehydes, and some esters and ethers (Ríos-Reina et al., 2017).

The loadings of first derivative transformed NIR spectra collected with the portable device (Fig. 4D) explained the differentiation among adulterated and authentic/low-level-adulterated samples; however, due to the derivative transformation, it is difficult to assign the signals to specific absorptions, which seems to be mainly related to OH first overtone (around 1400 nm) and phenolic and aromatic compounds (1450–1550 nm) (Ríos-Reina et al., 2018).

Figure 4 also shows the variables selected for the development of classification models. While PCA tends toward sample variance maximization, variable selection by the SELECT algorithm aims at maximizing class separation (authentic vs. adulterated). Thus, the variables selected for each spectroscopic technique did not match with the most relevant loadings. The selected UV–Vis variables (Fig. 4A) were related to the absorption of organic acids, with a peak around 285 nm (selected variables: 239, 288.5, and 329 nm). Furthermore, the variables selected around 215 nm (194, 195, 202, 206, 211, 215.5, and 221 nm) could be related to the absorption of free amino acids and nucleotides (Yalçın et al., 2021), which are expected to be more present in the authentic wine vinegar samples than in adulterated samples. The selected FT-NIR variables (Fig. 4B) resulted in the range $8188\text{--}7320\text{ cm}^{-1}$. Considering the variables

selected for the FT-IR range (Fig. 4C), the fingerprint region ($1500\text{--}800\text{ cm}^{-1}$) confirmed its relevance in discriminating samples, with a high number of selected variables around 1100 cm^{-1} (1169, 1146, 1111 cm^{-1}) and 950 cm^{-1} (964, 945, 922 cm^{-1}), related to alcohol compounds, aldehydes, and some esters and ethers (Ríos-Reina et al., 2017). The variables selected in the NIR spectra of the portable device (Fig. 4D) were in the regions 900–1200 nm (940, 942, 970, 972, 990, 1046, 1154, and 1172 nm), where no specific spectral signal was detected in the raw spectra, and in the region 1400–1600 nm (1400, 1408, 1428, 1444, 1488, 1518, 1548, 1586, and 1588 nm) characterized by a visible change in slope among the different adulteration levels.

Discrimination Models

After calibration and external set division, the fifteen selected variables for each dataset of UV–Vis, FT-NIR, and FT-IR spectroscopy were used to construct LDA models. At first, the whole set of samples was considered (i.e., 60 authentic and 100 adulterated at different levels), thus creating calibration sets with 110 spectra (40 from authentic and 70 from adulterated samples) and prediction sets with 50 spectra (20 from authentic and 30 from adulterated samples).

Really good correct classification rate was obtained for FT-NIR data (Table 1). Sáiz-Abajo et al. (2004), studying the adulteration of wine vinegars with alcohol vinegars by NIR spectroscopy, were able to correctly predict 100% of both authentic and adulterated vinegars (at 15, 30, 50, and 70% v/v levels); however, they failed in identifying the alcohol vinegar samples (0% of correct classification in prediction).

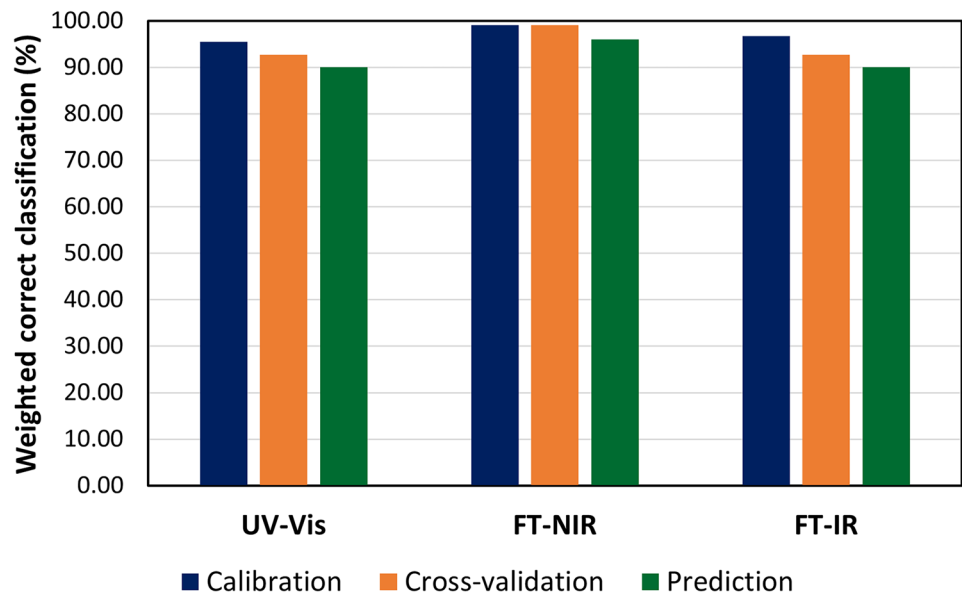
Good results were also achieved with UV–Vis and FT-IR data, with a lower correct classification in prediction for the authentic class (75.0% and 80.0%, respectively) than for the adulterated class (100.0% and 96.7%, respectively). This is, anyway, a good achievement as it means that no one adulterated sample will be identified as authentic. Similarly, Cavdaroglu and Ozen (2022) developed PLS-DA, OPLS-DA, and ANN models to evaluate the use of UV–Vis and FT-IR spectroscopy for vinegar authentication. However, to reach good classification rates in the OPLS-DA models (i.e., 95.16% and 96.8%, respectively, for UV–Vis and FT-IR spectroscopy), the removal of lowadulterated samples (5% adulterant levels) was needed. On the contrary, the LDA models here developed reached 90.0% of weighted correct classification in prediction (Fig. 5) for both the spectroscopic techniques considering vinegars with different acidity and a wide range of adulteration levels (from 5 to 25% v/v).

Even though a slightly better performance was achieved by the model developed with FT-NIR data, the McNemar test proved that there was no significant difference ($P > 0.05$) between the models developed for the three different spectral regions.

Table 1 Results of linear discriminant analysis for vinegar discrimination: correct classification percentages of models based on the 15 most informative variables selected for the different spectroscopic techniques

All samples		Calibration		Cross-validation		Prediction	
Technique	Range	Authentic	Adulterated	Authentic	Adulterated	Authentic	Adulterated
		<i>N</i>	40	70	40	70	30
	Pretreatment						
UV-Vis	700–190 nm	90.0	98.6	85.0	97.1	75.0	100.0
FT-NIR	9025–5430 cm ⁻¹	100.0	98.6	100.0	98.6	95.0	96.7
	4520–4320 cm ⁻¹						
FT-IR	4000–2500 cm ⁻¹	90.0	100.0	85.0	97.1	80.0	96.7
	2200–800 cm ⁻¹						
No 7.1% acidity							
		<i>N</i>	38	52	38	52	28
	Pretreatment						
UV-Vis	700–190 nm	89.5	100.0	81.6	94.3	100.0	100.0
FT-NIR	9025–5430 cm ⁻¹	86.8	96.2	76.3	90.4	100.0	96.4
	4520–4320 cm ⁻¹						
FT-IR	4000–2500 cm ⁻¹	92.1	100.0	84.2	98.1	100.0	96.4
	2200–800 cm ⁻¹						

Fig. 5 Results of weighted correct classification in calibration, cross-validation, and prediction for the LDA models developed for wine vinegar authentication with UV–Vis, FT-NIR, and FT-IR data



To evaluate if the sample variability reduction could affect the model performance, the vinegar samples with 7.1% acidity, together with their adulterated mixtures, were excluded and LDA models were recalculated. The results reported in Table 1 showed a better performance in prediction (> 96% of correct classification), and no significant difference among the spectroscopic techniques was highlighted (McNemar test: $P > 0.05$). These models could be applied when the end user is sure about the acidity of the tested vinegars; otherwise, the comprehensive models should be preferred; even if “less” performing, they guarantee to cover the high variability that might be present in vinegars.

Finally, the LDA models developed with the portable NIR data gave 100% correct classification in all the testing phases, considering both the complete dataset (i.e., 56 samples in calibration and 33 in prediction) and the dataset after the removal of samples with 7.1% acidity (i.e., 47 samples in calibration and 22 in prediction). The models should be tested with a higher number of samples and in real-life conditions, such as different lighting and operators to evaluate Polispec NIR™ application directly in the food industry (Gorla et al., 2023).

Conclusions

The adulteration of white wine vinegars with spirit vinegars (0–25% v/v) was successfully predicted by LDA models based on UV–Vis, FT-NIR, FT-IR, and NIR spectroscopy data suitably pretreated. No significant differences were calculated for model performances as a function of the specific non-targeted techniques applied, thus

demonstrating that all the spectroscopic ranges have good potential in authentication of wine vinegars. As hypothesized, the portable NIR device tested proved to be a valuable tool, with performances as good as the benchtop instruments. The variable selection approach can pave the way for the development of simplified and cheap instruments dedicated to fraud detection in the vinegar field. However, for a more robust validation, the portable device should be tested with a higher number of samples and under different operating conditions (e.g., different light sources and different operators).

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11947-023-03143-9>.

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Author Contribution Silvia Grassi: conceptualization, data curation, formal analysis, investigation, methodology, software, writing—original draft, writing—review and editing. Cristina Alamprese: conceptualization, funding acquisition, project administration, resources, supervision, writing—original draft, writing—review and editing.

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Data Availability The datasets generated and analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing Interests The authors declare no competing interests.

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